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Salminen, Reijo (ed.) 2001. International conference on practical applications in environmental geotechnology ***ecogeo 2000***. *Geological Survey of Finland, Special Paper 32*, 225 pages, 90 figures and 50 tables.

ECOGEO 2000 The International Conference on Practical Applications in Environmental Geotechnology conference concluded the Finnish five-year ecogeotechnology research programme, co-ordinated by Tekes, the National Technology Agency of Finland. The programme focused on the utilisation of industrial by-products in earth construction, on investigations and remediation of contaminated soils and groundwater protection technologies. The programme was financed by Tekes, private Finnish enterprises and R&D organisations. The total budget was about USD 10 million (FIM 60 million).

ECOGEO 2000 had the ambition to build a bridge between science and practical solutions to minimise the time lags in information and knowledge transfer. The scope of the conference was wide, as the reality in implementation projects.

ECOGEO 2000 provided a forum for transferring knowledge on practical applications. The presentations which are now published concern the technical and environmental aspects of industrial by-product utilisation, remediation of contaminated soils, and barrier structures for groundwater protection. The environmental risks connected with the use of industrial by-products are covered very thoroughly and the remediation methodology of contaminated soils is presented widely.

In addition to the reports from Ecogeotechnology research programme, a highly qualified collection of scientific papers from other countries were presented and are included in this Congress proceedings volume thus giving an excellent overview about the modern ecogeotechnological research.

Keywords: engineering, geology, by-products, earthworks, environmental geology, soils, pollution, remediation, ground water, protection, symposia

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Use of industrial by-products in earth construction

Contaminated soils

Barrier structures and ground water protection

PREFACE

Today, complex inter- and intra-relations between water, air and soil pollution, together with various time lags between the causes and effects, make practical decision-making in environmental matters very challenging. The temptation to postpone necessary decisions is in these circumstances perhaps understandable, but could be argued.

ECOGEO 2000 – The International Conference on Practical Applications in Environmental Geotechnology had the ambition to build a bridge between science and practical solutions to minimise the time lags in information and knowledge transfer. The scope of the conference was wide, as the reality in implementation projects.

ECOGEO 2000 provided a forum for transferring knowledge on practical applications concerning the technical and environmental aspects of industrial by-product utilisation, remediation of contaminated soils, and barrier structures for groundwater protection. The environmental risks connected with the use of industrial by-products were covered very thoroughly and the remediation methodology of contaminated soils was presented widely.

The conference was intended for corporate environmental managers, earth construction contractors, environmental experts and decision-makers from municipal and government sectors, entrepreneurs and private consultants as well as researchers.

The ECOGEO 2000 conference also concluded the Finnish five-year ecogeotechnology research programme, co-ordinated by Tekes, the National Technology Agency of Finland. The programme focused on the utilisation of industrial by-products in earth construction, on investigations and remediation of contaminated soils and groundwater protection technologies. The programme was financed by Tekes, private Finnish enterprises and R&D organisations. The total budget was about USD 10 million (FIM 60 million).

The results of the programme’s individual R&D projects were presented at ECOGEO 2000. They formed the major part of presentations in the congress but, however, in addition a highly qualified collection of scientific papers from other countries were presented and included in the Congress proceedings volume thus giving an excellent overview about the modern ecogeotechnological research.

This volume consists of 28 selected papers from 60 presented in the ECOGEO 2000 – The International Conference on Practical Applications in Environmental Geotechnology -congress. Mostly these selected papers deal with new scientific data but in addition some overviews as well introductions of some very practical products are included in order to give a cross section about the information delivered in the conference. Many of the papers the authors improved by adding the newest results, and they were thoroughly peer reviewed by the scientific advisors of the conference. All papers presented in conference are available in their original format in the conference volume:

International Conference on Practical Applications in Environmental Geotechnology. ECOGEO 2000
Tammirinne, Markku (ed.)
2000. VTT Communities and Infrastructure , Espoo. 477 p. + app. 2 p.
VTT Symposium : 204
ISBN 951-38-5701-8 951-38-5702-6
(http://www.inf.vtt.fi/pdf/symposiums/2000/S204.pdf)

From the offered papers the scientific advisors selected 67 papers for presentation at the conference. The Conference proceedings volume includes 60 papers of those accepted. The three main topics of the conference were:

1) Use of industrial by-products in earth construction
2) Contaminated soils
3) Barrier structures for landfills and groundwater protection

The papers in this volume are organised according to these main topics, too.

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THE QUALITY CONTROL AND GEOTECHNICAL PROPERTIES OF RECLAIMED CONCRETE IN EARTH CONSTRUCTION

by
Juha Forsman[1)], Helena Korjus[1)], Lauri Kivekäs[2)] and Antti Määttänen[3)]

Forsman, Juha, Korjus, Helena, Kivekäs, Lauri & Määttänen, Antti 2001. The quality control and geotechnical properties ofreclaimed concrete in earth construction. *Geological Survey of Finland, Special Paper 32, 7–13*, one figure and 4 tables.

Reclaimed concrete material is generated through the demolition of concrete elements and structures during building demolition operations. In this article the quality control and geotechnical properties of reclaimed concrete have been studied.

The quality control of by-products is even more important than for “traditional” aggregates in order to provide an acceptable material for utilisation in earth construction. The assorting demolition is a base for acceptable raw material for reclaimed concrete and the quality control secures the properties of the product. The geotechnical and environmental properties of reclaimed concrete have been studied in the field on test roads and in a laboratory over several years. Both geotechnical and environmental properties of the crushed material indicate that the use of reclaimed concrete in road construction is acceptable.

On the basis of the field and laboratory studies and experience, user guidelines for the use of reclaimed concrete have been made for the Finnish National Road Administration (FINRA) and for the Association of Finnish Local and Regional Authorities. Also the quality assurance standard of reclaimed concrete is under preparation in Finland.

Keywords: construction materials, concrete, crushing, by-products, quality control, engineering properties, highways, construction, Finland

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INTRODUCTION

Reclaimed concrete has been used in pavement construction since 1994 in Finland. Reclaimed concrete crushed by Lohja Envirotec Ltd is known by the name Betoroc™-crush, and it is classified in grades by the raw material and technical properties. In Finland Betoroc™ has been used in road and street construction mostly in sub-base and base layers. Reclaimed concrete has shown favourable geotechnical properties in field studies on test roads and in laboratory tests and is thereby suitable for use in road construction. The quality control of by-product is important in order to have an acceptable material for utilisation. The by-product utilised in earth construction must have adequate technical and environmental properties and permanent quality. The Finnish quality assurance standard of reclaimed concrete will contain the quality control demands, methods and schedule of controlling for each phase from demolition operation to earth construction.

CLASSIFICATION OF THE BETOROC MATERIALS

Betoroc™ is classified in grades by its raw material and technical properties. A material can originate from building demolition operations or, for example, from hollow concrete production as waste, which has been abandoned due to manufacturing process requirements. The classification by raw material is presented in Table 1. Table 2 presents the technical and environmental classification of the material.

Table 1. Classification of Betoroc*‘* by raw material.

Grade	Raw material
BeM I	Pure reclaimed concrete, which originates, for example, from concrete element manufacturing.
BeM II	Reclaimed concrete, which originates from demolition of concrete structures and buildings. It may contain some harmful materials.
BeM III	Reclaimed concrete, which originates from demolition of concrete structures. It might not possess any self-hardening properties.
BeM IV	Reclaimed concrete, which originates from demolition of concrete structures. It possesses no self-hardening properties and it may be frost susceptible.

Table 2. Basic properties of Betoroc*‘* (a) and content of harmful materials and the use on the ground water areas (b).

a)	Grain size distribution	Self-hardening properties	Frost susceptibility	E-modulus
BeM I	0–50 mm	Hardens	No	700 MPa
BeM II	0–50 mm	Hardens	No	500 MPa
BeM III	0–50 mm	Uncertain	No	280 MPa
BeM IV	Varies	No hardening	Varies	*£* 200 MPa*

* to be considered in each case

b)	max. content of bricks [weight-%]	Max content of other * materials [weight-%]	Leaching tests in crushing phase
BeM I	0	0,5	Not demanded
BeM II	10	1	Demanded
BeM III	10	1	Demanded
BeM IV	30	1	Demanded

* wood, plastics, etc. In addition to the weight-% demand, there may not be harmful amounts of special light materials (such as polystyrene and other insulation materials).

GEOTECHNICAL AND ENVIRONMENTAL PROPERTIES

The geotechnical properties of the crushed reclaimed concrete have been studied both in laboratory tests and in test roads and streets. Table 3 presents some test results of crushed reclaimed concrete materials. The values shown present the variation of values of testing (the real variation of the properties may be larger). The design values of E-modulus are presented in Table 2a. The determination of the E-modules on the basis of the field and laboratory tests is presented in detail in the article “Geotechnical Properties and Bearing Capacity of Reclaimed Concrete” (Forsman *et al*. 2000).

The bearing capacity of the reclaimed concrete structures has been measured using the plate load test or falling weight deflectometer (FWD). The first measurements have usually been done straight after compaction of the crushed reclaimed concrete layer and soon after paving. Later the bearing capacity has been measured in springtime and/or in summertime.

The E-modulus of the reclaimed concrete structure has been back calculated on the basis of the field measurement data using both the Odemark and APAS methods. APAS is a software developed by FINRA and Neste Ltd for analytical pavement design and it is based on the NOAH (Nynäs Overall Approach) calculation module developed by Nynäs in Belgium (Pienimäki 1994). Using this APAS program it is possible to fit the calculated bowl to the measured FWD-bowl and that way back calculate the E-modulus of road materials layer by layer.

Table 3. Geotechnical properties of reclaimed concrete based on some test results (Viatek 1999).

Property *	Unit	Betoroc*‘* I (hollow core slab waste)	Betoroc*‘* II (demolition waste)	Crushed concrete in general
Optimum water content	%	8…10	8…12	8…12
Maximum dry weight	kN/m^3	18…20	17.5…20.5	
Minimum dry weight	kN/m^3		12.7…14.5	
Specific gravity	t/m^3			2.55…2.65
Compression strength, 7 d	MPa	1.2…1.3	0.3…1.1	
Compression strength, 28 d	MPa	2.0…2.1	0.6…1.3	
Capillarity	m	0.25	0.20	
Permeability	m/s	1…7 × 10^{-5}		
pH		12.7…12.9		≥ 11
Segregation potential	mm^2/Kh	0.11…0.28		
Heat capacity (unfrozen)**	Wh/m^{3}K	485…590		
Friction angle	∞			40
CBR-ratio	%			90…140
Los Angeles -ratio		23	28	

* E-modules used in dimensioning are shown in Table 2a.
** approximated from the field observations by back-calculation.

The environmental properties of Betoroc™ have been tested at the Technical Research Centre of Finland (VTT)/Chemical Technology (VTT 1996, Wahlström *et al.* 1997). The results from these chemical analyses and from tests made later show that Betoroc™ is environmentally acceptable for road construction. On the basis of the chemical analyses the target values for leaching from mineral demolition waste have been given (shown in Table 3). The target values shown in Table 4 concern the replacing of class I (material that can be used without limits in base layers). VTT Chemical Technology (1996) has also proposed recommendations for the quality assurance system for the utilisation of reclaimed concrete. The quality control will be done by the manufacturer.

Table 4. Target values for leaching from reclaimed concrete proposed by VTT [1996]. Proposed test method is CEN-batch leaching test with protocol 3 (prEN 12457 1996).

Element	Sulphate	Chromium	Cadmium	Copper	Lead
Target value (mg/kg)	750	0.5	0.02	0.4	1.0

QUALITY CONTROL SYSTEM

In the case of by-products, the quality control is even more important than with a “traditional” aggregate in order to have an acceptable material for utilisation in earth construction. Both the geotechnical and environmental properties of the reclaimed concrete have to be studied by a detailed quality control plan and with the proper methods agreed in the user guidelines.

In Finland the quality assurance standard preparation of reclaimed concrete (VTT 2000) is in progress and is to be created in co-operation between VTT, Lohja Envirotec Ltd and Viatek Ltd as a part of a wider quality assurance standard. The draft of the quality assurance standard of reclaimed concrete contains the required quality controls and approved methods of control. The generation process and the quality control of reclaimed concrete is presented in Fig. 1.

The main phases in processing reclaimed concrete are as follows: the delivering of raw material, transportation, receiving raw material at operating site, stockpiling of raw material, crushing, grading, stockpiling of crushed material and supplying material to the utilisation site. In each phase the quality of the material is controlled. The studies the supplier does are grain size distribution, frost susceptibility (on the basis of the grain size distribution), maximum bulk density and optimum water content (if needed), compression strength, material purity and environmental qualifications.

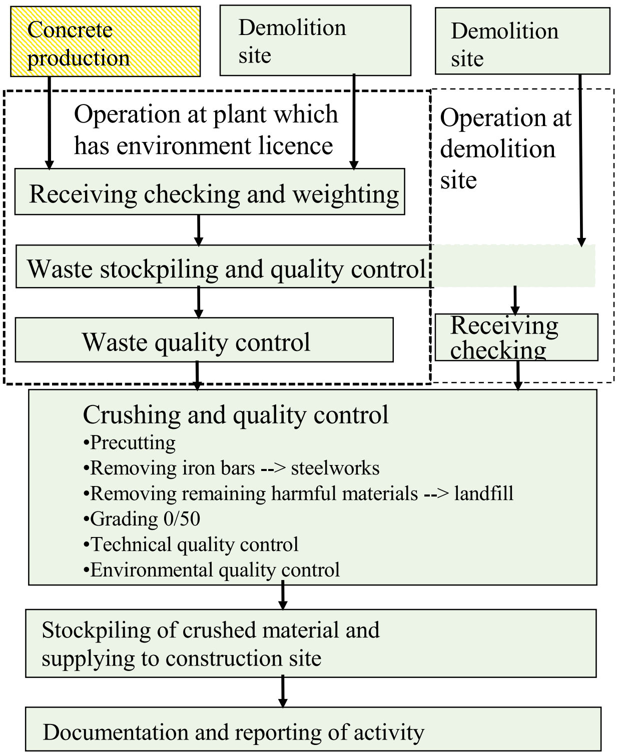

Figure 1. The generation process and the quality controlling of reclaimed concrete.

METHODS OF QUALITY CONTROL

Waste concrete

The first control of the waste concrete is visual checking of the material and its purity. The crushing stations of Lohja Envirotec Ltd have detailed guidelines for checking and classifying the received material. Materials containing harmful components such as PCB, creosote, asbestos, oil, heavy metals or sulphates are not accepted as raw material. The purity of the material is also checked visually throughout the manufacturing process from raw material until the utilisation site.

Grain size distribution and frost susceptibility

The grain size distribution is determined by grading (SFS-EN 933–1). One grading has to be done per 2000 tons of material, or at least twice a week. The grain size distribution curve must meet the requirements for the base layer grain size distribution presented in the FINRA’s guidelines (FINRA 1985). Frost susceptibility of the material is determined by the grain size distribution. The frost susceptibility of the material is monitored with the same sampling interval as the grain size distribution. The reclaimed concrete in groups I, II and III (Tables 1 and 2) must be non-frosting, in group IV the frost requirements depend on the case.

Maximum bulk density and optimum water content

If needed the maximum bulk density and optimum water content are determined by the improved Proctor test or the IC test. In the IC test the work pressure is 4.0 bar and the rounds are 160.

Compression strength

For determining the compression strength and to be able to follow the hardening, one sample set/10 000 t or at least one sample set/crushed parcel is created with specimens compacted by the IC-tester equipment. One sample set includes 7 specimens. The specimens are prepared in the same way as the maximum dry weight is determined. The compression is done following the guidelines of BY 15 Concrete Norms 1993. Three specimens are compressed for 7 days and three specimens are compressed for 28 days. The seventh specimen is a spare sample which is compressed if needed. The specimens are kept at +20ºC covered with plastic foil.

Material purity

Material purity must be determined sufficiently often, at least at 20 000 t intervals. The determination of the amount of organic impurities (wood, plastic etc.) and the amount of bricks is done following the standard NEN 5942 (the Dutch norm 1990) with the corrections as follows:

– The grains over 8 mm are divided approximately into three groups: concrete, bricks and others.
– The weight proportions (weight %) of each material are calculated from the total mass left on the 8-mm sieve.

The classification of the reclaimed concrete regarding impurities is presented in Table 2b.

The determination method of material purity has been developed by testing several methods and comparing the advantages and disadvantages of the different methods. Such methods as combustion, water weighing, photographs and picking have been tested for determining the amounts of organic impurities and bricks. Combustion, floating, blowing and photography have been used for determining the amounts of special light materials (such as polystyrene and other insulation materials).

Combustion

Combustion is a method which might be used for determining the amount of light materials but also for example wood, etc. Combustion was done in an asphalt furnace at a temperature of +500 ºC. The problem was that at the temperature as high as used causes loss of water of crystallisation in the concrete material and stones and the results are therefore unreliable. If combustion is used in determining the amounts of impurities, it needs the correction curves done for the loss of the water of crystallisation.

Water weighing and floating

The volume of the crushed concrete specimen was determined by sinking it into water. The specimen was weighed before sinking. The problem of this method was that the water intrudes into the concrete and brick grades and therefore the measured weight is not correct. Another problem was the relative close density of concrete and bricks which causes difficulties, especially in determining the amount of small proportions of bricks.

Floating is used for determining the amounts of special light materials (such as polystyrene and other insulation materials). When using floating, the material must be dehydrated after floating for further determining (amounts of bricks etc.).

Blowing

Blowing, like floating, is used for determining the amounts of special light materials (such as polystyrene and other insulation materials). Blowing loosens not only the special light materials but also cement from crushed material and is not therefore a reliable method for determining impurities.

Photography

Photographs were taken of materials which were made by mixing predetermined amounts of impurities into pure crushed concrete. The idea was to compare visually the crushed concrete specimens to the photographs and determine the amount of impurities by the reference photographs. The problem was that the small amounts of impurities gather on the surface of the pure concrete and the reference specimen looks less pure than it actually was. The method is suitable for a rough estimation but not for an exact determination of impurities.

Picking

In this method the impurities of the specimen are picked and sorted. The method was modified from the standard NEN 5942 (Dutch norm 1990). This method needs an experienced examiner, but it is by far the most exact and reliable method and therefore is chosen as the used method. The method used will continuously be developed by Lohja Envirotec Ltd.

Environmental qualifications

The environmental qualifications are determined following the principles shown in Fig. 1. Before the crushing of the specimen the purity and the content of harmful materials is estimated visually.

Studied sample taken from the crusher run is crushed to a grain size under 4 mm and then the sample is divided into laboratory samples (2 kg). The width of the testing depends on the raw material and the replacing site. The total content of harmful metals is determined using methods prEN 13657 or prEN 13656. It is recommended that PAH and PCB are determined using the methods Nordtest recommends. The defining of metals in solutions is performed using the prEN 12506 and prEN 13370 standards. The leaching of harmful compounds is determined by L/S relation 10 following test prEN12457–3 or using column test NEN7343 or NT ENVIR 002.

“USER GUIDELINES”

In Finland user guidelines for the use of reclaimed concrete and the guidelines for quality control have been made for FINRA and for the Association of Finnish Local and Regional Authorities with the financial support of The Finnish Technology Development Centre. The dimensioning parameters of reclaimed concrete and the construction procedures with reclaimed concrete have been studied on the basis of Finnish and foreign experiences. Dimensioning parameters have been reported in detail in a separate report “Design parameters of crushed reclaimed concrete” (Viatek 1999).

The bearing and frost dimensioning parameters have been presented in the first part of the user guidelines. The design considerations and ready-made dimensioning tables have also been presented. The E-modulus of reclaimed concrete (grade I-III) used in dimensioning has been 280–700 MPa depending on the properties of the material. By using highly classic reclaimed concrete in some cases, it is possible to make thinner bitumen bound surface layers. In the second part of the user guidelines the construction procedures of reclaimed concrete pavements have been presented following the format of FINRA’s guidelines.

The user guidelines for the use of reclaimed concrete and the guidelines for the quality control made for FINRA have been translated into English by Lohja Envirotec Ltd (Viatek 2000).

CONCLUSIONS

In the case of by-products, quality control is even more important than with “traditional” aggregates in order to have an acceptable material for utilisation in earth construction. Both the geotechnical and environmental properties of the reclaimed concrete have to be studied by a detailed quality control plan and with proper methods agreed in the user guidelines.

The sorting at the demolition site is a basis for acceptable raw material of the reclaimed concrete. The quality control is needed when receiving the material at crushing stations. Also the whole process from crushing to delivering of the crushed material is under continuous quality control for securing the properties of the product.

Reclaimed concrete has shown favourable geotechnical properties in field studies on test roads and in laboratory tests. The reclaimed concrete, Betoroc™, has self-hardening properties (grades BeM I-II) and it is not frost susceptible (grades

BeM I-III). The results from the chemical analysis show that Betoroc™ is environmentally acceptable for road and street construction outside the groundwater areas.

Crushed reclaimed concrete has been used several years in street and road structures mainly in base and/or sub base layers. The structures made with Betoroc™ can be constructed by using normal road construction methods. The most important difference is the need to maintain the necessary humidity for the hardening process of the concrete structure.

In Finland user guidelines for the use of reclaimed concrete have been made for FINRA and for the Association of Finnish Local and Regional Authorities. The guidelines contain information concerning material properties and classification of reclaimed concrete, dimensioning the structures with reclaimed concrete and guidelines for construction procedures. Preparation of the quality assurance standard of reclaimed concrete is in progress and is to be created in co-operation of VTT, Lohja Envirotec Ltd and Viatek Ltd as a part of a wider quality assurance standard in Finland.

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FINNISH GUIDANCE FOR THE USE OF SECONDARY PRODUCTS IN EARTH AND ROAD CONSTRUCTION

by
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Mroueh, Ulla-Maija, Mäkelä, Esa, Wahlström, Margareta, Kauppila, Jussi, Sorvari, Jaana, Puolanne, Juhani, Juvankoski, Markku, Tammirinne, Markku, Heikkinen, Päivi & Salminen, Reijo 2001. Finnish guidance for the use of secondary products in earth and road construction. *Geological Survey of Finland, Special Paper 32, 15–21*, one figure and 2 tables.

In the project "Secondary products in earth construction – assessment of applicability" guidance was developed for the assessment of the environmental and technical applicability of secondary products for use in earth and road construction. The project was a part of the Finnish Environmental Geotechnology Programme. The preparation of the guidance was a collaboration involving several research institutes.

The guidance presents the legislative requirements for the utilisation of secondary products in earthworks, recommendations for the investigation of environmental and technical applicability, recommendations for environmental and technical criteria of the utilisation in earthworks and recommendations for product quality control procedures.

A tiered system is presented for the assessment of environmental compliance. The assessment levels are 1) Concentrations of harmful components, 2) Leaching of harmful components from unpaved and paved constructions and 3) Risk assessment.

Keywords: engineering geology, earthworks, highways, construction, construction materials, by-products, utilization, risk assessment, Finland

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INTRODUCTION

About 70 million tonnes of natural mineral aggregate are used each year in Finland for earth and road construction. Depletion of the best materials, the need for resource conservation and increased transport distances have all stressed the need to introduce substitute materials for natural aggregates. At the same time, industry, construction and other similar activities produce large quantities of potentially usable secondary products. Current waste legislation also supports the use of waste as substitutes for natural materials. Usage of the secondary products of industry and other activities requires that they be proven to be environmentally friendly and technically suitable.

The usage of secondary materials has been hindered by uncertainty about their environmental impacts and technical performance. There is very little practical experience of the long-term performance of most of the materials. There has also been confusion about the procedures that should be used to assess the technical and environmental applicability of the products. The methods of investigation have been developed or changed, and the data on older projects is not comparable with the present investigation data. Up until the 1980s, the assessment of environmental acceptability was not considered as necessary as it is today.

To investigate the environmental, technical and legal preconditions of the use of secondary products, the project “Secondary products in earth construction – assessment of applicability” was started as a part of the Finnish Environmental Geotechnology Programme. The main objective of the project was to develop guidance for the assessment of the environmental and technical applicability of industrial by-products and other secondary products for use in earthworks and road construction (Mroueh *et al.* 2000). Several research institutes participated in the work. In addition, all the interested parties had an opportunity to contribute to the contents of the guidance through participation in the steering groups or project workshops and during circulation of the guidance for comment.

SCOPE OF THE GUIDANCE

The guidance applies to the utilisation of industrial by-products and secondary products in earthworks and road construction. Most of the potential materials are mineral products comparable to natural aggregates. Some organic-based materials may, however, also be used, for example in balancing structures. The pollutant content of these materials must be low or they must be bound to the material so that their migration into the environment is minimised.

Table 1 presents industrial by-products and other secondary materials that have been used in earth construction in Finland. These include fly and bottom ash of coal combustion, blast-furnace slag and some other steel and metal industry slag. The use of crushed concrete began a few years ago and has since grown considerably. Some organic or predominantly organic products, e.g. shredded tyres, are already being used in various types of construction. Asphalt pavement is being recycled into material for new pavement and other structures. Moreover, the prerequisites for the usage of several other types of material have been investigated in research and test construction projects.

The guidance covers conventional earth constructions, such as road and street constructions, various field structures and their foundations. It is recommended that, in the first place, secondary materials be used in large or medium sized cases, because control of the utilisation in small constructions is far more difficult.

The following applications are beyond the scope of the guidance:

- Landscaping, agricultural and forestry use, which are regulated by fertiliser legislation.
- The disposal of waste into the sea, which has a permit procedure of its own.
- The utilisation of secondary products in landfill constructions, for which the requirements clearly differ from those for road and field constructions.

THE BASIC STUDIES

The environmental, legal and technical preconditions for the use of secondary materials were extensively investigated during the preparation of the guidance. For example, the following aspects were studied during the project:

Environmental and health risks of the use of industrial by-products and the methodologies of risk assessment (Wahlstrom *et al.* 1999). The report deals with the effects of most common harmful components present in secondary products on human health and the environment, the migration of harmful components and the possibilities of exposure during various stages of construction. The risks caused by this exposure are also assessed. The risks considered are primarily related to human health because the data needed for ecological risk assessment is still under development. The proposed risk assessment concept was also tested in a case study on the use of coal fly ash in earth construction.

Life-cycle environmental impacts of the use of secondary materials and natural aggregates, and the methodologies of life-cycle environmental impact assessment of road construction (Eskola *et al.* 1999, Laine-Ylijoki *et al.* 1999). A life-cycle impact assessment procedure for the comparison and evaluation of alternative road and earth constructions was proposed. Additionally, a database containing the environmental burdens of the most significant construction materials and unit operations as well as the information required for the calculation of the data was constructed. In order to evaluate the applicability of the methodology, the use of coal ash, crushed concrete waste and granulated blast-furnace slag was compared with the use of natural materials in corresponding applications. An Excel-based computer application was compiled on the basis of the data.

Environmental criteria of the utilisation of industrial by-products in earth constructions (Sorvari 2000) Environmental criteria for the utilisation of secondary products in earthworks were drawn up during the project. For the basis of the study the environmental criteria and approaches used in various countries were studied. A tiered system is presented for the assessment of environmental compliance. The system is based on soil guideline values and on leaching values, which are calculated on the basis of Dutch leaching models.

Methods for laboratory-scale functional testing of the secondary products used in earthworks (Tammirinne *et al.* 1999, 2000). Guidance and descriptions of laboratory methods recommended for investigation of the technical performance of secondary products in road constructions were compiled. Reference values for the assessment of results were also presented.

The binding and transport of harmful components in soil (Heikkinen 2000). The factors that impact on the fate of harmful components in soil were studied based on the literature and a field investigation. The focus of the study was on the most common components leaching from secondary products, their migration and binding in Finnish soil. In the field study the migration of inorganic compounds from an older fly ash construction was investigated. On the basis of the study recommendations were given about the soil and bedrock conditions to be considered when planning the utilisation of by-products.

The Finnish environmental permit practice and suggestions for the development of more adaptable legal measures (Kauppila 2000). Those factors in the present legislation and administrative practice which hinder the use of secondary materials in earthworks were studied. The aim was to establish a knowledge base for deregulation of the present permit procedure and harmonisation of permit practices.

Quality control system for the production and use of the secondary materials in earthworks. The aim of the project is to draw up a quality control standard for the production of secondary materials used in earthworks and to draft material standards for three reference materials. These materials are crushed concrete, blast-furnace slag and bottom ash from pulverised coal combustion.

Landfill acceptability of industrial waste (Wahlstrom *et al.* 2000). Recommendations about the landfill acceptability of mineral by-products, such as parameters to be investigated and investigation methods suitable for these investigations, are presented. Some examples of criteria applicable for assessment of the landfill acceptability of typical industrial by-products are also given.

Table 1. Industrial by-products used in earth construction in Finland and their estimated annual consumption, 1998–1999.

Activity	Production, t/a	Amount used in earthworks		Usage in earth construction/(other usage)
		t/a	%	
Energy production				
Coal fly ash	350 000	190 000	40	Road and field construction, earth- fill/(Production of cement and concrete 30 %, asphalt filler 5 %)
Coal bottom ash	78 000	53 300	70	Road and field construction
Peat fly ash	180 000	78 800	60	Mainly earthfill
Peat bottom ash and slag	33 000	11 000	33	Mainly earthfill
Metallurgic industry				
Blast furnace slag	550 000			(Production of cement, use as fertiliser)
– unground sand and slag		200 000	36	Road constructions
– ground slag		120 000	22	Binder in soil stabilisation
Slag from LD steel production	170 000	18 500	10	Use as fertiliser
Slag from ferrochrome production	290 000	290 000	100	
Construction				
Crushed concrete	200 000	100 000	17	Road and field construction
Tyres	30 000	27 700	92	Road and landfill construction
Road construction				
Pavement materials	150 000	150 000		Recycling to pavements
Structural courses	160 000			
Forest industry				
Fibre and paste suspensions	128 000		55	Landfill construction
Ash	210 000			Landfill construction/(forest fertiliser), total usage 55 %
Chemical industry				
Ferro sulphate gypsum	70 000			Binder in soil stabilisation

PERMIT PROCEDURE

An environmental permit is required if an industrial by-product or secondary material is regarded as waste, which is usually the case. According to the Finnish Environmental Act, the permit is issued by the local environmental authority if the amount of waste being used is less than 5 000 t. For larger amounts the permitting authority is the regional environmental centre. It usually takes more than four months to obtain the permit. This is one of the biggest barriers to the use of secondary products, because the use of natural aggregates is much less complicated.

On the basis of the study of the legislative barriers, the Ministry of the Environment has started to prepare a decree of the Council of State. The decree covers the use of selected waste materials in earth construction. The aim is to give general regulations concerning the conditions on which the use of the materials is permitted and thus to release their use from the permit obligation.

INVESTIGATIONS FOR THE ASSESSMENT OF ENVIRONMENTAL COMPLIANCE

When industrial by-products or secondary products are used in earthworks, the migration of harmful compounds from the material is considered to be the most significant environmental hazard. Besides leaching, it is also important to consider other properties of the material and to check the environmental impacts of the entire utilisation chain. Before assessing environmental compliance, the testing agency has to ensure that all the potentially hazardous compounds and properties of the material have been measured or investigated.

Besides the conservation of natural materials, the utilisation of by-products may have other positive effects, such as reduction of energy use or emissions. Up to now, the legislation has not required consideration of these impacts during the permit procedure for waste utilisation in earthworks. According to the new environmental legislation, however, more weight is put on integrated environmental assessment.

The main phases of investigations for the assessment of environmental compliance of industrial by-products or secondary products are presented in Fig. 1.

The basic characterisation of industrial by-products is usually a demanding task. The testing must be planned on a case by case basis. At least the following compounds and parameters will be selected for further investigation:

- Compounds with a concentration that is expected to be at least on the level of Finnish soil target values
- Compounds on which there is insufficient information about their concentration or harmfulness
- Compounds with a concentration that may exceed the Finnish occupational exposure limits.

When the investigations are planned, one must ensure that all the significant parameters are studied, that the methods used are reliable and suitable for the investigation and that the expertise of the investigator of the study is sufficient.

Sampling is one of the most critical stages of experimental investigations, because the sample has to reliably represent the material or batch of material under investigation. The sampling plan must be based on the objectives of the study and on the properties of the material to be investigated. For example, for environmental characterisation it is recommended that at least 20 samples be taken systematically, one sample per day over a period of four weeks.

The report also presents the methods recommended for basic characterisation, which include determination of the concentration of inorganic and organic compounds in the material, determination of leaching behaviour using the column leaching test (NEN 7343 or Nordtest ENVIR 002), the diffusion leaching test (NEN 7345) and the pH-stat leaching test, and methods for analysis of test eluates.

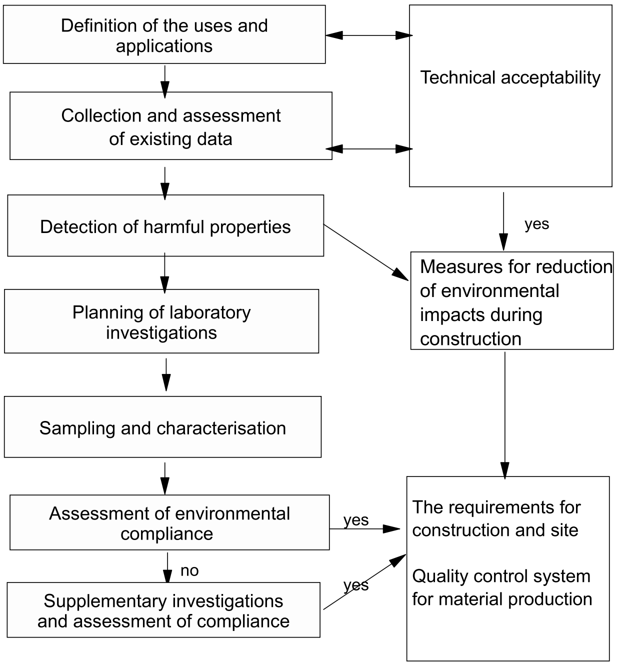

Figure1. Environmental assessment of secondary products: the main phases of investigation.

ASSESSMENT OF ENVIRONMENTAL COMPLIANCE

A tiered system is presented for the assessment of environmental compliance. The assessment levels are as follows:

1. *Concentrations of the harmful components.*

If the concentrations of all the inorganic compounds are below the Finnish soil target values and the concentrations of the hazardous organic compounds are below the detection level, the material is suitable for use in earthworks without restrictions. In the case of these values being exceeded, the use of material in ground water areas suitable for water supply is not recommended without a risk assessment.

2. *Leaching of harmful components from constructions.*

The leaching of inorganic compounds from the material is compared to the leaching values which were drafted as a part of the project by the Finnish Environment Institute. The recommended leaching tests are the column-leaching test, NEN 7343, and the diffusion-leaching test, NEN 7345.

Long-term performance is assessed by using a test simulating varying pH conditions or on the basis of expert assessment.

The leaching values (Table 2) are presented for unpaved constructions and for constructions paved with slightly water-permeable material, such as asphalt. The material must be placed above the upper ground water level. Sites on which the ground is from slightly to moderately permeable are considered as most suitable for the use of these products.

3. *Risk assessment.*

In the case of non-compliance with points 1 or 2 above, the material may be used on the basis of material- or site-specific risk assessment.

TECHNICAL COMPLIANCE

In most cases it is recommended that the technical compliance be investigated step by step. If the material is totally ”new” the basic parameters to be investigated usually include grading, compactibility and hardening. In most cases it is possible to make a preliminary screening of potential applications based on the above-mentioned data, the mode of production and chemical content of the material.

Further studies are then scheduled on the basis of the planned application. The report presents recommendations for the laboratory investigation of industrial by-products which are planned for use in road construction.

PRODUCTION CONTROL OF THE MATERIALS

In most cases it is not necessary to repeat the extensive basic characterisation of materials. In connection with the characterisation, a production control system for the assurance of conformity of the materials with environmental and technical specifications is established. Only in the case of significant changes of the quality of the material, such as changes in the production process or raw materials, it may be necessary to renew some or all the characterisation tests.

The production control system is material-specific. The technical characteristics that are significant for the application and the most critical environmental characteristics are selected for periodic control. Homogeneous materials with well-known basic characteristics require less monitoring than more complicated and less investigated materials.

Table 2. Suggested Finnish leaching values for the mineral materials used in earth construction (Sorvari 2000).

	Granular material	Granular material	Monolithic material
Substance	Unpaved	Paved	
	Emax, mg/kg	Emax, mg/kg	Emax, mg/m^2
As	0.14	0.85	58
Ba	10	28	2 800
Cd	0.011	0.015	2.1
Co	1.1	2.5	280
Cr	2.0	5.1	550
Cu	1.1	2.0	250
Hg	0.014	0.032	1.6
Mo	0.31	0.50	70
Ni	1.2	2.1	270
Pb	1.0	1.8	210
Sb	0.12	0.40	36
Se	0.060	0.098	14
Sn	0.85	3.1	280
V	2.2	10	700
Zn	1.5	2.7	330
F	11	25	2 800
CN, free	0.060	0.098	14
SO_4	1 500	N/A	N/A
Cl	250	N/A	N/A

SUMMARY

Based on an extensive study of various factors affecting the acceptability of secondary products for earth construction, guidance was developed for the assessment of the environmental and technical applicability of these materials. The guidance was prepared as a collaboration involving several research institutes.

The guidance presents the legislative requirements for the utilisation of secondary products in earth construction, recommendations for the investigation of environmental and technical applicability, recommendations for environmental and technical criteria of the utilisation in earthworks and road construction and recommendations for product quality control procedures.

There are many material- and site-specific factors that affect the environmental and technical applicability of the materials. At the same time, practical experience in the use of the materials is mostly quite limited. Therefore the recommended general requirements are conservative. The objective is to ensure the technical performance, to safeguard human health and to protect the environment. Exceptions can be made if it is possible to prove the applicability of a material at a determined site or at sites of a determined type, based on material or site-specific risk assessment or on technical investigations and construction requirements.

Adequate certainty about the applicability of secondary products will improve the prerequisites of utilisation, because it is possible to avoid negative experience resulting form the improper use of the materials.

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LIFE CYCLE INVENTORY ANALYSIS PROGRAM FOR ROAD CONSTRUCTION

by
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Eskola, Paula, Mroueh, Ulla-Maija & Laine-Ylijoki, Jutta 2001. Life cycle inventory analysis program for road construction. *Geological Survey of Finland, Special Paper 32, 23–30,* two figures.

The Finnish LCA-database and inventory analysis program for road construction was developed as a part of the Technology Development Centre's (Tekes) Environmental Geotechnology Programme. In the first stage of the study a life-cycle impact assessment procedure for the comparison and evaluation of alternative road and earth constructions was proposed. Additionally, a database containing the environmental burdens of the most significant construction materials and unit operations and the information required for the calculation of the data was constructed. In order to evaluate the applicability of the methodology, the use of coal ash, crushed concrete waste and granulated blast-furnace slag was compared with the use of natural materials in corresponding applications.

All the work stages, from material production to road management, as well as the materials most commonly used in the structural courses of road constructions, are covered in the analyses. The environmental loadings dealt with have been limited to those assessed as being the most important.

During the later phase of the work, a practical calculation model based on the developed methodology and existing data was formulated. The data obtained in the first stage was also augmented to the extent necessary for this purpose. For example, leaching tests of natural aggregates were performed. The Excel-based life cycle inventory analysis program created is suitable for the routine calculation of the environmental loadings of the most common road constructions and for their comparison.

Keywords: highways, construction, construction materials, by products, environmental effects, life cycle assessment,

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DEVELOPMENT OF THE LCA-METHODOLOGY

Background

About 70 million tons of natural mineral aggregates are used each year in Finland for road construction and earthworks. Depletion of the best materials, the need for resource conservation and lengthened transport distances have all increased the need to introduce substitute materials for natural aggregates. At the same time industry, construction and other similar activities produce large quantities of secondary products, which may be suitable for use.

One of the barriers to the wide-ranging utilisation of the secondary products of energy production and industry in road construction has been uncertainty about the environmental impacts. In order to prevent potentially hazardous effects it is important to identify all the positive and negative impacts of these materials. Life cycle impact is also being increasingly used as a selection criterion for products and materials both in industry and in other activities.

Describing the total environmental impact of activities and products reliably and in such a way that alternatives can be compared is no simple task. The ”cradle-to-grave” life cycle always involves numerous stages and activities that give rise to a number of different environmental loadings. In order to keep the amount of work within reasonable bounds, the assessments must always be limited and efforts must be made to identify the critical stages of the life cycle and those factors responsible for environmental loading. This requires not only adherence to the basic principles of life cycle analysis, but also knowledge of the product or activity in question.

Scope

The basic aim of the study was to provide a clear and functional procedure for the life cycle impact assessment of road constructions and for the comparison of alternative structural solutions. It was hoped that the assessment procedure would be so simple to use that it could easily be applied by road planners and designers. However, the assessment should cover the main life cycle phases of the constructions as well as the most important environmental impacts, and it should also meet the other basic requirements set for life cycle analysis. One premise was also that the assessment procedure should be applicable as a part of road planning, and that the results could be used as selection criteria for alternative constructions and materials.

The assessment procedure should also take into account the special features of road constructions, which are the large volumes of materials used, the long service lives of the finished products, the need to examine constructions as a whole rather than comparing alternative materials, and the significant effect of the constructions' longevity and need for repair on their life cycle environmental loadings.

The work was carried out in two stages so that in the first stage a proposal was made for a procedure suitable for the life cycle impact assessment of road construction (Eskola & Mroueh 1998, Eskola *et al*. 1999). In order to evaluate the applicability of the procedure, the use of coal ash, crushed concrete waste and granulated blast-furnace slag in road construction was evaluated in case studies. The use of these industrial by-products and waste materials was compared with the use of natural materials in corresponding applications. The necessary data was also collected during the studies. The aim of the work's second stage was to transfer the assembled data for utilisation as a practical model by creating an inventory analysis program to calculate and compare the life cycle impacts of the most common road constructions. The data obtained in the first stage of the study was augmented to the extent necessary for this purpose (Laine-Ylijoki *et al*. 2000, Mroueh *et al*. 2000).

Methodology

The basic phases of life cycle impact assessment are goal definition and scope determination, inventory analysis, i.e. calculation of the material and emission flows, impact assessment and, if necessary, improvement assessment. In this study the life cycle assessment methodology was adapted to meet the requirements of road construction. The material and emission flows were determined at all stages of the life cycle, and the most important environmental impacts as well as their associated factors were identified. The following general procedural guidelines on life cycle assessment were applied: SETAC's (Society of Environmental Toxicology and Chemistry) 'Code of Practice' (1993), Nordic Guidelines on Life-Cycle Assessment (Lindfors *et al.* 1995) and ISO standards (ISO 14040, ISO 14041).

System boundaries

Functions and work stages

The analysis included all the significant life-cycle stages covering the production and transportation of materials, their placement in the road structures and the use of the construction. The situation after the use of the construction was not included in the analysis because the structures most commonly remain in place after they have been withdrawn from service. The structures were examined as entities because in road construction the selection of a material often influences the quality and quantity of other materials used, the work methods employed, the need for upkeep, and so on. Pavement and sub-grade structures were analysed separately and can be combined when necessary. The environmental loading data was calculated for each individual structural component and work stage, so that it is possible to examine flexibly the alternative constructions under study at any given time. The principal road construction and usage phases, which were taken into account when comparing structures and materials are shown in Fig. 1.

If industrial by-products are used in the constructions, the environmental burdens of landfill disposal can also be assessed, as an alternative to their use in road construction. This requires that landfill disposal is a real alternative to utilisation.

Those stages of road construction and use that have no significance for the comparison of constructions were ruled out of the analysis. These include:

- Site clearance,
- Functions associated with road use (e.g. lane markings, the installation and use of traffic signs and lights),
- Regular or seasonal maintenance (e.g. snowploughing, road salting and sanding)
- Traffic emissions.

Environmental loadings

The environmental loadings assessed as being essential during the life cycle of road constructions were selected on the basis of the case studies for inclusion in the analysis. The included environmental loadings were the following:

1. *Use of resources*: natural materials, industrial by-products, energy and fuel consumption
2. *Atmospheric emissions*: carbon dioxide (CO_2), nitrogen oxides (NO_x), sulphur dioxide (SO_2), volatile organic compounds (VOC), particles and carbon monoxide (CO)
3. *Leaching into the ground*: heavy metals, chloride and sulphate
4. *Other loadings*: Noise, dust and land use

On the basis of a preliminary assessment of the quantity and significance, the following environmental loadings were excluded from the inventory: water use, discharges of COD and nitrogen to water, emissions of PAH, heavy metals and methane, ordinary and hazardous waste and accident risks.

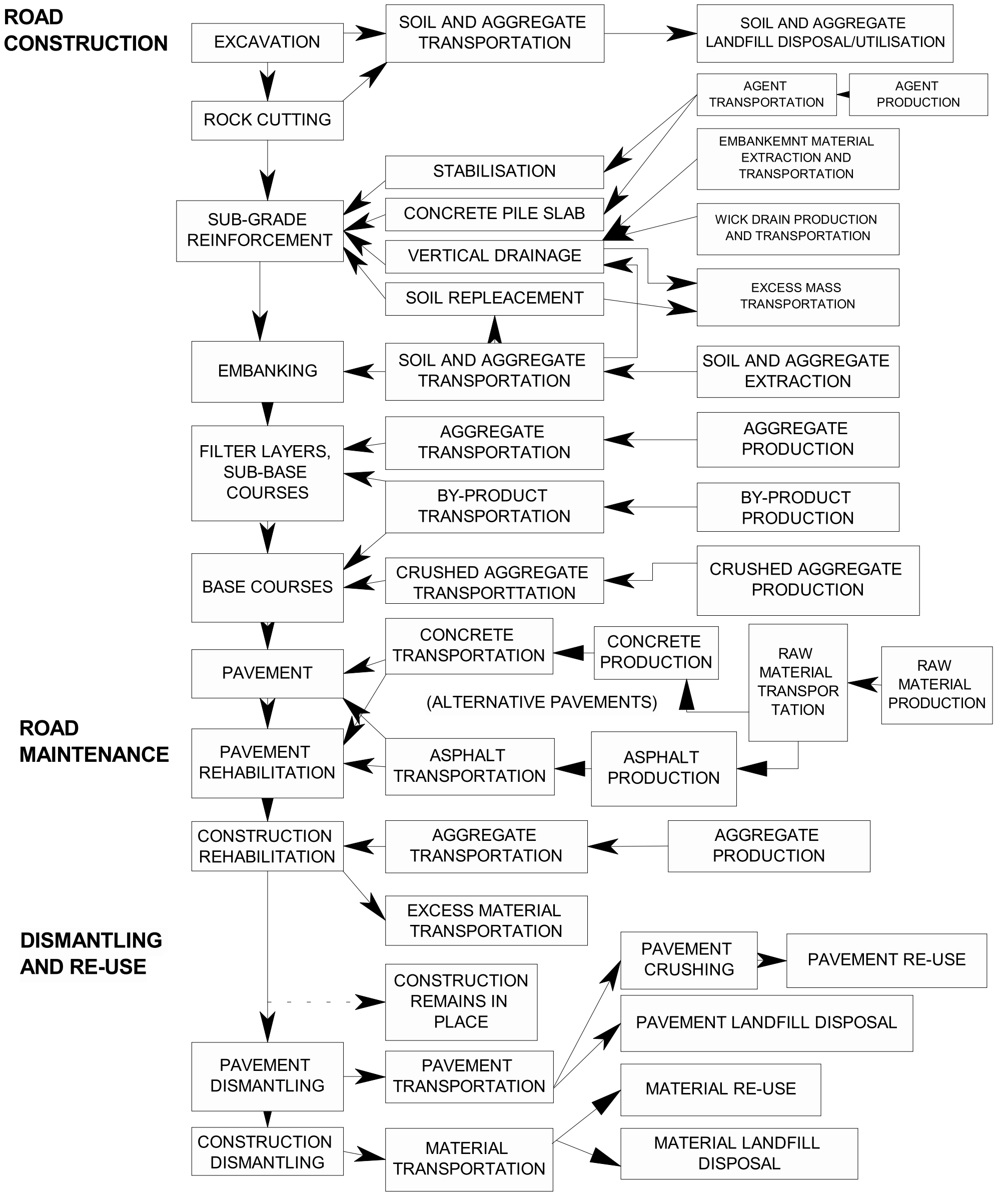

Figure 1. The principal road construction and usage phases.

Material production chains

The production chains of natural aggregates started from bedrock excavation or excavation from the ground. For cement and lime the starting point was extraction of raw materials and for rubber the production of crude oil. The production chains of industrial by-products were limited so that the environmental loadings of the by-product production process were not included in the analysis. By-products are defined in waste legislation as wastes for which no loadings are allocated in life cycle analyses. An alternative to the use of most by-products as recycled fill is disposal of the inert waste in landfills.

Other boundaries

Functional units: When comparing constructions, the functional units should always be structures of the same length that meet the same performance requirements and are designed for the same site. In practical cases, the entire construction can also be the functional unit.

Period of analysis: The period of analysis should include the entire life cycle of the material or product from raw material extraction to withdrawal from service and final disposal. In the life cycle assessment of a road construction the period of analysis must be sufficiently long to include the impacts of its service life.

Machines and equipment: The loadings caused by the manufacture of work machines and lorries and by the maintenance of machines were excluded from the analysis. The manufacture and transportation of blasting materials and fuels were also excluded.

Situation after use: It was assumed in the analysis that the construction would remain in service for 50 years. As road constructions usually remain in situ after use, no cases in which a construction was dismantled were examined in this report.

Landfill disposal: The program can give an approximate estimate of the environmental loadings avoided by using industrial by-products as recycled fills. The program calculates the minimum avoided loadings, i.e. the landfill volume needed for the inert waste and the emissions that would be released in transporting the material to the landfill site. The loadings caused by any sealing and covering of the landfill have not been included in the analysis because of their site-specific variability. Neither has leaching into the soil on the landfill site been assessed.

ENVIRONMENTAL IMPACT ASSESSMENT

Significance assessment

The aim of the life-cycle environmental impact assessment is to convert the inventory results into a form that can be more easily interpreted and compared. The impact assessment procedure is performed in three stages:

1. *Classification*, i.e. sorting the environmental loadings into impact categories on the basis of potential impacts;
2. *Characterisation*, i.e. weighting the emissions and other environmental loadings within the category according to possible impact potential; and
3. *Valuation*, i.e. weighting the environmental loadings or impacts in relation to each other.

The advantage of the valuation methods is the simplicity of result interpretation. The final outcome is either one environmental loading index or an impact matrix of mutually comparable effect scores. Further exploitation of the results does not require any expertise on the part of the user.

The creation of significance factors that are as reliable and widely applicable as possible is one of the greatest problems of valuation. Effect scores generated by different methods also make it more difficult for the users themselves to assess the significance of the results. They often also arouse suspicion, because valuation means an increase in the influence of experts in decision-making, which some fear will lead to the influence of citizens being reduced and one-sided views being over-emphasised.

Several assessment methods have been proposed for the results of life cycle analyses, none of which can be regarded as being generally accepted or even applicable in all situations (Lindfors *et al.* 1995). The best known are the Swedish Environmental Priority Strategy (EPS) system, the impact categorisation method, which has been developed in Holland and Sweden, and the ecopoints method developed by BUWAL of Switzerland.

In this study the effect scoring was made on the basis of expert assessment. The environmental loadings of the constructions were converted into relative values using the reference construction made of natural materials as the base level. After the relative environmental loading levels had been calculated, the impact categories were proportioned to one another by multiplying the effect scores by the comparative scores obtained on the basis of expert assessment. Because the construction and the transport distances always affect the outcome of such comparisons, the results as such cannot be generally applied to the comparison of materials. When comparing the alternative constructions it is notable that in all respects the differences between the constructions are not particularly great, and that the differentiating factors act in opposite directions.

Data sources and uncertainties

Because of the local nature of the effects of road constructions, primarily local or material-specific data was used. Use was also made of general Finnish know-how, which was supplemented by international sources of data where necessary.

The availability of data on by-products is limited by the fact that their utilisation is not yet well established. For this reason it is not always easy to determine the most usually employed working methods and the most general implementation methods of the work stages. As yet there is still relatively little experience- or measurement-based data on the work stages and their environmental loadings.

The release of dust emissions from materials during the different stages of production, transportation and construction is a significant environmental loading factor due to the comfort and heath risks that they pose. However, little measurement data on the release of dust emissions was found and its conversion into a comparable form was problematic. In practice, small particulate matter (SPM) can be more significant than dust particles. SPM emissions remain airborne for a very long time and are carried long distances by winds. Moreover, they pose a more serious health risk than dust particles. Because SPM emissions have attracted attention only recently, there is even less data available on them than on dust emissions.

The quantities of substances leaching out of secondary materials were simulated on the basis of laboratory-scale leaching tests. In practice, numerous factors affect leaching from construction materials. There can also be significant differences between the same material when produced under different conditions. Because there was no leaching data for natural aggregates, a few materials were tested during the project. The tests used were CEN pr EN 12457 (1996) and a pH-static test. The leaching of heavy metals from natural aggregates was found to be very small.

Because it is necessary to make many assumptions when assessing the environmental loadings, the uncertainties and ranges of the results are quite large. However, the fact that the same assumptions have been made when examining the various alternatives improves the reliability of the results.

THE INVENTORY ANALYSIS PROGRAM

In the second stage of the project, an Excel-based life-cycle inventory analysis program for road construction was created and the suitability of the program for calculation of environmental loadings of various constructions was tested.

Characteristics of the program

The inventory analysis program created on the basis of the developed methodology is suitable for the routine calculation of the environmental loadings of the most common road constructions, and for their comparison. Most of the data needed for calculations is included in the program. Only the dimensions of the construction, materials and thicknesses of the structural courses and transport distances of materials are required as input data.

The environmental loadings are presented in numerical form or using various standard graphical presentations included in the program. The program enables comparison of pavement structures as such, in relation to fixed reference construction or as effect scores.

The environmental loadings of constructions or structural courses (embankment materials, filter layer, sub-base, base course and pavements) can be presented by principal work stage or as total loading.

The pavement structure and the sub-grades are analysed separately and the results can be combined if necessary. The program includes the following alternative sub-grades: soil replacement, soil stabilisation, deep stabilisation, vertical drainage + drainage course + temporary loading berm and concrete pile slab.

If necessary the program can be extended to include new materials, structural components and alternative sub-grades.

Case studies

For testing of the life-cycle inventory program, the environmental loadings of five actual road planning cases were calculated (Laine-Ylijoki *et al.* 2000). The calculation of environmental loadings of case studies proved to be a relatively simple and fast task. The use of the comparison worksheet ena-

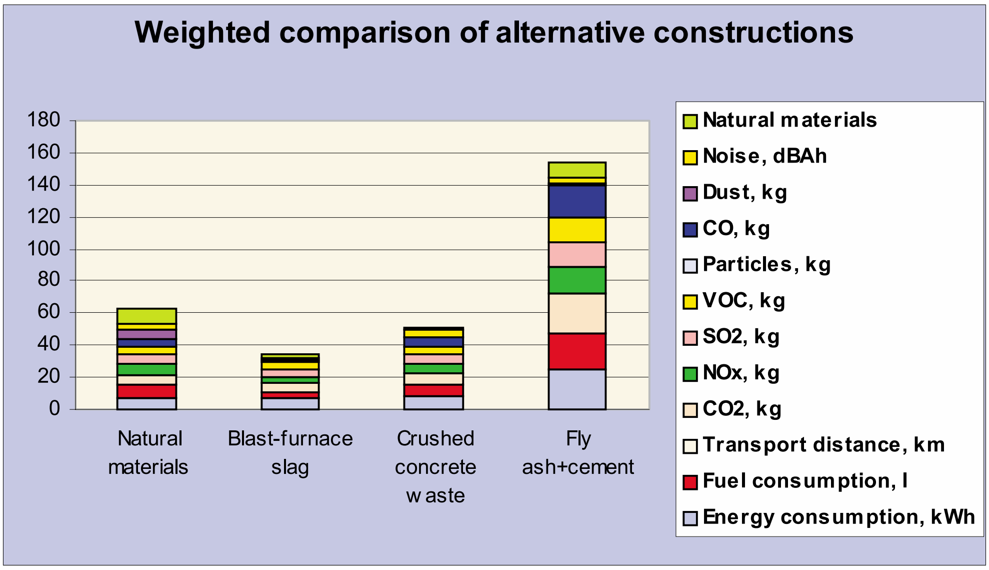

Figure 2. Example of the graphical presentations included in the program. Weighted comparison of alternative pavement constructions.

bled straightforward comparison of the alternatives and presentation of the results of calculations. An example of the graphical presentations included in the program is presented in Fig. 2.

CONCLUSIONS

A life-cycle impact assessment procedure for the comparison and evaluation of alternative road and earth constructions was developed. The procedure focused especially on the comparison of industrial by-products and natural aggregates, but the assessment of other constructions is possible as well. An Excel-based life cycle inventory analysis program for road constructions was developed on basis of the procedure. The program covers all the work stages from material production to road maintenance as well as the materials most commonly used in the structural courses of road constructions. The environmental loadings of the constructions and structural components made from the materials within the scope of the program can be calculated simply using only the dimensions of the construction, the thicknesses of the structural courses and transport distances as input data.

The program is suitable for routine calculations of environmental loadings of different road constructions. The environmental loadings are presented in numerical form or using various standard graphical presentations included in the program. The environmental loadings dealt with in the program have been limited to those assessed as being the most important. However, the loading factors in question described the total environmental loadings quite well as long as the life cycle inventory pertains to complete constructions. The environmental loadings regarded as being the most important for road construction in the expert assessment made when creating the inventory analysis procedure were the use of natural materials, energy and fuel consumption, the leaching of heavy metals into the soil, and atmospheric emissions of NO_x and CO_2.

The simplicity of the inventory analysis program makes it suitable for use by structural designers and other groups not so familiar with LCA methodology. The program has an extensive basis database and extensions with new materials, structural components and sub-grades are possible.

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INDUSTRIAL SLAG USE IN GEOTECHNICAL ENGINEERING: SLAG IN THE GEOTECHNICAL ENGINEERING PROJECT

by
Marko Mäkikyrö

Mäkikyrö, Marko 2001. Industrial slag use in geotechnical engineering: slag in the geotechnical engineering project. *Geological Survey of Finland, Special Paper 32, 31–37,* seven figures.

The use of industrial slag has for decades been based on experience with no dimensioning instructions being available until the 1990's. Attention was not paid to the systematic development of slag before the late 1980's, at which point the use of air-cooled blast-furnace slag was so extensive that there was an evident need for drawing up instructions in order to avoid structural problems possibly arising from its incorrect use. The first set of instructions were completed in 1989, accompanied by the introduction of new applications particularly for the use of granulated blast-furnace slag both in structural road layers and in stabilising the base course. The new applications and the stricter technical requirements placed on the products were one reason for launching the five-year "Slag in geotechnical engineering" project. According to the aim set for the project, design, dimensioning and construction instructions should be drawn up for all the slag produced at the Rautaruukki Group's works in Finland with regard to their use for earthwork and road construction purposes. To achieve this aim, the project was divided into three sub-fields, that is the use of slag 1) in structural road layers, 2) as a binding agent in stabilising structural layers and 3) as a binding agent in mix and deep stabilisation.

Keywords: construction materials, by-products, slag, utilization, earthworks, highways,

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INTRODUCTION

A remarkable number of different kinds of slag are produced in the different phases of the steel-making process. In Finland, total slag production from the Rautaruukki Raahe steel works and Fundia Wire Koverhar steel works is 1 000 000 tonnes. Even in a very modern integrated steel works, like the Rautaruukki Raahe steel works, the amount of slag per tonne of steel produced is more than 300 kg. This amount of slag is a total amount, which consists of blast-furnace slag, desulphurizing slag, converter slag and slag from secondary metallurgy (Fig. 1).

The first step in slag utilization is separating the metallic part, i.e. scrap from slag. The most valuable part of slag utilisation is the returning of this separated scrap back to iron- and steel-making processes. However, ever-increasing attention is paid to non-metallic slag and its utilisation. The main reasons for this kind of development are cost savings in storaging and avoidance of environmental penalty fees. A very essential part of the main product, i.e. steel, is the fact that the steel producer must show how it is taking the environment into consideration. It has been recognised that by systematic R&D work and effective marketing, by-products utilisation may also be a good business.

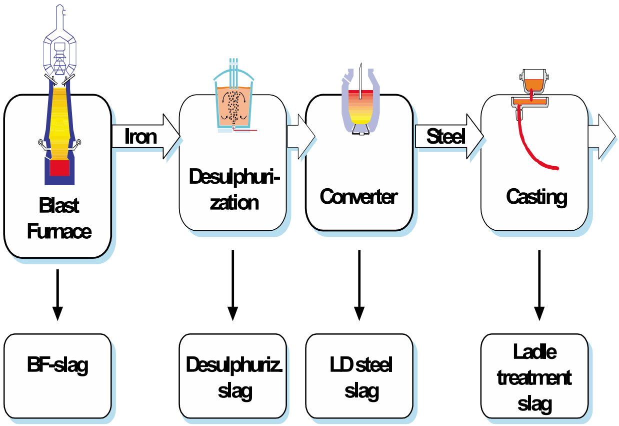

Figure 1. Slag types from integrated steel-making processes.

Over recent years, more and more emphasis in road building has been placed on cost saving. As a result of this, the recycling of used road materials and the use of cheaper industrial by-products has been developed. Environmental factors have also played their part in this development.

R&D PROJECT IMPLEMENTATION

Preliminary preparations and project organising

The principal responsibility in the whole project management and implementation was carried out by SKJ Ltd, which is a subsidiary of Rautaruukki Oyj, the biggest steel producer in northern European countries. Prior to the actual planning of the project a seminar was held in order to get new ideas and perspectives for the project. Experts from a wide variety of interest groups were invited and representatives from research institutes, universities and road constructors were present in the seminar.

After comprehensive analysis and several smaller work group meetings, the actual project organising and detailed planning were started. Contacts with the research institutes, universities and road constructors played an important role and thanks to effective netting, SKJ Ltd managed to create a project, which would be able to work technically efficiently.

Financial support was received from the Technology Development Centre of Finland (Tekes). Rautaruukki Oyj and the Finnish Road Administration also played an important role; the former by offering R&D services and the latter by constructing the experimental road sections. The University of Oulu took care of most of the laboratory work. The Geotechnical laboratory carried out a huge number of various kinds of laboratory tests. Some research work was also done in the Department of Geology and the Department of Chemistry. The Research Centre of Finland (VTT) had an important role as a coordinator of several associated projects, which were financed by Tekes and by-product companies like SKJ Ltd.

Actual project work

The actual project work was started at the beginning of 1995 and the project was carried out according to the diagram shown in Fig. 2. Total implementation time for the project was five years with one additional year being needed to finish all the instruction work. The most important instructions were made during the project.

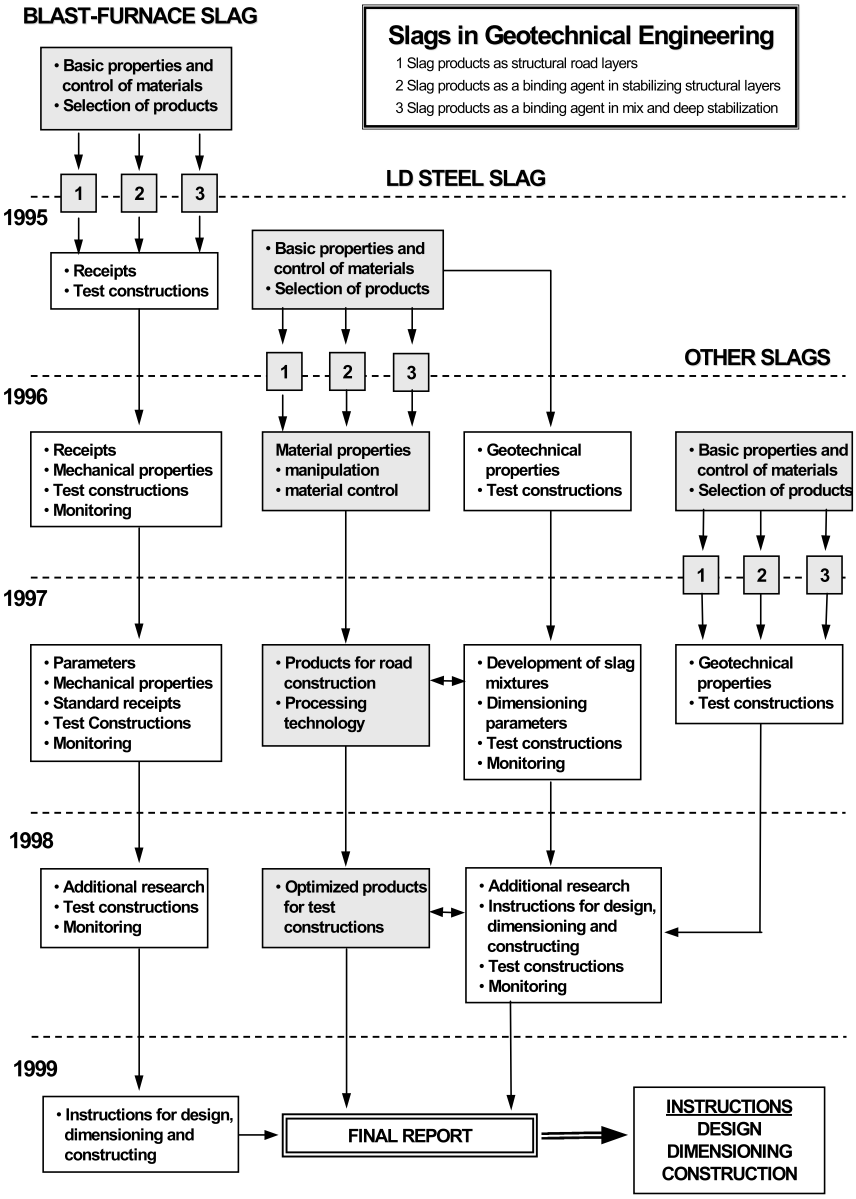

Figure 2. Research sectors in actual project work according to slag types under research.

The project was divided into three parts. These were: 1) slag products as structural road layers, 2) slag products as a binding agent in stabilising structural layers and 3) slag products as a binding agent in mix and deep stabilisation. A number of smaller studies on the use of slag as structural materials in road construction was also implemented, and a number of slag products with greatly varying grain-size distributions were examined and developed.

In all these areas of research, there was in progress an effort to increase knowledge of the basic properties of these materials. For the LD steel slag, desulphurizing slag, and ladle treatment slag, this means determining the technical properties, and for the blast-furnace slag and LD steel slag, the binding properties.

Area 1 research concentrated upon developing LD steel slag and blast-furnace slag products. In addition, the suitability of desulphurization and ladle treatment slag was studied. The main goal for area 1 research work was to increase understanding concerning material behaviour of each slag type.

In area 2 studies water cooled blast-furnace slag, refined in various degrees, for use as a binder in the stabilising of road courses, was developed. The products were granulated blast-furnace slag, preground granulated blast-furnace slag and ground granulated blast-furnace slag. In addition, included in this area of the research was LD slag, which was used in its original form or as an activator refined to various degrees. Cement was also used as an activator.

The main emphasis in area 3 was on the binding properties of ground granulated, cement-activated BF slag, primarily in the stabilisation of clay materials.

There are two possible ways of implementing the research and development work in order to improve the quality of the final product. One way is to change the properties of the product (Fig. 3). This would happen for example by grinding coarse slag gravel into a finer product. The other way is to develop a new way of using the product. For example granulated blast furnace slag has conventionally been used in Finland as massive layers in road construction. In the early 90’s its utilisation was developed in such a way that nowadays exactly the same product is used for a more demanding purposes, that is as a binding agent in road stabilisation.

For the blast-furnace slag the research concentrated on binder techniques of stabilisation, and its uses in massive layers (unbound layers).

In relation to the study of stabilising binder materials, a comprehensive investigation was already done on factors affecting the strength of stabilised materials (Mäkikyrö 1995). The development of binder material mixes, and determination of technical properties with the aim of producing procedures and instructions, was done on the basis of this previous study. These studies were focused on stabilisation of recovered road materials and till. In parallel with these, studies were made in connection with the stabilising of crushed rock. Materials to be used as binder were cement-activated granulated blast-furnace slag, preground granulated blast-furnace slag and ground granulated blast-furnace slag.

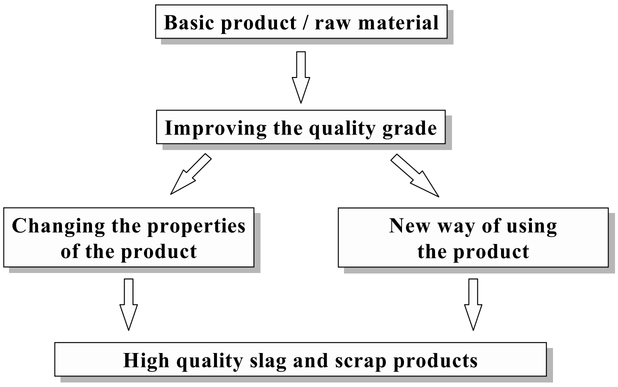

Figure 3. Two ways of implementing R&D work.

Mix- and deep stabilising studies were done on the stabilising of clays using cement activated ground granulated blast-furnace slag.

For LD steel slag the research concentrated on two main sections: 1) the use of LD steel slag as a replacement for natural materials and air-cooled blast-furnace slag in massive constructions and 2) the use of LD steel slag in slag binders for the stabilising of road courses. The use of LD steel slag in slag mixtures was studied only in some minor extent.

The emphasis in research was to put on producing LD slag products of a more valuable nature. The overall aim of the research was to determine the design parameters for construction courses using slag, and those using slag binders. The determination was made in relation to bearing capacity and frost resistance properties.

The use of LD steel slag as a replacement for natural materials and air cooled blast-furnace slag in massive constructions means in practice determining the properties of LD slag aggregates and comparing those properties to natural aggregates and blast-furnace slag. Possible changes in technical properties over the course of time were also taken into consideration.

The slag products, which were used in the binder material research, were granulated blast-furnace slag, preground granulated blast-furnace slag, ground granulated blast-furnace slag (400 m^2/kg, Blaine), LD steel slag (0–3 mm), preground LD slag, ground LD slag (150 m^2/kg, Blaine) and cement. The aim of the binder material research was the replacing of the cement, used as activator, by LD slag wherever possible. In addition, the binding properties of LD slag itself were studied. The first stage of the research was to develop more reactive slag binders. In the second stage the regulation of the ratio of binder materials in relation to the natural materials used for binding was investigated.

One of the most important studies regarding LD steel slag was that of determining the expansion properties, and in particular, the means of controlling expansion. The primary aim was to find a structural solution to stability.

Only some minor laboratory studies has been done concerning desulphurization slag and ladle treatment slag. The aim was to determine the basic properties of these slag types and get some idea of their technical behaviour.

The amount of laboratory tests performed and material used during the whole project were huge. Several thousands of single tests were made and about 15 tonnes of material were handled in the laboratory alone. Testing methods were the so-called ’basic geotechnical tests’, i.e. the number of results is remarkable and the methods were quite simple.

A central part of all areas of the research was the constructing of trial road constructions in co-operation with the Finnish National Road Administration. The design of the trial constructions was based on the results of the laboratory tests. A very important part of the monitoring of the trial road courses was that of measuring both bearing capacity and any possible changes occurring due to frost.

In the summer of 1995 seven trial stretches (each 100 m) of on-site stabilised road using cement-activated granulated blast-furnace slag stabiliser were built. Stabilisation was carried out so that the materials of the old road pavement were utilized as aggregate of the new layer, which was stabilised (Fig. 4). The existing natural rock material and the old bituminous pavement materials were used for improving the road. New material was used only for laying the new wearing course. The typical thickness of stabilised layer was 150–200 mm and the wearing course was bituminous asphalt concrete, thickness 50–100 mm.

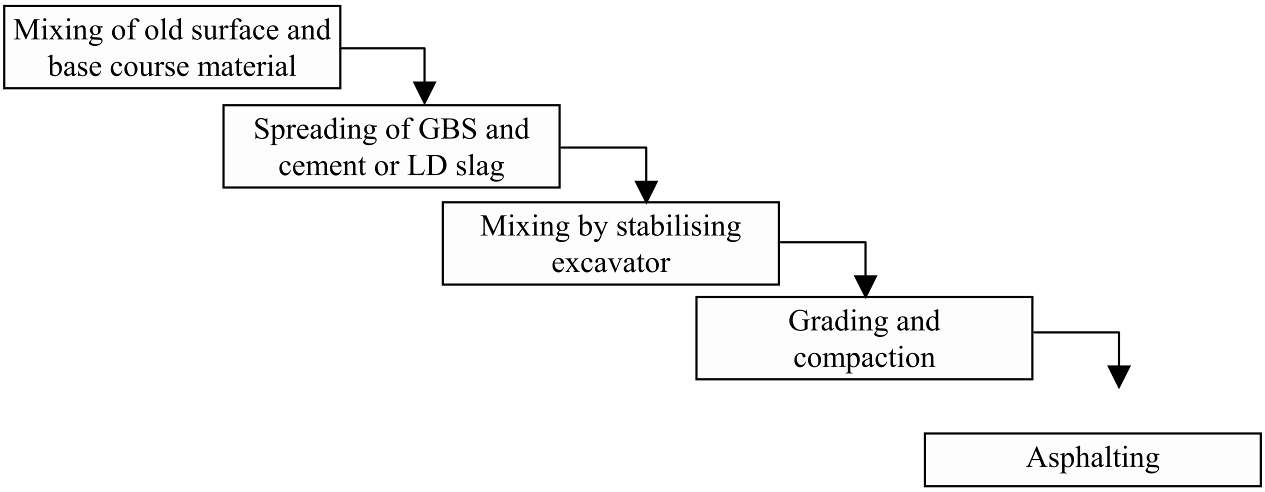

Figure 4. Different stages in so-called on site stabilisation technique.

At the end of 1995, a trial in connection with pillar type, deep stabilisation was carried out, using cement-activated ground granulated blast furnace slag. This test construction was made as a part of the Road Administration’s research project TPPT. In 1996, the first instrumented trial constructions using LD slag were built, in connection with massive constructions. In the summer of 1997, a total of 10 trial sections were built. Five sections were made with the on-site stabilisation techniques and five were made as massive layers. A schematic picture of the test sections with massive slag structure is presented in Fig. 5.

Instrumented constructions included the following measurements: frost penetration, moisture content in different layers, thermal regime from 10 different depths (10 cm to–350 cm), thickness of snow in road slope and thermal conductivity. Measurements have been performed 6 times a year and load bearing measurement with a falling weight deflectometer three times a year. Monitoring of test sections will be continued several years from now on.

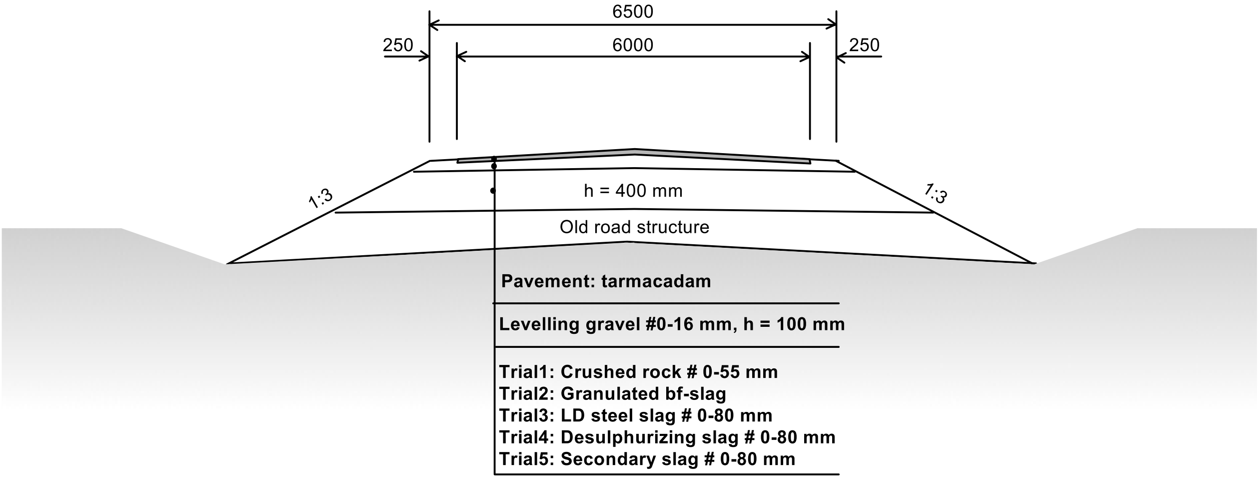

Figure 5. A schematic cross-section of test constructions with massive slag structures.

Results

During the whole project thousands of tests were performed in the laboratory. The majority of these tests were so called basic laboratory tests, the aim of which was to produce as much measured data as possible in order to get a reliable idea of the material behaviour of different slag types, especially those for LD steel slag. Research work was done for various product fractions from fine powder up to very coarse crushed aggregates.

The main result of the entire project was the development of instructions for the use of slag products in road construction. Some of the instructions were drawn up during the project implementation (Tielaitos 1998; Finnra 18/1999, Siira *et al.* 1999) and others (Slag products in earth structures, Slag products in construction on weak subsoils and Stabilisation with granulated blast-furnace slag) are still under preparation and will be finished by the end of 2001.

Fig. 6 shows one example of development work for slag-based binders. The effect of the amount of slag components in slag-based binder was tested by mixing selected slag components and using them as binder for stabilising of crushed rock material. LD3 is ground LD steel slag (150 m^2/kg, Blaine) and GBS is normal granulated blast-furnace slag produced in the Rautaruukki Raahe steel works. In this kind of binder LD slag is used as an activator instead of cement. Results indicate that the increasing amount of activator results increasing compression strength. However, the degree of strength should be higher in order to fulfil the demands set for stabilised road base.

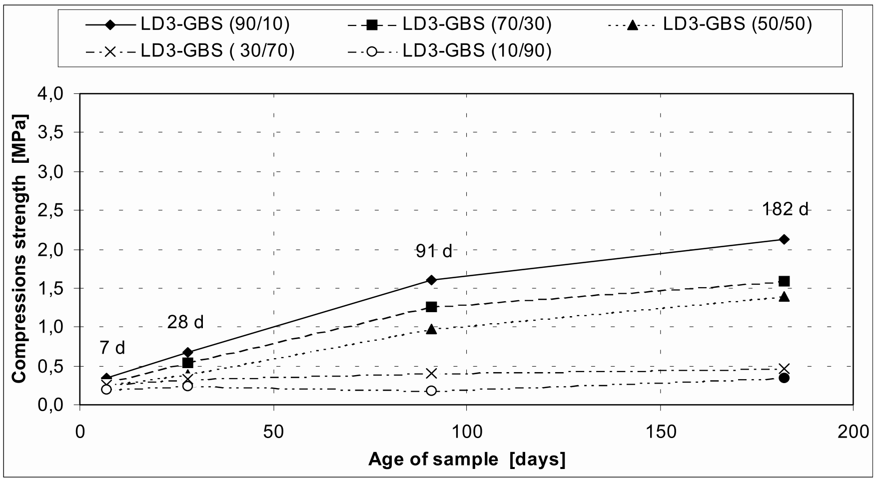

Figure 6. The effect of the amount of slag components in slag-based binder. Stabilised material was crushed rock.

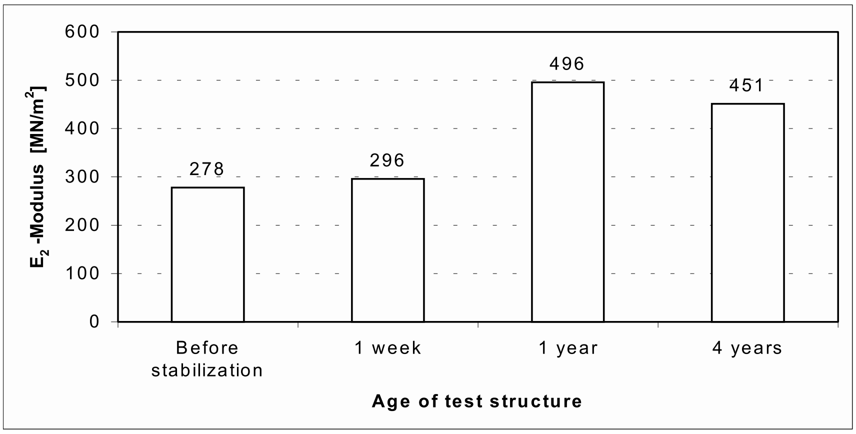

Figure 7. The improvement of bearing capacity of one test structure made in 1995. Values are mean values from ten different measuring points.

The measurement of bearing capacities of stabilised roads was an essential part of monitoring test road sections. Fig. 7 shows the development of the bearing capacity for one test section made by stabilising old road base with granulated blast-furnace slag, which was activated by cement. A remarkable increase for E_2 -modulus was achieved during the first year of monitoring, after which the value has remained at the same level.

SUMMARY AND CONCLUSIONS

The main goal for the project was to prepare instructions for the use of slag products in road construction. To achieve this result a lot of research work was done both in the laboratory and in actual conditions, i.e. on test constructions. The project was implemented according to a time schedule, which was set at the beginning and only some changes were made to the plans during the project. The main result, that is the making of new instructions, was also achieved and in this way one big step was taken forward in upgraded slag utilisation in Finland.

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FIELD HYDRAULIC CONDUCTIVITY OF A PAPER MILL SLUDGE HYDRAULIC BARRIER USING TWO STAGE BOREHOLE TESTS

by
Juan D. Quiroz[1], Thomas F. Zimmie[2] and Benjamin D. Rosenthal[2]

Quiroz, Juan, D., Zimmie, Thomas F., & Rosentha, Benjamin D. 2001. Field hydraulic conductivity of a paper mill sludge hydraulic barrier using two stage borehole tests. *Geological Survey of Finland, Special Paper 32, 39–45,* one figure and 2 tables.

This paper deals with the field hydraulic conductivity evaluation of a paper sludge landfill cover located in Corinth, New York that was completed in 1995. This landfill cover, a demonstration project, consists of compacted clay and paper sludge hydraulic barrier sections. Laboratory flexible-wall hydraulic conductivity tests were performed on in-situ samples obtained adjacent to in-situ two-stage borehole (TSB) test locations. The TSB test is a field infiltration test that has gained popularity over the last several years for the evaluation of hydraulic barriers. The objectives of this study were to evaluate the paper sludge long-term field hydraulic conductivity, and compare relative differences between laboratory and field hydraulic conductivity values. Results indicate that paper sludge is a suitable hydraulic barrier material alternative since laboratory and field hydraulic conductivity values were near or less than 1×10^{-9} m/s (the typical regulatory requirement), similar to compacted clays. Overall, the paper sludge field hydraulic conductivity values were about half an order of magnitude to one order of magnitude greater than those measured in the laboratory. In contrast, the compacted clay field hydraulic conductivity values were slightly lower than the measured laboratory values. The main difference between laboratory and field hydraulic conductivity values for each hydraulic barrier material type is attributed to effective stress sensitivity. Paper sludge is very sensitive to effective stress relative to compacted clay. Additional factors, advantages and disadvantages were discussed for laboratory and in situ TSB tests.

Keywords: landfills, disposal barriers, paper industry, by-products, sludge, hydraulic conductivity, boreholes, testing, Corinth, New York, United States

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INTRODUCTION

Paper mill sludge landfill covers

In recent years, paper mill sludge has emerged as an alternative landfill cover hydraulic barrier material instead of compacted clay. Paper mill sludge is the residual material from the paper making process produced by a wastewater treatment plant. Recycling paper sludge often creates a win-w*in situ*ation for the landfill owner (often a municipality) who saves the cost of obtaining clay that may not be readily available, and the paper mill which saves the cost of sludge disposal.

Paper sludges are high water content materials that typically have high organic contents, high compressibilities and low shear strengths. Several researchers have investigated the geotechnical and geoenvironmental properties of paper sludge (e.g., Zimmie *et al*. 1995, Moo-Young and Zimmie 1996, Kraus *et al*. 1997, Benson & Wang 1999 and Quiroz 2000). These studies have shown that paper sludge hydraulic barriers can achieve hydraulic conductivity values of 1×10^{-9} m/s or less, comparable to compacted clays. Moreover, the unique geotechnical field behavior of paper sludge is a result of its geotechnical properties, which are in a different range when compared to typical soils (e.g., sands, silts and clays). For example, after construction, paper sludge barriers undergo high settlement stains due to primary and secondary consolidation ranging from 18% to 35%, even under the low overburden pressures of a typical landfill cover. These settlement strains induce large reductions in void ratio causing decreases in hydraulic conductivity. This behavior is especially important in cases where marginal hydraulic conductivity values are measured, since the paper sludge hydraulic barriers improve with time. Thus the long-term evaluation of in-situ hydraulic conductivity is an important issue and should be considered in post-closure paper sludge landfill cover monitoring strategies.

Objectives

The purpose of this project was to evaluate the long-term field performance of a paper sludge landfill cover constructed in 1995, and compare the differences between laboratory and field hydraulic conductivity values. Currently, little information exists on the in-situ measurement of hydraulic conductivity for paper sludge landfill covers. The measurement of hydraulic conductivity using flexible-wall triaxial cell permeameters is probably the best method of laboratory testing (Zimmie 1981, Daniel *et al*. 1984). However, in general, one problem with laboratory tests is the amount of disturbance applied in the sampling process. This disturbance is significant in the retrieval of paper sludge samples due to high fiber contents and low shear strength ranges. In addition, the stress condition applied in a triaxial cell is different from actual, in-situ stresses (i.e., $\sigma_1 = \sigma_3$ versus K_0 stress condition). Therefore, the measured laboratory hydraulic conductivity values may be different from the in-situ values. An alternative is the in-situ measurement of hydraulic conductivity using the Two-Stage Borehole (TSB) test, also known as the Boutwell test (Daniel 1989, Trautwein & Boutwell 1994). The comparison between *ex situ* and *in situ* hydraulic conductivity testing will provide additional hydraulic conductivity behavior information essential to the application of paper sludge hydraulic barrier technology.

TOWN OF CORINTH LANDFILL

The paper sludge landfill is located in Corinth, New York and has a footprint of about 5.2 ha. Since paper sludge was considered experimental and nonconventional within the State of New York, this landfill closure was approved as a demonstration project. Thus the landfill cover had a compacted clay and paper sludge barrier layer section to provide a direct comparison between each barrier layer type. Approximately 4.1 ha is covered with a 1.2 m thick paper sludge layer, while the remaining 1.1 ha is covered with compacted clay about 45 cm thick. An overburden sand layer about 75 cm to 100 cm was placed on the paper sludge hydraulic barrier as a barrier protection and top soil layer. The landfill closure was completed in 1995.

The paper sludge for the hydraulic barrier was obtained from the International Paper (IP) Co. located in Corinth. Fresh paper sludge produced by the wastewater treatment plant and old paper sludge harvested from the paper mill's sludge landfill was used for the barrier layer. Moo-Young and Zimmie (1997) and Floess *et al.* (1998) have presented the construction aspects and geotechnical properties for the Town of Corinth Landfill and IP paper sludge.

IP paper sludge geotechnical properties

The initial geotechnical properties of the IP paper sludge were measured during the construction phase of the Town of Corinth Landfill cover (Moo-Young & Zimmie 1996). In general, water contents, organic contents, and specific gravities varied from 150% to 220%, 42% to 56%, and 1.80 to 1.97, respectively. These geotechnical properties are in a completely different range when compared to typical compacted clays. High water contents and high organic contents account for the large compression index (C_c) values, 1.27 to 1.96, measured for IP sludge. As a result, large settlement strains due to consolidation are expected after construction of the barrier layer.

METHODOLOGY AND APPROACH

A comparative testing program was developed to measure laboratory and field hydraulic conductivity values for the Town of Corinth Landfill cover. Four testing locations were selected on the landfill hydraulic barrier, three in the sludge section and one in the clay section. The tests were conducted during the summer and fall of 1999.

Sample retrieval

Hydraulic barrier specimens were obtained using 73 mm diameter thin-walled Shelby tubes. Compacted clay samples are typically retrieved using a slow, steady insertion effort such that a continuous, intact sample is obtained. However, this method is not very effective for paper sludge sampling due to the low shear strengths and high organic contents of the sludge (Moo-Young & Zimmie 1996, Benson & Wang 2000, and Quiroz 2000). A slow insertion and resistance to shearing by the organic fibers often compresses the sludge, greatly disturbing the soil matrix. Therefore, paper sludge samples should be retrieved using a clean, dynamic effort to obtain a more "undisturbed" sample and higher recovery rates. The most common method is to simply place a Shelby tube on the paper sludge then apply a dynamic effort, say with a sledge hammer, to quickly insert and cut through the fibrous sludge. A block of wood placed on top of the tube during hammering is often used to protect the Shelby tube. Shelby tubes samples were obtained adjacent to the two-stage borehole test locations. These samples were extruded and trimmed for laboratory hydraulic conductivity testing.

Laboratory hydraulic conductivity tests

Laboratory flexible-wall triaxial hydraulic conductivity tests were performed on *in situ* specimens in accordance with ASTM D-5084. The procedures were modified when testing the paper sludge specimens to account for its unique behavior. Large volume changes can be expected due to the compressible nature of the sludge, and gas generation has been encountered in some cases during hydraulic conductivity tests (Benson & Wang 2000). The effective stress applied during testing was 34.5 kPa, a typical landfill cover stress, which was accomplished using a cell pressure of 310.5 kPa and a back pressure of 276 kPa. A gradient of about 21 was applied across the specimen during the permeation phase of the test.

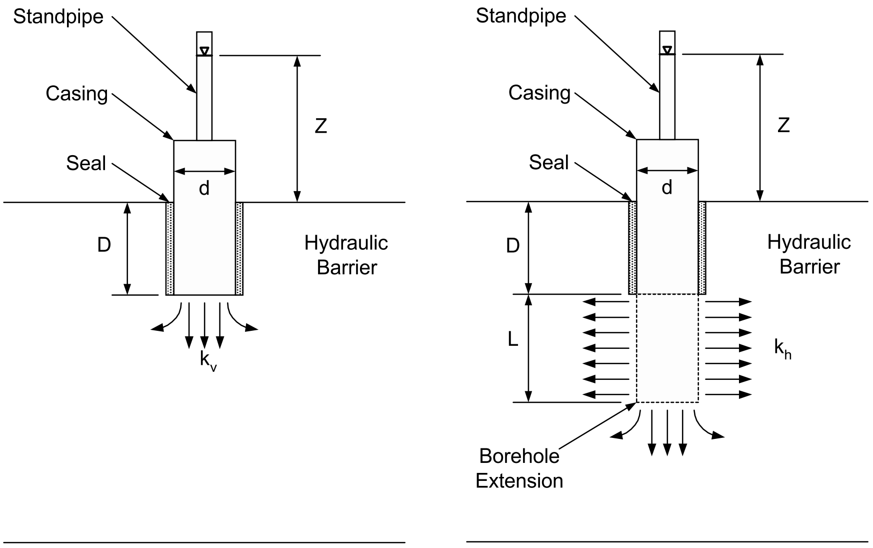
One important aspect of laboratory paper sludge hydraulic conductivity testing is effective stress specification. Since paper sludge is highly compressible, it is sensitive to testing pressures. Thus, as the applied effective stress increases, the hydraulic conductivity decreases. For example, by simply doubling the applied effective stress from 34.5 kPa to 69 kPa, a one order of magnitude decrease in hydraulic conductivity is not unusual for paper sludges. In practice, laboratory hydraulic conductivity tests are conducted at two effective stresses, 34.5 kPa and 69 kPa, for predictive purposes (Moo-Young & Zimmie 1995, Quiroz 2000). The 34.5 kPa test results represent current hydraulic conductivity values or those immediately after construction, while the 69 kPa results are used to provide long-term hydraulic conductivity values (i.e., a decreasing hydraulic conductivity trend) after subsequent consolidation.

Field hydraulic conductivity tests (two-stage borehole tests)

The *in situ* hydraulic conductivity was measured using the two-stage borehole (TSB) test following the procedures outlined in ASTM D-6391. This field test is often used to evaluate compacted clay hydraulic barriers within the United States due to its lower cost and shorter testing times in comparison to other field methods such as the sealed double ring infiltrometer test (SDRI) (Trautwein & Boutwell 1994). Holes about 1.3 m × 1.3 m were excavated down to the paper sludge barrier layer for TSB testing.

The TSB test method uses a sealed, cased borehole and monitors the water level in a standpipe. Using the falling head method, the rate of flow into the soil is measured. The first of the two stages is conducted with the casing flush with the bottom of the borehole. In this phase, the vertical hydraulic conductivity, k_v, is dominant (Fig. 1 a.). Flow into the soil is monitored until it is constant, then the first stage hydraulic conductivity, k_1, is determined. The subsequent second stage involves extending the borehole below the bottom of the casing and monitoring the falling water level (Fig. 1 b.). During this phase, the horizontal hydraulic conductivity, k_v, is dominant. Again, once the inflow is constant, the second stage hydraulic conductivity, k_2, can be determined. Finally, knowing k_1, k_2, and various test parameters, the values of k_v and k_h can be uncoupled.

From a practical standpoint, if the test is performed to determine if k_v is less than some specified value, sometimes only the first stage may be conducted. If the "apparent hydraulic conductivity" (arithmetic time-weighted average hydraulic conductivity) for the first stage is constant and below the specified value, then the test may be terminated (ASTM D-6391).



a.) STAGE 1 b.) STAGE 2

Figure 1. Two-stage borehole test for hydraulic barriers: a.) first stage of infiltration, b.) second stage of infiltration (after Trautwein and Boutwell 1994).

When conducting TSB tests on paper sludge, Benson and Wang (2000) recommended keeping Z less than D. This prevents hydraulic fracturing due to near zero effective stress at the bottom of the borehole, which can cause a rapid head loss in the standpipe, yielding incorrect hydraulic conductivity values. This was generally not a problem for the tests conducted in this study since the barriers had an overburden sand layer about 75 cm to 100 cm, resulting in relatively high effective stresses. The estimated effective stress at the bottom of the casing was about 18.9 kPa and test times lasted about 1 month to acquire constant k_1 and k_2 values.

RESULTS AND DISCUSSION

The results of the laboratory hydraulic conductivity tests conducted on *in situ* samples of the Town of Corinth Landfill cover are presented in Table 1. In general, under an effective stress of 34.5 kPa the hydraulic conductivity values were less than the typical regulatory limit of 1 × 10^{-9} m/s. The paper sludge proved to be an effective hydraulic barrier material similar to the compacted clay.

Table 1. Laboratory flexible-wall hydraulic conductivity test results for the Town of Corinth Landfill cover.

Sample	Effective Stress (kPa)	k_{lab} (m/s)
clay-1	34.5	2.0×10^{-10}
sludge-1	34.5	5.2×10^{-10}
sludge-2	34.5	7.9×10^{-10}
sludge-3	34.5	5.2×10^{-10}

Table 2 shows the results of the *in situ*, two-stage borehole tests conducted on the clay and paper sludge sections of the Town of Corinth Landfill. The k_v value is the main parameter of interest for landfill covers and liners. For an average effective stress of 18.9 kPa, the paper sludge k_v values (including the “apparent hydraulic conductivity” value for sample sludge-3) were about half an order of magnitude or less greater than 1 × 10^{-9} m/s; while the compacted clay exhibited a k_v much lower than 1 × 10^{-9} m/s. Since the paper sludge is a highly compressible material that can experience a large degree of secondary consolidation (Quiroz 2000) it is expected that continued barrier layer settlement will further decrease the vertical hydraulic conductivity to meet regulatory requirements.

Table 2. Two-stage borehole, field hydraulic conductivity test results for the Town of Corinth Landfill cover.

Sample	Effective Stress (kPa)	k_h (m/s)	k_v (m/s)
clay-1	18.9	1.0×10^{-8}	9.1×10^{-11}
sludge-1	18.9	2.3×10^{-9}	2.3×10^{-9}
sludge-2	18.9	1.3×10^{-8}	2.2×10^{-9}
sludge-3[a]	18.9	–	–

[a] Only stage one was completed (“apparent hydraulic conductivity” = 5.2 × 10^{-9} m/s).

For the paper sludge, the laboratory and field results indicated that field k_v values (including the “apparent hydraulic conductivity” value for sample sludge-3) were about half an order of magnitude to one order of magnitude greater than those measured in the laboratory. This is primarily due to differences in effective stress, 34.5 kPa in the laboratory versus about 18.9 kPa in the field. Since paper sludge is highly compressible, it is sensitive to confining pressures, i.e., as the effective stress increases the hydraulic conductivity decreases due to reductions in void ratio and/or constriction of microscopic flow channels. Therefore, for the paper sludge hydraulic barrier, greater hydraulic conductivity values were measured by the *in situ* two-stage borehole tests. In contrast, for the compacted clay samples, the difference in hydraulic conductivity between laboratory and field values was less than half an order of magnitude, which is not surprising since typical compacted clay barriers are not very compressible. LaPlante & Zimmie (1992) and Othman & Benson (1992) showed less than a half an order of magnitude decrease in hydraulic conductivity for compacted clays subjected to effective stress levels increasing from about 15 kPa to 200 kPa. Testing considerations which also affect hydraulic conductivity results are sample size or test volume affected, soil saturation levels, and quality of soil tested (how many defects present, micropores versus macropores, etc.). An additional hydraulic conductivity testing aspect specific to paper sludge is sample disturbance since it is a low strength and highly compressible material. The *in situ* TSB test for paper sludges may be a more suitable method for evaluating field performance since it minimizes sample disturbance. However, as discussed previously, laboratory tests may provide additional data since you can vary effective stress levels to acquire an indication of sensitivity to confining pressures and potential long-term predictions after subsequent consolidation.

SUMMARY AND CONCLUSIONS

Hydraulic conductivity analyses was performed on the Town of Corinth Landfill cover located in the State of New York. This unique 5.2 ha landfill cover demonstration project had a 4.1 ha paper sludge hydraulic barrier layer section and a 1.1 ha compacted clay barrier layer section. A total of four locations (one on clay and three on paper sludge) were tested via laboratory flexible-wall triaxial hydraulic conductivity tests and *in situ* two-stage borehole tests. The objective of this study was to evaluate the long-term performance of the landfill cover, especially the paper sludge barrier layer section, and compare the differences in laboratory and field hydraulic conductivity values.

The laboratory hydraulic conductivity tests performed using an effective stress of 34.5 kPa showed that the paper sludge and compacted clay samples had hydraulic conductivity values less than the typical regulatory limit, 1×10^{-9} m/s. This is not surprising for compacted clay barriers, however, it does show that paper sludge is a suitable hydraulic barrier material alternative. The *in situ* two stage borehole tests conducted at an average effective stress of about 18.9 kPa showed that the compacted clay k_v value was much less than 1×10^{-9} m/s, however, the paper sludge k_v values were half an order magnitude or less greater than 1×10^{-9} m/s. Also, the differences between laboratory and field hydraulic conductivity values were about half an order of magnitude to one order of magnitude. The differences between the compacted clay and paper sludge k_v values, as well as the laboratory and field hydraulic conductivity values, was a result of the high compressibility of paper sludge. Since paper sludge is very sensitive to effective stress (i.e., as the effective stress increases the hydraulic conductivity decreases) relative to clay, higher hydraulic conductivity values are expected from the tests conducted at lower effective stresses. Nonetheless, it should be noted that for all practical purposes the paper sludge hydraulic barrier is an effective hydraulic barrier. Moreover, due to high water contents and high organic contents, the paper sludge barrier layer is expected to undergo a considerable amount of consolidation which will continue to reduce void ratios and decrease hydraulic conductivity.

It was also noted that sample disturbance can play an important role in sludge hydraulic conductivity values due to its low shear strengths and high organic fiber contents which can impede the retrieval of quality “undisturbed” samples. Typically, a dynamic sampling method is best for sludge to minimize sample disturbance. As an alternative, *insitu* hydraulic conductivity tests such as the two-stage borehole test can give a more accurate estimation of field performance. However, the flexibility of laboratory tests which can be used for predictive purposes by testing under two confining stress levels (see Section 4) is an advantage. Overall, hydraulic conductivity monitoring strategies should be closely examined for paper sludge hydraulic barriers since there are several differences, advantages and disadvantages between laboratory and *in situ* hydraulic conductivity tests.

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FINNISH NATIONAL REPORT ON THE ALT-MAT PROJECT

by
Leena Korkiala-Tanttu and Hans Rathmayer

Korkiala-Tanttu, Leena. & Rathmayer, Hans 2001. Finnish national report on the ALT-MAT project. *Geological Survey of Finland, Special Paper 32, 47–55,* seven figures and 3 tables.

The environmental aspects strongly necessitate more extensive utilisation of the alternative materials. This utilisation needs more information on the mechanical and leaching properties, functional requirements and long-term stability of the materials. The European ALT-MAT project defines methods by which the suitability of alternative materials for use in road construction can be evaluated under appropriate climatic conditions. The Finnish national work in the ALT-MAT project concentrated on the development of the climate chamber test. The purpose of these tests was to simulate the field conditions of road structures (miniembankment) without a pavement. The test consisted of 20 wetting-drying and freezing-thawing cycles. Miniembankments were wetted with salt solution. The leaching of contaminants due to these accelerated climatic cycles and added salt solution was studied. The tested materials were two types of slag: ferrochrome slag and blast furnace slag. The test results showed that the leached amounts of many metals were very low, usually below the limit of detection. These results were verified by other leaching test results. All test data showed that the leaching of contaminants for both types of slag is very moderate. The climate chamber test proved to be a good, but expensive method to test the environmental suitability of alternative materials.

Keywords: construction materials, highways, by-products, slag, pollutants, leaching, experimental studies, Finland

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INTRODUCTION

Every year the Finnish steel industry produces over one million tonnes of by-products, mostly different kinds of slag. This slag has mechanical properties that satisfy ideally the material requirements of road construction layers. In some respects, such as low weight and stress- and deformation resistance, slag is even better than natural aggregates. During the last few decades, a part of the annual production of blast furnace and ferrochrome slag types has been utilised as road construction materials. This utilisation has occurred mainly in the vicinity of steel factories. The growing awareness of environmental aspects and the lack of high quality road materials in some regions have increased the interest in utilising slag more efficiently.

The greatest obstacles to more efficient utilisation of slag lie in the uncertainties of environmental impacts. The environmental impacts of by-products are normally studied with leaching tests. To study varying climatic conditions and their effect on the leaching, we developed a new test method called the climate chamber test. The aim of climate chamber tests is to simulate field conditions of a road construction without a pavement. This method provides opportunities to bridge the gap between small-scale laboratory experiments and full-scale trial embankments. The test is designed to study the release of contaminants (leaching) due to accelerated climatic cycles and salt solutions used for winter maintenance in Finland.

The development of the climate chamber test method is a part of the ALT-MAT project. The main objective of the ALT-MAT project is to define methods to evaluate the suitability of alternative materials in road construction. The methods evaluate the suitability under appropriate climatic conditions. We also compared the results of the climate chamber test with other leaching test results to exhibit leaching under various circumstances.

As test materials we selected ferrochrome slag and blast furnace slag, because their general use as road construction material is always subject to special permission from the Finnish Environmental Institute.

MATERIALS AND METHODS

Test materials

The two materials studied are blast furnace slag (BFS) and ferrochrome slag (FCS). Their chemical composition is displayed in Table 1.

Blast furnace slag is a by-product obtained from the manufacturing of pig iron in a blast furnace. Blast furnace slag is formed by the combination of the rock constituents of the iron ore with a limestone flux. Because the constituents of slag are important for the function of the blast furnace, they must be kept as constant as possible. The quality of slag produced in a particular steelwork is highly consistent and its variability matches that obtained in the exploitation of conventional aggregates (Lee 1974). The production sites of BFS in Finland are the steel factories in Raahe and Koverhar.

The temperature in the blast furnace is over 1 300°C. Lighter slag floats on pig iron. Slag is conducted away from pig iron to the cooling process. There are two possible cooling methods: air- or water-cooling. In the air-cooling method the melt slag is poured out onto a cooling embankment where it cools slowly. Because the cooling is slow, minerals have time to crystallise. In the water-cooling method a high-pressure water jet cools the slag. This rapid solidification produces a granulated glassy material. The internal structure of this glassy slag is more fractured, which means that it is more reactive than air-cooled slag. Nowadays, most of the BFS produced in Finland is water-cooled.

The major components of BFS are silica, sodium and magnesium oxides (together 83 %). The main minerals of BFS are melilite, oldhamite, mervinite and some glass.

In addition to the chemical composition, the glassy structure of BFS affects its binding properties. BFS binds hydraulically, but considerably slower than cement. The addition of cement or mincing of grain size accelerates the binding reaction. The binding reaction happens on the surface of grains. After crushing binding starts up again on a new surface. This crushing in structure is then self-reconstructive in the long-term.

The cooling method affects the mechanical and chemical properties of the slag. The water-cooled

BFS (granulated) is a porous, slowly binding material that looks like sand. The bulk density of BFS is low, 11–13 kN/m^3, which means that it can be used as a lightweight material. The air-cooled BFS is a little heavier and its grains are bigger than water-cooled slag. The hydraulic, deformation and strength properties of both BFS are excellent, and they fulfil ideally the material requirements of unbound road construction material. The thermal properties of BFS are also very good; therefore it is natural that BFS is also used as a thermal insulating material.

Ferrochrome slag is a by-product obtained from the manufacture of refined steel in Tornio. The manufacturing process resembles that of the BFS. Correspondingly, FCS can be cooled with air or with water. The mechanical and chemical properties of FCS are quite similar to BFS. Yet, there are two important differences: the abrasion values of FCS are much better than BFS's and FCS does not have the capability to bind itself.

The major components of FCS are silica, aluminium and magnesium oxides (together 77 %). FCS includes 6.5 % chrome, which exceeds the total recommended concentration of chrome (Table 1).

Table 1. The chemical compositions of ferrochrome slag and blast-furnace slag.

Ferrochrome slag (water cooled) (aqua regalis) (%)	Typical analysis of ferrochrome slag (%)	Blast furnace slag (air cooled) (%)	Blast furnace slag (water cooled) Koverhar (%)	Granite, Teisko (%
CaO 3–8	CaO 2.1	CaO 37	Mg 9.41 XRF	CaO 2.4
SiO_2 27–29	SiO_2 28.6	SiO_2 35	Al 6.14 XRF	SiO_2 69.7
Al_2O_3 23–28	MgO 24.3	MgO 11	Si 15.0 XRF	MgO 0.9
MgO 22–27	Al_2O_3 27.0	Al_2O_3 9	P 0.006 XRF	Al_2O_3 15.4
S 0.1	Ti 0.29	TiO_2 2.5	S 1.79 XRF	K_2O 3.3
Cr 6.5	Mn 0.15	MnO 1.0	K 0.43 XRF	Na_2O 3.8
Fe 4.0	K 0.08	K_2O 1.0	Ca 24.0 XRF	Fe 2.6
	Na 0.02	Na_2O 1.0	Ti 1.12 XRF	Ti 0.2
	S 0.16	S 1.5	V 0.057 XRF	Ba 0.074
	Fe 4.5	FeO 0.5	Mn 0.12 XRF	Mn 0.062
	P <0.0005	P 0.003	Fe 0.63 XRF	P 0.042
	Ba 0.01	Ba 0.038	Cr 0.004 XRF	S 0.029
	C 0.12	Ce 0.012	Ni 0.001 XRF	Sr 0.018
	Cr 7.7	Cr 0.003	Sr 0.047 XRF	Zr 0.016
	Cu 0.01	La 0.006	Zr 0.015 XRF	Cr 0.013
	Co 0.03	Sr 0.042	Nb 0.001 XRF	Zn 0.008
	V 0.020	V 0.030	Mo 0.000 XRF	V 0.004
	Zn 0.010	Zr 0.017	Sn 0.001 XRF	Cu 0.003
	Sn <0.0010	Sn 0.002	Ba 0.056 XRF	Ni 0.002
	Li <0.01	Nb 0.001	La 0.006 XRF	
	Ni 0.03	Ni 0.001	Ce 0.009 XRF	
	Mo <0.01	B 0.004 2	Ca_{ASTM} 39.7	
	Sb <0.001	Cu 0.000 8	Ca_{pH5} 6.9	
	As <0.001	As 0.000 67	Fe_{met} 0.3	
	Bi <0.001	Zn 0.000 2	F 0.1	
	Pb < 0.001	Pb < 0.000 05	Zn 0.000 4	
	Cd < 0.001	Cd < 0.000 005	As 0.000 5	
	Hg < 0.000 002	Hg < 0.000 002	Cd <0.000 005	
			Pb <0.000 01	
			Hg <0.000 005	
			B 0.0487	
			Cu 0.000 08	

Test methodology

The climate chamber test boxes were constructed to bear stresses from compacting, moving and freezing during the test. The width and length of a box were 700 mm each and its height was 300 mm. The body of a box was made of plywood and its inside was covered with a plastic membrane of HPDE. A filter cloth was placed between the specimen and the plastic membrane. In the bottom of the box there was a lead-through for draining and leachate collection. The boxes were placed on movable carriages. Fig. 1 illustrates the construction of the miniembankment.

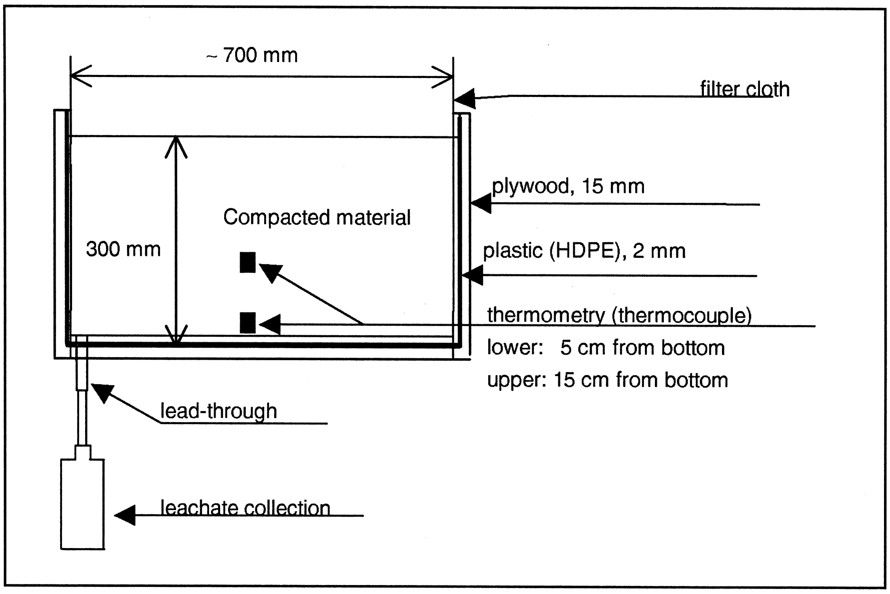

Figure 1. The construction of the miniembankment.

SKJ Company Ltd provided us with both types of slag. The tested materials were: air-cooled ferrochrome slag and water-cooled blast-furnace slag.

The test material was compacted into the box to a density of 95 % of the maximum density value of the Proctor compaction test. We had two concurrent specimens of both materials. In total we had four specimens. The technical data of these specimens are shown in Table 2.

Table 2. The technical data of specimens.

Data	Ferrochrome slag		Blast furnace slag		Remarks
	Specimen 1	Specimen 2	Specimen 3	Specimen 4	
Volume (cm^3)	152.70	150.37	152.80	159.31	95% of Proctor density
Dry density (g/cm^3)	1.83	1.83	1.37	1.37	
Weight (kg)	294.72	290.21	209.34	214.15	

The climate chamber test (CCT) consisted of three main phases: wetting-drying cycles, freezing-thawing cycles and leachate collection. The liquid, which is in contact with solid, is referred to here as leachant and the liquid leaving the solid is referred to as leachate. The flow chart diagram of the whole CCT leaching test is illustrated in Fig. 2.

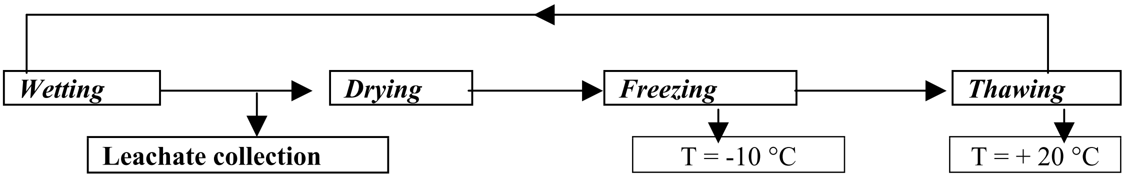

Figure 2. The flow chart diagram of the CCT test.

First, the specimens were wetted with a salt solution (leachant) composed of 10 % salt used by Finnish National Road Administration and 90% normal tap water. The typical chemical composition of salt is shown in Table 3. The total quantity of salt solution was about 25 kg in each wetting cycle. The wetting happened at a normal room temperature of +20°C. During the wetting, leachate was collected from the specimens into glass bottles. Leachate samples were analysed in the laboratory of VTT Chemical Technology. Leachate was collected:

- before first freezing
- after 2 freezing-thawing cycles
- after 10 freezing-thawing cycles
- after 20 freezing-thawing cycles

After the wetting and leachate collection, the specimens were dried for 1 day. Then, the lead-through was closed and carriages were moved to a cold room, whose temperature was –10°C. The temperature of the specimens during the freezing was monitored from two thermometers. When the whole specimen was frozen to –10°C, the specimens were taken to the thawing room (+20°C). In total, the specimens were treated with 20 freezing – thawing cycles.

In the beginning freezing – thawing cycles took at least 8 days each, thus to run all 20 cycles would have taken about five months. Since that was too long a period, the temperature of freezing room (-10°C) was be lowered to -30°C and the temperature of the thawing room raised to +40°C. With these test arrangements, one test cycle time could be lowered to 5 days. In spite of these accelerations, the climate chamber test is a very time-consuming test. In ideal conditions one whole test takes at least 3 months.

Table 3. The chemical composition of salt.

Component	Percentage (%)
NaCl	98.82
Ca	0.27
Mg	0.02
K	0.09
SO_4	0.80

RESULTS

Mechanical behaviour

We visually followed the behaviour of specimen during the test. The surface of the specimens cracked and heaved during freezing. These cracks and frost heave were due to the volume increase of the freezing water. After thawing some compaction occurred, but the structure did not get back to the original compaction ratio. The grain size distributions of specimens were tested before the first freezing and after the whole test (Fig. 3) to see whether any weathering had occurred during the test.

Chemical behaviour

We had two concurrent specimens of both slag types. The leachates were collected four times, so the total amount of analysed leachates was 16. We discussed with the chemists and chose together the most interesting elements from the environmental point of view to be analysed. The analysed elements were: Ca, Na, K, Al, As, Cd, Cr, Cu, Mo, Ni, Pb, Zn, Fe, Mg, Mn, sulphate and vanadium from blast furnace slag. The values of pH, conductivity and redox-potential were also measured. The concentrations of Ca, Na, K, Al, Cr, Cd, Mo, Zn, Fe, Mg, Mn and V were analysed using the atomic spectrometric technique (ICP-AES). The concentrations of Cu, Ni, Pb and As were determined using the GRAAS-technique and the concentration of sulphate was determined using the chromatograph – technique (FINAS T44/A1/95). The concentrations of many analysed elements were under the limit of detection. The accuracy of results varied from ± 10 % to ± 30% depending on the analysis technique.

DISCUSSION

Mechanical properties

The grain size distributions of the slag before and after the test cycles differ only a little. The largest grains are crushed a bit during the test (Fig. 3). The visual observations during the test cycles exhibit some loosening and frost heave.

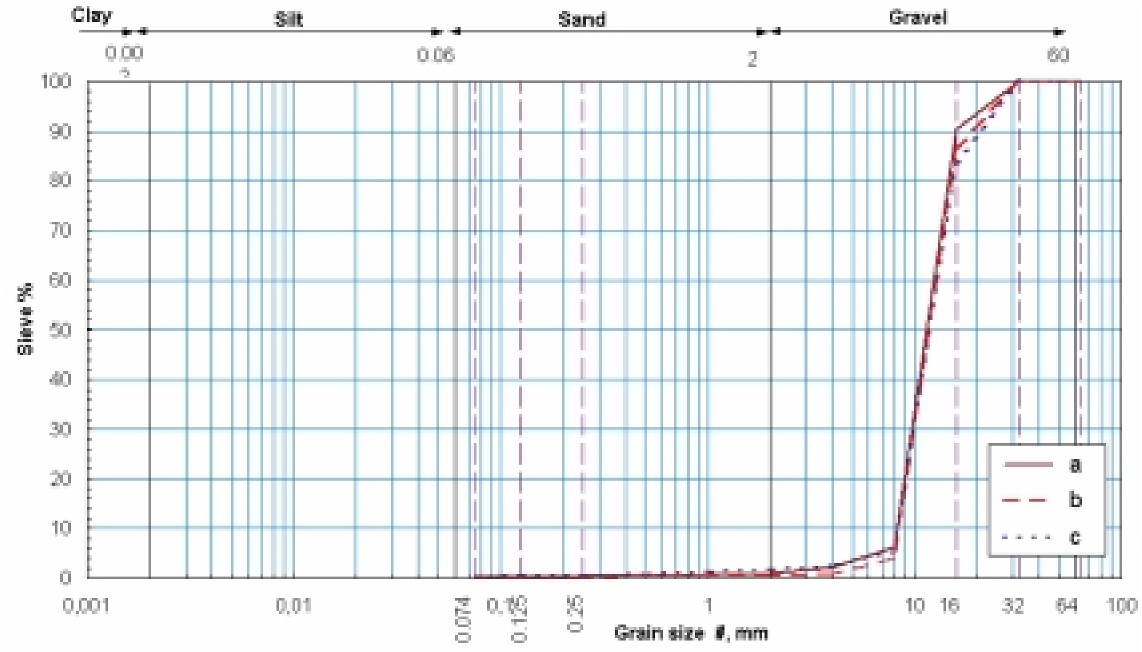

Figure 3. The grain size distribution of air-cooled ferrochrome slag (a = before test sample 1, b = before test sample 2 and c = after climate chamber test).

Leaching of blast furnace slag

The leaching of blast-furnace slag is very moderate in climate chamber tests. The test results indicate that freezing–thawing cycles and the use of salt solution do not accelerate the leaching process of any analysed element. Vanadium and sulphate, which are possibly the most harmful elements of blast furnace slag, stay in indissoluble form. Even though their total constituents are quite big, they leach very moderately in the climate chamber tests. We verified CCT test results with many other leaching tests successfully (Figs. 4 and 5). All test results confirm the impression that the leaching of blast furnace slag fulfils environmental requirements.

We changed all leaching test results to L/S-ratios, where L refers to liquid (the amount of used leachate flowing through a solid) and S to solid (the amount of alternative material). The use of L/S-ratio makes it easier to estimate leaching in relation to time, because the amount of liquid flowing through the solid increases with time. In many tests more than one L/S-ratio was analysed, so we summed up all these concentrations to get the cumulative concentration (mg/kg).

Most of the verification tests were different kinds of parametric tests, but we also included some simulation tests like the climate chamber, lysimeter, column and permeameter tests. In a parametrical test only one parameter, for example some leachant, is tested in ultimate conditions. Simulation tests, on the other hand, try to simulate the whole leaching process. That is why parametrical tests tend to overestimate real leaching. Leaching usually decreases with time, because the easily dissolvable elements leach out first. Thus, many illustrated test results, including climate chamber tests, exhibit a lower slope, when L/S-ratio is greater than 1 (Figs. 4 and 5).

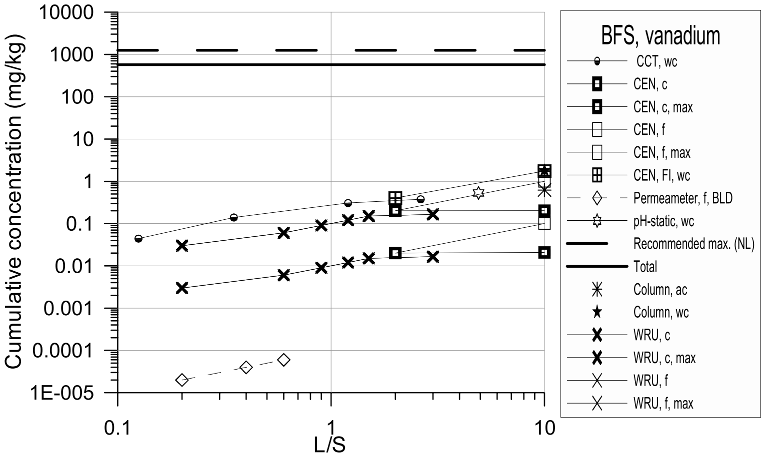

Figure 4. BFS. The leaching of vanadium.

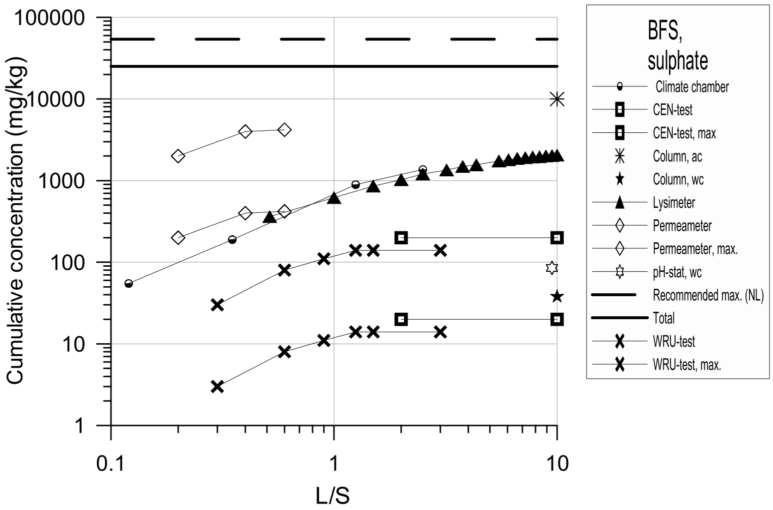

Figure 5. BFS. The leaching of sulphate.

The cooling method affects some leaching properties, especially the leaching of sulphate (Fig. 5). The leaching of sulphate from air-cooled (ac) slag in a column test is more than hundred times larger than leaching from the water-cooled (wc) slag (ac: about 10 000 mg/kg; wc: about 30 mg/kg). When slag is cooled with a high-pressure water jet, it is probable that sulphate components leach out with the cooling water. The leaching of sulphate in climate chamber tests increases up to 1000 mg/kg, even though the slag is water-cooled. This is because the salt solution contained sulphate and a great part of leached sulphate originates in salt solution.

Leaching of ferrochrome slag

The most interesting elements of ferrochrome slag from the environmental aspect are chrome and aluminium. The total constituent of chrome is 6.5 %, which is many times larger than the total concentration recommended by Dutch authorities. Because Finland does not yet have its own environmental recommendations, the Finns usually comply with Dutch recommendations. The chrome of FCS is mainly Cr^{3+} or Cr^{2+}, which are both difficult to dissolve. Hence, it is not surprising that no great leaching was detected from the climate chamber test (Fig. 6). Even though the recommended total constituent of chrome is exceeded, the leaching is very moderate and fulfils Dutch recommendations for a covered material. The other elements also leach moderately and fulfil Dutch recommendations. The most essential method of mitigating the environmental impacts of alternative materials is to decrease the amount of infiltrated water. Covering and bounding are common, effective and economical mitigation methods.

Wahlström & Laine-Ylijoki (1996, 1997) have studied widely the leaching of FCS in laboratory tests. Fig. 7 shows their test results of different kinds of leachants as a function of pH. The climate chamber test data agree with their test data. Ferrochrome slag's natural pH is over 10, but acidic rain water decreases its pH gradually. Yet, according to these test results, the leaching increases significantly only after the pH is less than 6. Wahlström & Laine-Ylijoki (*op. cit.*) have estimated that the time needed to neutralise a 0.3 m thick layer of ferrochrome slag from pH 10 to pH 7 takes 270 years. The pH in the climate chamber test is about 7.5.

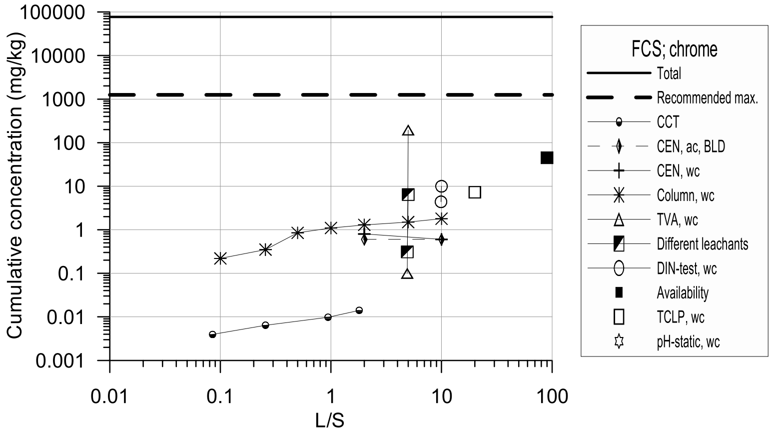

Figure 6. FCS. The leaching of chrome.

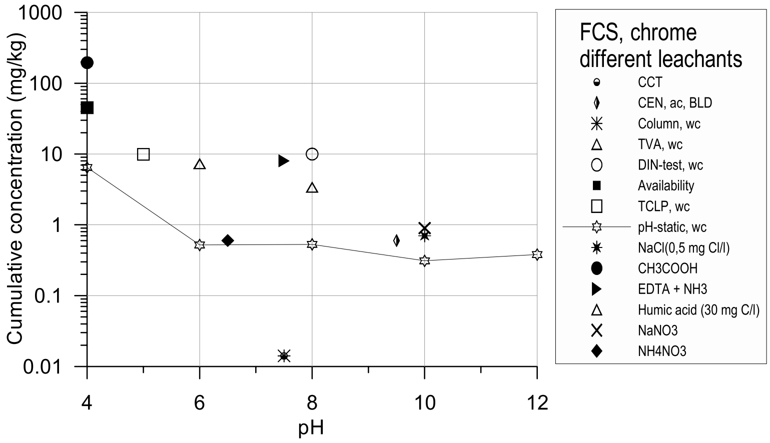

Figure 7. FCS. The leaching of chrome as a function of pH.

Future developments, limitations and errors

The climate chamber test is an expensive and time-consuming test. To make the test more efficient, more than two specimens should be handled at the same time.

The climate chamber test method has some limitations with leachate analysis. The first limitation is with the leachant. The use of salt solution as a leachant increases the leaching of sodium, potassium, calcium, magnesium and sulphate. The real leaching and its changes are difficult to distinguish from the initial constituency of these elements in salt solution. Because the parametrical and climate chamber tests have shown that salt solution does increase leaching, the use of fresh water instead of salt solution as the leachant would give a clearer picture of the leaching process. The second limitation is the analysis technique. The real scatter of test results is quite small. Yet the accuracy of analysis changed from ± 10 % to ± 30 %, which is quite big. The limit of detection with some elements is low, which hides the real leaching behaviour of that element. With more expensive analysis methods, the inaccuracy could be decreased and the limit of detection could be increased.

The simulated road construction is very simple: it has only one construction layer. This simplification causes some errors in test results. In a real road construction the flowing paths of water through the many construction layers are more complicated because of the covering asphalt layer. A new uncracked asphalt layer is practically impermeable. The ingress of water to a road construction occurs via the slopes, capillary rise from groundwater and through the cracks of an older asphalt layer. Thus, in the real road construction water infiltrates mainly horizontally, but in CCT water infiltrates vertically. The total quantity of water flowing through a road construction is also difficult to assess. In short, the flowing conditions in the climate chamber test are simplistic and they do not give the whole picture of the flowing in the sense of leaching.

CONCLUSIONS

The climate chamber test is a new, very promising test method to simulate the leaching of alternative materials. Even though it is expensive, it is the only test method which simulates the varying climatic conditions. The test includes twenty accelerated drying-wetting and freezing-thawing cycles. The tested materials were ferrochrome slag and blast-furnace slag. These climatic cycles did not increase the leaching of any element studied when we verified them with other leaching test results. Furthermore, the use of salt solution does not affect the leaching of the elements studied. On the contrary, the leaching in the climate chamber test, as in many other simulation tests, is clearly less than in small-scale parametrical tests.

The solubility of harmful elements of FCS is so small that the use of FCS is acceptable in every part of a road construction in which slag is covered or bound. In spite of the high total amount of chrome, the leaching of chrome is moderate. The leaching of harmful elements of BFS is also very moderate and fulfils today’s environmental requirements where slag is covered or bound.

The flowing conditions in the climate chamber test differ from real road construction, where water ingress happens via slopes, though the cracks of asphalt layer and capillary rise from groundwater. The greatest part of water infiltrates the road construction horizontally. In the climate chamber test the water infiltrates through the specimen vertically from top to bottom.

Besides the error in flowing conditions, there are some limitations to the CCT method. First, the leached amounts of many metals are very low – in many cases below the limit of detection. The limit of detection and accuracy of results depend on the analysis technique, which could be increased with more expensive analysis techniques. Second, the use of salt solution as a leachant increases the leaching of sodium, potassium, calcium, magnesium and sulphate. It is difficult to distinguish the real increase in leaching from the initial constituency of these elements in a salt solution. Therefore, we recommend the use of pure water as a leachant in the future. Another future development could be the measurement of frost heave or some other mechanical property.

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RISK ASSESSMENT OF INDUSTRIAL BY-PRODUCTS USED IN EARTH CONSTRUCTIONS

by
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Wahlström, Margareta, Eskola, Paula, Laine-Ylijoki, Jutta, Leino-Forsman, Hilkka, Mäkelä, Esa, Olin, Markus & Juvankoski, Markku 2001. Risk assessment of industrial by-products used in earth constructions. *Geological Survey of Finland, Special Paper 32, 57–63*, one figure.and 2 tables.

Industrial by-products can be used in earth constructions provided their environmental properties are acceptable; for example, not exceeding defined acceptance values. Risk assessment is needed in cases where the disposal conditions significantly differ from the conditions considered when the acceptance values were set for by-products suitable in earth constructions; for example, in the case of sensitive disposal environments or in cases where the construction consists of several different by-products. It might also be possible through risk assessment to accept higher emission values in cases where more information is available about the properties of the harmful compounds.

A procedure for the risk assessment of industrial by-products to be utilised in earth construction is proposed. The procedure is designed to be as simple as possible to use, yet cover the most important aspects and environmental risks in the assessment. Additionally, the special characteristics of construction and the use of earth constructions were also considered. The risks were primarily related to human health because the background data needed for an ecotoxicological risk assessment is still under development.

The proposed risk assessment concept has also been tested in a case study on the use of coal fly ash in earth construction.

Keywords: construction materials, risk assessment, by-products, ash, earthworks, construction

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INTRODUCTION

Industrial by-products may possess detrimental environmental properties, the impacts of which need to be considered before the products are used in earth constructions. Some examples of the most relevant properties to be considered for by-products are the following:

- Direct toxicity of by-product for humans, plants, and animals
- Leaching properties with special attention paid to all relevant conditions at the disposal site (as a consequence of the transport of toxic metals into the environment)
- Formation of toxic gas emissions, e.g. due to degradation, reactivity
- Dust problems
- Risk for ignition (fire)

Risk characterisation means that all the unfavourable properties or conditions are listed and described, and if possible quantified, for example as doses or emissions. The risk assessment also covers assessment of the impacts of undesired phenomena. The results of the risk assessment can be used to choose an appropriate way of utilisation where the undesired phenomena are minimised or can be totally eliminated by suitable measures.

Guidelines for the risk assessment of contaminated sites have been developed. These guidelines are often very broad and general, covering all kinds of exposure pathways to humans and the environment. In particular, the future use of the site is considered in the risk assessment.

The guidelines for the risk assessment of contaminated sites have been the basis for this work. It is not necessary to take into account many of the exposure pathways when industrial by-products are used in earth construction. The following exposure pathways, however, are considered relevant with regard to the use of by-products for such purposes:

Human exposure:
- inhalation of dust
- inhalation of volatile compounds
- dermal contact
- intake of by-products and water that has been in contact with the by-product

Environmental risks:
- influence on plants and animal species near the deposit
- influence on ground and surface water quality

The direct risks to humans are mostly related to the transportation and handling of the by-product. Examples of typical risks are the inhalation of dust, dermal contact, and with respect to children also ingestion of by-products. Risks to the environment may be caused by contamination of the ground and surface water as well as the soil near the site. The risks are significantly dependent on the sensitivity of the surroundings, the geological conditions at the site and the use of the site. For examples in an urban area, where drinking water or irrigation water is not taken from wells, the effects on humans from the intake of water or food can be regarded as insignificant. Examples of exposure routes in the utilisation of by-products in earth constructions are given in Table 1.

The study is part of a larger research entity aimed at developing a guide for the assessment of the environmental and technical suitability of by-products in the permission and product qualification process.

DESCRIPTION OF MODEL

Scope and field of application

The procedure is designed to be as simple as possible to use, yet cover the most important aspects and environmental risks in the assessment. The procedure is simplified and gives an overview of the general risk factors involved in using the by-product. The model includes several assumptions and it may not be applicable in all cases. Therefore, special attention needs to be paid to the differences between the conditions of real cases and of the model presented.

The model procedure is mainly based on the properties of the by-product. In particular, the risk factors involved in the handling and the typical use of the by-product are emphasised. The material

Table 1. Examples of work stages in the utilisation of by-products in earth construction and possible exposure pathways related to these.

Work stage	Exposure pathway (condition)	Target group for exposure and likelihood for occurrence
Transportation of by-product	Dust can be spread to the environment if the by-products are transported in open vehicles and if the moistening of the powdered by-product has not been sufficient.	Dust emission may cause exposure to the lungs. Furthermore, dermal irritation and ingestion via the mouth are possible. Detrimental compounds in the dust particles may leach out to the soil. The risks are dependent on the amount of dust and the place. The risks are casual and local.
Temporary storage (heap)	In the case of a powder-like by-product, which is stored in a heap without a covering, dust emissions to the environment are likely to occur. Detrimental compounds may leach out from the heap if the by-product is not covered with a water impermeable by-product.	Dust emission may cause exposure to the lungs. Furthermore, dermal contacts and ingestion via the mouth (small children) are possible. Detrimental compounds in the dust particles may leach out to the soil. The risks are dependent on the amount of dust and the place. The risks are casual and local.
Earth construction	During the construction work dust emission may occur, especially in the case of powder-like by-products. Also leaching from the construction needs to be evaluated if the construction is not covered with asphalt or other by-products for a long time.	Workers may be exposed to dust inhalation and dermal contact. If the construction is situated in a residential area, the inhabitants of the area may also be affected by dust and ingestion. The risks are usually occasional.
Use of road	In the case of a non-covered by-product, rainwater percolates into the construction layer. In the case of an asphalt construction, the water might find its way through cracks. In both cases detrimental compounds may also leach out to the environment.	Humans and animals may use the surface or ground water as drinking water near the construction. The background levels in soil and water increase. The increase in the soil near the deposit may have effects on the plant and animal species.
Maintenance and remedial actions at the road/construction	For the maintenance and the remedial actions of the construction, the asphalt layer is often removed and the earth layers are dug up. These activities may lead to dust emission, and the leaching of detrimental compounds to the environment is enhanced.	Humans and animals use the water as drinking water. The background level in soil and water is increased. Workers are also exposed to possible dust emission.
Time after the road/ construction has been used	The road constructions are usually not removed even if the road is taken out of use. In some cases the road is broadened or reconstructed in such a way that the old road is left beneath the new. Only seldom, is the road construction demolished and the by-products removed. This means that the leaching of detrimental components continues.	Humans and animals use the water as drinking water. The background level in soil and water is increased.
Accidents	Accidents need to be considered, especially for transportation. The by-product may be spread into the surroundings causing dust emissions and leaching of detrimental compounds.	People passing near the accident site are exposed to dust. Also skin contacts or ingestion is possible. The risk is usually very local.

properties are also important for the site-specific risk assessment where the site conditions (e.g. geology and hydrogeology) are considered. The sensitivity of the environment and the future use of the site are especially evaluated in an extensive risk assessment.

It is not possible to give general recommendations on how specific environmental conditions should be included in a site-specific risk assessment, and therefore a case-by-case approach is recommended. Only lists of general aspects to be considered can be given.

The procedure is developed for inorganic by-products mainly containing inorganic contaminants; that is metals and salts that influence the behaviour of the metals. The environmental assessment of organic pollutants needs to be studied separately taking into account their special properties (degradation, colloid formation).

Procedure

The risk assessment of the by-product includes the following parts:

1. Description of material properties (origin, processes, raw materials, variations in quality).

2. Identification of detrimental properties during the whole life cycle of the by-product including the most important handling steps.
3. Evaluation of the mobility of contaminants at the site, including the storage of the by-product when necessary.
4. Evaluation of the transportation of contaminants into the environment (surface and groundwater, soil).
5. Calculation of exposure (doses) for critical target groups.
6. Evaluation of the results of the risk assessment based on the material properties including presentation of the acceptable risk level used.
7. Uncertainty analysis.
8. Site-specific risk assessment (when necessary).
9. Conclusions and recommendations.

Identification of properties detrimental to humans and environment

The first step is to collect all the relevant background information of the material (usually through the material's origin and chemical analysis) in order to be able to identify the detrimental properties. The content of harmful components is usually compared to the trigger and limit values given for the soils in the assessment of soil contamination. If the concentration of the harmful component is below the trigger value for clean soil, no further assessment is usually needed. On the other hand, if the concentration of some harmful component significantly exceeds the limit values for contaminated soils or exceeds the maximum leaching values, a risk assessment is needed.

Transportation of detrimental compounds to the environment

Usually the leaching of detrimental compounds needs to be considered in the risk assessment. This means that the mobility of the compounds from the by-product layer and also the factors controlling the leaching need to be assessed usually through a full characterisation of the leaching behaviour. Suitable test methods for the assessment are now under preparation in the standardisation organisation of CEN (CEN TC 292). The leaching is either governed by percolation of rainwater (in the case of a non-isolated construction), or through diffusion (in the case of an isolated construction). The leaching rate as a function of time can be modelled by mathematical equations. The proposed time-scale to consider in the model is 50 years, which is the lifetime of a road. During this period, the leached amount during some time intervals of 1, 3, 10 and 50 year is calculated. The magnitude of the water flow in a non-isolated construction is an important parameter and it can be estimated to vary between 70 and 300 mm/year.

The transportation of contaminants in the water to the surroundings of the site from the earth construction containing the by-product can be modelled in many ways. The simplest way is to calculate with dilution factors. Dilution factors of between 1 and 4000 have been suggested in the model.

Estimation of exposure

The most relevant exposure pathway for many powder-like by-products is via inhalation of dust. Unfortunately, suitable models were not found. For this reason, field measurements of dust emissions during different working stages are helpful.

The exposure pathways (e.g. dermal contact, leaching) are evaluated for human, plant, and animal species during the handling of the by-product, and the construction and maintenance of the earth construction (see Table 1). The doses to humans from other exposure pathways are calculated using simple mathematical models developed for the risk assessment of the contaminated site. Several models are available with different kinds of assumptions for the exposure. Usually, intake through e.g. drinking water, and in the case of playing children, direct inhalation of the by-products is estimated. The calculated intake values can be compared to tolerable daily intakes (TDI). In the assessment, the background exposure also needs to be considered, and in some cases other sources may be significant.

The direct contact (intake by animals or uptake by plants) of the by-product can be ignored because the earth constructions are usually covered and compacted. The environmental risk is due to harmful compounds in the leachates from the construction.

Evaluation of risks

The significance of the emissions from the considered scenario is estimated through comparisons to guideline values that indicate minimal risks for human beings and indicator organisms The assessment of ecotoxicological risks can only be made roughly, because only limited suitable reference values for no-observed effect concentrations (NOECs) are available. Moreover, the values are usually derived for certain laboratory conditions and certain chemical compounds. The applicability of these values may be poor, and therefore they should be used with caution. The ecological effects on plant and animal species are evaluated by comparing the calculated values with the toxicological values reported in literature.

The environmental risks can also be evaluated by comparing the calculated values in the leachate to the background concentrations or ecotoxicological target values, when available. This gives information on the need for dilution to reach the background concentrations in the environment.

In the risk assessment, the exposure of several different pathways (e.g. human exposure to dust and one or several heavy metals) should also be considered. However, today no guidelines are available for assessing the synergetic effect of several exposure pathways.

Uncertainty analysis

An important part of the risk assessment is to evaluate the impact of critical parameters on the results. Variations in input data and checking the influence of critical assumptions give important information on the sensitivity of the results.

Examples of typical parameters to be checked are:

- thickness of material
- chemical composition and leaching behaviour
- assumptions in the calculation of water flow (rate, percolation, surface wash)
- dilution factor for leachate from the earth construction.

Sit- specific risk assessment

Site conditions are taken into account in a more extensive risk assessment. The geology and hydrogeologial conditions of the site are especially considered. The risk assessment may include leachate handling, permeability of surrounding material and climatic and biological conditions. The background levels of detrimental compounds can also be considered in certain cases (e.g. in urban areas). The future use and the sensitivity of the site are important in the risk assessment.

Conclusions and documentation

The results of the material-specific risk assessment give information about which risks are most relevant. This information can be used to classify by-products into different classes (no risks – some risks in the earth utilisation). The risk assessment gives valuable information on the appropriate ways of utilisation in which all risks can be minimised. In the site-specific risk assessment, which is more extensive, especially the sensitivity of the site (hydrogeology, geology) and its future use are considered.

It is important that all the steps and assumptions in the risk assessment are well documented and transparent. The conclusions and the chosen acceptance criteria need to be carefully explained. Usually the risk assessment involves a broad expert group.

A CASE STUDY

The developed model has been tested for coal fly ash in earth construction. Two construction-types consisting of a one-metre material layer were studied: a construction isolated by asphalt and a construction covered by soil.

For the most relevant properties to be considered, dust was chosen as well as the following contaminants: chloride, sulphate, chrome, molybdenum and vanadium. The potential exposure routes and target groups for exposure were evaluated for typical handling procedures during the whole lifecycle of the by-product. Also the consequences of possible accidents were checked.

In the risk assessment it was concluded that dust emission might cause problems especially for workers if the fly ash is not moistened and handled according to the instructions given by the power plant. The most relevant risk factor was the mobility of contaminants into rainwater and transportation of the contaminant with water to the environment.

The assumptions of some parameters in the scenarios are presented in Table 2. An example of the estimated concentrations in the leachate before dilution, which presents the most critical scenario, is illustrated in Fig. 1. The predicted values have been compared to the Finnish background values, guideline values or reference values reported in the literature. In particular, no-observed effect concentrations (NOECs) for water organisms were used when possible. Only a few ecotoxicological values have been presented in the literature. Most values are set for the protection of human health.

The conclusions of the case study were the following:

- For the isolated construction: The concentrations of contaminants in the leachate from the construction were generally near the background concentrations of the Finnish environment or near the guideline values for human health. Because the leachate amounts were small, it could be concluded that the environmental impacts on surface and groundwater can be regarded as insignificant. This means, of course, that the isolation layer is without cracks during the lifetime of the construction.
- For the covered but non-isolated construction: The concentrations of contaminants may cause some kind of risk to the environment. The contaminant concentrations in the leachate were momentarily high and clearly exceeded the guideline values, e.g. the drinking-water values given for the protection of human health. A site-specific risk assessment is needed for the evaluation of impacts. Exclusion of the non-isolated construction in sensitive areas is recommended.

Table 2. Examples of input data in the scenarios considered in the case study.

	Non-isolated construction	Isolated construction
Properties of by-product		
Density	2000 kg/m^3	2000 kg/m^3
Porosity of the by-product	0,45	0,45
Content of some elements		
– Cr	120 mg/kg	120 mg/kg
– Mo	6,4 mg/kg	6,4 mg/kg
– V	200 mg/kg	200 mg/kg
Road dimensions:		
Width	10 m	10 m
By-product thickness layer	1 m	1 m
Leaching mechanism	Percolation of rainwater (300 mm/a)	Diffusion (D_e 2 10^{-9}m/s)
Leaching characteristics:	Results from column test at L/S 10	Pore water concentrations:
– Cr	11.6 mg/kg	150 mg/l
– Mo	3.3 mg/kg	15 mg/l
– V	4.9 mg/kg	49 mg/l

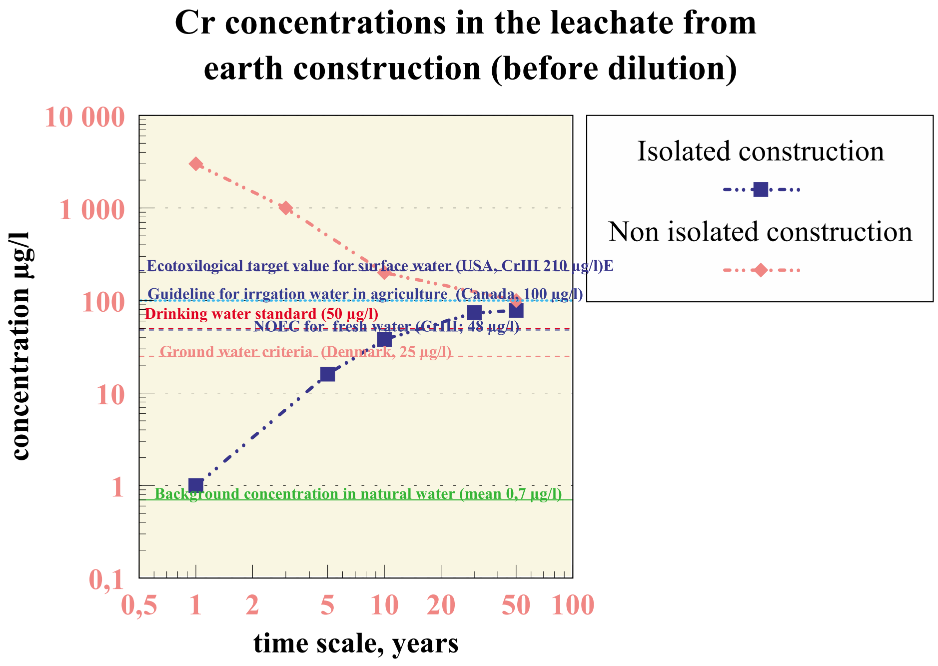

Figure 1. Estimated Cr concentrations in the leachate from two earth constructions during the lifetime of the road. The values can be compared to the following reference values: drinking water standard 50 *m*g/l, ecotoxicological target value for surface water 210 *m*g/l (USA), Canadian guideline for irrigation water in agriculture 100 *m*g/l, Danish ground water criteria 50 *m*g/l and Finnish background concentration in natural water 0,7 *m*g/l (mean).

CONCLUSIONS

The aim of the risk characterisation and assessment is to find an environmentally safe way to use by-products in earth constructions. The risk assessment is needed for by-products possessing properties that fall outside the general guidelines for by-products in earth constructions.

Documentation of the input data and assumptions is important for the evaluation of the results, especially if supplementary information should later become available, e.g. from field studies. The report of the work should be as transparent as possible.

The conclusions should at least contain the following information:

- Which risk factors, exposure routes, target groups have been considered
- Time frame of the evaluation
- Which risk level is assumed to be acceptable (motivations)
- Which environmental conditions (e.g. dilution of leachate) are included in the study
- Limitations of the study
- Assessment of the reliability of the input data and reference values (suitability for comparison)
- Expertise record of the experts.

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Edited by Reijo Salminen.
Geological Survey of Finland, Special Paper 32, 65–74, 2001.

TESTING OF LONG-TERM GEOTECHNICAL PARAMETERS OF BY-PRODUCTS IN THE LABORATORY

by

M. A. Juvankoski, R. J. Laaksonen and M. J. Tammirinne

Juvankoski, M. A., Laaksonen, R. J. & Tammirinne, M. J. 2001. Testing of long-term geotechnical parameters of by-products in the laboratory. *Geological Survey of Finland, Special Paper 32, 65–74*, seven figures and 2 tables.

The properties of four by-products were determined in this research project. These by-products were cement stabilised fly ash, blast-furnace slag sand, a mixture of fly ash and de-sulphuration product and cement stabilised wood or peat ash. The properties considered critical in relation to long-term stability were determined in the laboratory. The results obtained were compared to the requirements set for by-products used in road construction. One of these tested materials has also been used in a test section built before these tests were started.

All the materials tested here are in principle capable of producing adequate compressive strength and stiffness to be used in road construction. In practice some problems may arise particularly concerning the choice of parameters for design due to poor environmental durability. In other words, stabilised or self-strengthening materials do not necessarily last intact after repeated freeze-thaw cycles. Although the stiffness of stabilised material decreases significantly due to structural failure, it may still possess reasonably large stiffness to be of use in the sub-base of a road structure.

Keywords: construction materials, by-products, ash, slag, engineering properties, durability, laboratory studies, road tests, Finland

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INTRODUCTION

The properties of four by-products were determined in this research project. These by-products were cement stabilised fly ash, blast-furnace slag sand, a mixture of fly ash and desulphuration product and cement stabilised wood or peat ash. The properties considered critical in relation to long-term stability were determined in the laboratory. The results obtained were compared against the requirements set on by-products used in road construction. One of these tested materials has also been used in a test section built before these tests were started. This research project has been one project in the Tekes Ecogeotechnology Research &Development program.

TEST MATERIALS

The tested materials were cement stabilised fly ash from Fortum's Meri-Pori power plant, SKJ's blast-furnace slag sand, a mixture of fly ash and desulphuration product from Helsinki Energy and cement stabilised wood or peat ash from UPM-Kymmene's Kaipola factory. Blast-furnace slag sand was different from the other materials studied in that it was granular unbound material. The properties and behaviour of slag sand are well known and it has been used as a reference material for the test methods in this study. The test results obtained with slag sand are not presented in this article. Cement stabilised fly ash from Fortum's Meri-Pori Power plant has been used in a test road section in the Pori area built under the test construction program of the Ecogeotechnology R&D program.

A typical gradation curve of fly ash of Fortum's Meri-Pori Power plant is presented in Fig. 1. This fly ash was used in the sub-base layer of the Pori test road. Also shown in Fig. 1 is the typical gradation curve of the bottom ash, which was used as one component in the base layer of the test road. Typical gradation curves for fly ash from Helsinki Energy are shown in Fig. 2. Typical gradation curves for peat ash created by burning mixed peat, wood and waste wood at the UPM Kaipola factory are shown in Fig. 3. The mix ratios, quantities of additives and optimum water content together with maximum dry densities determined for mixtures are presented in Table 1.

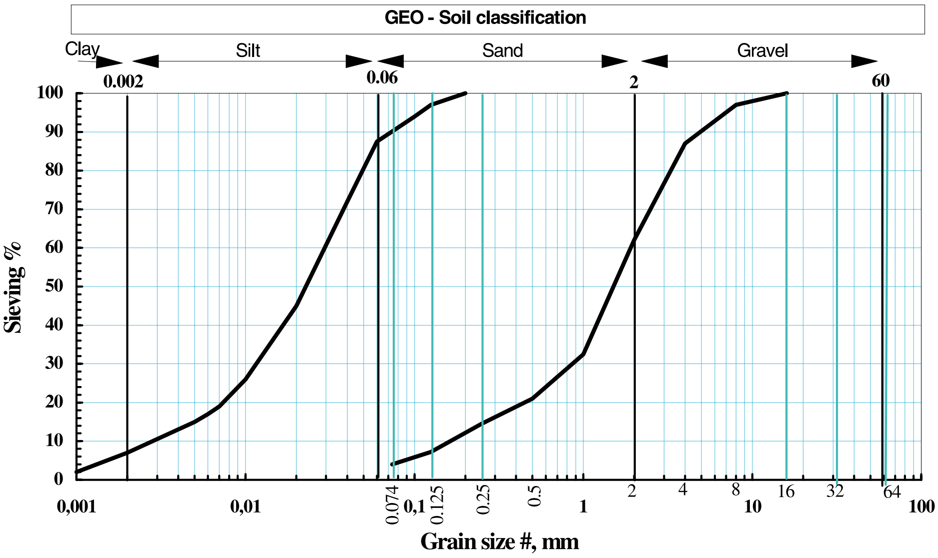

Figure 1. Typical gradations of fly ash (finer) and bottom ash (coarser) of Fortum's Meri-Pori power plant.

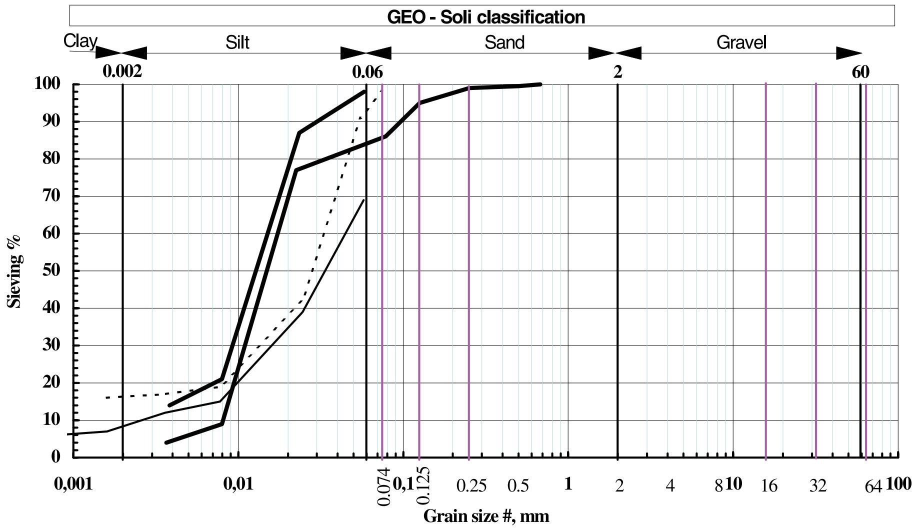

Figure 2. Typical gradation curves for fly ash from Helsinki Energy. The gradation of dry fly ash (thick lines, samples taken 26.3.97–7.5.98), desulphuration product heaped 30 days out of doors (broken line) and fly ash heaped 180 days out of doors (thin line).

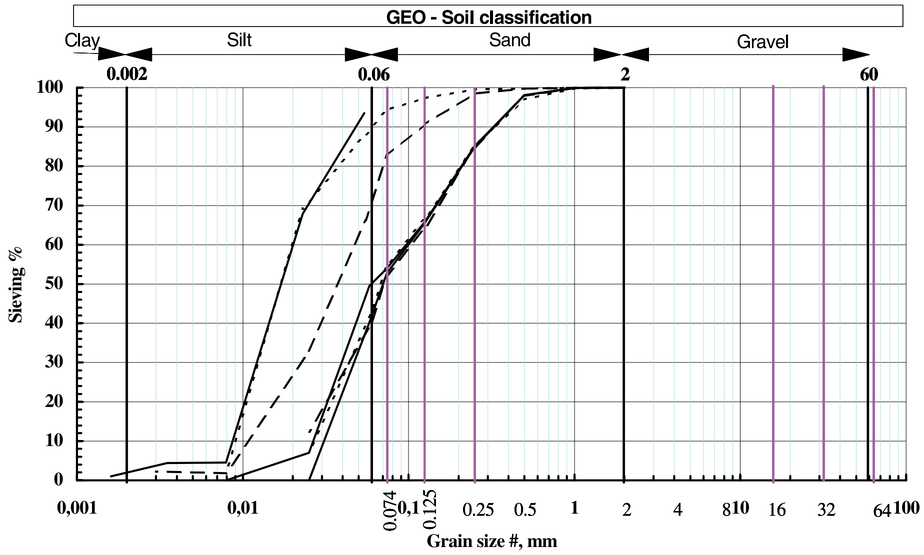

Figure 3. Gradations of Kaipola wood ash. The finer samples are dry ash and the coarser (four lines nearly on top of each other) heaped ash.

Table 1. Materials, mix ratios and basic properties.

Property	Stabilised fly ash	Fly ash/ desulphuration product	Stabilised peat ash
MIX RATIO OR CEMENT CONTENT, %	6 %	50 % / 50 %	4 %
w_{opt}, %	25.0	25.0	36.2
$\gamma_{d\ max}$, kN/m^3	12.9	13.3	11.6
Design relative density, D, %	90	95	92

TESTING PROGRAM AND TEST RESULTS

The testing program included determination of the dynamic stiffness modulus, compressive and tensile strength, thermal conductivity and permeability together with freeze thaw durability and frost heave parameters. In addition, a specific forced bending test with beam specimens (h * b * l : 100mm*100mm*500mm) was carried out.

The test specimens used in the tests were compacted under the optimum water content specific to each material to the desired densities. These densities are presented in Table 1. The stabilised fly ash specimens were manufactured to correspond to the density obtained in test road. The specimens of other materials were manufactured to correspond to the designed use of the material. The specimens were usually tested at the age of 28, 90 and 180 days.

The test results are presented in Table 2. Table 2 contains also the requirements submitted for materials used in the upper part of the road section. The requirements submitted for by-products with their complete justifications are presented in reference 2.

Table 2. Test results for each material and requirements in the upper layer of road section.

Property, parameter	Stabilised fly ash	Fly ash/desulphuration product	Stabilised peat ash	Requirement
Stiffness modulus, MPa				> 300
(water content at compaction/saturated)				
28 d	1400/2100	1100/700	4800/ 3300	
90 d	-/-	1200/2700	2300/ 3700	
180 d	2300/2300	2300/1000	-/-	
Compressive strength, MPa				> 1.5
(wc at compaction/ saturated)				
7 d	1.5/-	1.7/1.1	3.1/ .0	
28 d	2.2/2.5	2.6/2.0	5.4/4.8	
90 d	3.2/-	3.5/3.1	8.5/9.6	
180 d	3.5/2.4	4.8/2.9	-/-	
Tensile strength, kPa	290	247	715	-
Freeze-thaw durability,				≥ 12
cycles (end cycles)	3	2	12 (30)	
Frost heave parameters				< 0.5
SP (2 kPa), mm^2/Kh	0.9...1.1	0.05	0.0	
Water permeability, m/s	$2.0*10^{-7}$	$1.1*10^{-8}$	$1.2*10^{-9}$	> $3*10^{-7}$
Bending capacity,				R_{longt} < 300
R_{min}, m	94	108	84	R_{shortt} < 150
Thermal conductivity,				< 1/1
W/Km, recommendation	0.69/0.76	0.55/0.95	0.57/0.89	
(frozen/thawed)				

REVIEW OF THE RESULTS

All materials fulfilled the compressive strength and dynamic stiffness requirements at the age of 28 days, both under optimum water content and saturated state submitted for the upper part of the road structure.

Also the beam specimens of all materials sustained the required long-term and short-term bending test. On the other hand, none of the materials (excluding the slag sand) fulfilled the required permeability value or criteria.

Stabilised fly ash

The value of the frost heave parameter, the segregation potential (SP), did not fulfil the requirement submitted for the upper part of the road section. The requirement submitted for the lower part (SP < 1.5 mm^2/Kh, depth > 1.0 m) was on the other hand fulfilled.

The specimens did not survive the freeze-thaw test due to specimen failure after the third cycle. Also the permeability value determined failed to meet the required value.

Mixture of fly ash and desulphuration product

The value of the segregation potential determined met and exceeded the required value for the upper part of the road section, but the specimens failed to pass the freeze-thaw test due to specimen failure after the second cycle. The permeability value determined failed to meet the required value.

Cement stabilised peat ash

The value of segregation potential determined met and exceeded the required value for upper part of the road section. The specimens also passed the freeze-thaw test, which was carried out until 30 cycles were completed

THE ESTIMATION OF LONG-TERM DURABILITY OF MATERIALS

Stabilised fly ash

According to the test results, the cement stabilised dry silo fly ash is capable of developing reasonable compressive strength and high stiffness values. This leads to good structural bearing capacity and better thermal insulation properties than can be found with natural materials. Saturation was found to decrease the compressive strength and static stiffness modulus value.

The stabilised fly ash appeared to be mildly susceptible to frost in the frost heave test made according to the test program and it also failed to pass the freeze-thaw test simulating the climatic effects. Based on these results, the long-term stability of the cement-stabilised fly ash in question appears to be poor. Because of this the properties do not remain constant but they may change considerably when the structure fails. This result means that it is not possible in the pavement design to use the high stiffness level measured (of the order of 2400 MPa). One must estimate the structural modulus value level, which can be reached for the failed material. The modulus value of the layer of failed bound material may still be of the same order as in similar layers built with natural materials.

The properties of bound materials are always strongly dependent on the internal alteration of quality of the product. The test specimens were compacted to the density obtained in the test road. Before the test road was constructed a set of preliminary laboratory tests were carried out. In these tests material did not possess frost heave problems (larger degree of density) and it passed the freeze thaw tests, although it lost some of the compressive strength. The properties of fly ash studied during the test program and during the preliminary tests differed to some extent also in relation to compactability properties.

Mixture of fly ash and desulphuration product

The mixture of fly ash and desulphuration product was behaving almost like the cement stabilised fly ash. The mixture was found to be non-frost susceptible but it failed to pass the freeze thaw test simulating the environmental impact/stress.

Cement stabilised peat ash

Cement stabilised dry peat ash can produce very high compressive strength and a good stiffness value. At the age of three months the level of compressive strength is 8 to 9 MPa and the value of dynamic stiffness modulus varies between 2000 and 4000 MPa. The effect of saturation on these properties is small and inconsistent. The material is non-frost susceptible and it passed the freeze thaw test simulating the environmental impact/stress. Based on the test results one can estimate that the long-term durability and also the bearing capacity stability are reasonably good. The thermal insulation properties of the material are better than those of natural materials.

It remains a slight problem to foresee whether the behaviour of material is due to quality variations (proportion of wood and peat) and at least in this case the extreme reactivity of used peat ash. Because of the latter property the ash behaved as a binder. At most, the ratio of stiffness values of comparable specimens was up to 7. This strong strengthening property was not observed during the earlier studies. If one wants to make use of the beneficial properties of this material one must check and verify the properties of the material and construction process with dry material on a case specific basis. Even a small amount of moisture beforehand reflects strongly on the strengthening of the material and a long-term wetting alters also the other material properties to a large extent. The permanent strengthening property disappears and the peat ash may become frost susceptible.

EXPERIENCE GAINED FROM THE FLY ASH TEST ROAD

The stabilised fly ash test road (Mt 272 Ämttöö-Poikeljärvi) has been closely monitored since the construction phase. The subject was one of the test road structures constructed under the test construction programme of Tekes's Ecogeotechnology R&D program and it is currently being monitored under the TPPT test road project. The results obtained since spring 2000, which are presented later in this document, are based mainly on this ongoing monitoring work (Tammirinne *et al.* 2000). This monitoring has included sampling, surveying, measuring of evenness (IRI) and bearing capacity determinations.

Cement stabilised (6 %) fly ash from Fortum's Meri-Pori power plant was used in the sub-base layer of this test road. Based on preliminary tests the stiffness modulus value of this stabilised fly ash in design was 400 MPa. The relative density was planned to be D = 92 %. The amount of binder was chosen to produce a compressive strength of 3 MPa at the age of 28 days to limit the effect of freezing on the material. According to the preliminary tests, the fly ash was non-frost susceptible. The bearing capacity requirement for the final fly ash structure was 416 Mpa, and the bearing capacity requirement for the reference (traditional structure) structure was 263 MPa.

The densities required in design were not fully achieved. The mean relative density of the sub-base layer was ca. 90 %. One reason for the lower density was probably the variation in fly ash properties during the construction phase, compared to the fly ash used in preliminary tests. The maximum dry density of fly ash varied between 12.8 and 15.8 kN/m^3 and optimum water content between 17.2 and 28.2 % during the construction. The variation in compactability properties of fly ash may, especially in lower relative densities, reflect other geotechnical properties of the material. The optimum water content of the material used in the preliminary tests was 15 % and the compacted specimens (D = 92 %) were non-frost susceptible. In the laboratory tests made after the construction (i.e. in this project) the optimum water content of fly ash was 25 % and relative density D = 90 %. In this case the material was slightly frost susceptible (SP = 1 mm^2/Kh).

In spite of a lower relative density than planned, the bearing capacity measured on top of the fly ash layer in-situ gave modulus values between 700 and 800 MPa. Samples taken in the autumn before the freezing season gave a compressive strength of 3.1 MPa.

Surveying

During the winter of 1997–98 frost heave values of 40 to 50 mm were monitored on the fly ash sections. These measured values corresponded well to the calculated frost heave value of ca. 40 mm determined based on winter's freezing index (9500 h°C) and laboratory and in-situ parameter values. The calculated frost depth (61 cm) corresponded reasonably well to the measured values (39 to 49 cm) from the road shoulder.

During the winter of 1998–99 (freezing index F = 14789 h°C) the maximum frost penetration of 1.09 m was reached in April 1999, at which time the frost had penetrated through both the stabilised fly ash layer (500 mm) and underlying filter sand layer (300 mm) into the subgrade. The frost penetration depths varied between 1.36 and 2.00 m in the reference structure.

The maximum frost heave of the fly ash structure varied between 28 and 88 mm. The largest frost heave (88 mm) was measured at pile number 750 on the right shoulder in March 1999. At that time the frost had just penetrated through the fly ash layer, which means that the frost heave measured is due to frost heave of the stabilised fly ash itself. The frost heave has been unevenly distributed in the section and the road shoulders have larger frost

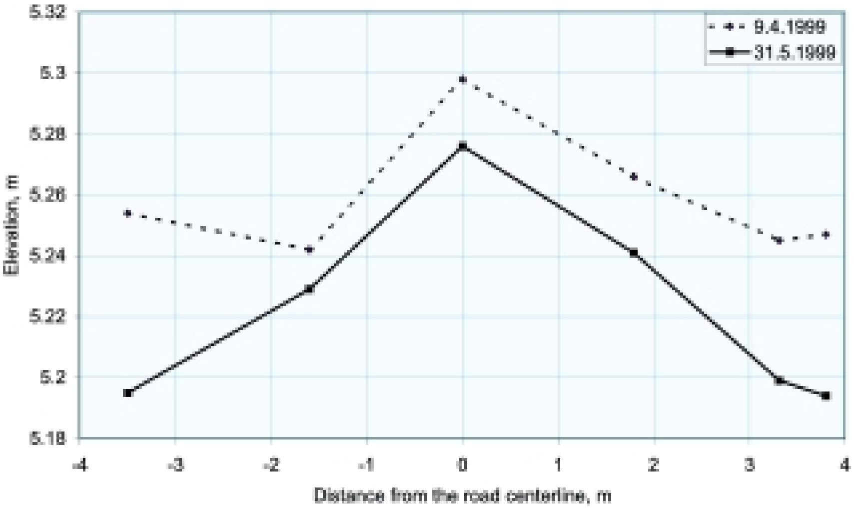

Figure 4. Cross-section surveying at pile number 750 during the maximum frost heave period in spring 1999 (9.4.1999) and early summer 1999 (31.5.1999).

heave than the central part of the road. The frost heave values of the reference structure varied between –10 and 11 mm.

The survey results at pile number 750 are shown in Fig. 4. From the cross-section one can deduce that the largest frost heave values appear to occur at the shoulders. The smallest frost heave values appear within 1.5 to 2 metres of the centre line, partly on both sides, where they form depressions collecting water. Pooling due to uneven frost heave of the stabilised fly ash is shown in Fig. 5.

In-situ samples

A second set of samples was bored in autumn 1998 at the same pile number as in autumn 1997. The age of the material was ca. 14 months when the samples were taken. Visual inspection revealed plenty of cracks and disintegration of stabilised material. The most intact specimens were chosen for laboratory tests.

Compressive strength, density and water content in in-situ condition were determined from these samples. Between the samplings (ca. 9 months), the

Figure 5. Pooling due to uneven frost heave of fly ash structure in spring 1999.

compressive strength of the intact parts of sub-base layer decreased from 3.1 MPa down to 2.1 MPa. The in-situ water content also increased by 4 to 8 % -units.

The third set of samples were bored, and this time from the frozen layer, in March 1999. The samples were taken at pile numbers 460 and 900 from the right lane. The lengths of these samples varied between 25 and 110 mm.

Water contents were determined for all the samples and the compressive strength from two specimens (the only ones long enough). In addition to that, thaw settlement and frost heave tests were carried out with the longest specimen. The compressive strength had decreased down to 25 to 50 % of the value determined in autumn 1997. The average values of compressive strength as a function of time are shown in Fig. 6, together with the average compressive strengths determined in the laboratory.

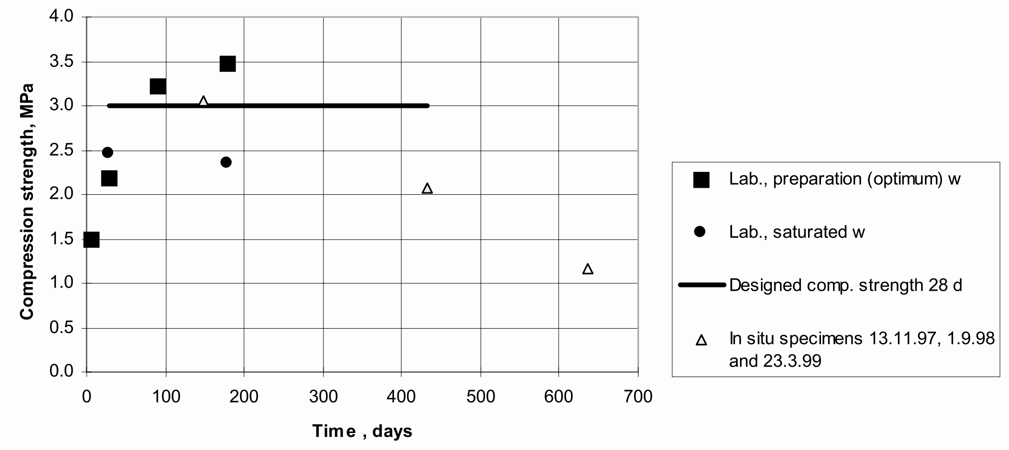

Figure 6. The compressive strength (average values) of specimen taken from test road at different times and the compressive strength (average values) determined at different ages in the laboratory.

A thaw settlement test was carried out with a specimen taken from pile number 460, which had been taken frozen from the depth of 0.32–0.43 m. This depth corresponds to the upper part of the sub-base layer. The settlement of the specimen was ca. 2 %. After the thaw settlement test a normal frost heave test was carried out, which gave the segregation potential of SP = 0.5 mm^2/Kh for the fly ash specimen.

A segregation potential value SP = 1.0 mm^2/Kh was determined earlier for a laboratory compacted stabilised fly ash specimen. Using temperature, frost depth and frost heave values measured during winter 1998–1999 as input, the back-calculated segregation potential value for the stabilised fly ash structure varied between 0.5 and 2.0 mm^2/Kh, depending on the absolute in-situ frost heave. According to the frost susceptibility criterion based on segregation potential SP, the material is slightly frost susceptible if SP varies between 0.5 and 1.5 mm^2/Kh and medium frost susceptible if SP varies between 1.5 and 3.0 mm^2/Kh.

The bearing capacity

In autumn 1998, about one year after the construction, the average bearing capacity level at the road surface of the fly ash structure measured with FWD was 476 MPa. The average bearing capacity of the reference structure was 430 MPa. The average bearing capacities for both structures were in excess of the design value.

In spring (May) 1999 the average bearing capacity of the fly ash test structure was 415 MPa, which corresponded to the design value. The average bearing capacity of the reference structure was 430 MPa. The bearing capacity values for reference sections are well above the design values (263 MPa). The results of these FWD measurements are shown in Fig. 7.

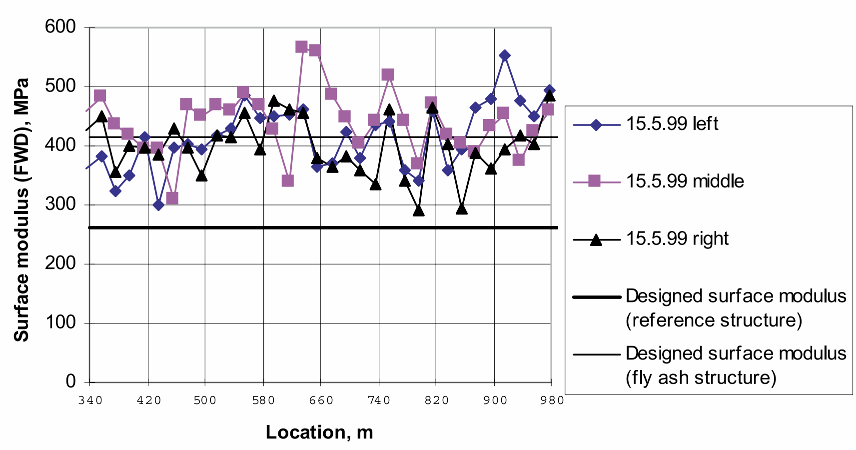

Figure 7. Temperature corrected FWD results, May 1999.

Wearing course damage

The first longitudinal crack was observed after the first winter (1998–1999) between pile numbers 442.4 – 445 on the centre line of the road. The width of the crack was ca. 5 mm. In spring 2000 there were found to be five cracks in separate places along a 70 m length. In addition, there was one 5–10-mm wide crack on the left lane at 2-m distance from the centre line. The frost behaviour during winter 1999–2000 is quite similar to that during the previous winter.

Summary of the test road

As a summary of the behaviour of the fly ash structure one can conclude that the structure as a whole has behaved reasonably well, despite the unevenness and pooling during late springtime. In spite of uneven frost heave, cracking of stabilised fly ash and decreased compressive strength, the fly ash structure still had the design bearing capacity during spring 1999 and the bearing capacity was better than that of the reference structure. Frost heaving, material cracking and decrease in strength probably even down to less than half of the design value had no detrimental effects on the bearing capacity of the stabilised fly ash layer. This badly cracked stabilised fly ash layer behaves similarly to unbound crushed aggregate.

If the modulus value of the other layers are in agreement with design values, then the modulus of stabilised fly ash is also in line with the design value (400 MPa). This value is only 19 to 15 % of the dynamic modulus value determined with saturated intact specimens at the age of 28 and 180 days. In previous studies with cement stabilised glacial till it has been found that the average dynamic modulus values of broken specimens are ca. 16 to 17 % of that of corresponding intact specimens. With moist, broken stabilised glacial till the modulus value is ca. 21 % and with saturated and broken specimen ca. 13 % of the modulus value of the corresponding intact specimen.

SUMMARY

All the materials tested are in principle capable of producing adequate compressive strength and stiffness to be used in road construction. In practice, some problems may arise especially concerning the choice of parameters for design due to poor environmental durability. In other words, stabilised or self-strengthening materials do not necessarily last intact in repeated freeze thaw cycles. Although the stiffness of stabilised material decreases significantly due to structural failure, it may still possess a reasonable degree of stiffness to be used as sub-base of a road structure.

The second phenomenon strongly influencing the use of such by-products is the internal variability of properties of the by-products. These variations are mainly due to variations /changes in origin and composition of the fuel used in relatively small power and production plants. This variation in quality causes uncertainty in the properties obtained in the structural layer in two ways. The material used in laboratory test may sometimes differ quite remarkably from the material used in the actual structure. In addition, the variation during construction phase makes it difficult to carry out the compaction control

By-products can be successfully used for road construction as long as special properties of these materials and the precautions governing their use are taken into consideration in testing, planning and construction.

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THE USE OF FLOTATION SAND IN EARTH CONSTRUCTION

by
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Kujala, K., Ahonen M. & Koivisto, H. 2001. The use of flotation sand in earth construction. *Geological Survey of Finland, Special Paper 32, 75–81,* four figures and 5 tables.

Flotation sand, comparable to natural sand in its material properties, is a by-product of calcite flotation. The capillary rise of flotation sand is lower than 90 cm, the material is non-frost-susceptible (SP < 0.5 mm^2/Kh) and its hydraulic conductivity is higher than 10^{-6} m/s. Its friction angle is approximately 44° and therefore a little greater than that of sand with a corresponding grain size. In its technical properties, flotation sand is a suitable material for the filter layer of road construction as well as for filling structures that require a frost resistant material with a high bearing capacity. Refined with mixture materials, such as bentonite, it is also a suitable material as a hydraulic barrier in landfill covers. In addition, flotation sand is greatly strengthened when bound with bark ash, for example, thus fulfilling the quality requirements set for bound materials of road structures. Flotation sand does not contain any materials that are harmful to the environment.

Keywords: construction materials, flotation sand, by-product, engineering properties, earthworks, Finland

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INTRODUCTION

Approximately 70 million tons of natural rock material is used annually in earth construction in Finland. The decline in good quality rock material resources, the need for protection, and the growing distances in transportation have all increased the need for applying substitute materials for sand and gravel. Simultaneously, various fields of industry generate a great number of mineral by-products that can be utilised in earth construction. Also, waste management legislation requires replacing natural materials with waste and by-products. Nevertheless, if these materials are to be utilised in earth construction, certain technical properties and environmental qualifications are required of them. In the initial stages of using recycled material, the material properties have to be examined more comprehensively compared to traditional materials, whose use is based on results achieved during long-term utilisation. Defining long-term behavior often requires test constructions and monitoring of the structures before actual utilisation.

This article deals with the material properties of the by-product of calcite enrichment at the Lappeenranta mine of Partek Nordkalk Oyj Abp, and its suitability as an earth construction material. A total of 100 000 tons of flotation sand is generated annually.

MATERIALS AND METHODS

Materials

The grain size distribution of flotation sand varies in the different sections of the disposal area. The coarse fraction is deposited close to the discharge pipe, and the finer fraction is carried along in water, further away from the pipe. The grain size distribution of the coarser fraction equals that of sand, and the finer component is equivalent to silty sand (Fig. 1). Both materials are even-grained. The average grain size d_{50} of the fine flotation sand is 0.08 mm and of the coarse flotation sand, 0.16 mm.

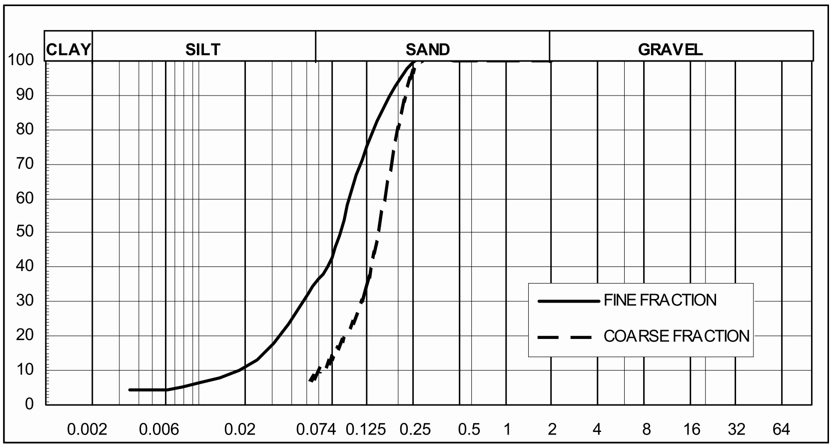

Figure 1. Grain size distribution of flotation sand in the disposal area. Partek Nordkalk Oyj Abp’s Lappeenranta mine.

Bark ash from Enso Oy’s Imatra works was used to strengthen the flotation sand and calcium bentonite was used to decrease their hydraulic conductivity.

Methods

The material properties of flotation sand was determined by methods commonly used in geotechnical research. Only principles of the most essential test methods used in this research are presented in Table 1.

Table 1. Test methods used in flotation sand research.

Property, parameter	Principle of test method
Environmental validity	
*total elemental content	ICP-AES
Hydraulic properties	
*hydraulic conductivity	*flexible-wall perimeter
*capillary rise	*capillarymeter
*water retention capacity	*pressure plate apparatus
Frost	
*segregation potential	*frost heave test
*freeze-thaw durability	*change of length, compressive strength test
*water durability	*water immersion test
Thermal properties	
*thermal conductivity	*thermal needle probe
Mechanical properties	
*shear strength	*triaxial test
*resilient modulus	*dynamic triaxial test
*tensile strength	*bending beam test
*bearing capacity	*CBR-test

Sample preparation

When using a mixture that includes binding materials, cylindrical test samples of the flotation sand were prepared. Water was added to the mass to achieve the optimum water content. The masses were then mixed together mechanically. The process of mixing was continued until the binding material, based on visual examination, was completely blended with the dry aggregate. Compaction of the samples tested (compressive strength test, freeze – thaw test, water immersion test), using uniaxial compression test equipment was performed using a gyrator compactor into a 4” mold. The target degree of density was 95 %. The height of the compacted samples was 100 ±±2 mm. Compaction of the samples tested using dynamic triaxial equipment and bending capacity equipment was performed with a hand rammer. After the test samples were prepared, they were stored in a closed plastic box on top of a steel grating at room temperature. There were 10 centimeters of water at the bottom of the plastic box.

Calcium bentonite and flotation sand were mechanically mixed together to form a material of uniform quality, after which the target amount of water was added. The degree of density of the samples was set either at 90 % or 95 %. The samples were compacted with a slightly higher water content than the optimum water content.

RESULTS AND DISCUSSIONS

Environmental validity

The mineralogical composition of flotation sand is wollastonite, quartz, calcite, and dolomite. The results of elemental analyses of flotation sand are presented in Table 2. The major elements are Ca, Mg and Fe. Because all trace elements are lower than the proposed threshold values set for materials used in earth construction (Assmuth 1997), the material can be used without limit (Table 1). Only the beryllium content (1.3 mg/kg) of the coarse fraction slightly exceeds the lower limit value (1.0 mg/kg), but it is still clearly lower than the upper limit value (10 mg/kg). The beryllium content of the fine fraction is lower than the defining value (< 0.5 mg/kg). It must be noted that the beryllium content is lower than the background content in Finnish soil.

Table 2. Elemental contents of flotation sand. The table also presents the suggested values for concentrations of harmful materials in soil. The results are reported in milligrams per kilogram of dry matter (mg/kg).

	Flotation sand Fine fraction (mg/kg)	Flotation sand Coarse fraction (mg/kg)	Judgement criteria Upper limit value (mg/kg)	(Assmuth 1997) Lower limit value (mg/kg)
Al	2430	5280		
As	<10	<10	13	60
B	<5	5	5	50
Ba	29.4	39.7	600	600
Be	<0.5	1.3	1	10
Ca	233000	205000		
Cd	<0.5	<0.5	0.3	10
Co	<1	1.8	50	200
Cr	3.2	7.2	80	500
Cu	6.6	8.7	32	400
Fe	1850	3910		
K	361	929		
Mg	4020	4000		
Mn	52.6	99.4		
Mo	<3	<3	5	200
Na	466	899		
Ni	<2	4.0	40	300
P	431	358		
Pb	<5	<5	38	300
S	359	830		
Sb	<15	<15	5	40
Sr	296	277		
Ti	184	354		
V	1.1	4.5	50	500
Zn	8.4	13.6	90	700

Material properties of flotation sand

The physical properties of flotation sand is presented in Table 3. Its values of friction angles correspond to the values of sand. The cohesion of the fine fraction flotation sand is greater than that of the coarse fraction flotation sand. According to the CBR value (CBR = 40 %), the resilient modulus of the materials is appr. 185 MPa, calculated with an empirical formula (Table 3).

$Mr=17.6\ xCBR^{0.64}$ (MPa)

Both fractions are non-frost-susceptible (SP<0.5 mm^2/Kh), and their hydraulic conductivities are ($k>10^{-6}$ m/s). Their capillary rise is lower than the values (0.9 m) set for filter layers in the conditions for quality control of the Finnish National Road Administration. The thermal conductivity of flotation sand is equal to that of sand in the same degree of density and water content (Table 3).

Table 3. Material properties of flotation sand determined by laboratory tests.

Property	Coarse fraction	Fine fraction
Specific gravity ρ (t/m^3)	2.89	2.88
PH	8.6	8.6
Maximum dry density $*_{g*dmax}$ kN/m^3	17.6	17.7
Optimum water content w_{opt}	13	14
Capillary rise h_c cm	78	86
Hydraulic conductivity k (m/s)	$10^{-5.0}$	$10^{-5.6}$
Friction angle ϕ	44.7°	43.7°
Cohesion c (kPa)	3	15
CBR-value	42	51
Segregation potential SP_0 (mm^2/Kh)	0	0.4
Maximum frost heave h (mm)	0.5	0.1
Thermal conductivity λ (W/Km)		
– unfrozen (+22°C)	1.17	1.13
– frozen (–15°C)	1.50	1.41

Material properties of bound flotation sand

Compressive strength and modulus of elasticity

After 28 days of curing, the compressive strength of flotation sand bound with bark ash varies between 1.3 and 5.5 MPa, depending on the grain size of the flotation sand and the amount of bark ash (Fig. 2). As the amount of bark ash increases, the compressive strength increases in a linear manner. After the first 28 days the increase is minor. The modulus of elasticity varies between 76 and 221 MPa. The changes in the modulus of elasticity are similar to the changes in compressive strength.

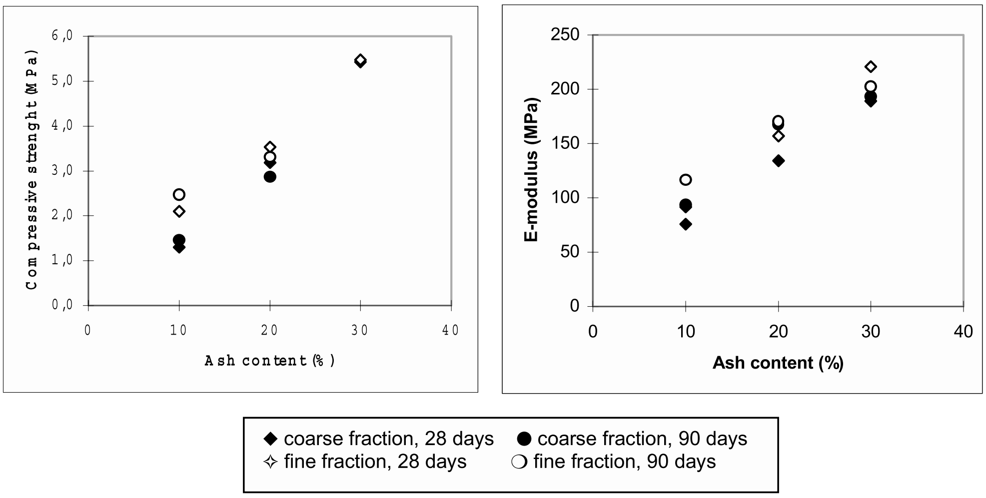

Figure 2. Compressive strength and modulus of elasticity of flotation sand bound with bark ash as a function of time and amount of bark ash.

Resilient modulus

The resilient modulus of flotation sand bound with bark ash increases as a function of the sum of principal stresses, the grain size of the flotation sand, and the amount of bark ash. For example, with the sum of principal stresses of $\theta = 50$ kPa, the resilient modulus varies between 548 and 787 MPa (Fig. 3 and Table 4). The values of the resilient modulus of sand are equal to the values of high quality crushed aggregate.

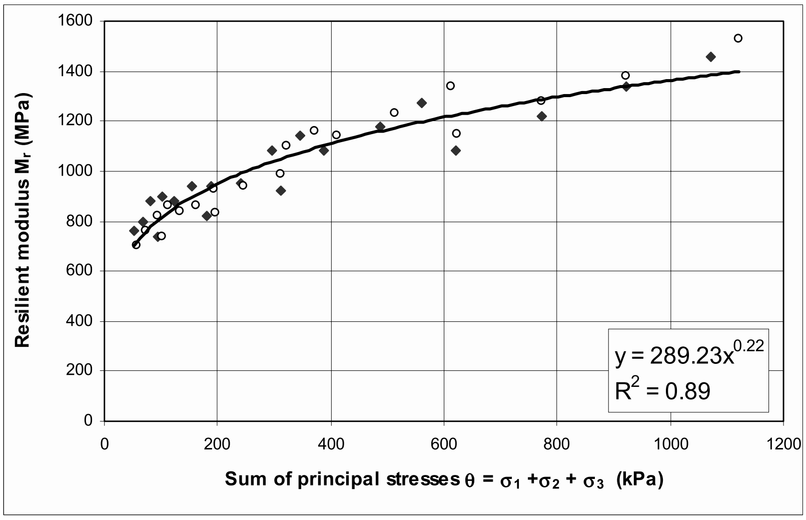

Figure 3. Resilient modulus of flotation sand bound with bark ash as a function of the sum of the principal stresses and the power curve fitted to measurement observations. Curing time 90 days, amount of bark ash 30 %, degree of density D = 95 % ($*_{g*dmax.}$ = 17.6 kN/m^3, $w_{opt.}$ = 14 %).

Table 4. Resilient modulus of flotation sand bound with bark ash as a function of the sum of the principal stresses θ = 50 kPa, θ = 100 kPa and θ = 200 kPa.

Flotation sand + bark ash	R^2	Resilient modulus M_r (MPa) θq = 50 kPa	θq = 100 kPa	θq = 150 kPa
Coarse fraction + 10 % bark ash	0.86	584	660	872
Fine fraction + 10% bark ash	0.91	685	524	623
Coarse fraction + 30 % bark ash	0.85	651	685	734
Fine fraction + 30 % bark ash	0.94	787	540	728

Freeze-thaw durability and water immersion

The compressive strength of flotation sand bound with bark ash does not decrease significantly after freeze-thaw cycles or after immersion in water (Fig. 4). In the samples that were visually examined, there were no signs of ruptures or brittle fractures after freeze-thaw tests and water immersion tests. Also, changes in length were minor in these tests. The test samples absorbed only <10 % water during the tests.

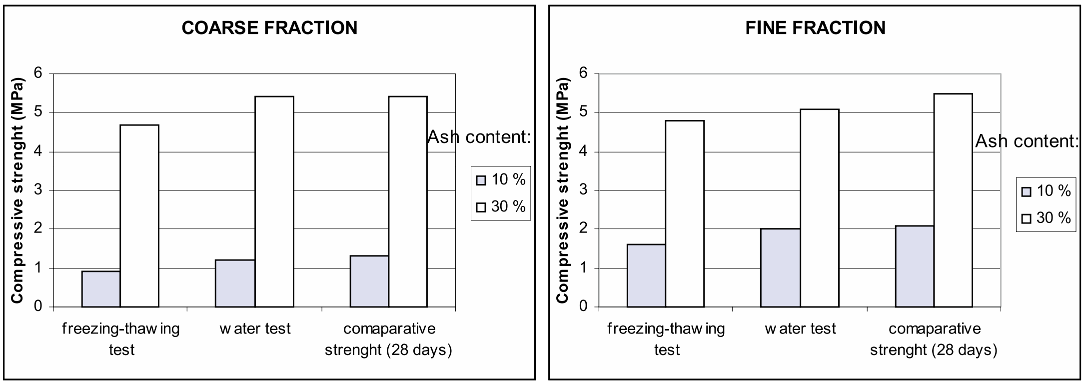

Figure 4. Compressive strength of the test samples after freeze-thaw tests and a water immersion test. The figure presents the compressive strength of the samples without freeze-thaw cycles. Curing time (28 days).

Material properties of flotation sand and bentonite mixture

The frost heave test was repeated as four freeze-thaw cycles. The segregation potential was defined after the first cycle. After the frost heave tests, the hydraulic conductivity of the samples was measured using a flexible-wall permeameter.

The flotation sand was found to be non-frost-susceptible, excluding the material fine fraction flotation sand + 4 % bentonite, which was low frost-susceptible (Table 5). After the four freeze-thaw cycles that were also frost heave tests, the hydraulic conductivity of each samples was < $10^{-9.0}$ m/s.

Table 5. Frost susceptibility and hydraulic conductivity of the mixture of flotation sand and bentonite after frost heave tests.

Material	Number cycles	Degree of density [%]	Water content [%]	Segregation potential [mm²/Kh]	Classification of frost susceptibility	Hydraulic conductivity [m/s]
Fine fraction + 6 % bentonite	4	94.9	16.2	0	Non-frost-susceptible	$10^{-9.0}$
Fine fraction + 6 % bentonite	4	89.8	16.2	0	Non-frost-susceptible	$10^{-9.0}$
Fine fraction + 4 % bentonite	4	94.7	16.1	1.0	Low-frost-susceptible	$10^{-9.1}$
Coarse fraction + 6 % bentonite	4	94.7	16.2	0	Non-frost-susceptible	$10^{-9.4}$
Coarse fraction + 6 % bentonite	4	89.7	16.2	0	Non-frost-susceptible	$10^{-9.9}$
Coarse fraction + 4 % bentonite	4	94.7	16.0	0	Non-frost-susceptible	$10^{-9.0}$

Utilization of flotation sand in landfill structures

The suitability of flotation sand as a filling material for landfill cover was ascertained in a test structure carried out at the factory landfill site of Partek Nordkalk Oy. The test structure was built particularly to study the feasibility of using flotation sand as the hydraulic barrier layer of landfill when its additive materials were bentonite and other commercial additives. Clay was used as a reference material in the hydraulic barrier. Flotation sand was also used on the surface of the waste layer to smooth out inequalities. The follow-up measurements of the test structures includes the amount and quality of the water infiltrating through the cover, the settlement, temperature and volumetric water content. The details of the results are beyond the scope of this paper.

SUMMARY AND CONCLUSION

The concentrations of harmful elements in flotation sand are very low and below limit values used in the evaluation of environmental suitability. Therefore, the material can be used without limit in earth construction.

The grain size distribution of flotation sand is even-grained and equals that of silty sand and sand. The capillary rise is lower than 90 cm, the material is non-frost-susceptible (SP<0.5 mm^2/Kh), and its hydraulic conductivity is greater than 10^{-6} m/s. Its friction angle is approximately 44° and therefore a little greater than that of sand with a corresponding grain size. According to the CBR value, the load-bearing capacity of flotation sand is as good as the material of a good base course. The coarse fraction of flotation sand fulfils the requirements set for a material used in a filter layer in the guidelines for quality control of the Finnish National Road Administration.

Flotation sand bound with bark ash fulfil the quality requirements for the material for the base course layer in road structures, among others.

The hydraulic conductivity of flotation sand can be effectively reduced with bentonite. With a bentonite content of approximately 6 %, the hydraulic conductivity is $>10^{-9}$ m/s, which fulfils the hydraulic barrier requirements set for landfill cover structures, for example. The mixture of bentonite and flotation sand is non-frost-susceptible and at the most, low frost-susceptible. Freeze-thaw cycles do not increase the hydraulic conductivity of the flotation sand-bentonite mixture.

According to the research results, the potential uses of flotation sand are as filter layers of road and street constructions, environmental structures, and pre-filling and surface layers of the landfill covers. Refined with admixture materials like bentonite, flotation sand can also be used in the barrier layer of landfill covers. As a bound material, with binding materials like bark ash, for example, flotation sand is suitable for other pavement structures of road and street constructions. To resolve the long-term behaviour of the material, test structures should be used particularly to find out the environmental and mechanical durability of the bound material.

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THE USE OF PEAT ASH IN EARTH CONSTRUCTION

by
Eero Huttunen [1)] and Kauko Kujala[2)]

Huttunen, Eero & Kujala, Kauko 2001. The use of peat ash in earth construction. *Geological Survey of Finland, Special Paper 32, 83–90,* four figures and 4 tables.

This paper reviews the results of peat fly-ash utilisation at a peat power plant in Rovaniemi. Peat ash has been tested as a filler material in asphalt pavement, as a sub-base material of a road structure and as layers of landfill cover structures. On the basis of the results obtained, peat ash is suitable for these applications in both technical and environmental aspects. At the moment, peat ash is classified as waste and this retards the progress of extensive use of peat ash in earth construction.

Key words: engineering geology, peat fuel peat, ash, utilization, construction materials, highways, pavements, fillers, landfills, Rovaniemi, Finland

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INTRODUCTION

The Suosiola peat power plant of Rovaniemen Energia uses peat ash as its main fuel. In addition, oil and wood chips are used. In August 1995, a new modern combustion unit was brought on stream. Approximately 13 000 m+ of peat fly-ash is generated annually. At present, the peat ash is disposed 30 km away, at the Suksiaapa swamp.

The utilisation of peat ash in earth construction applications has been studied in several research and development projects, which have proceeded through the classification of material-related properties to designing and constructing test structures. At present in the research, the monitoring phase of the test structures is in progress. Because of the restricted availability of suitable natural raw materials in Rovaniemi and its surroundings, technical and economic needs for the development of the utilisation of peat ash exists. Because of the geographical location, special climate conditions and, especially, the freezing of the soil, has to be taken into consideration in structures and materials.

ENVIRONMENTAL ASPECTS

The basic precondition for extensive use of peat ash is the environmental qualification. In Finland, no such official standards exist which could be applied in evaluating the environmental impact of by-products applicable for earth construction. However, in general a three-step evaluating procedure is followed. In the first stage, the environmental impact is estimated on the basis of composition. If the total concentration exceeds a certain maximum concentration, the environmental impact is evaluated on the basis of solubility and risk analysis.

Table 1. Total concentration and solubility of peat ash in maximum solubility test NEN 7341 and column test NEN 7343. The results are given in milligrams per kilogram of dry matter (mg/kg dm).

Element	Total concentration	Max. leaching test NEN 7341	Leaching test (Column test) NEN 7343	Guideline values for total concentrations	
				Lower Value (mg/kg)	Upper value (mg/kg)
Al	47500	699.4	109		
As	54	20.7	0.04	13	60
B				5	50
Ba	142			600	600
Be				1	10
Ca	106000	29060			
Cd	4.3	0.99	0.004	0.3	10
Co	33	<5		50	200
Hg	–	<0.02			
Cr	212	<5	80	500	
Cu	86	4.93	0.01	32	400
Fe	244000	<100			
K	1410	285.7			
Mg	12500	1132.9			
Mn	1700	51.2			
Mo	<5	<5		5	200
Na	1090	275.8			
Ni	85	<5	0.11	40	300
P	13200	492.6			
Pb	35	<10		38	300
S	9450	9851			
Sb		<50		5	40
Sr	353	89.6			
Ti	815	5.9			
V	356	18.7		50	500
Zn	126	9.9	0.76	90	700

< concentration is below the detection limit

The previously mentioned values are interpreted so that the by-product that has total concentrations below the lower value can be used without limitations for almost any purpose. Material that has total concentrations above lower value, but below the upper value, can be used with some limitations or with some protective precautions, but only in those applications that pose no risk to human beings. If the upper value is exceeded, the use of material requires a detailed risk assessment before use. This procedure is not, however, officially in use.

The total concentrations of the peat ash were determined by extracting it in nitrohydrochloric acid (3:1 hydrochloric acid: nitric acid 90^{o}C). The analysis was performed using ICP-AES- method. The concentration of the leaching elements were examined using maximum leaching test NEN 7341 and with leaching test NEN 7343.

The total concentration and the maximum leaching values and leaching values of peat ash are presented in Table 1. Total concentration of all the elements are below the upper value. The lower value is exceeded by elements As, Cd, Co, Cr, Cu, Ni, V and Zn, but their solubility is slight according to the maximum leaching test. In the leaching test (column test), the concentration of these elements was smaller than the maximum concentration when the placement criteria was group 1, i.e. application in an unisolated form.

THE USE OF ASH AS ASPHALT FILLER

In the laboratory, the material properties of ash as filler, as well as the material properties of the bitumen-filler mixtures, and asphalt pavements that included ash, were studied. On the basis of the laboratory results, a test pavement was constructed, where peat ash was used as filler for a SMA-12 pavement. In this research, PANK-norms (PANK 1995) that correspond the European standards were applied.

Laboratory Phase

The peat ash fulfilled the requirements of the filler, except the void content, which was higher than the maximum value presented in PANK-norms. Because the fine aggregate of asphalt pavement generally includes a mixture of sand and crushed rock as fine aggregate, the void content in the fine aggregate is smaller than that of the peat ash alone. Hence the norm requirement can be achieved. To fulfil the norm requirement, an individual estimation of each case is required. The peat ash that was examined met the PANK-norm requirements in terms of grain size distribution and ignition loss for the additive filler.

The effect of peat ash on the stiffness of the pavement was studied by determining the softening temperature of bitumen (B120) and filler mixtures for aged and unaged samples. According to these results, the softening temperature of mastic that included peat ash as filler was clearly higher than the softening point of the mastic that included lime or a mixture of lime and peat ash. On the basis of the softening temperature, it can be estimated that when using peat ash as additive filler, the modulus of stiffness is greater than when lime filler is used. However, using only peat ash as filler is limited by the void content that can raise the total amount of mineral aggregate voids content in asphalt mass to be too high.

In laboratory experiments, pavement type AB 16 was used. Bitumen B120 was used as binder. In the test pavements, peat ash was used as additive filler. For the reference pavements, lime powder was used as additive filler. According to the determinations, the optimum binder content of those pavements that included peat ash is from 0.6 to 0.8% higher than that in the reference pavements. The increase in binder content is apparently caused by the higher void content in the ash. The modulus of stiffness of the test pavement was higher than that of the reference pavement; for example, at +20 °C, a 1.45 times higher value was achieved. The deformation values of the test pavements represent the deformation class I in 1995 Asphalt norms. All test and reference pavements were easy to compact.

Field Results

The test asphalt pavement is located in the centre of Rovaniemi, on Lapinkävijäntie road. The proportional class that is determined on the basis of the traffic flow, is B. The SMA 12 test pavement included 9% peat ash of the total mass of mineral aggregate. In the reference pavement, lime powder was used as filler. Core samples were taken from the test and reference pavements, and the mass density, void content, indirect tensile strength and sensitivity for deformation were measured. The void values of the test pavement, which included fly ash, varied slightly more than the void values given in the 1995 Asphalt norms. On the basis of the tensile stress (–2ºC), the test pavement is classified in the highest freeze-proof class. On the basis of the cyclic creep stress test, the test pavement is classified in deformation class II, which corresponds to the proportional classification (Table 2).

Table 2. Material properties of the test and reference pavements (SMA 12) at the Lapinkävijäntie road site.

Pavement	Pavement density kg/m^3	Mass density kg/m^3	Voids %	Indirect tensile strength (kPa)		Creep test %
				−2°C	+10°C	
Reference	2410…2566	2566…2604	4,1…5,2	2060…2491	1018…1488	
Test	2460 – 2482	2586 – 2618	0.9 – 6.1	1819 – 2491	1068 – 1163	2.7 – 3.1

THE USE OF ASH IN SUB-BASE LAYERS

Peat ash reacts with water in a pozzolanic manner. This property is viable in structures that require high bearing capacity, such as road, street and yard structural layers. On the other hand, problems caused by the strength gain of the material have to be taken into consideration; for example in municipalities' technical maintenance work. In order to strengthen the structures as far as possible, peat ash should be stored dry and moisturised only immediately before the constructing or during the work. Repeated freeze – thaw cycles decrease the compressive strength of peat ash.

In the test structure (Fig. 1) peat ash replaced the 900-mm thick sub-base layer of crushed rock or gravel crush. The compacting of peat ash was done in November 1997, at a temperature of -7 ºC. Compacting was successful with ash that was still warm after the burning process. Peat ash was moisturised at the power plant immediately before transportation to the construction site. The base course and the pavement layers above peat ash were constructed during summer 1998. At the same time, monitoring equipment was installed and samples were taken from the peat ash layer. More information will be obtained from the freeze-thaw weakening, from the relationship between thermal conductivity and the water content, and from the frost susceptibility of ash, when the behaviour of the structure is observed in situ conditions. This information and other material parameters (Table 3) are essential requisites for establishing instructions for the use of peat ash in the future.

The results of laboratory tests of the samples taken from the test structure are presented in Table 3. Significant in these results is the low frost susceptibility (segregation potential), which indicates that when ash is well compacted and its strength is increased, the material is classified as non-frost susceptible.

The maximum depth of frost during the winter of 1998–1999 was 156 cm, and during the winter of 1999–2000, 138 cm. The freezing index was 36 300 Kh and 30700 Kh, respectively. In reference structures, the frost depth during the winter of 1998–1999 in the Rovaniemi region was approximately 250 cm. The insulation effect of peat ash is clearly illustrated in the temperature gradient (Fig. 2). During wintertime, peat ash retards frost penetration and the temperature decreases in the top part of the base course. On the other hand, in springtime the layer of peat ash slows down the thawing process. Both low thermal conductivity and high heat capacity affect the frost behaviour of ash.

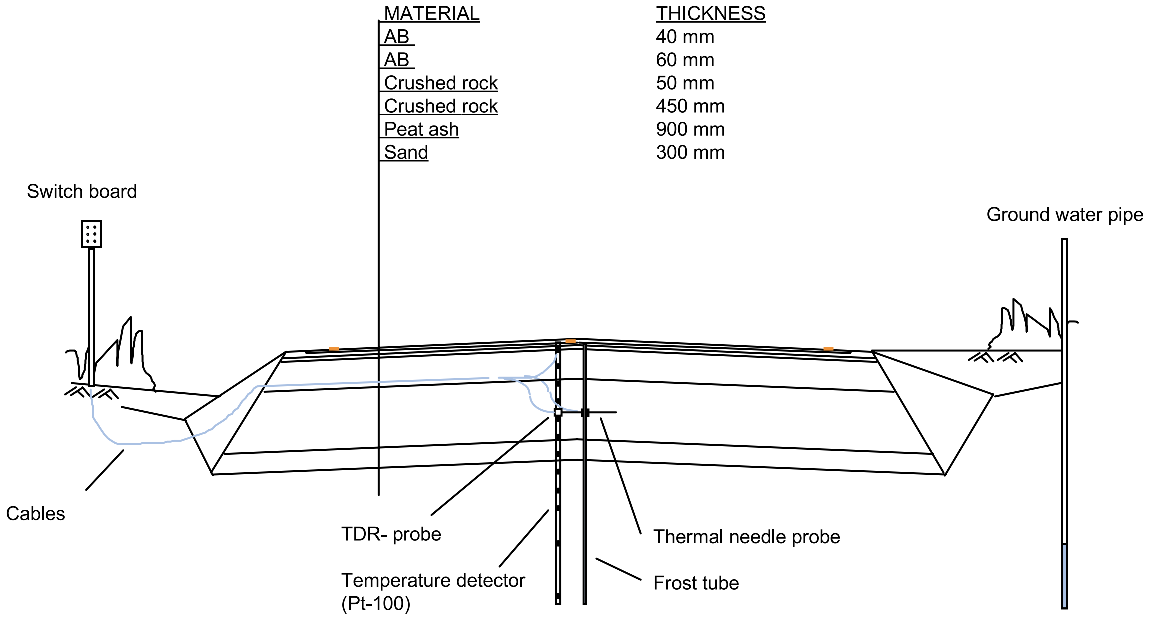

Figure 1. Cross-section of the peat ash test structure of Teollisuustie Road, Rovaniemi.

Table 3. Material properties of peat ash layer of Teollisuutie Road, Rovaniemi.

Properties	Value	Unit
Density*	1.59	kg/m^3
Water content*	35	%
Porosity*	61.9	%
Compressive strength*	1.14	MPa
CBR-value	34.8	%
Segregation potential	0.2	mm^2/Kh
Thermal conductivity (+20°C)	0.75	W/mK
Hydraulic conductivity	$10^{-6.1}$	m/s

* Average values of the samples

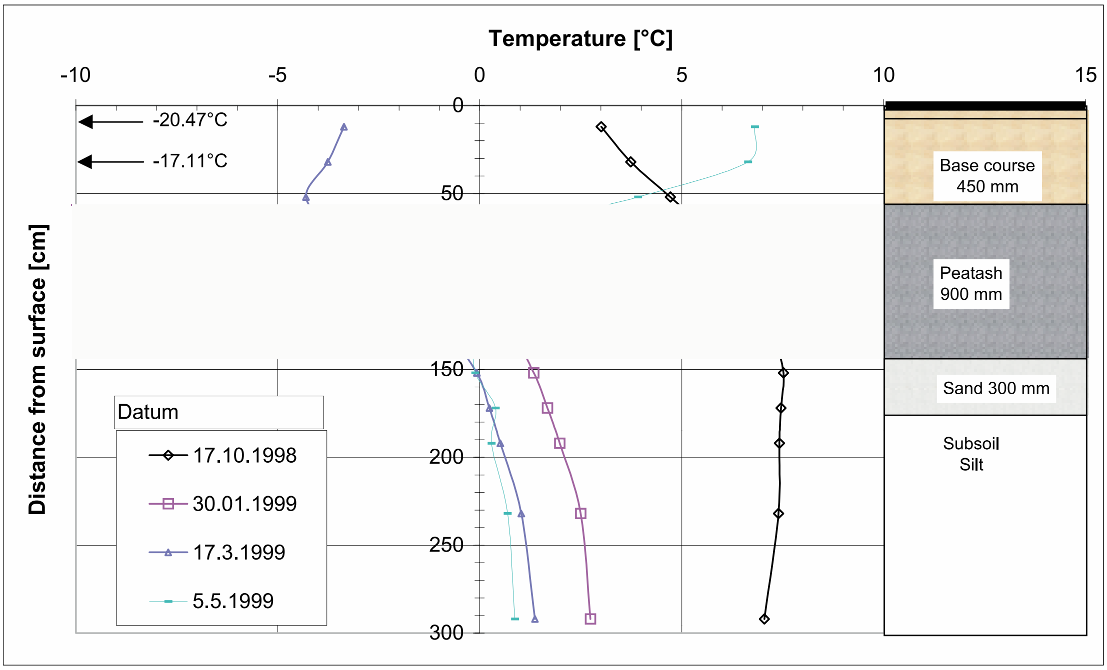

Figure 2. Temperature profiles of the Teollisuustie peat ash structure in Rovaniemi, winter 1998–1999.

UTILISATION OF PEAT ASH IN LANDFILL STRUCTURES

The cover of Mäntyvaara landfill was chosen for test constructing purposes. In the laboratory, the peat ash index properties, compaction properties, frost susceptibility, freeze-thaw behaviour, air permeability and erosion properties, as well as mechanical, hydraulic, and thermal properties, were studied. In addition, the amount of water infiltrating through the test structure was estimated by numerical modelling. On the basis of this research material, a plan for the test structure was made. The plan includes two test structures. In the thinner structure, the idea is to allow the frost to penetrate the hydraulic barrier, and to determine the effects of frost on the material properties of the hydraulic barrier, as well as on the functioning of the whole structure.

The monitoring of the test structure includes measuring the amount and quality of the water infiltrating through the structural layers, settlement, temperature and peat ash water content measurements (Fig. 3).

The settlement of the test structures from June 1999 to May 2000 was between 0.22 and 0.32 m. In both test structures, the greatest settlement was in the middle of the structures. In test structure 1, the maximum frost depth was approximately 0.8 m in February, when 0.6 m of the peat ash surface layer was frozen. During November – April, a significant temperature gradient (approximately 5 – 7°C) in the peat ash surface layer was noticed. The temperature difference is related to the low thermal conductivity, which is about 0.35 – 0.55 W/Km at the present density and water content (Jauhiainen 1999). The temperature of the waste fill during the period varied between 6.5 and 14.4°C (Fig. 4).

In test structure 2, the maximum depth of frost was approximately 0.8 m in February. At the time, the 500-mm thick peat ash surface structure was completely frozen. The temperature of the waste fill in test structure 2 during the period varied slightly more (between 4.8 and 14.1 ^{o}C) than the temperature of the waste fill in test structure 1 (Fig. 4).

No significant changes occurred in the volumetric water content of the peat ash layers in test structures 1 and 2. In both test structures, the volumetric water content of the hydraulic barrier (46 to 48 %) was higher than the volumetric water content of the surface layer (32 to 42) during the observing period June 1999 to May 2000.

In December, it was observed that through the surface structure of the test layer 1, about 0.5 l of water had infiltrated. In other lysimeters no water was noticed during the observation period. The rainfall during June-December was 454 mm in total in Rovaniemi. On the basis of the analysis, the pH of the water was high (11.97). The concentration of heavy metal in the infiltrated water were extremely small (Table 4).

Table 4. Results of the analysis of water infiltrated through the surface layer.

Properties	Value	Unit
pH	11.97	
Electrical conductivity	766.0	mS/m
Copper	267	µg/l
Arsenic	0	µg/l
Nickel	45	µg/l
Chrome	649	µg/l
Zinc	4	µg/l
Cadmium	0	µg/l

CONCLUSIONS

On the basis of the results and experience obtained, both technical and environmental properties enable peat ash to be used in the tested applications. In order to determine and ensure the long-term durability, continuing the monitoring of the test structures is essential. The long-term behaviour of peat ash in different applications will finally determine the application-specific life cycle costs, which determine the technical and economical competitive ability of different utilisation alternatives.

According to the present environmental legislation in Finland, peat ash is classified as waste. The research and development projects implemented, as well as the monitoring measurements in progress will enable the analysing the physical and chemical functioning of the structures with numerical models. The analysis of the water infiltrated through the peat ash layer showed that the solubility of heavy metals *in situ* was very low. To help in decision making, risk analysis can be performed on the basis of these preliminary results and experiences.

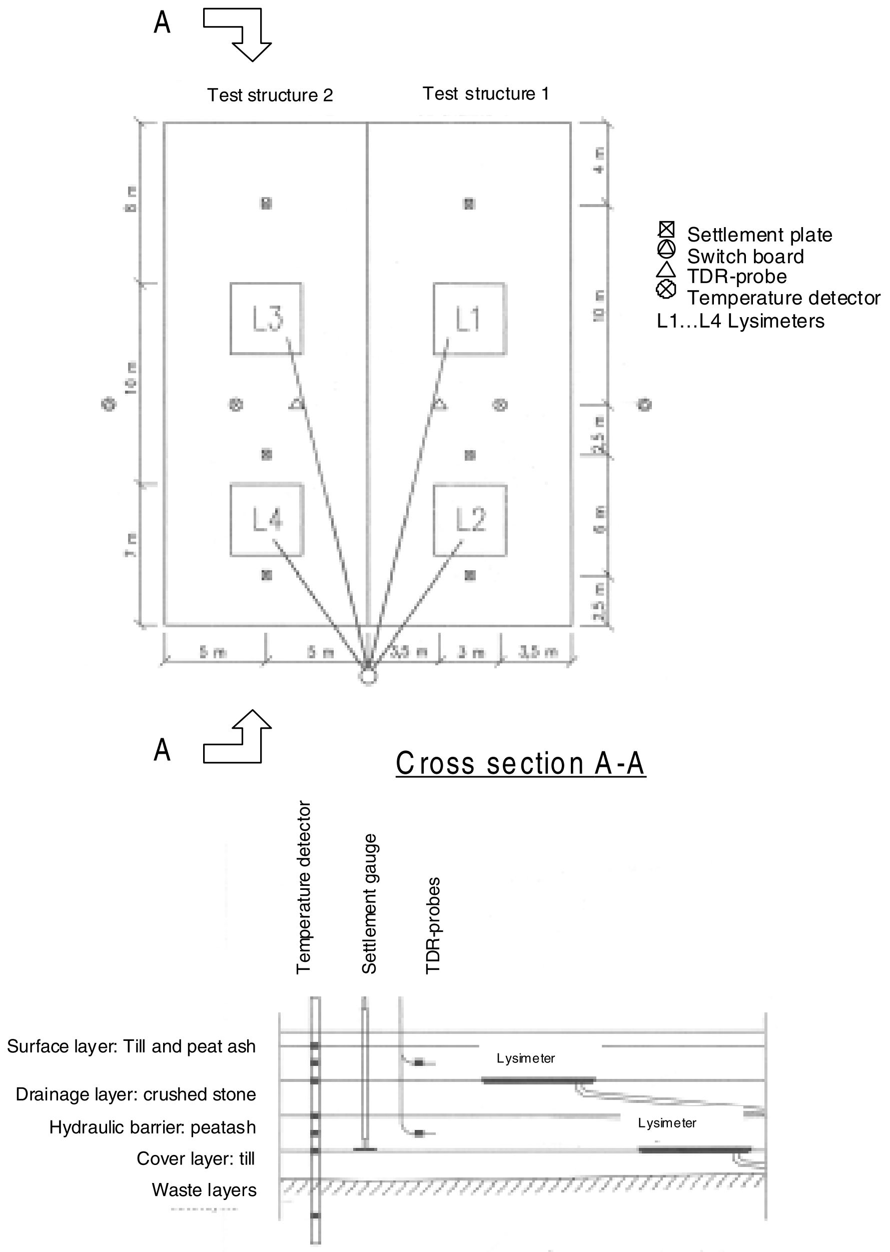

Figure. 3. Instrumentation of the test structure.

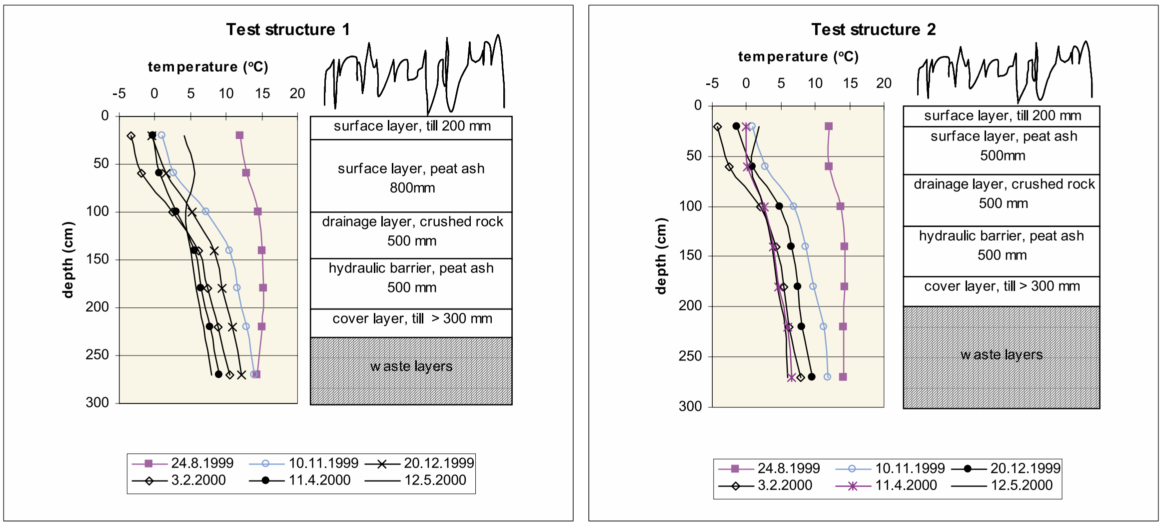

Figure. 4. Temperature profiles of test structures 1 and 2 during the observation period.

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Geological Survey of Finland, Special Paper 32, 91–98, 2001.

DEFINITION OF ENVIRONMENTAL CRITERIA FOR INDUSTRIAL BY-PRODUCTS USED IN EARTH CONSTRUCTION

by
Jaana Sorvari and Jyrki Tenhunen

Sorvari, Jaana & Tenhunen, Jyrki 2001. Definition of environmental criteria for industrial by-products used in earth construction. *Geological Survey of Finland, Special Paper 32, 91–98,* one figure and 2 tables.

At present in Finland, Dutch methodologies and criteria are frequently employed in the environmental assessment of by-products used in earth construction. A study was initiated in order to develop a national code of practice. A hierarchical procedure including composition and solubility standards was elaborated. Derivation of solubility criteria (MPEs) was based on Dutch leaching models and Finnish target values of soil. Conservative risk assumptions were used as a starting point in the definition of parameter values. Different MPE alternatives were created by altering the values of the parameters defining an acceptable load to soil. A risk-benefit analysis was executed by using a decision analysis (the SMART method) in order to identify the best MPE alternative. Land use, uncertainties in analytical methods and non-uniformity of the target values (soil) were considered in the definition of the final MPE values. The proposed solubility standards differ slightly from the corresponding Dutch values. On the basis of the results of previous leaching tests, water-cooled blast-furnace slag and crushed concrete can be considered as readily recyclable, whereas most of the fly ashes from coal combustion could be used only in a monolithic i.e. prefabricated form.

Keywords: construction materials, by-products, earthworks, risk assessment, solubility, standards, Finland

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INTRODUCTION

Significant amounts of natural mineral aggregates such as rock, gravel and sand are used annually in earth construction in Finland. In industrial activities, several wastes having suitable technical properties for use as a substitute for primary materials are produced. Materials employed traditionally in earth construction include blast-furnace slag (BFS), crushed concrete from demolition works and fly ash from energy production. Environmental compliance is a key factor in the assessment of reusability of different by-products.

In Finland, the use and disposal of by-products is mainly regulated by the Waste Act. All industrial by-products are considered to be wastes and normally an environmental permit must be obtained in order to reprocess, reuse or recycle them in a large scale. As an exception to this practice, a simpler notification procedure exists for experimental construction projects. Depending on the amount of a by-product, the permit is granted by a regional environmental centre or by a municipality. The permits have been granted on a case-by-case basis and the process has been regarded as time-consuming and laborious. Since for the time being there has not been any official code of practice concerning the definition of environmental suitability of by-products, practices of acceptance as well as foundations of the decisions have varied. Different leaching tests and environmental criteria based mainly on the 100-fold quality standards of domestic water have been employed. During recent years, Dutch test methodologies and criteria have been widely adopted. The Dutch guidelines and standards are based on the Dutch legislation and environmental conditions and as such are not directly applicable to Finnish conditions. The definition of unified national environmental guidelines has been considered an important development need.

DEVELOPMENT OF A CODE OF PRACTICE

Principles

The basis of the development of environmental criteria was the existing or prospective Finnish legislation. Following the legislation, priority was given to the prevention of ground water contamination. This protection is accomplished by limiting the use of by-products outside the classified aquifer areas. The second principle was the prevention of soil contamination. This is achieved by issuing substance-specific limit values, which define an acceptable, negligible emission to soil.

The primary condition for the reusability of a by-product is that it may not be hazardous, which means that it must be disposable in a landfill designed for non-hazardous wastes.

Structure

A hierarchical procedure has been proposed for the definition of environmental compliance of by-products (Sorvari 2000). According to this model, environmental acceptability can be assessed on different levels depending on the characteristics of a by-product. Numeric criteria have been issued for each of these levels. These criteria are, however, applicable only to non-sensitive areas which receive background load *e.g.* traffic areas, commercial districts and equivalent regions in general.

At the highest assessment level, the composition of a by-product is compared with the composition standards corresponding to the target values characterizing a clean soil. If these values are exceeded, as is normally the case, the solubilities of harmful substances must be examined with leaching tests. The NEN 7343 column test and the NEN 7345 diffusion test should be used as primary test methods. Solubilities in various, relevant pH conditions should be studied as well. The test results are compared with the solubility standards, i.e. Maximum Permissible Emission (MPE) values. If these are exceeded, the by-product in question can be studied further by using a case-specific risk assessment (RA) procedure. In the RA, environmental conditions (*e.g.* transportation and exposure routes, recipients) and the long term stability of the material are taken into account. This study can be material-specific or site-specific. If it can be stated that there is no harm to human health or to the environment, it is allowable to use the by-product at a site equivalent to the one characterized in the RA. The methodologies, methods and contents of risk assessments are not defined in this context since separate national guidance has been prepared elsewhere (Sorvari & Assmuth 1998, Wahlström *et al.*, 1999).

No MPE values are given for organic substances, because there is no consensus concerning suitable leaching test methods. If organic substances are present, environmental suitability can be defined only by a comparison with the target values of soil and a detailed risk assessment procedure.

The composition of a by-product is also compared with the limit values of soil. If these values are exceeded, the definition of environmental suitability should not be exclusively based on solubility, but the stability of the material (possible changes in properties), additional, relevant transportation routes and possible direct exposure routes (*e.g.* inhalation of dust during construction etc.) must be taken into account.

DEFINITION OF SOLUBILITY STANDARDS (MPE VALUES)

MATERIALS AND METHODS

Dutch leaching models presented by Aalbers *et al.* (1996) were used in the derivation of the MPE values for metals. These models are based on extrapolation and interpolation of laboratory scale measurements and the definition of a maximum permissible load to soil (Eqs. 1…4).

Granular materials:

$$E_{max} = E_{soil} + \frac{I_{max}}{d_c \times h \times f_{ext,n}} \qquad (1)$$

$$I_{max} = \frac{\alpha}{100} \times T_s \times \rho_s \times h_s \qquad (2)$$

$$f_{ext,n} = \frac{1 - e^{-\kappa \times \frac{J \times N_i}{d_c \times h}}}{1 - e^{-\kappa \times 10}} \qquad (3)$$

Monolithic materials:

$$E_{max} = \frac{I_{max}}{f_{ext,v} \times f_{temp}} \qquad (4)$$

Where

E_{max} = maximum allowable emission (of an individual substance) determined by the NEN 7343 column test (mg kg $^{-1}$) or the NEN 7345 diffusion test (monolithic materials)
E_{soil} = substance-specific correction factor corresponding the leaching of an individual substance from soil (mg kg^{-1})
I_{max} = maximum allowable immission (of an individual substance) in soil during J years (mg m^{-2})
d_c = dry density of a waste material (kg m^{-3})
h = total thickness of a waste material (i.e. the sum of separate construction layers consisting of the same waste material) (m)
$f_{ext,n}$ = factor for the extrapolation from the time representing the duration of the leaching test to J years
α = factor representing the marginal burdening of soil (i.e. acceptable increase in the concentration of an individual substance in soil) (%)
T_s = target value of soil (mg kg^{-1})
ρ_s = dry density of soil (kg m^{-3})
h_s = thickness of the soil layer underneath a by-product structure (m)
J = duration of the emission to soil (yr)
N_i = infiltration through the structure made of waste (mm yr^{-1})
κ = substance-specific slope of the regression line representing the leaching function which has been transformed to a linear model
$f_{ext,v}$ = factor for the extrapolation from the time representing the duration of the diffusion test to J years; wetting period, exhaustion and changes in the diffusion coefficient are taken into account in the definition of the factor
f_{temp} = correction factor for the difference between the temperature in the laboratory and the temperature at site.

The definition of an acceptable load is founded on the target values of soil. Parameter values describing conditions in Finland, *e.g.* soil characteristics, weather conditions and the properties of by-products were employed in the calculations. The information on the variables was gathered from several sources and calculations were performed by using a Monte Carlo technique. Separate MPE values were calculated for different applications (granular vs. monolithic material, unpaved vs. paved structure). The time scale was fixed to 100 years and the thickness of a by-product layer to 0.7 m. MPE alternatives were created by modifying the values of those parameters which describe the "permissible load", i.e. the acceptable increase in the concentrations of contaminants in the soil underneath a by-product structure (á, % compared with the target values of soil) and the depth of a soil layer below a by-product structure (h_s, m). Conservative values such as á = 1 % and h_s = 0.2 m were used as a starting point.

Finnish soil is both horizontally and vertically very heterogeneous. The main soil type is sandy till, which represents 75 % of all tills (Koljonen 1992). Altogether 54 % of the total land area is covered by till in thick layers. Therefore, variables describing the soil type till were selected for the calculations.

A sensitivity analysis was carried out in order to test the sensitivity of the calculated MPEs to the variability of parameters. For some metals, the national target values of soil are defined according to the content of clay and organic matter in soil. For this reason, the sensitivity analysis had to be performed separately for each individual substance.

For those contaminants lacking a soil target value *e.g.* sulphate and chloride, MPE values were derived from the national quality standards issued for domestic water. The target value for chloride is 25 mg l^{-1} and the corresponding quality criteria for sulphate is 150 mg l^{-1}.

Sample size in the Monte Carlo calculations was defined according to an acceptable uncertainty in the medians of the MPE values. Here, the range of +/-2 percentiles was selected, leading to a sample size of 2500.

Decision analysis

For metals, the best alternative of the groups of MPEs was defined by using a SMART method (Simple Multi-attribute Rating Technique) and the methodology employed in a previous study on the selection of a water supply system (Tenhunen & Seppälä 1996). The SMART method has been described in detail by Von Winterfelt and Edwards (1986). Along with the environmental risk, social and economic benefits, i.e. savings in natural resources and landfill space, construction costs as well as social disadvantages were taken into account (Fig. 1).

Indexing (to a range from 0 to 100) of the values given for each decision criterion was executed on the basis of an assumption of linear regression. The sum of each individual indexed value (x_i) multiplied by the criterion-specific weighting factor (w_i), represents the total value of the corresponding group of MPEs. This total value describes the superiority of one alternative compared with the others

The analysis was performed for a 12 m wide road structure containing either natural mineral aggregates (sand, gravel), fly ash from coal combustion or crushed concrete in the base course. Information on the consumption of each by-product was taken from a previous life cycle assessment report (Eskola *et al.* 1999). Several experts representing material producers and waste management and construction companies were interviewed in the collection of the data concerning the costs of construction and landfill disposal.

In the definition of values for the decision criterion "amount of waste ending up in a landfill", it was assumed that all the material which is not environmentally acceptable (i.e. at least one of the substance-specific MPEs is exceeded) is disposed to a landfill. The value of the decision criterion "social disadvantages" was expected to correspond to the amount of the recycled by-product.

The definition of separate MPE alternatives was based on the variability of the parameters á and h_s (see above) and the prognosis of recycling potentiality (% of the total mass of the by-product produced) of the fly ash and crushed concrete in different structures (paved and unpaved structure, granular and monolithic material). The prognosis was estimated from the results of previous leaching tests collected from several information sources around the country.

Weighting of the decision criteria was carried out by representatives of the Ministry of the Environ-

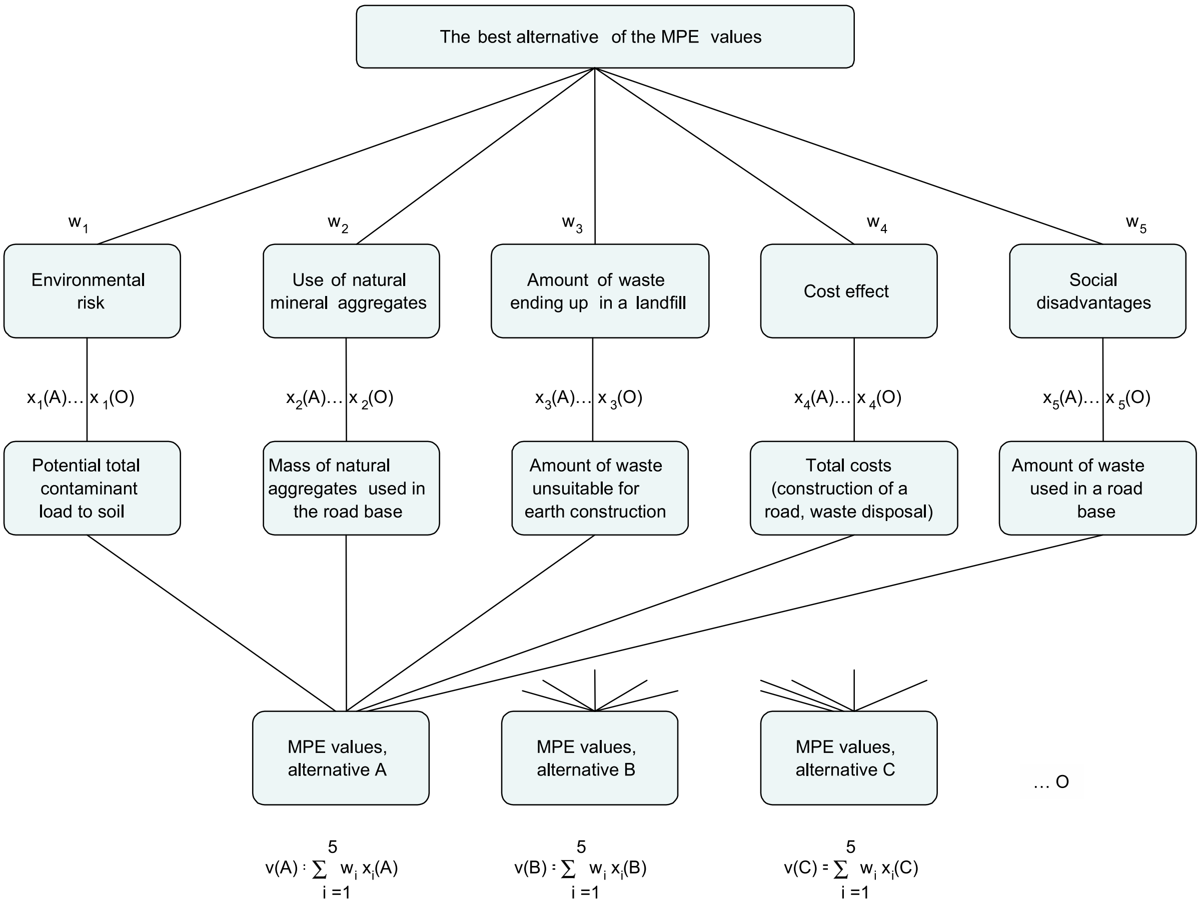

Figure 1. Decision criteria used in the decision analytical study of the different alternatives (A…O) of solubility standards i.e. MPE values (Maximum Permissible Emission). w_i = weighting coefficient of an individual decision criterion i (i = 1…5), $x_i(A)$… $x_i(O)$ = indexed numerical value of a decision criterion i (i = 1…5) corresponding to an alternative representing a separate group of MPEs (A…O), v(A)…v(O) = total value of an alternative representing a separate group of MPEs (A…O).

ment. Since the Ministry is responsible for the prescribing of national environmental criteria, the involvement of other stakeholders such as industrial representatives, users of by-products etc. was considered unnecessary.

Results

According to the results of the sensitivity analysis, the key parameters in the definition of MPE values are those which define the acceptable load to soil underneath a by-product structure that is parameters á, h and h_s. For paved structures, the time scale (J) was also critical and in the case of a monolithic material the density of soil was decisive as well. When all the variables defining the acceptable load (J, á, h, h_s) were fixed, density of soil and by-product, substance-specific constants describing leaching behaviour (ê) and infiltration rate (N_i, mm a^{-1}) were identified to be the key parameters. In the category of monolithic materials, correction factors taking weather conditions into account ($f_{ext,v}$ and f_{temp}) were important. The results of a sensitivity analysis depend significantly on the selection of parameter values (i.e. extreme values, statistical variables and the shape of the distribution).

In the decision analysis, the criterion “environmental risk” received a weighting coefficient of 0.75 and the criterion “use of natural mineral aggregates” a coefficient of 0.25, respectively. Coefficients of other decision criteria obtained the value 0, meaning that they were not regarded as important factors. Aspects such as the type of land use and uncertainties in analytical methods (soil and waste analyses, leaching tests) were considered in the final definition of the MPEs. Furthermore, the non-uniformity of the national target values was also taken into account. The resulting group of MPE values corresponds to the marginal load defined as “a load that can cause a 5 % increase of concentrations (clean soil) in the 1 m thick soil layer underneath a by-product structure”. Due to the non-uniformity of the national target values of soil, the corresponding Dutch guideline values were used in the calculation of the MPEs of Ba, Co, Ni, Sb, and Sn. The final solubility criteria are presented below (Table 1).

Table 1. Proposal for the solubility standards for by-products used in earth construction.

Substance	Granular material Uncovered, mg kg^{-1}	Granular material covered, mg kg^{-1}	Monolithic material mg m^{-2}
As	0.14	0.85	58
Ba	10	28	2800
Cd	0.011	0.015	2.1
Co	1.1	2.5	280
Cr	2.0	5.1	550
Cu	1.1	2.0	250
Hg	0.014	0.032	1.6
Mo	0.31	0.50	70
Ni	1.2	2.1	270
Pb	1.0	1.8	210
Sb	0.12	0.40	36
Se	0.060	0.098	14
Sn	0.85	3.1	280
V	2.2	10	700
Zn	1.5	2.7	330
F	11	25	2800
CN, free	0.060	0.098	14
SO_4	1500	–	–
Cl	250	–	–

CONCLUSIONS

On the basis of this study, new national environmental criteria for the use of by-products in earthworks were proposed. The final solubility standards differ from the corresponding Dutch values and are mainly less strict than the solubility standards issued in some other countries (Table 2). This distinction is due to differences between 1) analytical methods (especially leaching tests) 2) the bases, foundations, methods and principles of the assessment and definition of risks/load and 3) the risk management and waste management policies.

Disparities between the proposed Finnish criteria compared with the corresponding Dutch standards are mainly due to the differences in the national target values of soil, in the definition of an acceptable load and in environmental conditions. Especially the standards issued for paved structures differ from the corresponding Dutch solubility criteria. This is due to the difference in infiltration rates. In this study, a value of 34 mm a^{-1} was employed for paved structures, representing 10 % of the infiltration rate through unpaved structures. In the Netherlands the corresponding value has been 6 mm a^{-1}.

In respect of the proposed solubility criteria, the use of granular fly ash (from coal combustion) will be limited. Especially the concentrations of soluble selenium and molybdenum usually exceed the corresponding MPE values. Further studies are needed in order to determine the significance of Se and Mo loads. However, solubility can be minimized by using a binding material such as cement. On the other hand, according to the results of some previous leaching tests, water-cooled BFS and crushed concrete can be considered environmentally acceptable without any additives.

The Dutch method, which was used for the derivation of MPEs, has the disadvantage of being very sensitive to variables for which it is not possible to define scientifically incontestable and “correct” values (referring to the parameters which specify the acceptable load to soil). The definition of the MPE values for monolithic structures is based on simple equations and the results are very sensitive to the definition of some correction factors which characterize environmental conditions. Furthermore, mixture effects, speciation of contaminants and pH variations are not taken into account. The

Table 2. Some solubility standards issued in different countries (mg kg^{-1}). All values have been transformed to equivalent units by using the L/S-ratio of the corresponding leaching test. a) unpaved b) paved structures. (ÖNORM S 2072 1990, LAGA 1995, Aalbers 1996, Department of Natural Resources 1999, Rasmussen 1999).

Substance	Finland		the Netherlands		Germany, dw		fs	Austria	Denmark	USA, Wisconsin	
	a)	b)	a)	b)	a)	b)	b)	a)	b)	a)	b)
As	0.14	0.85	0.88	7.0	0.10	0.50	0.60	1.0	0.016	0.20	10
Ba	10	28	5.5	58	-	–	–	10	0.60	16	40
Cd	0.011	0.015	0.032	0.066	0.02	0.10	0.10	0.05	0.0040	0.020	0.10
Co	1.1	2.5	0.42	2.5	-	–	1.5	1.0	–	–	–
Cr	2.0	5.1	1.3	12	0.30	–	3.0	1.0	0.020	0.40	2.0
Cu	1.1	2.0	0.72	3.5	0.50	2.0	–	10	0.090	5.2	26
Hg	0.014	0.032	0.02	0.076	0.0020	0.020	–	0.020	0.00020	0.008	0.040
Mo	0.31	0.50	0.28	0.91	-	–	1.5	-	–	–	–
Ni	1.2	2.1	1.1	3.7	0.50	1.0	2.0	1.0	0.020	0.80	2.0
Pb	1.0	1.8	1.9	8.7	0.40	1.0	–	1.0	0.010	0.06	0.30
Sb	0.12	0.40	0.045	0.43	-	–	–	–	–	–	–
Se	0.06	0.098	0.044	0.10	-	–	–	0.50	–	0.4	0.10
Sn	0.85	3.1	0.27	2.4	-	–	–	10	–	–	–
V	2.2	10	1.6	32	–	–	6.0	2.0	–	–	–
Zn	1.5	2.7	3.8	15	1.0	4.0	10	30	0.20	100	200
F	11	25	13	100	-	–	–	30	–	32	80
CN, free	0.06	0.098	0.013	0.076	-	–	–	0.20	–	1.6[1]	4.0[1]
SO_4	1500	–	750	22000	1500	6000	–	varies[2]	500	5000	10000
Cl	250	–	600	8800	200	1500	–	varies[2]	300	5000	10000

dw = demolition waste, fs = foundry sand; [1]total cyanide; [2]depends on the electric conductivity

definition of permeability in the case of a paved road structure is problematic because of possible crack formation and infiltration through ramps.

The quantitative data on chemical properties of Finnish by-products was somewhat inadequate. The national target values of soil should be unified. The knowledge concerning adaptation of soil organisms, mixture effects, accuracy of leaching models and long-term behaviour of soluble substances is also insufficient.

The proposed environmental criteria have been used as a basis in the preparation of the Council of State Decree, by which some by-products can be exempted from an environmental permit process. Along with this preparation work, the development of quality assurance procedures is continuing.

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PROBABILISTIC RISK ASSESSMENT OF A CONTAMINATED SITE

by
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Kuusisto, S. M. & Tuhkanen, T. A. 2001. Probabilistic risk assessment of a contaminated site. *Geological Survey of Finland, Special Paper 32, 99–105,* one figure and 5 tables.

The results of deterministic and probabilistic risk assessment at a contaminated site were compared. The deterministic point estimates of the incremental lifetime cancer risk following exposure to dioxin/furan and PCB-contaminated recreational site were 1×10^{-4} for dioxins/furans and 3×10^{-5} for PCBs. The point estimates located at the 99.8 and 97.3 percentiles in the probabilistic range of risk. The distribution means were 4×10^{-6} for dioxins/furans and 4×10^{-6} for PCBs. The point estimates highly overestimated the risk, whereas, the probabilistic approach revealed valuable information regarding the possible distribution of risk. Clean-up levels corresponding risk level of 10^{-6} were also calculated. The point estimates were 5.5 ng TCDD-equivalents/kg soil and 0.5 mg PCBs/kg soil. The 5th percentile values, which are protective for 95 % of the population, were 11.6 ng TCDD-equivalents/kg soil and 0.4 mg PCBs/kg soil. The sensitivity analysis revealed that the soil concentration data is the dominating parameter when it comes to the uncertainty in this assessment.

Keywords: risk assessment, soils, pollutants, dioxins, PCBs, sensitivity analysis

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INTRODUCTION

The traditional way of doing risk assessment is to use single value best estimates for each parameter in risk calculation. This deterministic approach rarely represents the real life situation. Most of the parameters are variable in nature; for example, people who are *exposed* weigh different amounts. Furthermore, many of the exposure assumptions are unknown. The major shortcoming in deterministic approach is the ignorance of the elementary features of exposure parameters, which are the variability and uncertainty.

Because of this variability and uncertainty, most of the input values are really random variables, which can take any value in a range of values, with a certain probability of occurrence. This probabilistic nature of parameters is taken into account in probabilistic risk assessment. (Burmaster 1995)

This paper presents the use of probabilistic risk assessment in a case of contaminated site. Results of the probabilistic approach are compared to the results of the traditional deterministic approach. Carcinogenic risk and risk-based cleanup levels are calculated.

The most commonly used probabilistic method in health risk assessment is the Monte Carlo simulation. In the Monte Carlo simulation, numbers for parameters are generated according to their distributions and risk equation is solved repetitively.

METHODS

The point estimates for the risk where calculated using a deterministic approach recommended by the U.S. EPA (U.S. EPA 1989). Probabilistic risk assessment was done by Monte Carlo Simulation with Latin Hypercube Sampling using Crystal Ball® 4.0 spreadsheet program (Decisioneering). Exposure parameters were approximated by probability density functions (PDFs) found from the literature. PDFs for contaminant concentration data was evaluated using the Kolmogorov-Smirnov goodness-of-fit test and the Maximum Likelihood Estimation in Crystal Ball program.

Sensitivity analysis was done calculating a contribution to variance for each exposure parameter. Contribution to variance is determined by squaring the rank correlation coefficient and then normalizing to 100 % (Decisioneering).

EXPOSURE ASSESSMENT

The contaminated area is a part of a recreational area planned to serve the inhabitants of nearby residential areas. A wide variety of industrial and commercial activities has taken place in the area. The current contamination is mainly due to dumping of fly ash from a waste incinerator. The major contaminants in fly ash are polychlorinated dibenzodioxins and dibenzofurans (PCDD/PCDF), polychlorinated phenols (PCBs), lead (Pb), zinc (Zn), and copper (Cu). Cancer risk following exposure to PCDDs/PCDFs and PCBs is assessed in this case study.

Probability density functions for soil concentration data were approximated from the site data. Soil concentration data from earlier years was used (Paavo Ristola 1988, 1997). A lognormal distribution was chosen to represent concentration data based on goodness-of-fit data and personal judgement. The 95th percentile concentrations were selected as the point estimates. Distribution parameters for soil concentrations are given in Table 1.

Table 1. Point estimates and distribution parameters for soil concentrations.

Chemical	Units	Point estimate	Distribution type and distribution parameters[a]
PCDD/PCDF as TCDD-equivalents	(ng/kg)	643	Lognormal (μ = 174 σ = 392 95% = 643)
PCB	(mg/kg)	12	Lognormal (μ = 3,69 σ = 42,2 95% = 12,16)

[a] Distribution parameters of underlying normal distribution $X = \ln[\underline{X}]$.

The contaminated fly ash is covered with clean sand and gravel. The goal of this paper is to evaluate the usefulness of probabilistic risk assessment compared to the traditional deterministic approach. Therefore, in order to simplify the exposure assessment the risk was assessed as if no clean cover exists.

Ingestion of soil, dermal contact, and ingestion of mother's milk where chosen as possible exposure pathways. Inhalation and ingestion of particles in air contributed less than 1% to the total exposure, and were therefore not included here. The lifetime average daily doses for each pathway was calculated using equations 1, 2 and 3, adapted and modified from the U.S. EPA guidelines (U.S. EPA 1989). The equation for the exposure via mother's milk was adapted from CalTox (CalTox 1993a).

$$DI = \left[\left(\sum_{i=m}^{n} \left(\frac{IR_i}{BW_i} \times \frac{t_i}{10} \right) \times EF_c \times U \times ED_c \right) + \left(\frac{IR_a}{BW_a} \times EF_a \times ED \right) \right] \times CS \times CFS \times \frac{1}{AT} \tag{1}$$

$$\begin{aligned} DD = & \left[\left(\sum_{i=m}^{n} \left(SA_i \times \frac{t_i}{10} \times FSA_i \right) \times EF_c \times ED_c \times RT \right) + \left(SA_a \times FSA_a \times EF_a \times ED_a \right) \right] \\ & \times CS \times CF \times AF \times FC \times \frac{1}{AT} \end{aligned} \tag{2}$$

$$DM = C_{bmilk} \times \left[\frac{IR_{bm}}{BW} \right] \times \frac{EF \times ED}{AT} \qquad C_{bmilk} = I_{mo} \times BW_{mo} \times B_{bmk} \tag{3}$$

Where

DI = lifetime average daily dose following ingestion of soil (mg/kg-day)
DD = lifetime average daily dose following dermal contact with soil (mg/kg-day)
DM = lifetime average daily dose following ingestion of mother's milk (mg/kg-day)
IR = the soil ingestion rate (mg/day)
BW = the body weight (kg)
ED = the exposure duration (years)
U = the amount of days when the soil is unfrozen (days/year)
CS = the contaminant concentration in the soil (mg/kg)
EF = the exposure frequency (days/year)
CFS = the conversion factor (10^{-12} kg/ng)
AT = the averaging time (days)
SA = the skin area as a function of body weight (cm^2/kg)
FSA = the fraction of skin area exposed
AF = the soil adherence factor (mg/cm^2)
FC = the fraction of soil that is contaminated
C_{bmilk} = the chemical concentration in mother's milk (mg/l)
IR_{bm} = the amount of mother's milk ingested (l/d)
I_{mo} = the mother's total exposure (mg/kg-d)
B_{bmk} = the ratio of chemical in milk and mother's exposure (d/kg-milk)

Subscript *c* denotes a child, and *a* denotes an adult. Input parameters are shown in Table 2.

The results of exposure assessment are shown in Table 3. Only the pathways contributing more than 1 % to the overall exposure are shown.

Table 2. Variables and constant used in exposure assessment.

Parameter	Point estimate	Distribution parameters and type		Reference
IR (mg/d)				
< 6 years	179	μ = 179 95% 208	LN	U.S. EPA 1996
Adult	480	–	–	U.S. EPA 1996
BW (kg)			LN	Burmaster & Crouch 1997
1–2 years	11.8	μ = 11.8 σ = 1.9		
7–8 years	25.1	μ = 25.1 σ = 3.9		
35–44 years	80.9	μ = 80.9 σ = 13.4		
U (d/year)	273	min = 204 max = 355 likeliest = 273	TR	Huttunen & Soveri 1993
EF_c (d/year)	1	min = 0.2 max = 1.0	U	Personal judgement
EF_a (d/year)	40	min = 20 max = 60	U	Personal judgement
ED_c (years)	10	μ = 11.36 σ = 13.76 truncated at 10 years	LN	Israeli & Nelson 1992
ED_a (years)	10	μ = 11.36 σ = 13.76	LN	Israeli & Nelson 1992
AT (years)	75	min = 73 max = 79.5	U	U.S. EPA 1996
SA (cm^2/kg-d)			LN	Burmaster 1998
1–2 years	5.77	μ = 5.77 σ = 1.30		
7–8 years	14.84	μ = 14.84 σ = 2.26		
35–44 years	52.90	μ = 52.90 σ = 9.14		
AF (mg/cm^2)	0.52	μ = 0.52 σ = 0.9	LN	Finley et al. 1994c
FC	1	–	–	–
FSA	0.19		U	U.S. EPA 1996
1–2 years		min = 0.057 max = 0.48		
6–7 years		min = 0.047 max = 0.52		
adult		min = 0.052 max = 0.56		
IR_{bm} (kg/kg-d)	0.11	0.11 CV 0.2	LN	CalTox 1993b
EF (d/a)	365	365	–	–
ED (year)	1	–	–	–
BW_{mo} (kg)	64.2	μ = 64.2 σ = 15	LN	Burmaster & Crouch 1997
B_{bmk} (d/kg-milk)				
(PCB)	0.52	μ = 0.52 CV=10	N	CalTox 1993b
(TCDD)	0.92	μ = 0.92 CV=10	N	

μ, mean; σ, standard deviation of underlying normal distribution X = ln [X]; CV, correlation coefficient; LN, lognormal; W, weibull; U, uniform; TR, triangular

Table 3. Exposure to TCDD-equivalents and PCBs through different pathways. Lifetime average daily dose following DI = ingestion of soil, DD = dermal contact with soil, and DM = ingestion of mother's milk (mg/kg-day)

	TCDD-equivalents		PCB		
	DI (mg/kg-day)	*DM* (mg/kg-day)	*DI* (mg/kg-day)	*DD* (mg/kg-day)	*DM* (mg/kg-day)
Deterministic					
Point estimate	6.68×10^{-10}	5.91×10^{-11}	1.25×10^{-5}	2.35×10^{-6}	6.23×10^{-7}
% of total exposure	91.9	8.1	80.7	15.2	4.0
Percentile [a]	98.73	93.19	98.1	93.4	87.8
Probabilistic					
Mean	6.77×10^{-11}	1.36×10^{-11}	1.40×10^{-6}	5.97×10^{-7}	6.73×10^{-7}
Standard deviation	1.97×10^{-10}	7.01×10^{-11}	1.02×10^{-5}	5.68×10^{-6}	1.05×10^{-5}

[a] The location of point estimate at the probabilistic range of exposure.

RISK CHARACTERIZATION

The risk is characterized by considering the exposure, bioavailability and cancer slope factor. The conservative estimate of oral TCDD bioavailability is 100 %. 0.5 – 50 % of TCDD is found to be absorbed in experimental animals (Copeland 1993). For PCBs the oral absorption is assumed to be 90 % (RAIS 1999). The dermal bioavailabilities are 0.1–3 % for TCDD and 0.6– 6 % for PCBs (U.S. EPA 1992). Bioavailabilities were assumed to be uniformly distributed.

Carcinogenic slope factors (CSFs) were entered as point values, so their effect on variability in the risk estimate was not estimated. The used CSFs were 160 000 kg-day/mg (oral) (U.S. EPA 1997), 300 000 kg-day/mg (dermal) (RAIS 1999) for TCDD-equivalents and 2 kg-day/mg for PCBs (IRIS 1999).

Incremental lifetime cancer risks (ILCRs) were calculated using equation 4. The results are shown in Table 4.

$$ILCR = \left[CSF_o \times (B_o \times (DI + DM)) \right] + [CSF_4 \times B_d \times DD] \quad (4)$$

Where

CSF = the cancer slope factor (kg-day/mg)
B = the bioavailability
DI = lifetime average daily dose following ingestion of soil (mg/kg-day)
DD = lifetime average daily dose following dermal contact with soil (mg/kg-day)
DM = lifetime average daily dose following ingestion of mother's milk (mg/kg-day)

Subscript *o* denotes oral and *d* denotes dermal

Table 4. Risk estimates for TCDD-equivalents and PCBs.

	TCDD-equivalents	PCBs
Deterministic		
Point estimate	1.16×10^{-4}	2.65×10^{-5}
Percentile[a]	99.82	97.33
95th percentile	1.33×10^{-5}	1.28×10^{-5}
Probabilistic		
Mean (μ)	3.51×10^{-6}	4.19×10^{-6}
Median	8.82×10^{-7}	2.58×10^{-7}
Standard deviation (σ)	1.28×10^{-5}	3.83×10^{-5}
Coefficient of variability	3.66	9.14
Range minimum	6.02×10^{-10}	2.72×10^{-11}
Range maximum	6.86×10^{-4}	2.49×10^{-3}
Range width	6.86×10^{-4}	2.49×10^{-3}

[a] The location of point estimate at the probabilistic range of risk.

Point estimates located at the high end of probability distribution and were therefore conservative and overprotective. Mean and median estimates of risk were smaller than the generally acceptable risk 10^{-6}. The probability distributions of estimated risks were found to be lognormally distributed. Lognormal distribution is a positively skewed distribution, that is most of the values are at the low risk end of the distribution.

RISK-BASED CLEANUP LEVELS

Risk-based cleanup levels corresponding to a risk level of 10^{-6} were calculated. One in a million risk is generally though to be acceptable risk. The soil concentration parameter was solved using combined risk and exposure equations. The results are presented in Table 5.

The deterministic estimate of TCDD-equivalent cleanup level is located at the 0.03 percentile, meaning that for only 0.03% of the exposed population the risk will be higher than the acceptable risk of 10^{-6}. The range minimums gives the conservative estimate of soil concentration where the risk is smaller than 10^{-6} in all cases.

Table 5. Risk-based cleanup levels corresponding to 10^{-6} risk.

	TCDD-equivalents (ng/kg)	PCB (mg/kg)
Deterministic		
Point estimate	5.53	0.45
Percentile[a]	0.03	7.56
5th percentile	11.64	0.38
Probabilistic		
Mean (μ)	129.22	1.47
Median	54.56	1.10
Standard deviation (σ)	294.75	1.31
Coefficient of variability	2.28	0.89
Range minimum	2.91	0.12
Range maximum	10 583.30	35.77
Range width	10 580.39	35.77

[a] The location of point estimate at the probabilistic range of risk.

SENSITIVITY ANALYSIS

Sensitivity analysis was done by calculating the contribution to variance for each exposure parameter. Contributions to variance in dioxin/furan risk assessment are shown in Fig. 1. The most important parameter in PCB risk assessment was also the chemical concentration in the soil (contribution 90.7 %). Less important parameters in PCB risk assessment were exposure duration in childhood and adulthood (contributions 4.0 and 1.3 %, respectively).

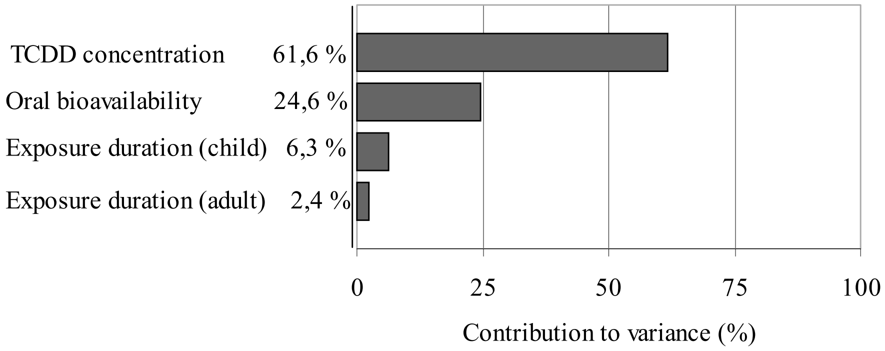

Figure 1. Contributions to variance in dioxin/furan risk assessment. Parameters contributing more than 1% are shown.

CONCLUSIONS

Relatively high deterministic estimates of risk were achieved. However, the probabilistic assessment revealed that these estimates were conservative. The 95th percentiles (for 95 % of the exposed population, the risk is smaller than this value) for both TCDD-equivalent and PCB exposure were 10^{-5}. The medians (50 % of the population experience higher risk) were two orders of magnitude smaller (10^{-7}).

Due to the lognormal shape of risk distribution, majority of the exposed population experience low risk and only a few people are at higher risk. The small high-risk minority will be protected if management decisions are made based on the deterministic assessment. This becomes questionable in cases where resources are scarce but will remain an acceptable approach if resources are not the limiting factor.

ACKNOWLEDGEMENTS

The support of Helsinki University Development Services Ltd, Environmental Projects is kindly acknowledged.

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DEVELOPMENT OF SIMPLIFIED PROCEDURES FOR THE SAMPLING OF CONTAMINATED SOILS

by
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Kimura, Takashi, Iwamoto, Hiroshi,Hatanaka, Munenori & O-hara, Junryo 2001. Development of simplified procedures for the sampling of contaminated soils.*Geological Survey of Finland, Special Paper 32, 107–113,* ten figures and 2 tables.

Three simplified procedures for sampling contaminated soils were developed. They are portable, easy to set up and can be operated under space and time restrictions. They are as follows:

a) A sampling method without using drilling mud: A sampler with a spiral cutter on its outside surface like a screw was developed. Compared with a conventional sampler not using drilling mud, this sampler needs about half the time for boring.

b) A shallow depth boring device: An electrical rotary type boring machine using the newly developed screw-type sampler was developed for obtaining soil samples from a depth of about 10 to 15 m below the ground surface without using drilling mud.

c) A simple method for surface soil sampling: This method for surface soil sampling was achieved by attaching a newly developed screw-type sampler with a smaller size to a electric hand drill.

These newly developed tools were successfully applied to investigate a TCE contaminated site together with leaching tests by using PID-gas chromatography.

Keywords: environmental geology, soils, pollution, sampling, methods, samples, drilling, Japan

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INTRODUCTION

In Japan, remedial measures to improve the water quality of lakes, rivers and coastal areas have been in place for several decades; and they have been gradually bearing fruit. However, the recognition for pollution of soil and groundwater has been slow so far. Soil pollution has only recently become a serious social problem. At present, related activity has grown to a fever pitch. In addition, investigations and remedial measures for soil and groundwater pollution are based on trial and error in many municipalities and private enterprise.

Investigations are often carried out at sites that are still in use for other purposes and where space and time are restricted. In such cases, conventional sampling methods may cause problems due to noise and vibration during the operation; they also need considerable space. In order to overcome these disadvantages, three new sampling methods were developed and are as follows.

One basic problem related to the sampling of contaminated soil is to prevent contaminated material from spreading to surrounding non-contaminated areas. For this reason, the drilling mud usually applied in conventional sampling methods should not be used. A second problem consists in the generation of heat caused by the drilling. In the case of sampling soils contaminated by volatile organic compounds (VOCs), the generation of heat should be kept to a minimum in order to prevent volatilisation. This led to the development of a sampler with a spiral cutter on its outside surface like a screw. In this way, the drilling ability was improved, and the generation of heat was decreased as the friction of the periphery surface of the sampler was decreased.

For contaminated areas, it is necessary to determine the distribution of contaminated zones with respect to depth. Soil samples have usually to be collected and analysed down to a drilling depth of several tens of metres below the surface of the ground. In this case, a rotary-type boring machine (see Fig. 3) is typically used. This method allows deep sampling in any kinds of soil, and all-core sampling is possible if an appropriate sampler, e.g. a triple tube sampler, is used. Soil samples obtained by this method have excellent quality showing neither disturbance nor shrinkage of the sample. However, since the main body of the boring machine is large and heavy, it cannot operate on narrow sites and the mobility of the machine is not good either. Moreover, the required drilling time and costs per site are high. Therefore, percussion style boring methods like the SCSC (Yamamoto 1994) and Geoprobe (PB Reports 1998) used in Europe and America for the investigation of contaminated soils have been introduced in Japan. However, because they are percussion style, problems of noise and vibration of hammer and compression of the soil samples have been reported (Suzuki 1999). To counter these problems, a simple electrical rotary-type boring machine was developed for investigation of contaminated soils. Using the newly developed screw-type sampler, soil samples from a depth of about 10 to 15 m below the ground surface can be obtained at an excellent quality without using drilling mud.

Initial investigation of contaminated sites usually requires specification of the areas of contamination by taking a large number of soil samples at shallow depths of about 1 to 2 m below the surface. It would therefore be convenient to have an easy and quick method for surface sampling. By attaching the newly developed screw-type sampler with a smaller size to an electric hand drill, such a method for easy collecting was developed.

Finally, a case study using these new tools for a soil contaminated by VOCs (TCE) is reported, together with soil leaching tests and analysis using the photo ionisation detector-gas chromatography (PID-GC) and gas detecting tubes.

SAMPLING METHOD WITHOUT USING DRILLING MUD

The drilling mud used for usual boring method has the function of preventing a collapse of the bored wall. Other functions are to remove and discharge the drilling chips, to reduce the friction resistance force, to cool of drilling bit and so on. However, drilling mud cannot be used in the case of contaminated soils, so it is necessary that there is a substitute for the drilling mud. A casing pipe with an inner diameter larger than the sampler's outer diameter may be installed in order to keep the bored wall stable. In order to compensate for the other functions of the drilling mud, a spiral cutter was added to the surface of the soil sampler, as shown in Fig. 1. With this spiral cutter, discharg-

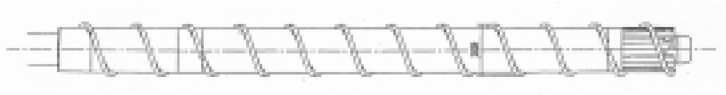

Figure 1. Sampler with a screw-like spiral cutter on its outer surface.

ing and removing of the drilling chips become easy and the generation of heat is decreased. Moreover, the penetration ability in the ground is improved by converting rotary forces into axial forces like a screw nail.

The features of this newly developed screw-type sampler without drilling mud was then tested and compared to those of a conventional sampler. The sampler was improved on the basis of a triple tube sampler system with 99mm outer diameter designed for general use. Because the height of the spiral cutter adds 5mm to either side, the outer diameter of the improved sampler is 109mm. The diameter of the collected soil sample is 71mm. The boring machine used in this sampler test is an oil feed type usually used in boring investigation, and its 100m class of boring ability is a popular type. The test ground is composed of a Kanto loam layer with an N-value of about 5 and a Narita sand layer with an N-value of about 10, shown in Fig. 2, which makes it a typical ground in the Greater Tokyo area. In the test, it was drilled to GL-10m. The situation of the test is shown in Fig. 3.

The length of sampled soil per process is basically 500mm, but lengths of 700mm and 900mm were also tested. 15 soil samples were collected in total. As a result of the test, the length of time required for drilling is only 5 minutes on average for a 500mm sampling length. In the case of 700mm and 900mm sampling lengths, the time required almost doubled. The silty clay with a gravel layer from GL-4.3m to GL-5.7m required about twice the time, but it was still possible to drill to GL-10m. However, below GL-8m the power of the boring machine proved insufficient for the casing pipe of 149.8mm outer diameter that was used to prevent the bored wall from collapsing. This indicates that the outer diameter of sampler and casing pipe should be reduced to make drilling more efficient. There was no temperature increase observed in freshly collected soil samples.

From previous boring investigation carried out near this test site using drilling mud, it was known that conventional boring using drilling mud requires about the same time for soil sampling as the screw-type sampler. Without drilling mud, it takes about the twice the time. It was further assumed that sampling lengths over 500mm would not work because of the friction resistance getting too large.

In order to assess disturbance and shrinkage of

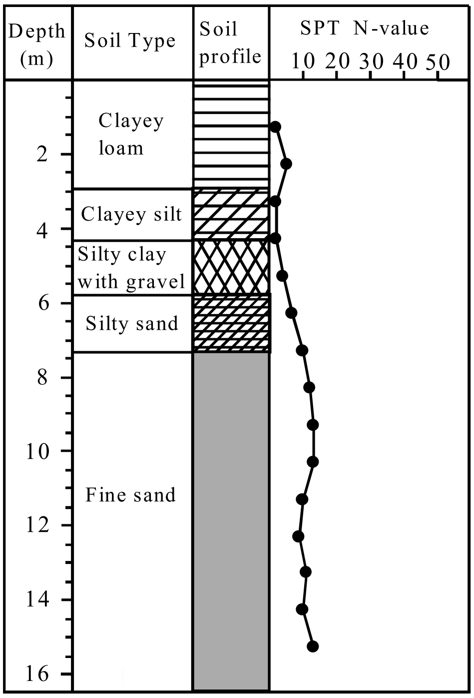

Figure. 2. Soil profile of the drilling ability test site of the screw-type sampler.

Figure. 3. Drilling ability test using the screw-type sampler (Example of a rotary-type boring machine).

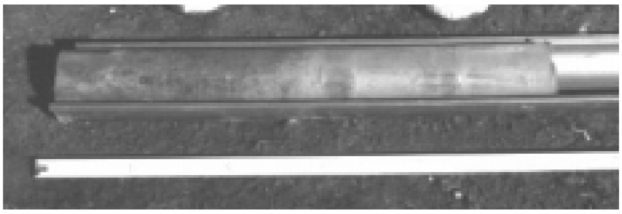

Figure. 4. Example of a soil sample collected by the screw-type sampler.

the soil samples, the ratio of the sampled soil length to drilling length was calculated. In the case of the 500mm sampling length, this sampling ratio remained 100% and hence showed no signs of shrinkage. Fig. 4 shows the example of a collected soil sample. But, in the case of the 700mm and 900mm sample lengths, the ratio varied between 93% and 100%. This smaller ratio is assumed to be caused by the inner tube rotating together with the outer tube. One reason may be that the drilling chips fill in the void between both tubes, causing greater friction than with the usual drilling mud in the void. Furthermore, the drain hole in the sampler was disturbed by the drilling chips; this may have hampered the progress of the sampling tube. This problem was solved by installing a sealing mechanism between the inner tube and the outer tube at the top of the sampler.

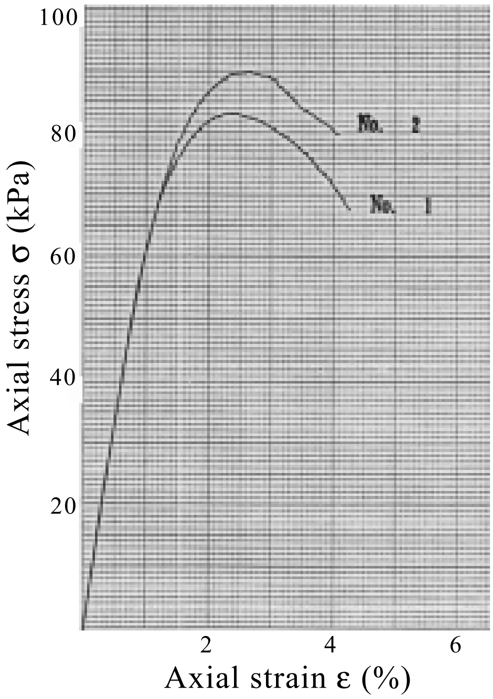

Figure. 5. Example of the uniaxial compression test result for the soil sample collected by the screw-type sampler.

Several soil tests were carried out using soil samples collected by the screw-type sampler. For the clayey soil, a uniaxial test, consolidation test, and physical test were carried out. In the case of the sand, a liquefaction test, dynamically deformation test and physical test were carried out. On the basis of these test results and the data of ealier tests, it was confirmed that the soil samples collected by the screw-type sampler were undisturbed. An example of the uniaxial compression test result is shown in Fig. 3.

By installing the spiral cutter on the surface of the soil sampler it could be shown that boring without drilling mud, which is the optimum method for investigation of contaminated soils, could be efficiently carried out. Furthermore, it was confirmed that no disturbance occurred, and that soil samples of excellent quality could be obtained. In addition, this sampler makes drilling more efficient and can also be employed in conventional boring using drilling mud.

SHALLOW-DEPTH BORING MACHINE

For the purpose of investigating contaminated soils, a simplified rotary-type boring machine was developed that allows sampling at shallow depths but with excellent mobility. The drilling rod mounted on a metal table moves up and down along a frame as shown in Fig. 6. For the trial, rotation of the rod as well as lifting of the table were powered by two electric motors of 1.0kW and 0.4kW, respectively. The inverter motor allows for smooth regulation of rotational frequency and upward/ downward velocity. Noise impact during the drilling was very small, and there was absolutely no vibration either. The machine can be handled by one person because of the wheels attached to the frame. With the described features, the overall weight of the machine was about 160kg.

Drilling tests were carried out on the loam ground with an N-value of between 3 and 5. The drilling depth was 4m. Instead of drilling mud, the newly developed screw-type sampler was used. Because of the small N-value of the sampling ground, there was no problem for drilling using only the dead

Figure. 6. View of the shallow-depth boring machine.

Figure. 7. Shallow depth boring machine mounted on a mini caterpillar.

weight of the boring machine. Soil samples at 600mm sampling length took about 3 minutes. Total time for drilling up to 4m depth required about 90 minutes.

However, once the ground became harder or the drilling bit hit small gravel, it became apparent that the counterweight of the machine's dead weight was insufficient and the boring machine bounced up. Also, although the boring machine was equipped with wheels, the moving of the boring machine was difficult on uneven grounds. To solve these problems, the boring machine was mounted on a compact caterpillar, as shown in Fig. 7. This improvement prevented the machine from bouncing; and operating on uneven grounds became easy. The result of the drilling test under these improved conditions is shown in Table 1.

Drilling mud was not used until a depth of about 6m was reached. Since there was gravel at the depth of from 6m to 7m, the drilling was changed to using drilling mud below this depth, but boring and sampling of soils was possible down to about 15m depth even at N-values of over 10. As Table 1 shows, it took about 3 to 4 minutes to collect soil samples of 700mm length, and there was no shrinkage of the sample. Two persons were sufficient for handling the drilling operation.

This confirmed that the newly developed boring machine was suitable for the purpose of the contaminated soil investigation.

Table 1. Drilling test results of the Shallow Depth Boring Machine.

Sampling Depth (m)	Soil Type	N-Value (estimated)	Sampling Time (minutes)	Sampling Ratio (cm/cm)
0.0 – 0.7	Loam	5	4	65/70
0.7 – 1.4		5	2	65/70
1.4 – 1.8	Clay	5	1	40/40
1.8 – 2.1		5	1	30/30
2.1 – 2.8		5	2	70/70
2.8 – 3.5	Clayey sand	8	2	70/70
3.5 – 4.2		11	3	70/70
4.2 – 4.5		12	5	30/30
4.5 – 5.2		12	9	60/70
5.2 – 5.5		12	6	30/30
5.5 – 5.8		14	5	25/25
6.0 – 7.0		9	6	40/100
7.0 – 7.7		10	3	50/70
9.0 – 9.7		12	5	60/70
11.0 – 11.6		9	4	60/70
14.5 – 14.9		39	3	40/40

Note: sampling ratio = sampling length/boring length

A SIMPLE METHOD FOR SURFACE SOIL SAMPLING

During initial investigation of contaminated sites, identifying the zones of contamination requires that rather large numbers of soil samples be taken at shallow depths of about 1 to 2 m below the ground surface. Hand augers, for example, have been in use for this purpose. However, drilling by manpower requires much time per sampling point as well as considerable input of labour. To overcome this, a simple method for collecting soil samples was developed by making the diameter of the screw-type sampler small enough to be installed on an electric hand drill. The sampler for trial was the improved triple tube sampler, the outer diameter being 50mm and the diameter of the soil sample 23mm. The length of the soil sample was 400mm. This sampler was installed on a 100V electric hand drill, as shown in Fig 8. Drilling and soil sampling was possible as long as the ground was not hard. However, it was impossible to drill through small gravel, and handling below a depth of 1m demanded increased efforts. However, the sampler proved effective for collecting soil samples in the initial stage of investigation. Further improvements are planned to render this sampling method more convenient.

Figure. 8. Surface soil sampling with electric hand drill.

CASE STUDY ON TCE CONTAMINATED SOILS

An investigation of a ground contaminated by volatile organic compounds (TCE) was carried out as the flow chart shows in Figure 4. Since the scope of the contamination had been previously determined by other methods, this part deals with the detailed investigation of the site.

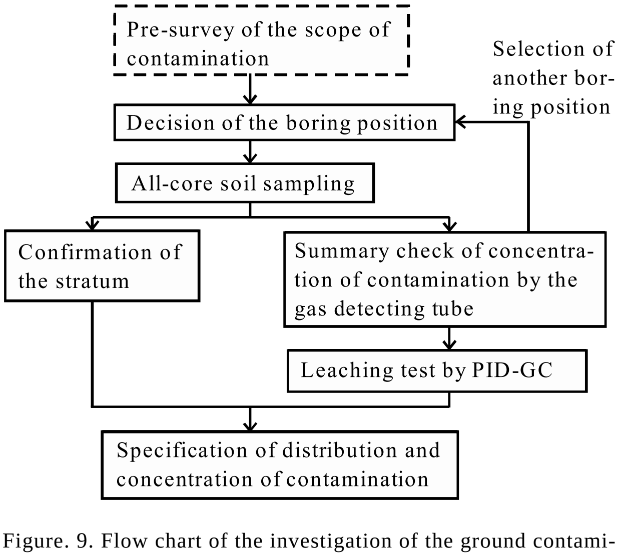

Figure. 9. Flow chart of the investigation of the ground contaminated by TCE.

First, the stratum condition was determined from all-core samples using the above-mentioned boring machine and the screw-type sampler. Simultaneously, gas-detecting tubes were used to check whether or not TCE was present in the soil samples. If soil samples were found to be contaminated, contaminated parts of the soil were picked up and subjected to leaching tests using PID-GC set up in a working hut near the site. Boring positions are displayed in Fig. 5 and the results of the leaching tests are shown in Table 2. It can be concluded that soil parts with higher concentrations found by the gas detecting tube also resulted in high values with the PID-GC analysis. At this site, the concentration of TCE exceeded the standard for environmental quality (0.03mg/l leachate or less from soils) in Japan. Because the boring machine was able to be moved easily from boring point to boring point, this site investigation could be accomplished in just one day.

Table 2. Leaching test results using Photo Ionisation Detector-Gas Chromatography (PID-GC).

Boring Number	Sampling Depth (m)	TCE by PID-GC (mg/l)	TCE by Gas Detecting tube (ppm)
B-1	0.8	n.d.	n.d.
	1.3	0.002	n.d.
	1.7	0.358	10
B-2	0.7	n.d.	n.d.
	1.5	0.230	9
	1.8	0.170	7
	2.3	1.374	42
	3.0	0.129	7
	3.6	0.002	n.d.
B-3	1.2	0.007	n.d.
	1.8	0.052	2
B-4	1.3	0.733	23
B-5	1.5	0.240	14
B-6	1.4	0.333	18
	1.8	0.009	0.5

Note: n.d. – not detected

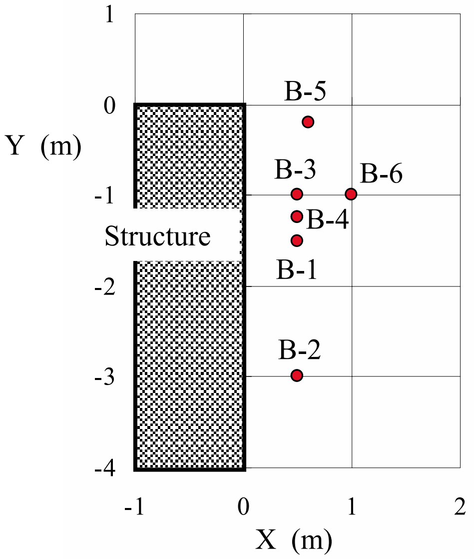

Figure. 10. Position of boreholes at the investigated site.

CONCLUSIONS

The three newly developed methods for investigating contaminated soils reported here have proven to be very effective tools. Future improvements remain to apply these methods to various kinds of ground, e.g. sandy ground and harder ground, and to further rationalise their handling.

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AIR INJECTION WELLS – JUST ANOTHER PUMP AND TREAT METHOD OR A COST EFFECTIVE, INNOVATIVE TOOL FOR *IN SITU* REMEDIATION?

by
M. Luber¹⁾ and J. Brauns, J.²⁾

Luber, M & Brauns, J. 2001. Air injection wells – just another pump and treat method or a cost effective, innovative tool for *in situ* remediation? *Geological Survey of Finland, Special Paper 32, 115–121,* seven figures and 1 table.

The remediation of groundwater has been one of the most urgent tasks concerning environmentalism for several years. Experience has shown that heterogeneous aquifer structures and sorption onto soil material are the reasons for long decontamination times. An Air Injection Well (AIW) is a simple and low cost method for the remediation of dissolved volatile organic compounds and for the enhancement of natural attenuation, e.g. the support of biodegradation by generating an aerobic environment in, or by adding nutrition to, the saturated zone. The possibilities for the application of an AIW are illustrated by means of two large scale experiments and one field application. The Advantages and disadvantages of the AIW are discussed.

Keywards: environmental geology, ground waters, pollution, volatile organic compounds, remediation, methods, air stripping, Germany

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INTRODUCTION

The remediation of the groundwater, i.e. its decontamination, has been an aim that has turned out to be very costly and time intensive. After numerous discussions, two groups were formed with different approaches to the whole problem; on the one hand, the complete decontamination and restoration of the environment was the one and only aim. On the other hand, there was a proposal to leave contaminated sites as they are and trust in natural (and cheap) degradation processes to solve the problem in the near future.

The combination of these two objectives seems to be the only solution accepted by a majority of people. But this will only succeed if low cost and effective techniques can be found for the remediation. Experience has shown that an AIW seems to meet these different requirements for certain contaminants (such as volatile DNAPLs and aerobe biodegradable compounds).

AIW PRINCIPLES AND CIRCULATION FLOW

An AIW is a combination of two common construction or remediation techniques. These include an air lift pump and a strip reactor. With an air lift pump, water can be pumped upwards in a vertical tube using pressurised air as the energy supply. The air is injected at the base of the tube. Due to the buoyancy, the air moves upwards and transports water in the same direction. In the case of an AIW, the well tubing is screened below the water table. The aim is not to pump the water out of the aquifer but to create a groundwater circulation area in the aquifer (Fig.1). The driving force of the groundwater flow is the difference in hydraulic head between the AIW and the surrounding aquifer. It is induced by the air injection.

The second technique, stripping, is a common method used for removing volatile compounds from water. The contaminated water is aerated and because of the difference in equilibrium concentrations the volatile compound is transferred to a certain amount into the air.

For these reasons the injected air is not only the driving force for the groundwater circulation but also acts as a stripping medium.

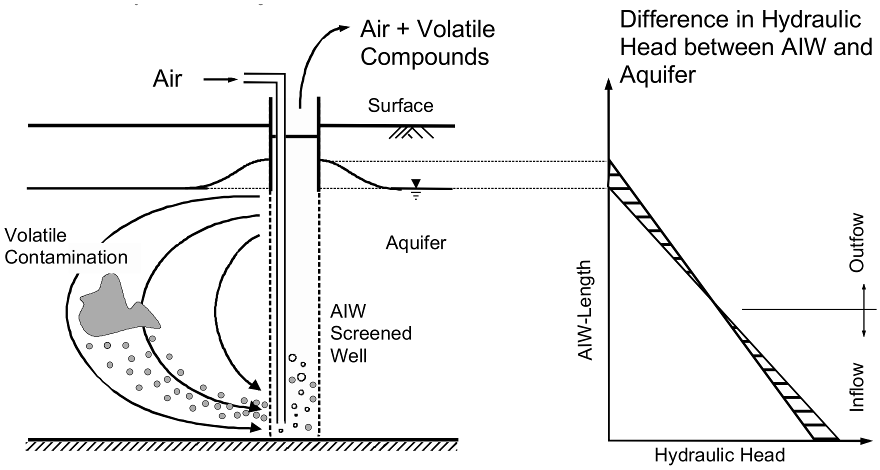

Figure 1. AIW and the differences in hydraulic head between the well and the surrounding aquifer due to air injection.

AIW COMPONENTS

The main part of an AIW structure is the well itself. It normally consists of a screened metal tube with a diameter of 2”. Assuming there are suitable underground conditions, structures of this diameter can be driven into the soil, whereas larger wells always have to be drilled. This means there is the advantage of simple, cost-effective installation of the AIW or, if necessary, of several AIW, if the efficiency has to be increased. This should be the preferred strategy instead of using one AIW with a larger diameter.

The upper part of the AIW consists of a normal tube in order to avoid the loss of possibly contaminated air into the vadose zone.

At the upper end of the AIW, the air injection pipe passes through the well tube and is lowered down to the lower end of the AIW, where the air injection takes place.

A compressor is necessary to produce the pressurised air. Additional periphery components are: water, a separator, activated carbon filters or other cleaning devices and a flow meter. In recent years, theoretical and experimental work has been done to investigate the functional principles and to determine design rules regarding the hydraulic and required air injection rate. They can be found in Luber (1999).

LARGE SCALE EXPERIMENTS

The first full scale application of AIW took place in two remediation experiments under controlled conditions at the so-called VEGAS facility in Stuttgart, Germany. Two aquifer models, each with a different subsoil structure, were installed in vessels there. The utilised aquifer materials in both experiments were coarse-to-medium sands with a hydraulic conductivity between 3.2 and 34 x 10^{-4}m/s (see Fig. 2).

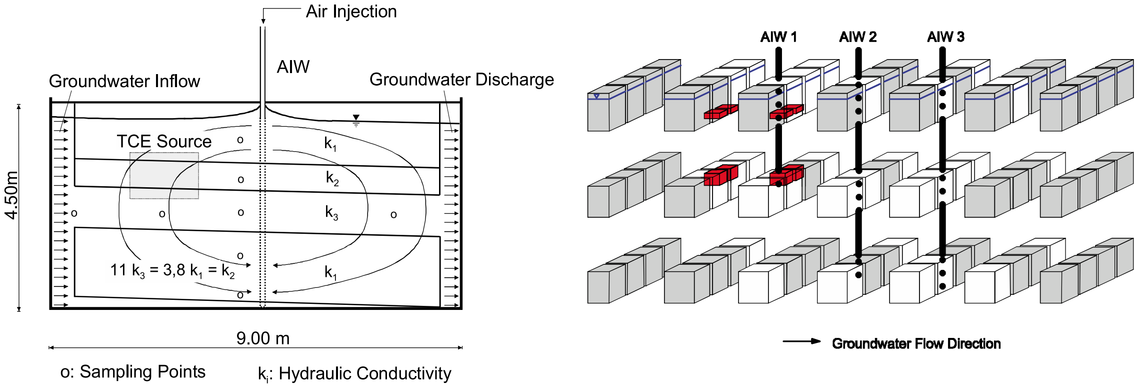

Fig. 2. Set-up of the artificial aquifers at the VEGAS facility Left: Vertical cross section of the layer-model, k3=3.2 x 10^{-4}m/s Right: For better visualisation open composition of the block-model; approx. volume of each block: ~ 2.8m+; grey coloured blocks: k=12 x 10^{-4}m/s; others: k=34 x 10^{-4}m/s

In each of the models with a size of 4.5mx6mx9m (heightxwidthxlength), two remediation experiments with a duration of several months were carried out. In both cases, the aquifers were contaminated with around 30 kg Trichlorethylene (TCE). The volatile organic compound was placed in residual saturation in a soil volume of approximately 1.6 m+. The unavoidable loss of TCE during the contamination was measured and taken into account in later considerations.

Through edgeways-placed vertical distribution layers, the aquifers could be fed with groundwater (Q-controlled). On the other side of the vessels the water flowed through a similar layer into a water treatment plant. The remediation experiments were carried out with different groundwater flow rates (Table 1).

The relatively high groundwater velocities were necessary in order to avoid any influence of the vessel walls on the hydraulics of the AIW. The lower velocities were chosen for the examination of the AIW capability in different groundwater flow situations.

The main difference between the two experiments was the number of AIW used for the remediation process (besides the different aquifer structures). In the first experiment, that was carried out on the layer model, the focus was on the determination of the efficiency of one AIW only. The second experiment on the block model, should

Table 1. Average groundwater velocities at different times after the beginning of the experiments

Layer Model		Block Model	
Time (d)	Velocity* (m/d)	Time (d)	Velocity* (m/d)
0 – 24	0.55	0 – 8	1.20
24 – 62	0.10	8 – 66	0.30
62 – 145	0.45	60 – 70	0
		70 – 111	0.30

*Averaged Darcy velocity of the inflow cross section

evaluate the benefit of the use of more than one AIW.

Immediately after the groundwater flow was started, the remediation activities began. The TCE-concentrations of the AIW discharge (air) and of the groundwater discharge were measured continually in order to balance the mass removal through from the aquifers. The results of these determinations are shown in Fig.3.

In both cases, a significant amount of TCE was lost through the groundwater discharge. But this loss occurred only at times of relatively high groundwater velocities (high gradients of the groundwater flow). Regarding those periods of time, where the groundwater velocities where low, almost all the TCE mass was removed from the aquifer by the AIW. In addition, the efficiency of the 3 AIW in the block model was much higher, even though the groundwater flow was all together higher in the block model than in the layer model.

Increasing the groundwater flow again after 62 days in the layer model resulted in a short-term TCE mass removal only. This was probably due to the dissolved TCE mass during the lower groundwater flow. Since the AIW still removes TCE at significant rates during times with low groundwater flow rates, whereas the mass loss through the groundwater outflow is very low, one can see the comparatively higher remediation capability of the AIW at lower groundwater flow rates. Not until the groundwater flow is increased again does the contaminant removal by the AIW drop close to zero. A total TCE mass of 23.0 kg was removed from the layer model. Sampling the aquifer material after the experiment gives reason for the assumption that all of the TCE input was removed in the one or other way from the aquifer. That suggests a loss in the mass balance of 4.4kg (16.4 %) TCE, which is a very good result taking the scale of the experiment and the compound characteristics into account.

The increase of the groundwater velocity or the shut down of the experiment at times in the block model, did not show any effect on the removal of TCE at all, since most probably no more TCE was in the aquifer at this time. Due to the higher hydraulic conductivity, the total TCE mass removal took place comparatively quickly. After 66 days no significant amount of TCE was removed from the aquifer via the AIW or the groundwater discharge. The mass balance in the block model resulted in a mass loss of 25.4 %, which still is a fairly good result.

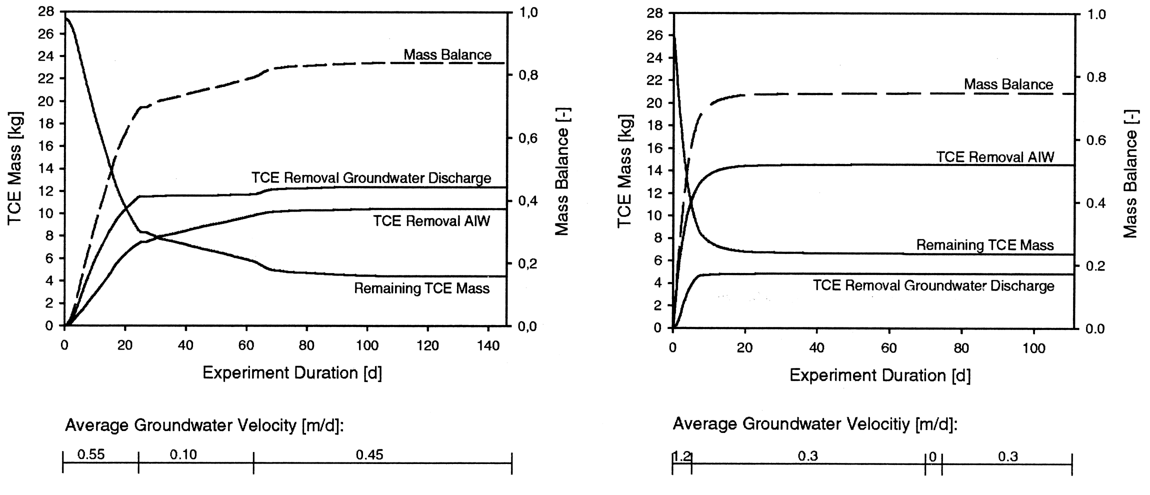

Figure 3. TCE removal and mass balance from the layer model (left) and the block model (right)

FIELD APPLICATION

At the large-scale experiments at the VEGAS facility the emphasis of the AIW application was on the stripping of the volatile contaminant. But in this case the aim is to distribute a compound – namely oxygen – in the aquifer, in order to facilitate the microbiological degradation of the contaminants in the subsoil.

On site, organic lead compounds (Tetraethyllead, TEL) had been produced in the past. The TEL accidentally infiltrated into the vadose zone together with hydrocarbons that were used in the production processes. Because of the density of the mixture, the phase liquid spread on the groundwater surface. Due to the evaporation of the volatile compounds of the hydrocarbons, the mixture became more and more viscous and was distributed over long periods with the natural movement of the water table in a layer with a thickness of 2 to 3 m. Since the water table has risen in recent years, the contaminants are more or less below the water table nowadays.

The TEL is transformed to mineral lead in the presence of oxygen via several steps (which include Tri- and Diethyllead). This was proved in small scale laboratory tests by Mulroy & Ou (1998). Until now, it has not been clear whether the organic lead is degraded microbiologically or if it is transformed by hydrolysis.

Due to the low concentrations of the organic lead compounds, the hazardous influence on the environment is removed as soon as the organic lead is transformed into mineral lead, which is immobile.

The subsoil at the site is formed by fluvial deposits, mostly sand with a hydraulic conductivity of 3.5×10^{-4} m/s. The natural gradient of the groundwater flow is comparatively low with $i = 1‰$. The whole aquifer thickness is about 50 m, but as already mentioned, contamination is found a little below the water table which is located at a depth of 3 to 4 m.

In order to saturate the groundwater in the relevant zone, 4 AIW with a screened length of 4 m below the water table were installed on the site at a highly contaminated spot. The distances between the AIW were 5 m. A ground plan with the AIW and monitoring wells is given in Fig. 4.

Even though TEL is a volatile organic compound, the emphasis of the AIW application in this case was not so much on the stripping of the contaminants but on in the supply of the relevant soil volume with oxygen. The oxygen concentrations in the groundwater at different times after start up are presented in Fig. 5.

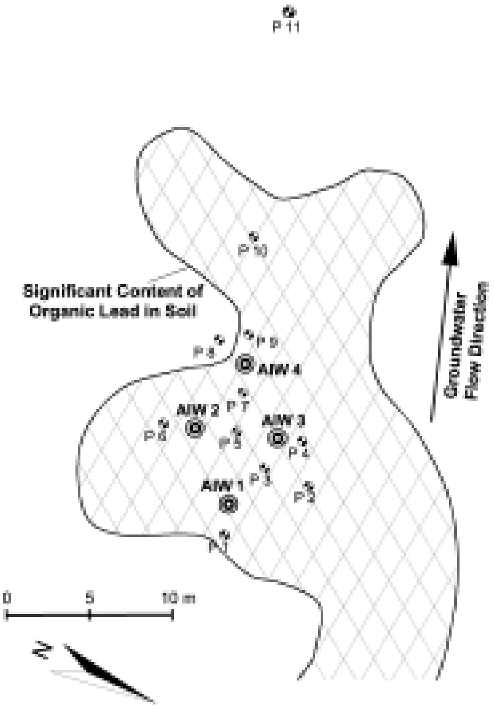

Figure 4. Ground plan of the site with AIW, monitoring wells and distribution of the organic lead contamination

In the first four weeks, only one AIW was operated in order to monitor its influence. Then the oxygen concentrations increased approximately by one order of magnitude starting from values around 0.4 mg/l. The range of influence seemed to be about 4 m (radius), showing that the number of monitoring wells was too small for an exact determination. With regard to the results of the oxygen measurements after 63 and 272 days, the oxygen concentrations increased continually, even though a certain amount should have been consumed for the degradation of the organic lead compounds.

The degradation of the TEL started shortly after the beginning of the AIW operation. Whereas the low soluble TEL had appropriate low and more or less constant concentrations before and after the operation of the AIW, the content of the more soluble metabolic compounds increased significantly due to the changed flow situation around the AIW and the degradation process (Figs 6 and 7). Most probably formerly dead end pores now release some quantity of stored organic lead, that is degraded during the further progress of the AIW operation. This results in higher concentrations of Tri- and Diethyllead initially. The continuing supply of oxygen decreases the amount of all the compounds. Taking P 5, which is positioned in the centre of

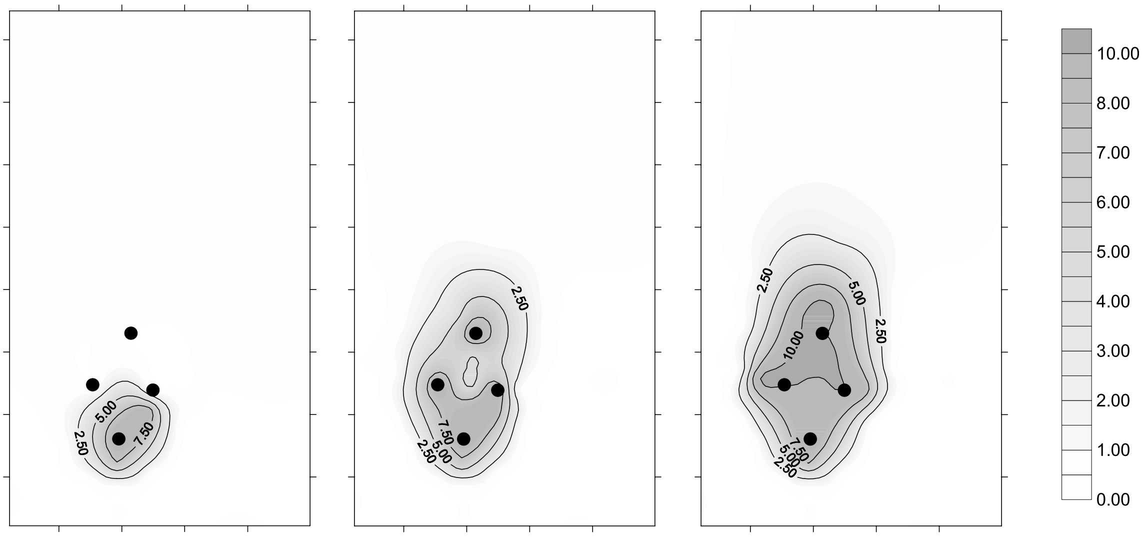

Figure 5. Oxygen concentration [mg/l] in the groundwater adjacent to the AIW at three different times (left: 27 days, one AIW only; middle: 63 days, operation of all four AIW for 36 days; right: 272 days, four AIW for 245 days; distance between two ticks: 5 m)

the four AIW, as an example, this phenomena can be followed up in Fig. 6.

The other monitoring wells showed similar reactions. The development of the total organic lead (that consists of Tri- and Diethyllead and a negligible amount of Tetra- and Monoethyllead) in the groundwater in three monitoring wells is shown Fig. 7. After the increase in the concentration in the first 100 days, the concentrations dropped to a certain level which is a little higher than originally – apart from an increase during the last but one measurement, which cannot be explained at the moment. It is possible that higher rainfall caused an infiltration of organic lead from the vadose zone. The more or less constant concentrations of the organic lead in water indicate a constant degradation process from the TEL (low soluble) via the Tri- and Diethyllead. A further reduction of the concentrations can be expected at the time as the supply from TEL decreases; that is, the main part of the low soluble TEL, that acts as a storage for the metabolic compounds, is degraded.

The organic lead concentrations downstream of the natural groundwater flow direction (monitoring wells P 10 and P 11; see Fig. 4) show no significant reaction, although the flow time between the AIW and the wells had long been completed, which was demonstrated by a tracer test. So far, this circumstance is the most definite indication for the degradation of the organic lead, since other parameters (germ population, CO_2 evolution) did not change in a significant way. Due to a huge heterogeneity of TEL concentrations in the soil at short

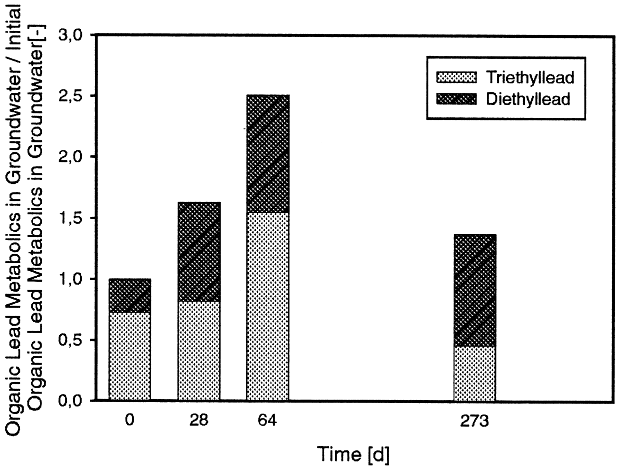

Figure 6. Concentration of Organic Lead Metabolics in Groundwater at Monitoring Well P 5.

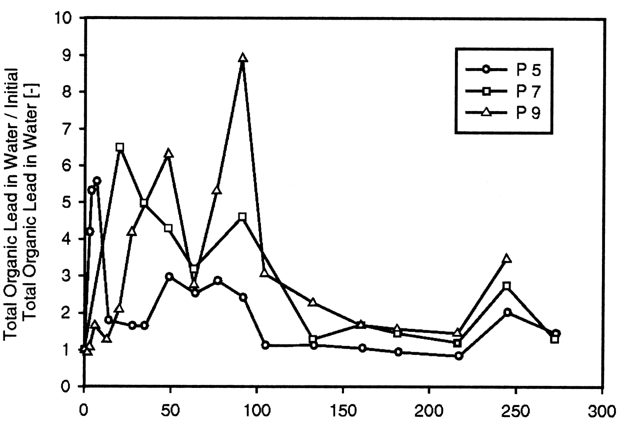

Figure 7. Total organic lead concentrations in the groundwater versus time

distances, this data cannot be interpreted at moment.

For the enhancement of the degradation process, two steps will be taken in the future: a supply with nutrients is being considered and additional AIW will be installed.

PROS AND CONS

The remediation experiment at the VEGAS facility in Stuttgart with the AIW was the first application under controlled conditions on a true scale. The AIW showed good results in removing TCE from the aquifer in the low groundwater flow situation. If the groundwater flow increases, a considerable amount of TCE mass is “lost”. A possible solution to this problem could be the installation of further AIW in order to increase the removal capability.

The application of the AIW in the field showed very good results with regard to the oxygen supply of the groundwater. It can be predicted that the distribution of liquid substances in the groundwater zone close to an AIW would work in a similarly satisfactory manner. Some factors point towards the ongoing degradation of the organic lead.

Summarising the advantages (+) and disadvantages (–) of the AIW on the basis of:

– the large scale experiments carried out at the VEGAS facility, Stuttgart, where the stripping effect for volatile compounds was in the foreground

– the field application emphasising the capability of the AIW to support a degradation process (natural attenuation)

one ends up with the following points:

+ The AIW are produced with conventional parts, and due to their simple construction, the investment is exceptionally low. The installation of the four AIW in the field application cost Euro 14 000,-, including the compressor and the drilling.

+ Operation costs are low too because of the use of injected air as a stripping medium as well as a driving force for the induced circulation flow.

+ In terms of the “tempered” hydraulic behaviour of the AIW, the costs for the investigation of the site can be reduced significantly.

+ Due to the low costs and the simple installation it is easy to install several AIW according to the actual extent of the site and the subsequent progress of the remediation process.

+ The application of the AIW is not limited to volatile organic compounds; it can also be applied for the support of biodegradation.

+ The distribution of compounds into the aquifer (oxygen, liquid nutrients) works very well. It is carried out in a controlled and predictable manner.

+ The removal capacity of the AIW is good concerning low concentrations, low groundwater flow gradients and shallow aquifers.

– However, the AIW cannot be recommended for deep aquifers, high concentrations or high gradients of the natural groundwater flow (concerning the stripping of volatile contaminants).

– Remediation of compounds takes a relatively long time due to the gentle hydraulic activity of the AIW.

– High concentrations of iron in the groundwater necessitate the use of nitrogen instead of air as a process gas. This increases the costs.

– The hydraulic conductivity of the aquifer should be higher than 10^{-6} m/s.

Taking these points into consideration and bearing in mind the limitations, the method can be regarded as innovative, simple and applicable for a variety of different cases. However, like every other method, the application of the AIW cannot be recommended in all cases. The presence of high natural groundwater gradients seems to be the most important restriction; by contrast, the low costs for operation and installation are probably the biggest advantage.

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POLYCHLORINATED BIPHENYL (PCB) CONTAMINATION OF APARTMENT BUILDING AND ITS SURROUNDINGS BY CONSTRUCTION BLOCK SEALANTS

by
Sannamari J. Hellman and Jaakko A. Puhakka

Hellman, Sannamari J. & Puhakka, Jaakko 2001. Polychlorinated biphenyl (PCB) contamination of apartment building and its surroundings by construction block sealants. *Geological Survey of Finland, Special Paper 32, 123–127,* four figures.

Because of their superior properties polychlorinated biphenyl (PCB) containing sealants were used in the construction industry in the 1960’s and 1970’s. The PCB content and distribution of individual PCB-congeners in sealant and soil samples were determined using gas chromatography (GC) with selective-ion monitoring mass spectrometry (MS-SIM). In the present case study, the total concentration of extractable PCBs in sealant samples ranged from 8 300 mg/kg to 108 700 mg/kg. The concentrations were highest on the northern side of the building studied. In soil samples the concentrations ranged from 0.03 mg/kg to 6.62 mg/kg. The concentrations were the highest in the immediate vicinity of the building and declined with distance.

The risk assessment carried out was based not only on the total PCB concentrations but also on the congener specific analyses. Toxicity of specific congeners was estimated on the basis of 2,3,7,8-TCDD equivalents. The highest deterministic risk value with congener specific information and exposure pathway as soil ingestion was 4.90 x 10^{-5}. Using probabilistic assessment along with lognormal distributions for the parameters the 95 percentile value for the toxicity risk was 3.02 x 10^{-7}. For the purpose of this study, only PCB exposure by dermal contact and ingestion of soil was evaluated. There was no significant toxicity risk to the children.

Keywords: buildings, construction materials, PCBs, soils, pollution, risk assessment, toxicity, Tampere, Finland

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INTRODUCTION

Several million kilograms of polychlorinated biphenyls (PCBs) have been used worldwide since their invention in 1929. Since then millions of kilograms of PCB have been released into the environment (Zwiernik *et al*. 1998) and adsorbed in organic soils and sediments where they still persist (Bedard & May 1996). Because of their chemical properties PCBs are extremely difficult for living organisms to excrete (Lanting et al. 1998). Thus they tend to bioaccumulate in the food chain. Despite their ban in the1970s, PCBs have still been found in the environment for decades afterwards.

Sediments are the main sink for PCBs (Hansen 1987). In Finland, for example, PCB- contaminated lake sediments have been found to be very persistent (Hurme & Puhakka 2000). A new type of PCB source for environmental contamination has been observed from apartment building houses (Jansson et al. 1997, Pyy & Lyly 1998, Hellman 2000). PCB mixtures were used as sealing paste additives in prefabricated constructions from the 1960's to the late 1970's. PCBs may pose a risk to people, especially to children living in houses with PCB-containing sealants.

The aim of this work was to study the problems in apartment buildings in Tampere. The concentration and composition of PCBs in sealants and in soil next to the building were determined followed by toxicity risk assessment.

MATERIALS AND METHODS

Site description and sample collection

The building studied was located in the city of Tampere, Finland. It was built in 1971 and had the original sealants. Sealant samples were collected from each quarter of the building using cluster sampling. Soil samples were taken using systematic grid sampling. The distances were 0.1 m, 0.5 m, 1 m, 2 m, 5 m and 10 m away from the vertical joint of elements.

Extraction of soil and sealant samples

Hexane was used as a solvent with the sealant samples. Samples were cut into small pieces and placed in 15-ml test tubes sealed with teflon lined rubber septas and screwed-caps. The solvent was added and the test tubes were mixed with a vortex mixer. The test tubes were placed into an ultrasonic bath for one hour. The procedure was repeated twice. Finally, the elutes were diluted with hexane.

Hexane-acetone (1:1) was used as a solvent for the soil samples. The pretreated soil samples were placed into test tubes. The solvent was added. After vortex mixing, the test tubes were placed into an ultrasonic bath for one hour. The tubes were centrifuged and the elutes were transferred to clean test tubes. The procedure was repeated twice. Acetone was removed from the extracts by reverse partitioning into distilled water. The remaining hexane fractions were concentrated under nitrogen stream and passed through a Florisil solid-phase extraction column.

PCB analysis

PCBs were analysed in a HP 6890 Series GC equipped with a HP-5MS (length 30 m, diameter 250 μm, film thickness 0.25 μm). Helium served as the carrier gas (1 ml/min). The initial oven temperature was 80°C from which it was ramped at 30°C/min to 150°C, 5°C/min to 250°C, 15°C/min to 300°C and finally held for 1 minute. PCB-congeners were identified by using Aroclor 1260 mixture as an external standard. The twenty-three different PCB-congeners were used in quantitative analysis.

The concentrations of individual congeners in standard samples were calculated from the total PCB content and the weight-% distribution of congeners in the Aroclor 1260 mixture according to Schulz *et al*. (1989). The relationship between the response area and concentration was determined. The total PCB content for each sample was the sum of the peak concentrations.

Risk assessment

The risk assessment considered children from 1 to 6 years of age who are exposed to surface soil while playing on the ground. According to the U.S. EPA (1996) procedure, cancer dose-response assessment was performed. The parameters were chosen from the U.S. EPA's Exposure Factors Handbook (U.S. EPA 1997). In quantitative risk assessment the deterministic approach, which uses a single point value for each estimate of the input variables, thus yielding a single output value, was used. This seldom represents a real life situation. Most of the input variables are random variables. If the input variables are random then the output variable is random as well.

The probabilistic approach was used to incorporate the uncertainty into the exposure assessment calculation. Using commercial software (Crystal Ball with Monte Carlo simulation) the distribution of the health risk for the case study was estimated. The distribution was compared to the point estimate of the risk. The 95th percentile value was estimated.

In order to perform a specific risk assessment the toxicity of specific congeners was estimated on the basis of 2,3,7,8-TCDD equivalents. The total PCBs, but also the congener specific results, were included in exposure assessment. Exposure parameters having the greatest impact on the result were identified.

RESULTS AND DISCUSSION

Quantification of PCBs

The sealant samples were analysed for PCBs and the results are presented in Fig. 1. Total concentrations of extractable PCBs in the sealant samples ranged from 8 300 mg/kg to 108 700 mg/kg. The PCB concentrations were highest in samples from the northern side of the building. This indicates that the climate conditions, including sunlight, have less effect on PCBs in the sealants on the northern side of the building.

The distribution of PCBs in the surroundings of the building were also studied. The total PCB concentrations in the soil samples ranged from 0.03 mg/kg to 6.62 mg/kg. The PCB concentrations were highest adjacent to the building and declined with distance. Fig. 2 shows an example of a sampling line of soil samples on the southern side of the building.

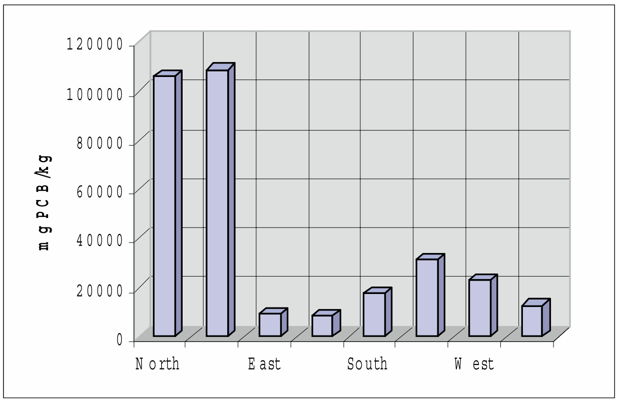


Figure 1. The PCB concentrations in apartment building sealant samples.

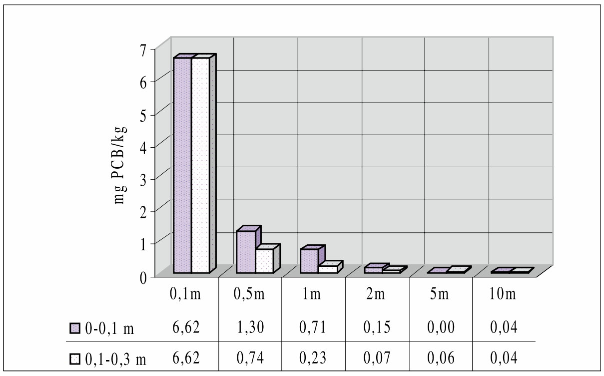


Figure 2. The PCB concentrations of the soil samples collected from the southern side of the apartment building.

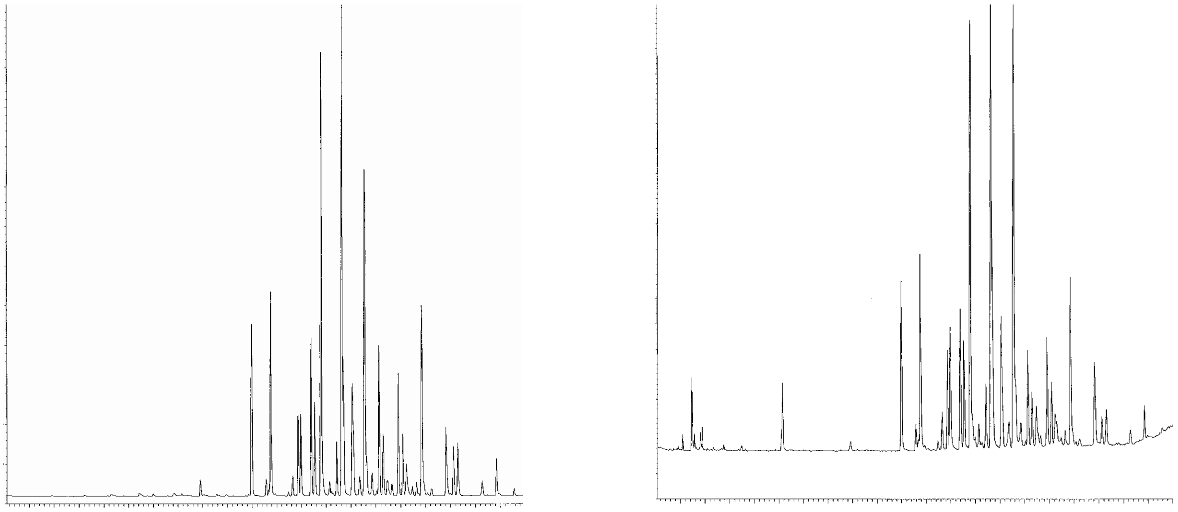


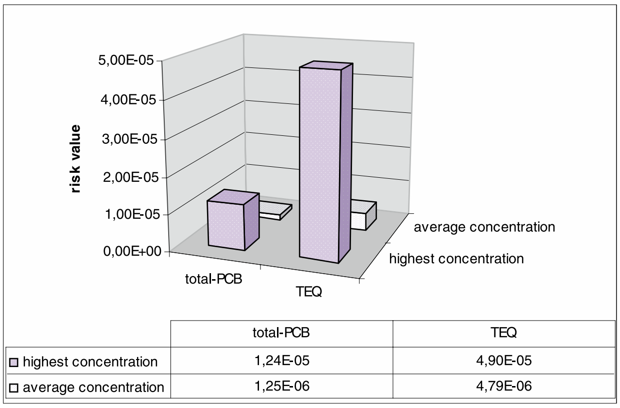
Figure 3. Ion chromatograms for a soil sample (on the right) and the Aroclor 1260 –standard. The scale in Y-axes is more accurate with the soil sample than with the Aroclor standard that causes the background to be higher on the right hand side ion chromatogram.

The composition of the PCB product did not change significantly over the years in the sealant; the peak patterns followed closely that of Aroclor 1260. Examples of the ion chromatograms of one soil sample and the Aroclor 1260 standard are shown in Fig. 3.

The most toxic coplanar congeners (IUPAC Nos. 77, 126, 169) were studied but not detected. The most persistent congeners (IUPAC Nos. 153, 180) differentiated well in all ion chromatograms.

The results from the risk assessment when exposed by soil ingestion are shown in Fig. 4. Fig. 4 shows the difference between the risk assessment based on the total PCBs and the one calculated with the congener specific information.

The highest risk value with the congener specific information and the exposure pathway as soil ingestion was 4.90 x 10^{-5}. Using lognormal distributions for the assumed parameters the 95th percentile value for the toxicity risk was 3.02 x 10^{-7}. These results show that there is no significant risk to the children in this case.



	total-PCB	TEQ
☐ highest concentration	1,24E-05	4,90E-05
☐ average concentration	1,25E-06	4,79E-06

Figure 4.The risk values for the children from 1 to 6 years of age when exposed to PCB by soil ingestion from the apartment building surroundings.

A risk based cleanup goal was calculated using the case specific parameters. The risk based cleanup goal in the case was 0.56 mg PCB/kg. It is very close to the given limit value for PCBs in soil (0.5 mg PCB/kg), which shows that the limit value is applicable in the case.

CONCLUSIONS

Apartment building sealants may contain high concentrations of PCBs. The sealant itself used in the case study building was classified as hazardous waste.

Apartment block PCBs are spread from the construction into soil next to the building. In this case study the concentrations in soil were lower than those in sealants but exceeded the given limit value for PCBs in soil (0.5 mg PCB/kg).

The case study building did not exhibit a significant toxicity risk to the children. The risk assessment was based on the specific exposure parameters (*e.g.* exposure factors and pathways, average weight of the children, average soil ingestion volume) which were then approximated by probability distributions found in the literature. The most important parameter for the assessment was the PCB concentration of soil (56 %). The assessment was case specific and the results do not apply to all cases. With good site-specific knowledge the results from the risk assessment can be improved.

The risk assessment protocol for apartment building sealants and soils contaminated by these materials should not be based on the total PCB. The congener specific information with chosen standard congeners is needed for the assessment. How detailed the required information is and how accurate the assessment should be depends highly on the case but the basic rules should be open to discussion.

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Geological Survey of Finland, Special Paper 32, 129–133, 2001.

ADVANCED OXIDATION OF PCDD/FS, PCBS AND PAHS IN CONTAMINATED SOIL/ASH MATRIX

by
Tuula Tuhkanen

Tuhkanen, Tuula 2001. Advanced oxidation of PCDD/Fs, PCBs and PAHs in contaminated soil/ash matrix. *Geological Survey of Finland, Special Paper 32, 129–133,* one figure and 3 tables.

Two different, innovative remediation techniques were compared for the treatment of cross-contaminated incinerator ash, which contained polychlorinated dibenzo-dioxins and furans (PCDD/Fs) and polychlorinated biphenyls (PCBs). The goal of the treatment program was to achieve the proposed Finnish guideline levels for both compounds (PCDD/Fs, 20 ng/kg and PCBs, 50 µg/kg). The amount of oxidants needed and the total treatment costs were estimated.

Ozonation in alkaline conditions and Fenton treatment were selected in laboratory-scale experiments. When the initial PCDD/Fs concentration was 240 ng/kg measured in International toxicity equivalents (I-TEQ) and PCB concentration was 395 µg/kg, ozonation removed 50 percent of the PCDD/Fs and 40 percent of the PCBs. Fenton treatment removed 36 percent of the PCDD/Fs and 6 percent of the PCBs. Even though the proposed Finnish target levels were not reached in these experiments, ozonation under alkaline conditions appears to offer a viable alternative for the treatment non-biodegradable compounds in the soil matrix.

Key words: soils, ash, polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans, PCBs, remediation, oxidation

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INTRODUCTION

A variety of organic compounds are refractory, toxic or inhibitory in nature. They can not be treated by conventional biological treatment methods. The existing method for the treatment of hazardous substances such as polychlorinated dibenzodioxins and furans (PCDD/Fs) and polychlorinated biphenyls (PCBs) has been excavation and off-site treatment by incineration or encapsulation (LaGrega *et al.* 1994). Some innovative treatment methods including advanced oxidation techniques (AOTs) have been studied recently for the decomposition of such refractory compounds, with promising results (Watts *et al.* 1991). Removal efficiencies higher than 90% for PCDD/Fs have been achieved in laboratory scale experiments when dioxin-spiked or soil-free samples are used (Isosaari *et al.* 1997; Watts *et al.* 1991)

Advanced oxidation techniques are ambient temperature processes for the treatment of contaminants, which involve the generation of highly reactive intermediates, usually hydroxyl radicals (Glaze *et al.* 1987). Hydroxyl radicals react with most organic compounds with rate constants of 10^7 to 10^{10} $M^{-1}s^{-1}$ (Buxton *et al.* 1988). The list of various AOTs is presented in Table 1. Some of these AOT combinations are suitable for the in-situ or on-site treatment of contaminants in soils.

Table 1. Advanced oxidation techniques

Ozone	UV	
Ozone	alkaline pH	
Ozone	Hydrogen peroxide	
UV	Hydrogen peroxide	
UV-vis	TiO_2	
Fe^{2+}	Hydrogen peroxide	Fenton's reagent
e- beam		

MATERIALS AND METHODS

Soil matrix studied

The aim of this project was to investigate innovative chemical oxidation techniques for the risk reduction of a PCDD/F- and PCB- contaminated site. The ash/soil material originated from an old landfill site containing bottom ash from a waste incineration plant. The soil/ash matrix contained about 9–12 % organic material, and high concentrations of metals including iron, lead and zinc. The pH of the material was slightly more than 8.

The concentration of PCDD/Fs varied between 200–300 ng/kg I-TEQ. The PCB-concentration was 310–480 µg/kg in the study area. Chemical analyses were conducted by the National Public Health Institute, Kuopio, Finland. The proposed limit values for PCDD/F and PCBs are 500 ng/kg (I-TEQ) and 500 µg/kg of dry weight, respectively. The target values for PCDD/Fs and PCBs are 20 ng/kg and 50 µg/kg (Assmuth 1997), respectively. The contamination of these compounds in the sampling area was between these two guideline values.

Chemical analyses

The samples were treated with 1 M HCl and dried and then extracted in an ultrasound bath in toluene, which was then replaced with hexane. The samples were purified in silica, activated carbon and aluminumoxide columns. The PCDD/F standards used contained sixteen PCDD/F congeners, which were ^{13}C labeled. The PCDD/F congeners were analyzed by a Hewlett-Packard 6890 gas chromatograph equipped with a high resolution mass selective detector (VG 70-250SE) which was operated on a single ion recording (SIM) mode. The separation column was J&W Scientific DBDXN. The method used at the National Public Health Institute has been accredited. The detection limit for a single PCDD/F congener is 0.5 ng/kg in and for PCB congeners 50 ng/ kg of dry weight.

Treatment techniques

In this study the following AOTs were used: Fenton treatment (Fe^{2+} + H_2O_2 in acidic pH) and ozonation of ash/soil slurry in alkaline pH. Experiments were conducted at laboratory scale.

Fenton treatment

Thirty grams of contaminated material were collected from well-mixed sub-samples taken from the contaminated area. The pH was adjusted to 3 using 4 M H_2SO_2. Since the ash contained 46 g/kg total iron, no addition of $FeSO_4$ was needed. At the desired solution pH, sufficient Fe^{2+} ions should be present. Thirty-four ml of 30 % hydrogen peroxide was added in three replicated doses at 2-hour intervals. The amount of reagents selected was based on the experiments reported for pentachlophenol spiked soil (Kwon *et al*. 1999, Watts *et al*. 1990).

Ozonation

Ozonation of ash was conducted in a gas washing bottle. Ozone was generated from pressurized air with a Sander Ozonizer Molel 200. The ozone concentration in the gas stream was measured by potassium iodine method (IOA 001/87). The pH of the sample was adjusted from the initial pH 8.1 to 10 using CaO. The extent of ozonation and the ozone dose required was estimated by using the organic matter content of the sample. Sampling took place when the ozone dose reached the value 0.5g of ozone to one gram of organic matter in ash.

RESULTS AND DISCUSSION

The initial concentrations of identified congeners are presented in Table 2.

The PCDD/Fs in this material consisted mainly of dioxins. The PCDD/PCDF ratio was 3:4, which

Table 2. The initial PCDD/Fs concentrations and the reduction efficiencies in Fenton treatment (1) and in the ozonation under alkaline conditions (2). The sum of tetra, penta, hexa, and octa forms of dioxins and furans are marked with bold text.

	Initial ng/kg	1) Fenton red-%	2) Ozone red %
2378-TCDF	65.0	1.2	66.1
TCDF	**583**	**49.4**	**48.7**
2378-TCDD	13.1	0	44.5
TCDD	**65.3**	**56.2**	**62.6**
12378-PF	63.6	5.9	39.2
23478-PF	130	20.0	48.5
PCDF	**679**	**30.8**	**9.1**
12378-PD	74.5	10.2	30.3
PCDD	**737.5**	**42.8**	**33.7**
123478-HF	87.6	-15.3	29.2
123678-HF	102.5	4.9	42.6
234678-HF	120.0	16.7	49.7
123789-HF	6.9	21.2	48.1
HxCDF	**798.5**	**-4.6**	**28.5**
123478-HD	77	30.0	60.7
123678-HD	266.5	25.9	58.1
123789-HD	288	56.3	77.4
HxCDD	**2715**	**29.7**	**56.9**
1234678-F	504	8.5	50.8
1234789-F	35	21.1	50.9
HpCDF	**715.5**	**8.3**	**49.3**
12345678-D	1248.5	35.7	61.0
HpCDD	**2755**	**46.3**	**66.4**
OCDF	**195**	**46.2**	**71.9**
OCDD	**3855**	**57.5**	**74.3**
Toxic PCDD/Fs	7130	43.5	66.5
Total PCDD/Fs	13050	40.0	58.0
I-TEQ ng/kg	**242.5**	**21.3**	**50.1**
WHO-TEQ	276.5	19.3	47.2

is typical of PCDD/Fs formed in combustion processes. The most dominating congeners were octa-, hepta, and hexachorinated dioxin, representing 30, 21 and 20 percent of the total PCDD/Fs, respectively. The most toxic congener, 2,3,4,7,8-TCDD constituted just 0.001 percent of the total PCDD/Fs.

The ozonation under alkaline conditions removed 50 percent of the PCDD/Fs measured as I-TEQ. The disappearance of various congeners varied between 10 and 75 percent, being higher for dioxins and for highly chlorinated congeners. Fenton treatment gave less promising results. The removal of PCDD/Fs was just 20 percent measured in I-TEQ. Figure 1 shows the initial concentration and the concentration of the PCDD/Fs after Fenton's reagent treatment and ozonation under alkaline conditions.

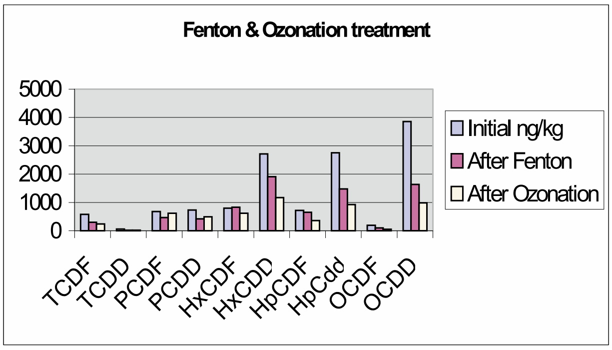

Figure 1. Initial concentrations of PCDD/Fs (ng/kg) and the concentrations after Fenton treatment and ozonation under alkaline conditions.

Table 3. Initial concentration of PCBs and removal efficiencies of Fenton treatment and ozonation under alkaline conditions.

	µg/kg	1) Fenton red %	2) Ozone red %
Total PCBs	395	6.1	36.2

The removal of PCBs was 36 percent using ozonation and 6.1 % in Fenton treatment. Besides the lower reduction efficiency, the Fenton treatment of this particular sample matrix was problematic. The initial high pH and buffer capacity of the ash caused especially high acid demand in order to lower the pH to the suitable range for the Fenton reaction (pH 2–3). In real-scale remediation projects the treated material should be neutralized which increases the chemical consumption. The acidification step of the treatment train can also mobilize the lead compounds from the ash matrix.

CONCLUSIONS

There were no data available for the calculation of engineering costs of either Fenton or ozonation treatments. Both of those techniques could be scaled up by using standard equipment and unit operations of soil handling and ozonation. In order to get an idea of the costs compared to traditional disposal methods of PCDD/F contaminated soil, the costs of oxidizing chemicals were calculated.

The cost of chemicals needed to obtain the purification results were 1000 FIM/ton and 450 FIM/ton for Fenton treatment and ozonation, respectively. The estimated cost for achieving the proposed Finnish target level is about 800 FIM/ton in this particular case by ozonation. The costs of treatment could be reduced by combining biological post-treatment to the pre-oxidation in a slurry reactor (Lee *et al.* 1992; Mckensie *et al.* 1999). The more suitable application of Fenton treatment can be a soil matrix which is acidic initially, has a low buffer capacity and has no problem with the mobilization of heavy metals. Modifications of the Fenton technique which involve the stabilization of Fe^{2+} by chelating agents (Sun 1993) or addition of solvent (Lee *et al.* 1998) can offer a viable alternative to iron and hydrogen based remediation alternatives.

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THE RISK ASSESSMENT OF PCB CONTAMINATED SOIL AND WATER – CALCULATED AND PERCEIVED RISK

by
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Dack, S. & Loxham, M. 2001. The Risk Assessment of PCB Contaminated Soil and Water – calculated and perceived risk. *Geological Survey of Finland, Special Paper 32, 135–142,* one figure.

Polychlorinated Biphenyls (PCBs) are both toxic and carcinogenic contaminants that bioaccumulate within the food chain and are persistent for long periods of time. This means that the receptors (or targets) identified as at risk from a particular source can be numerous over the time period that the PCBs remain in the environmental media. A Risk Assessment therefore has to consider a wide range of pollutant linkages and complex scenarios as the PCBs are transported through the environment.

The perceived risk from PCBs is heightened in relation to many contaminants as they are banned substances under UK law and EC Directive 76/464/EC on Dangerous Substances, where they are classified as List 1, Black List chemicals, and therefore "require clean-up". However, remedial action has few choices, so is both difficult and expensive with most PCB contaminated sites having to undergo exsitu incineration. The calculated risk may show that although present, PCBs do not require action, but this must be balanced by the perceived risk which may actually become the driving force for clean-up. Therefore a sensible balance has to be sought between industry, the Regulatory Authority and local people who all identify risk differently.

The problems associated with PCB risk assessment and the interpretation of source-path-receptor will be illustrated with reference to studies taken from the UK.

Keywords: environmental geology, soils, water, pollution, PCBs, risk assessment, remediation, United Kingdom

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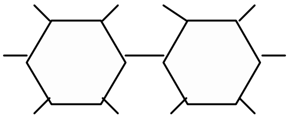
INTRODUCTION

Polychlorinated Biphenyls (PCBs) are banned substances in the UK, due to their ability to bioaccumulate and remain persistent in the environment, as well as due to their toxic and carcinogenic effects. PCBs are man-made chlorinated aromatic hydrocarbons, which historically, have been used as dielectrics in transformers & capacitors, used in hydraulic systems, in lubricating oils, and as plasticisers and solvents in paints and plastics. The environmental implications of PCBs were first identified in Sweden in 1966, and in the UK sale of "open application" PCBs was banned in 1972 (*e.g.* plastics, varnish, cutting oils). Production in the UK ceased in 1977, and in 1979, all uses of PCBs were banned. However, PCBs are now found world-wide, including Polar Regions thousands of miles from the original sources, due to the persistence and mobility of these compounds.

COMPOSITION OF PCBS

PCBs are aromatic chemicals, manufactured by the chlorination of biphenyl. The chemical formula is $C_{12}H_{10-n}Cl_n$, where the chlorine number ranges from 1 to 10.



There are theoretically 209 congeners (individual PCBs), although only 130 are generally found to occur in commercial products. PCBs were generally sold in mixtures of a number of different isomers and congeners, generally under the trade name "Aroclor"; the Aroclor number varying according to percentage by weight Chlorine, *e.g.* Aroclor 1260 contains 60% chlorine.

Individual congeners are colourless, often crystalline compounds, but commercial PCB mixes form yellow oily liquids to resins, with properties varying with the mixes. PCBs have a high degree of chemical stability under normal conditions, (19 PCBs are stable at room temperature), with low water solubility. PCBs however are lipophilic; they have high fat solubility, and are also soluble in organic matter. These properties govern PCB fate and transport in the environment – coupled with their resistance to breakdown by a wide range of oxidants and other chemicals – this means that metabolism in an organism is very slow, therefore bioaccumulation occurs in the adipose (fatty) tissue.

TOXICOLOGY

PCBs are classified as both toxic chemicals and carcinogens for humans as well as other animals, with 12 of the 209 congeners considered dioxin-like. A further 9 are considered highly toxic, and 25 have been commonly identified bio-accumulated in milk or fish. Less research has been carried out on vegetation, although this is contaminated from soils generally due to adsorption onto the outer surface of the plant, although some absorption may occur onto the waxy outer cuticle of the plant. There is no evidence for PCBs being particularly phytotoxic (except for a slowing of growth at very high concentrations); however this is an uptake route for herbivorous animals, and thus to higher mammals. Aquatic life is especially prone to bioaccumulation, with the PCB concentrations increasing up the food chain. In the UK the Ministry of Agriculture, Fisheries & Food (MAFF) has measured concentrations of PCBs in fish-liver oils for human consumption as well as PCB concentrations in many other foodstuffs (MAFF 1989). Cod liver oil was found to contain between 0.9 and 1.2mg/kg PCB, and Halibut Liver oil 7mg/kg. It should be noted that for a risk level of 1 in 1 million, the US Food and Drug Administration (FDA) consider the concentration of PCBs in water should be below 0.1 µg/l and the safe limit in foodstuffs to be between

0.2 and 3mg/kg. There are no limits for PCBs in food stuffs within the UK.

Human studies relate to commercial mixtures, rather than individual congeners, and are generally based on those that worked in the manufacture or use of PCBs, and those that have ingested high levels of PCBs through contaminated foodstuffs. Two highly publicised incidents in particular, both in Japan, (Yusho and Yu-Cheng), involved contaminate rice bran oil. Victims showed acute toxic effects, such as Chloracne, pigmentation and death; long term chronic effects, *e.g.* liver cancer, respiratory problems and Central Nervous System Damage, and finally developmental damage to children born of the victims. However, it should be noted that in the two incidents PCBs plus other impurities such as Polychlorinated Dibenzo-Furans (PCDFs) (which are known carcinogens), were present.

RISK ANALYSIS

As demonstrated in Sections 2 and 3 above, the main driving forces behind PCB focussed environmental assessment are the toxicity and carcinogenity of PCBs, and their ability to bioaccumulate and remain persistent in the environment. Due to their hydrophobic and lipophilic nature they remain relatively immobile in soil, but due to media attention as Polar contaminants', PCBs are perceived as highly dangerous chemicals, achieving the same reputation that pesticides such as DDT did, after "*Silent Spring*" was published.

Source characterisation

The calculated assessment of risk is complex for PCBs firstly due to their persistence. This means that firstly the receptors identified as at risk from a particular source can be numerous over the time period that the PCBs remain in the environmental media. A Risk Assessment therefore has to consider a wide range of pollutant linkages with greater uncertainties in the risk analysis model data for historic and potential future pollutant linkages. Secondly, it is difficult to accurately characterise the PCB source. This is due to several factors; certain PCB congeners are difficult to analyse for, the original Aroclor used is not known, and commercial PCB mixtures, have, through transformation, partitioning and biological metabolism, altered greatly in congener profile. Unfortunately much of the laboratory work with regard to toxicity has been carried out on "pure" commercial mixtures, and a few selected individual congeners. In many cases, the environmental mixture analysed will be relatively more toxic overall than the commercial mixture, since it is generally the most toxic and most dioxin-like congeners that have the greatest persistence.

Therefore for a credible risk assessment, with limited uncertainties, a number of points have to be considered to assess the environmental mixture. Firstly the composition of the mixture as individual congeners will vary in toxicity and carcinogenity (required for human and ecological risk assessment), and secondly the mobility and migration rate of individual congeners additionally varies (the less chlorinated congeners are more water soluble, and volatile, whilst the higher chlorinated congeners sorb more to particulates and sediments). Therefore although calculating risk based on Aroclor type is acceptable, a more accurate estimation of risk can be calculated using individual congeners as part of an environmental mixture.

In a water environment it is therefore often expected that given the properties of PCBs there should be a "chromatograph" like effect away from the original source. However, work done by IERC for the Environment Agency (EA) (EA 1997) and GeoDelft on contaminated streams and estuaries in the UK, has shown that although a few of the more soluble congeners may be found in the water near the source, the pattern of congeners does not seem to relate strongly to chlorination. For the site in Figure 1 below, (a cable and transformer manufacturer), there was a slight increase of the more toxic, more chlorinated, congeners with distance from the source, and a slight decrease of the lightest, most soluble congeners (di & trichlorobiphenyls such as IUPAC congener numbers 4 and 30). Many of the mid range congeners showed no pattern at all (in-

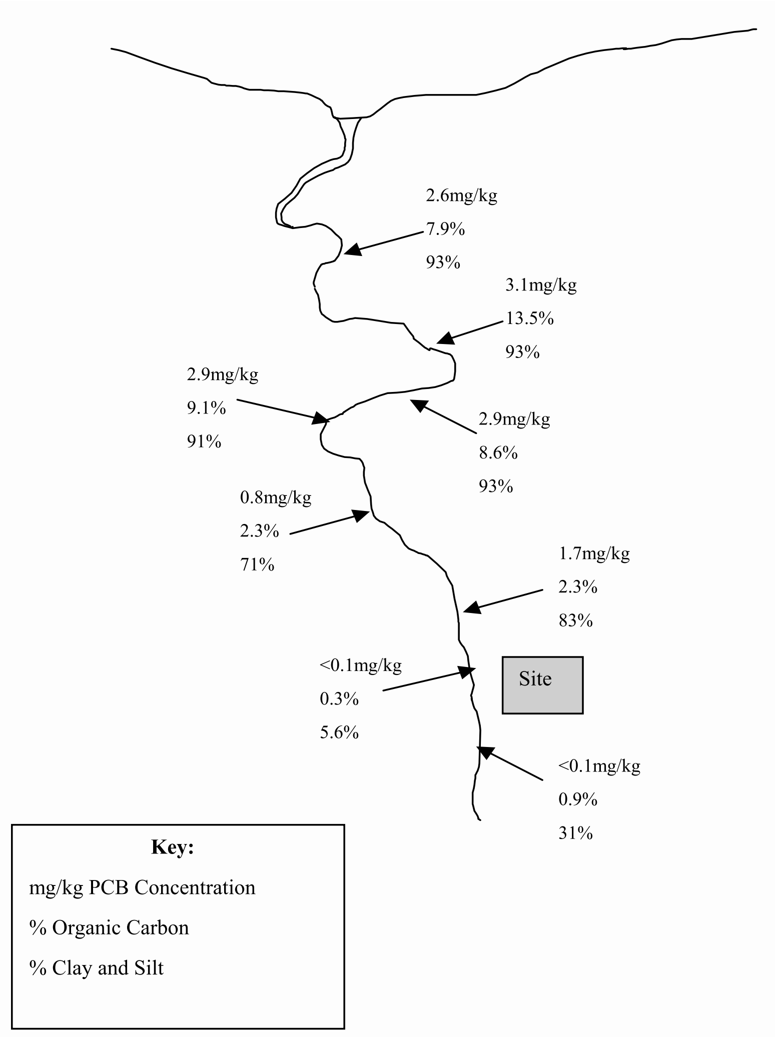


Figure. 1. Showing relation between PCB sediment content & organic content and fine sediment.

cluding one of the most toxic congeners, IUPAC Number 77), although they did show more direct correlation with other environmental factors.

For mid-range congeners and overall total PCB concentration, there was strong correlation against organic matter and to some extent clay content in stream sediments. Thus PCB contamination may be found away from the source, concentrated in slow moving/depositional environments and/or organic environments. This can lead to confusion in the interpretation of the source area (See Figure 1). As can be seen in the accompanying figure, the concentrations of PCBs were low close to the main source due to the stream sediments being mostly sand. The highest concentrations correlated to the organic content. This organic matter was highest in a large bend in the water-course, where flow rates were low, and in addition there was nutrient input from a fertiliser factory. This lead to the increase in organic content. However, although the bend was the area of highest concentration, it was not linked with the industry on this bend.

It should also be noted that for the three sample locations upstream of the alleged site, that the background total PCB content of the sediment varied from 1.3 µg/kg to 21.9 µg/kg. This, it can be assumed, is from aerial deposition, via run-off from the land. This highlights the fact that the presence of PCBs on a site does not necessarily indicate a site source, but may in fact indicate the local or national background concentration, due to volatility and particulate transport.

A second example concerning the difficulty of identifying a historic PCB source area, also occurred in the UK, where a major river and estuary was contaminated at low levels by PCBs. However, prior to the site being identified, elevated concentrations of PCBs had been found in eels by the Regulatory body, upstream of the input from the site. No PCB input was identified near the eel sampling area, and downstream was not considered. However, due to the migratory habit of eels, the PCB input did not have to be upstream of the sampling area.

Finally, the identification of PCBs on a contaminated site may be missed altogether and therefore a source not identified. This is because PCBs may not have been used directly on site. PCBs were often contaminants of reclaimed oil, and therefore may be found in scrap yards and oil reclamation centres where there is no obvious site-held or desk study evidence of their presence. Therefore sites contaminated with PCBs do not just include the more obvious sites, such as electrical sub-stations, cable and capacitor manufacturers, but also railway depots, scrap yards, paint and paper manufacturers, for example.

3.1 Transport Pathways

Given their chemical properties, it is often believed that PCBs will be attenuated by soil and sediments, and transported via surface water. However, even with such low volatility, PCBs are mainly distributed within the environment via air, particularly due to incineration, the drying out of sewage sludge and the burning of scrap and waste at low temperature. Once in the atmosphere, vapour phase PCBs adsorb onto particulates or water droplets, and although the mono and dichlorobiphenyls have relatively short half lives (days/weeks) in sunlight, most are transported and deposited at distance from source, commonly within 2–3 days from the area of origin. Work carried out to characterise the background PCB and Dioxin contamination across the UK by the HM Inspectorate of Pollution (HMIP 1989, 1995), found PCBs at all sampling points on 50km grid squares with concentrations between 1 and 141 µg/kg. The national and local background around a contaminated site requires consideration when carrying out a risk assessment. For a PCB contaminated industrial site, with particular reference to nearby residents, the overall *current* risk from the nearby site may pose a low overall risk, but the low level chronic exposure from “background” historically deposited in and around their property, may be a lot higher. They are therefore at risk from their own environment, contaminated historically, rather than current contaminant conditions at the site. However, the risk to local residents from historic contamination is generally perceived to be from the site only, by both the site and the residents, since long-term effects are rarely noticeable and obviously attributable; most long term health effects can be attributed to a number of other factors.

The main other transport pathway is by sorption to clay-rich suspended solids or colloids (especially organic acids) in surface water. Drains are a “special case” of this pathway, but often ignored. In fact on the majority of PCB contaminated sites investigated by GeoDelft in the last 3 years, storm water drains and foul sewers have all contained PCBs often in significant concentrations. This is mainly due to the partial silting of drains and the relatively high organic matter content often associated with them – therefore any PCBs washed in as soils or dust, or spilt directly into the drains, often collects there at significant concentrations. This can additionally then become a disjointed source characterisation problem, when elevated concentrations are identified further down the drainage network at sewage works.

Groundwater transport is not usually significant due to the attenuation of the PCBs by soil, the solubility of PCBs and due to pore sizes/connectivity of the soil media. Sorption processes include adsorption, cation exchange and chemisorption. The amount of sorption is dependant on the amount of organic matter and clay within the soil, the effective porosity and bulk density of the soil and rock material. However, on contact with water, sediments will act as a reservoir of PCBs and not just as a sink, as the PCBs desorp into the clean water. Desorption will also occur if there is subsequent contamination of PCB contaminated soils and sediments by other organic solvents such as BTEX hydrocarbons, alcohols and bipolar esters.

Exposure pathways

As with the majority of environmental contaminants, the main exposure routes to humans and other animals are via inhalation, ingestion and dermal contact. For humans, the pathways are relatively simple, and exposure normally considered as a chronic cancer risk. Inhalation of vapours from historic contamination is extremely low, with inhalation of particulates higher, although proportional to the amount of bare soil and the surface concentrations. Ingestion can vary greatly and is effected by the behaviour patterns of the individual for on and off site contamination. On site, this is less of a concern, as occupational hygiene protocols can be enforced. However, at least one study, summarised in WHO 1993, showed that wives of workers exposed to PCBs, had raised levels of PCBs in their blood due to bad industrial hygiene and cross contamination of the home. However, it should be noted that many foodstuffs contain PCBs, although there has been a steady drop in concentration over the last 20 years. PCBs identified in foodstuffs are mostly sourced from bioaccumulation up the food chain, packaging material, and cross-contamination. The average intake of PCBs is approximately 0.01 to 0.04mg per day, which should be considered as part of the Total Daily Intake of PCBs alongside that sourced from contaminated land. In the UK there are no limits set for PCB content of food, although there are limits set in the US by the FDA (See Section 3).

Dermal Adsorption, along with ingestion is the main exposure route for uptake from contaminated soils and waters. Quantitative studies have been carried out on shaved rats etc (WHO 1993), and qualitative studies on capacitor workers. Uptake is believed to be due to the lipophilic nature of PCBs, and thus the adsorption to the uncovered skin surface area. However, it should be noted that the degree to which the skin comes into contact with the soil (*e.g.* digging, rather than walking on it) affects the amount of PCBs adsorped, and therefore behaviour patterns of receptors require consideration.

Non-human exposure pathways, particularly for grazing animals, have to account for soil and herbage uptake, since the grazing pattern (whether the animal cuts through the grass stalk or whether grass is pulled up with soil on the roots) is significant. For sheep, the main exposure route, given their coat helps prevent elevated dermal contact, is through ingestion of soil rather than grass. On contaminated sites studied by GeoDelft in the UK, the PCB concentration of grass is generally about 4% of that of soil. The HMIP (1989, 1995) studies of UK contamination identified grass concentrations of between 7–16 μg/kg in rural areas. The implications of this mean that for a full risk assessment to be carried out, the assessor has to consider not only the internal uptake processes of PCBs within the animal but also the grazing habits and patterns of the specie of concern.

Although uptake by grazing animals is not generally considered of importance to the animals themselves, it is of consideration to the human food chain, as meat or milk.

Receptor characterisation

The significance of the receptor is identified by legislation and “shareholder value”. Legislation prevents contamination or demands clean-up of certain receptors. In the UK the PCB contamination of surface and groundwater is legislated at zero concentration. Therefore any contamination likely to cause “significant harm” of receptors, or impact water resources is prosecutable and has to be cleaned-up, although in practice the interpretation of the law usually takes into account the historic nature and past good working practices. As a result areas in the UK where PCB contamination is significant *e.g.* Mersey Estuary/Liverpool Bay, remedial actions are unlikely to be considered as sites where the “polluter pays”, but instead will be considered and assessed from the public purse. This is because the sources of the PCB present are many, discharge followed past good practice, and much of the contamination is historic. Sewage sludge too contaminated to be spread to land was disposed of within the Bay (approximately 0.5 to 1.8 million tonnes/year), contaminated industrial waste (0.05 to 0.3 million tonnes/year), dredged spoil from docks and harbours (3.5 to 5.7 million tonnes/year), (Norton *et al.* 1984) plus there are historical manufacturing sites in the area that used PCBs and discharged many other contaminants, notably mercury. Fish in this area contain concentrations of PCBs in the 23 to 130 μg/kg range (Leah *et al.* 1997), being a receptor for contamination in their own right, and as a pathway to PCB uptake to human health. Therefore although there is harm being caused to receptors, the positioning of blame becomes difficult to assess or prove with so many potential sources. It is therefore unlikely that remedial measures will be taken.

Shareholder value is the main driver to clean-up and the identification of other receptors. This is the perceived and actual risk of PCB contamination becoming interlinked. The bad publicity of actual risk, or the perceived risk from a site to a nearby receptor such as a school, all cause bad publicity with an associated drop in share value and possibly a local/national boycott on the products manufactured at a site. Although clean-up is often associated with a calculated risk to human health, this is rarely done as simply a "duty" to the community, but a reaction to the likelihood of prosecution and the resulting publicity. Once off site risk is determined, risk communication is of the utmost importance both to the site owners, the Regulatory body, and the local community to keep the risk in context and to prevent the risk being perceived as a larger or smaller problem than it technically is. It should be remembered that any contaminant risk is an involuntary risk and therefore treated very differently to a voluntary risk, *e.g.* the risk of driving to work may be a higher risk than that of living next to a chemical dump, but the risk of driving is being controlled by the individual and thus is more acceptable. Therefore for clear communication work, it has been shown by Freudenburg *et al.* (1994), that the biggest barrier to risk communication is the "trustworthiness" of those communicating the risk.

RISK BASED CLEAN-UP

PCBs are classified as List 1, Black List chemicals, and therefore "require clean-up". However, remedial action has few choices, due to the physiochemical nature of PCBs, so this is both difficult and expensive with most PCB contaminated sites having to undergo exsitu high temperature incineration. A number of techniques have been studied such as bioremediation, soil washing, etc, but very few techniques have passed the laboratory or pilot stage, and most work under very site-specific conditions. Therefore most clean-up is based on soil excavation and landfilling (up to 50mg/kg) or incineration in the UK.

However, given the sorptive properties of PCBs to soil, the calculated risk of leaving the contaminated soil insitu, and the risk of the clean-up process has to be considered as part of the overall site management. Excavation produces dust, and excavation usually involves large quantities of soil that need to be moved on lorries away from the site. This may spread contaminated PCB dust, and poses an increased vehicular hazard, both directly, and indirectly from fumes. Finally, clean soil has to be brought in to replace what has been removed, posing a second vehicular hazard.

GeoDelft have been involved in two cases where the risk of clean-up was greater than leaving the contamination insitu. The first, involved the water course, discussed in Section 4.1, where dredging would have remobilised fairly static sediment, which increased in PCB content with depth. Secondly the increase in traffic across a recreational area of marshland near the estuary would have posed significant risk to the public.

A second site involved contaminated grazing land with potential access to the public (it was surrounded on two sides by housing and commercial properties), and was contaminated by PCBs from an adjoining site. However to excavate the contaminated soil would have produced bare earth (increasing the risk of dust), and the lorries would have had to leave the gazing land via a small cul-de-sac with residential housing, and then pass on through the village concerned. The increased traffic (approximately 200 lorry loads) posed a considerable risk to local residents. Since PCBs are strongly sorped to soil and the contamination was associated with drainage ditches in the field, comprising the top 0.3m of soil, it was agreed by the site and the Regulatory body that the overall risk of leaving the material in the field was less than transporting it a way. However, the level of contamination did pose a potential risk, albeit small, so the final remedial option for the field was deep ploughing (locking the PCBs at depth), with the end-use determined as wildlife habitat – thus it removed the grazing receptor and as a wildlife habitat reduced the likelihood of people accessing the field.

The calculated risk to local residents from the grazing land, prior to remediation, was such that, although a risk was present, it was low enough not to require action, but this was balanced by the perceived risk of both the site owner and the Regulator which actually became the driving force for considering clean-up, and after much consideration, the option of deep ploughing and biodiversifying the area. The risks were explained to local residents, but the perceived risks by the public were considered low, probably because many of the villagers had worked on the nearby manufacturing site where the PCBs had originated, were currently working on that site or had relatives there. Thus the problem became one of "rational selfishness", where the benefits of the manufacturing site appeared to outweigh the risks caused by the site, and because (due to the employment of local people), the site owners were seen as "trustworthy" and "credible" in the communication of risk. (Freudenburg *et al.* 1994).

CONCLUSION

Thus it can be seen that the risk assessment of any contaminated site that involves particularly a persistent chemical, has to take into account a vast range of considerations when studying the source-path-receptor scenarios; many of which are extremely complex. The technical assessment has additionally to take into account the perceived risk, which is the attitude to that risk from individuals. The scientific environmental community has progressed a long way into statistical risk assessment, with the risk to receptor from a contaminant or contaminants better understood than ever before. The fields of toxicology, chemistry and biology have been integrated in the last few years into the fields of environment and geotechnics and this has led to a greater understanding of the source-path-receptor linkage. However, it is now time that the scientific community integrated the social scientists, because particularly with contaminants with a public high profile, the calculated risk is not enough. This is especially true for "emotive" contaminants such as PCBs where the perceived risk is often of greater importance than the calculated technical risk, and as can be seen from the above examples, where the remedial strategy drivers have been direct legislation or perceived risk rather than just the calculated risk scenario. This does not make the assessment of risk any less scientific or important, it just has to be considered within the wider framework.

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REHABILITATION OPTIONS FOR A FINNISH COPPER MINE

by
M. A. F. Angelos

Angelos, M.A.F. 2001. Rehabilitation Options for a Finnish Copper Mine. *Geological Survey of Finland, Special Paper 32, 143–149,* two figures and 4 tables.

Numerous rehabilitation options for mined land exist. This paper will examine the basic structure of a remediation plan for a Finnish Copper Mine outlining the remediation techniques that are applicable to the geology and climate of the mine site. Techniques include proven and innovative methods such water covers, wet barriers, composite capping layers and in-pit disposal. Treatment methods for AMD will be discussed including the role of passive treatments, such as wetlands and permeable barriers. The role of risk assessment in the rehabilitation plan and the importance of an ecological risk assessment will be discussed.

Keywords: environmental geology, mines, copper ores, acid mine drainage, risk assessment, remediation, Luikonlahti, Finland

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INTRODUCTION

The Luikonlahti Copper mine is located near the town of Kaavi in eastern Finland, approximately 30 km northwest of Outokumpu. The copper sulphide ore deposit formed the northern most part of the Outokumpu assemblage. During 15 years of operation, between 1968 and 1983, a total of 33 km of underground tunnels and 5.5 km of underground shafts were excavated in the mining of 6.85 million metric tons of ore. The underground working are now flooded with 2 million m^3 of contaminated water and three open pits contain over 1 million m^3 of contaminated water. Five separate waste rock piles exist and are actively forming acid mine drainage (AMD).

ACID MINE DRAINAGE (AMD) GENERATION

Acid Mine Drainage is the product formed by the atmospheric oxidation (i.e. by water, oxygen and carbon dioxide) of the relatively common iron sulfide minerals pyrite (FeS_2) and pyrrhotite (FeS) in the presence of (catalysed by) bacteria (Thiobacillus ferrooxidans), and any other products generated as a consequence of these oxidation reactions (i.e. heavy metals solubilized by acidic solutions). The following chemical reactions describe the oxidation of pyrite (FeS_2) to the products that constitute the contaminants generically termed Acid Mine Drainage, although similar equations may be written for the oxidation of pyrrhotite (FeS) (Shaw & Mills 1998).

Table 1. Chemical reactions involved in the oxidation of pyrite to form AMD (Shaw and Mills 1998)

$$FeS_2 + 7Fe_2(SO_4)_3 + 8H_2O = 15FeSO_4 + 8H_2SO_4 \qquad (1)$$
$$FeS_2 + Fe_2(SO_4)_3 = 3FeSO_4 + 2S \qquad (2)$$
$$4FeSO_4 + O_2 + 2H_2SO_4 \text{ bacteria} = 2Fe_2(SO_4)_3 + 2H_2O \qquad (3)$$
$$2S + 3O_2 + 2H_2O \text{ bacteria} = 2H_2SO_4 \qquad (4)$$
$$4FeS_2 + 15O_2 + 2H_2O = 2Fe_2(SO_4)_3 + 2H_2SO_4 \qquad (5)$$
$$S^0 + 3Fe_2(SO_4)_3 + 4H_2O = 6FeSO_4 + 4H_2SO_4 \qquad (6)$$

The stable pH developed (2.5 to 3.0) and the products of sulphuric acid and ferric sulphate create conditions where the ferric iron ion itself can act as an oxidant (above about pH 3 the ferric ion is itself hydrolyzed to ferric hydroxide, which precipitates as the familiar rust-coloured stain associated with AMD). In the absence of ferric iron at pH 2.5–3.0, sulphuric acid will dissolve some heavy metal carbonate and oxide minerals, but has little reactive effect on heavy metal sulphides. However, ferric iron ion is capable of dissolving many heavy metal sulphide minerals, including those of lead, copper, zinc, and cadmium , by the general reaction:

$$MS + nFe^{+++} = M^{n+} + S + nFe^{++} \qquad (7)$$

Where: MS = solid heavy metal sulphide; Fe^{+++} = aqueous ferric iron ion; M^{n+} = aqueous heavy metal ion; S = sulphur; Fe^{++} = aqueous ferrous iron ion (Mills 1995).

It is by this process that significant amounts of heavy metals maybe solublized by AMD. It has been customary to call Acid Rock Drainage (ARD) with the dissolution of metals by equation 7, Acid mine drainage, even though this term implies that the process occurs only in association with mining activity.

PREDICTION OF AMD

The prediction of AMD generation can be broadly divided into three categories: static tests, kinetic tests and, mineralogy and surveys (Papastavrou & Georgopoulou 1997). The standard international approach for the prediction of AMD chemistry involves the “Wheel” approach (Fig. 1) in which types of static and kinetic tests are conducted and compared (Morin & Hutt 1998).

Static Tests: These attempt to define the balance between the acid producing components of the material (acid producing AP) which usually are sulphide minerals and the acid consuming components (neutralization potential NP) which usually are carbonate minerals (Morin & Hutt 1998)

Kinetic Tests: These consist of repetitive cycles of leaching and monitoring either under field or laboratory conditions, and they are used to predict long-term generation rates of metals, non-metals,

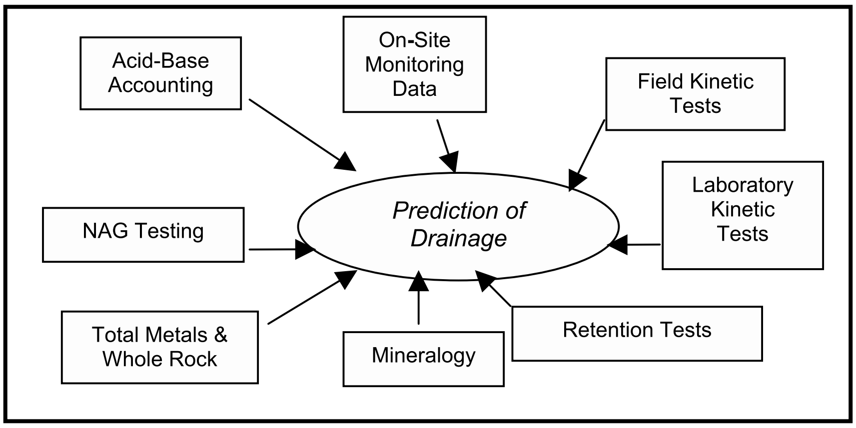

Figure 1. The "Wheel" for Acid Mine Drainage Chemistry (Morin & Hutt 1998).

and parameters such as acidity. These tests can also evaluate the effect of biological reactions (Lawrence & Sheske 1997).

Mineralogy and Surveys: Surveys such as magnetic surveys and IP/resistivity measurements may also be applied to locate zones of increased sulphide concentrations (Papastavrou & Georgopoulou 1997). A sample's mineralogy is a critical aspect in the prediction of its acid generation, acid neutralisation, and metal leaching (Morin & Hutt 1998).

RISK ASSESSMENT

Risk assessment is an analytical tool used to identify the potential adverse effects to humans and ecosystems resulting from exposure to environmental hazards. Health risk assessment deals with effects on humans and ecological risk assessment with effects on ecosystems. Risk assessment is a tool providing a basis for site-specific decisions (e.g. one criterion for choosing remediation/rehabilitation techniques, identifying chemicals of concern, prioritizing sensitive receptors). It can also be used to assess acceptable contaminant levels in the environment (Kuusisto 1999).

Health risk assessment (HERA) methodologies and techniques are firmly established. The most commonly used framework involves the following steps: hazard identification, exposure assessment, dose-response assessment and risk characterization (EPA 1989). HERA produces measures of risks, which contain the probabilities of the possible adverse outcomes and the uncertainties involved.

Ecological risk assessment: Ecological risk assessment (ERA) is the process of estimating the likelihood that adverse ecological effect (*e.g.* reduced species diversity) will occur as a result of exposure to a physical, chemical or biological stressor (*e.g.* elevated metal concentration). ERA includes three primary steps: problem formulation, analysis and risk characterization (EPA 1998). In problem formulation, goals are evaluated and assessment endpoints selected. Also, a conceptual model is prepared and an analysis plan developed. In the analysis phase, exposure to stressors is evaluated (exposure assessment) and the relationship between stressor levels and ecological effects is estimated (toxicity assessment). Finally, in risk characterization, risk is estimated through integration of exposure and stressor- response profiles (Kuusisto 1999).

AMD with elevated metal concentrations from the Luikonlahti mine drains to the nearby lakes and to groundwater. Water ecosystems are known to be susceptible to metals and, therefore, ecological risk assessment is warranted.

The risk assessment process in Luikonlahti case will contain the following steps (Kuusisto 1999):

- Establishing a planning group, which will meet prior to and during the risk assessment process. ERA planning team could include e.g. representatives of Mondo Minerals, environmental authorities, risk assessors, local communities and environmental groups.
- Data collection
- Risk assessment (health and ecological)
  1. Risks caused by the current state
  2. Risks caused by the different remediation options
- Reporting

Table 2. Data to be collected as a part of an ERA. (Adapted from VEPA 1997)

ERA COMPONENT	DATA THAT MAY BE COLLECTED	RESPONSIBLE PARTY
Problem formulation	Extent and degree of contamination	TUT / Mondo Minerals
	Identification of contaminants of concern	Risk assessor
	Distribution of biota of ecological value at the site (biological survey)	Ecologist/biologist?
Exposure assessment	Fate and transport modeling of chemicals of concern	Risk assessor
	Literature-based inhalation, ingestion and absorption rates	Risk assessor
	On-site environmental properties that affect contaminant mobility/availability (*e.g. foc*, pH, bulk density, porosity)	TUT / Mondo Minerals
	Bioavailability factors	Risk assessor
Toxicity assessment	Literature review of relevant toxicological studies Laboratory toxicity tests	Risk assessor
Risk characterization	Integration of preceding steps	Risk assessor

REHABILITATION OPTIONS

The rehabilitation options for the Luikonlahti site revolve around the principals of prevention and control. Regarding the fundamental processes governing the generation of AMD and metal release from mine water, methods for prevention and control of AMD in general aim at either:

- Changing the chemical properties of the waste prior to disposal by, for example, separation of pyrite or blending of waste rock with acid consuming material,
- restricting the transport of oxygen and/or water into the waste dump or
- treating the leachate (MiMi1998).

Prevention and control methods can be divided into four different groups:

1. Treatment of waste which includes altering the chemical properties of the waste but also altering the physical properties of the waste in order to limit the transport of oxygen or air into the waste dumps.
2. Flooding of the waste, i.e. a water table is established above the disposed waste as a barrier in order to limit the transport of oxygen or air into the waste.
3. Dry covering of the waste in order to limit the transport of oxygen and/or water into the waste dump.
4. Treatment of AMD with the purpose to reduce the metal concentrations and neutralized the water that is discharged for the site.

Water Covers: Water cover is considered the most effective of all AMD prevention and control measures (MEND 2.18.1, 1997). The maximum concentration of dissolved oxygen found in natural waters is approximately 25,000 times lower than that found in the atmosphere.. The solubility of oxygen in water is quite low (11 mg/L at 20°C) and the diffusion rate of oxygen through water is about 10,000 times slower than through air. This means that the rate of oxygen transported through water is sufficiently slow to be of no significance in acid generation.

Major factors influencing the application of a water cover include the availability of water and the cost of maintaining the required depth of water in the long term. The cover maybe achieved by the disposal of wastes into natural water bodies, into engineered impoundments (such as dykes or dams) or into flooded underground mine workings and open pits (ARD Tech. Guide 1989).

Sub-aqueous disposal: The principal of water cover applies if acid producing material is deposited in a deep water body such as lakes or marine places. Possible environmental impacts related marine or lake disposal are the toxicitry of heavy metals, the increased turbidity due to suspended solids and direct physical impact on inhabitants. (ARD Tech. Guide 1989).

In-pit disposal: Open pit mines that have ceased production are increasingly being considered for the permanent and environmentally acceptable disposal of mine waste rock and tailings that are, or have the demonstrated potential to become, sources of acidic drainage. There are four basic concepts for the placement of wastes in pits:

Option 1 – Underwater disposal
Option 2 – Elevated water tables
Option 3 – Dry disposal
Option 4 – Perched water tables (MEND 1995).

Soil Covers: Soil covers (or dry covers) are placed over mine wastes to reduce oxygen diffusion and water infiltration (MEND 1994). The effectiveness of a soil cover as an oxygen barrier is dependent on its moisture content. A cover that can be maintained in a saturated condition will be more effective because of the low diffusivity of oxygen in the pore water and absence of desiccation cracking (MEND 2.35.2b 1997). In general, soil covers can be classified according to their function as shown in Table 3.

In Finnish winter conditions, the effects of freezing and thawing on a saturated soil cover is a major design factor. Another major factor in selecting the appropriate cover material is the availability of local soil types. Cost is a limiting factor in the soil cover design process. Local soils are much cheaper to use than soils that have to be transported distances (MEND 2.35.2b 1997). The final factor in regard to soil cover design for the Luikonlahti site is climate. The net water balance delivered to the ground surface of the site is a positive value, and therefore the site can be considered a “wet” site. This is a key feature regarding the performance of the cover system that will be selected for use and allows for a system that utilizes a saturated condition.

Innovative Covers: Innovative “dry” cover research is indicating that several materials, including waste materials from other industries provide excellent potential at lower cost for generating moisture-retaining, oxygen-consuming surface barriers. Recent studies have shown that clean (non-acid generating) tailings, often available close to problematic sites, can be used as a lower cost alternative for the fine material in layered cover systems (MEND 1996, MEND 1999).

Single layer covers: To meet the requirements of an oxygen transport barrier, a single layer cover should be constructed of a material with mainly fine pores, which is generally true for fined grained materials (clays, clayey silts) and well graded materials with a high content of fines (clayey and silty tills) (MiMi 1998). A single layer cover can be an efficient oxygen transport barrier particularly at sites where the ground water table is close to the ground surface and conditions are favourable to maintain a high degree of saturation in the cover.

Composite covers: Composite cover systems consist of several layers (up to six) and may include synthetic membrane covers, compacted layers and drainage layers. Composite covers provide good medium-term control but they are sensitive to long term degradation (ARD Tech. Guide 1989, MiMi 1998).The basic cover design rationale for composite covers design is:

- Base granular material draining to residual saturation before fine grained till layer,
- Fine grained glacial till with low hydraulic conductivity as a moisture and oxygen barrier,
- Overlying coarse grained granular layer as a recharge layer to the underlying till layer, and
- Surficial coarse grained layer for erosion protection.

Table 3. Soil cover classification (adapted from MiMi 1998).

Cover Type	Primary Function
1. Oxygen diffusion barriers	To limit the transport of oxygen by acting as a Barrier against the diffusion of oxygen to the waste
2. Oxygen consuming barriers	To limit the transport of oxygen by consumption of which penetrates into the cover
3. Low permeability barriers	To limit the transport of oxygen and the formation of leachate by acting as a barrier against the diffusion of oxygen as well as the infiltration of precipitation
4. Reaction inhibiting barriers	To provide a favourable environment to limit reaction rates and metal release

TREATMENT OF AMD

Passive Treatment of Acid Mine Drainage: Passive treatment systems have been developed that do not require continuous chemical inputs and that take advantage of naturally occurring chemical and biological processes to cleanse contaminated mine waters. The primary passive technologies (Figure 2) include constructed wetlands, anoxic limestone drains (ALD), successive alkalinity producing systems (SAPS), limestone ponds, open limestone channels (OLC) and permeable reactive barriers (PRB).

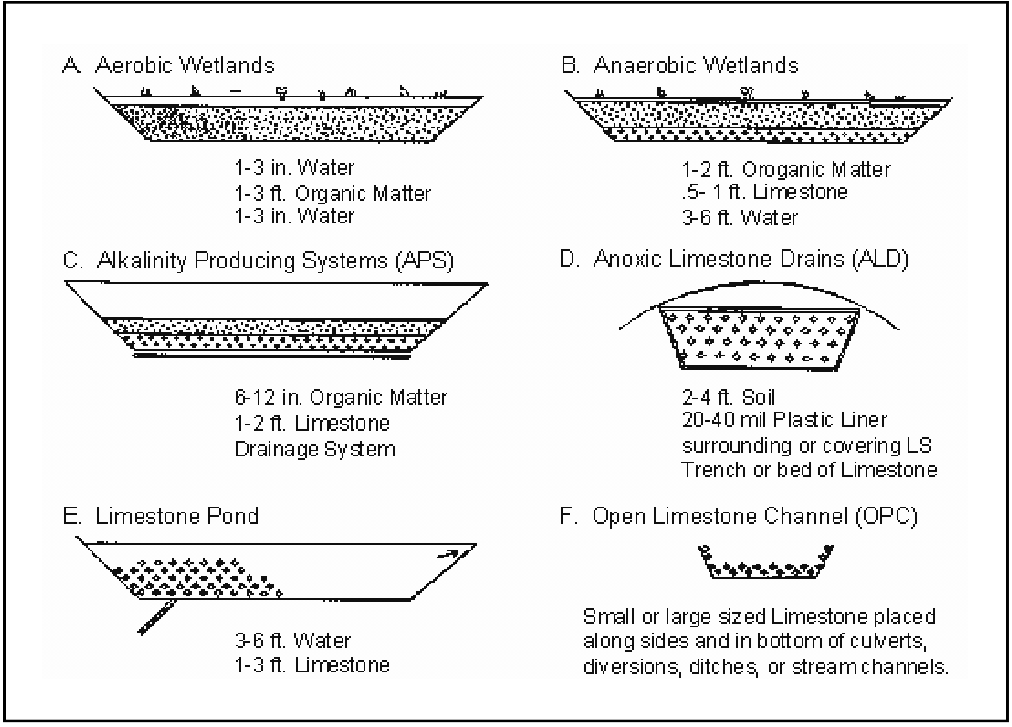

Figure 2. Passive treatment options for AMD (Skousen & Ziemkiewicz 1995).

Natural wetlands are characterised by water-saturated soils or sediments with supporting vegetation adapted to reducing conditions in their rhizosphere. Constructed wetlands are man-made ecosystems that mimic their natural counterparts. Often they consist of shallow excavations filled with a flooded gravel, soil, and organic matter to support wetland plants, such as *Typha*, *Juncus*, and *Scirpus* sp. Treatment depends on dynamic biogeochemical interactions as contaminated water travels through the constructed wetland

ALDs are abiotic systems consisting of buried limestone cells that passively generate bicarbonate alkalinity as anoxic water flows through. SAPS combine treatment concepts from both wetlands and ALDs. (Skousen & Ziemkiewicz 1995). PRBs are installed in aquifers down gradient of contaminated source areas and are designed to stimulate sulfate reduction and metal sulphide precipitation reactions (Papastavrou & Georgopoulou 1997).

Selection of an appropriate passive system is based on water chemistry, flow rate and local topography and site characteristics (Hyman & Watzlaf 1995), and refinements in design are ongoing. In general, aerobic wetlands can treat net alkaline water; ALDs can treat water of low Al, Fe^{3+}, and DO; and SAPS, anaerobic wetlands and OLCs can treat net acidic water with higher Al, Fe^{3+}, and DO.

Chemical Treatment of Acid Mine Drainage: Six primary chemicals have been used to treat AMD (Table 4). Each chemical has characteristics that make it more or less appropriate for a specific condition. The best choice among alternatives depends on both technical and economic factors. The technical factors include acidity levels, flow, the types and concentrations of metals in the water, the rate and degree of chemical treatment needed, and the desired final water quality. The economic factors include prices of reagents, labor, machinery and equipment, the number of years that treatment will be needed, the interest rate, and risk factors (Skousen *et al.* 1990).

Table 4. Chemicals available to treat AMD (Skousen et al. 1990).

Common Name	Chemical Name	Formula	Conversion Factor[1]	Neutralization Efficiency[2]
Limestone	Calcium carbonate	$CaCO_3$	1	30%
Hydrated Lime	Calcium hydroxide	$Ca(OH)_2$	0.74	90%
Pebble Quicklime	Calcium oxide	CaO	0.56	90%
Soda Ash	Sodium carbonate	Na_2CO_3	1.06	60%
Caustic Soda (solid)	Sodium hydroxide	NaOH	0.8	100%
20% Liquid Caustic	Sodium hydroxide	NaOH	784	100%
50% Liquid Caustic	Sodium hydroxide	NaOH	256	100%
Ammonia	Anhydrous ammonia	NH_3	0.34	100%

[1] The conversion factor may be multiplied by the estimated tons acid/yr to get tons of chemical needed for neutralization per year. For liquid caustic, the conversion factor gives gallons needed for neutralization.

[2] Neutralization Efficiency estimates the relative effectiveness of the chemical in neutralizing AMD acidity. For example, if 100 tons of acid/yr was the amount of acid to be neutralized, then it can be estimated that 82 tons of hydrated lime would be needed to neutralize the acidity in the water (100(0.74)/0.90).

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REMEDIATION OF SOILS CONTAMINATED BY LEAD- AND COPPER-CONTAINING RIFLE BULLETS

by
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Hintikka, Väinö, Parvinen, Pekka, Stén, Pekka, Laukkanen, Jukka & Leppinen, Jaakko 2001. Remediation of soils contaminated by lead- and copper-containing rifle bullets. *Geological Survey of Finland, Special Paper 32, 151–157,* one figure and 5 tables.

Mineral processing techniques were evaluated to see if they could be used to remediate rifle range soils. The soils studied originated from two Finnish rifle ranges, Viikinmäki and Utti, and were heavily contaminated by lead- and copper-containing bullets and their disintegrated fine-grained weathering products.

Most of the lead and copper in the soil samples is in the form of coarser particles, which can be easily recovered mechanically by combining wet sieving with consecutive gravity concentration stages. In this process, 83–85% of the total lead and 91–99% of the total copper can be recovered into a ca. 90% pure bullet product.

The fine soil fraction contains 14–17% of the total lead, mainly as fine-grained carbonaceous weathering products of the bullets, and 1–3% of the total copper. Due to the higher solubility of the lead carbonates and the high surface area of these particles, they form a significant source of contamination for the environment. Complex-forming leaching was used to recover the lead and copper from the fine soil fractions with recoveries of 95-98%. By combining the mechanical and leaching processes, lead and copper recoveries in excess of 99.4% can be reached.

Keywords: environmental geology, shooting ranges, soils, pollution, lead, copper, remediation, methods, Viikinmäki, Utti, Finland

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INTRODUCTION

As big cities expand, the lack of suitable areas for new housing is becoming a problem of ever-increasing importance. Abandoned industrial estates and shooting ranges often cover several hectares of prime building land in the vicinity of the new suburbs of growing cities, but due to the former activities these areas are often contaminated and call for remediation measures before being used for housing purposes.

Until now the most popular option in Finland, as in many other countries, for managing the problem of contaminated soils has been the encapsulation and isolation of the contaminated material. However, this solution does not meet the requirements of sustainable development and an active treatment to remediate the contamination is to be preferred. In an earlier work (Hintikka *et al*. 1999), we evaluated whether conventional mineral processing technologies–such as screening, heavy medium separation, and complex-forming leaching–could be used in the remediation of soils contaminated by lead-containing shotgun pellets. In this work, we extend the research to include rifle range soils, which along with lead are heavily contaminated by copper, but the contaminants are restricted to smaller areas. In addition to the concentration methods used for shotgun shooting range soils, in this work we also consider shaking table concentration. As all the studied methods are already highly developed and used on an industrial scale in extractive metallurgy, the treatment prices are expected to be low compared to the emerging, innovative technologies, *e.g.* electro-dialysis, for the remediation of soils polluted by heavy metals.

EXPERIMENTS

Research material

A typical rifle range consists of a firing line, target line, target berm, and impact berm, the distance from the firing line to the target line being normally from 50 to 300 m. The impact berm is designed to capture the fired projectiles containing mainly lead, along with copper, zinc and, sometimes, nickel originating from the jacket of the bullet. The soils studied in this work originated from the berms of the Viikinmäki rifle range in Helsinki and from the Utti rifle range in Kouvola.

The first step in the characterization of the soils was to screen them into a series of size fractions. The oversize fraction was separated (and rejected from further studies) by a screen with 20 mm aperture. The coarse fraction in the Viikinmäki case was made by screening on 2.8 mm aperture, while in the Utti case a screen with 5.1 mm aperture was employed. In both cases, the medium and fine fractions were separated employing a screen with 1 mm aperture. The distribution of the soils in each particle size fraction is given in Table 1, while the results of the chemical analyses of the soil fractions are given in Tables 2 and 3. The fine and medium size fractions were analyzed by XRF. However, due to problems with sampling and sample preparation of material containing particles as coarse as bullets, the metal contents of the coarse fractions were estimated as follows. The whole bullets were first separated by the heavy medium apparatus as described below. The purity (as bullets) of the product was estimated by density measurement and pure bullets were assumed to contain 80% Pb and 20% Cu. After grinding, the remaining bullet-free coarse soil fraction was analyzed by XRF to take into account the small metal particles and metal dust attached to the soil. Based on the analyzed and estimated metal contents, the calculated average concentrations in the bulk samples and the distribution of the metals in the various particle size fractions are also given in Tables 2 and 3.

The Finnish environmental authorities have proposed reference values for lead and copper concentrations in soils (Puolanne *et al*. 1994). In the case of lead, the figures are 60 mg/kg and 300 mg/kg, while in the case of copper the corresponding figures are 100 mg/kg and 400 mg/kg. Although discretion can be exercised, generally, concentrations below the lower values, referred to as guideline values, are considered harmless, while concentrations exceeding the higher values, referred to as limit values, call for clean-up measures. As can be seen from Tables 2 and 3, the lead and copper concentrations in the studied soils exceed the limit values by even a thousandfold.

Table1. Distribution of the studied soils in the various particle size fractions.

Sample	Mass of the soil in the oversize fraction (%)	Mass of the soil in the coarse fraction (%)	Mass of the soil in the medium fraction (%)	Mass of the soil in the fine fraction (%)
Viikinmäki	1.4	37.3	8.3	53.0
Utti	0.1	13.0	15.2	71.7

Table 2. Concentrations and distribution of lead and copper in the various particle size fractions of the Viikinmäki sample. Calculated average concentrations of the bulk sample are also given.

Element	Minus 20 plus 2.8 mm size fraction, the coarse fraction		Minus 2.8 plus 1 mm size fraction, the medium fraction		Minus 1 mm size fraction, the fine fraction		Calculated mass-weighed average concentration (mg/kg)
	Concentration (mg/kg)	Percentage of total amount	Concentration (mg/kg)	Percentage of total amount	Concentration (mg/kg)	Percentage of total amount	
Pb	237 000	73.3	135 000	9.4	39 000	17.3	120 000
Cu	59 000	87.6	34 000	11.2	600	1.2	25 000

Table 3. Concentrations and distribution of lead and copper in the various particle size fractions of the Utti sample. Calculated average concentrations of the bulk sample are also given.

Element	Minus 20 plus 5.1 mm size fraction, the coarse fraction		Minus 5.1 plus 1 mm size fraction, the medium fraction		Minus 1 mm size fraction, the fine fraction		Calculated mass-weighed average concentration (mg/kg)
	Concentration (mg/kg)	Percentage of total amount	Concentration (mg/kg)	Percentage of total amount	Concentration (mg/kg)	Percentage of total amount	
Pb	588 000	67.4	139 000	18.6	22 000	13.9	113 000
Cu	146 000	80.7	25 000	16.2	1 100	3.1	24 000

It has been shown (Tanskanen *et al*. 1991, Lin 1996, Jörgensen and Willems 1987) that lead from the bullets after shooting into berms begins to transform into secondary lead compounds, which may be found as surface coatings on the lead particles or as fine-grained weathering products in the surrounding soil. In our earlier work, we conducted detailed mineralogical studies in order to identify the lead-bearing solid phases in the fine fraction of the Viikinmäki sample (Hintikka *et al*. 1999) and found the lead to exist mainly as hydrocerussite, $Pb_3(CO_3)_2(OH)_2$, and cerussite, $PbCO_3$, associated with minor amounts of anglesite, $PbSO_4$, and massicot, PbO. Also in the Utti sample, the transformed lead is present mainly as carbonates. The mineralogy of the copper phases has not yet been studied.

By studying Tables 1–3, it is obvious that the majority of the metals in the berms exists as whole bullets or their coarse parts, which were concentrated in the coarse soil fractions containing 37.3% (in Viikinmäki) or 13.0% (in Utti) of the total mass, but 73.3 % (in Viikinmäki) or 67.4% (in Utti) of the total lead. However, 17% (in Viikinmäki) or 14% (in Utti) of the total lead was found in the fine soil fractions emphasizing the significance of the weathering and transformation processes.

The remediation methods used

Heavy medium separation

In this study, we used heavy medium techniques employing the so-called Erickson’s cones to separate whole bullets and coarse bullet parts from the coarse and medium soil fractions. This technique is a standard method for separating minerals of different specific gravity, particularly when a rather big difference between specific gravities occurs. Heavy liquids or suspensions of suitable density are used, so that minerals lighter than the medium float, while the denser minerals sink. In this study, the heavy medium was an aqueous suspension of finely ground ferrosilicon (an alloy of about 82% iron and about 16% silicon) having a pulp density of about 3.4 kg dm^{-3}. In this medium, the bullets and their coarse parts sink, while the metal-poor soil floats. During the heavy medium separation, the secondary lead and copper phases attached to the surface of the bullets partly loosened and the fine-grained weathering products ended up in the middling product, i.e. the medium suspension. The magnetic properties of the ferrosilicon medium are utilized to separate it from the nonmagnetic lead and copper phases, which can be combined with the original fine fraction and processed as described below.

Shaking table concentration

Shaking table is a gravity concentrating device consisting of a slightly inclined deck shaken with a differential movement in the direction of the long axis and washed at a right angle to the direction of motion by a stream of water. The minerals, fed as ca. 25% suspension, at the upper corner of the table are thus subjected to two forces – one due to the table motion and the other, at right angles to it, due to the flowing film of water – causing size and specific gravity classification. The net effect is that the smaller denser particles, in our case small metal particles and their transformation products, can be separated as concentrate from the larger lighter particles constituting the tailings. Along with a high-grade concentrate, denoted by C_1 below, and tailings, a middling product, denoted by C_2, is also often separated for further processing. In this study, shaking table was used in the treatment of the fine soil fractions and the float products of the heavy medium separation.

Complex-forming leaching

In hydrometallurgy, leaching is a standard method for extracting valuable metals from solids into an aqueous solution. It is necessary that the particle size of the solid is small enough (<1 mm) in order to liberate the leachable material, that the waste material has a solubility low enough to yield an acceptable separation during the leach, and that the leaching agent can be regenerated. In the practical leaching systems, complex ion formation in the solution is often utilized to increase the solubility of the metal concerned. Based on our earlier studies (Hintikka *et al.* 1999), we used a hot (70 °C) concentrated (470 g dm^{-3}) calcium chloride solution at low pH (1.0 achieved by HCl addition) to remove lead and copper from the fine soil fraction into the solution. After leaching, the lead- and copper-poor fine soil fraction, i.e. the remediation product to be deposited, is separated by filtration and washing. The solution then enters a subsequent process stage, in which the leaching agent is regenerated by sulfide addition liberating the chloride ions bound to the complexes and converting the dissolved lead and copper into corresponding solid sulfides. The metal sulfides are separated by filtration or flotation and recycled along with the bullet scrap recovered by the heavy medium separation.

RESULTS

Heavy medium separation

Heavy medium separation was used on the coarse and medium soil fractions obtained by screening in order to remove the whole bullets and their coarse parts as the sink product. As can be seen from Table 4, in the Viikinmäki case practically all of the lead and copper present in these soil fractions could be recovered in the sink product at a grade of 720 000 mg Pb / kg and 180 000 mg Cu / kg, which means that the sink product was 90% pure bullets. No copper could be analyzed in the float product, while the analyzed lead concentration of the float product was 300 mg/kg indicating the attachment of minor amounts of lead to coarse soil particles.

In the case of the Utti sample, the performance of the heavy medium separation was not as good as can be seen from Table 5. Although the sink product was also in this case 90% pure bullets, considerable amounts of copper and lead remained in the float product at significant grades. The float

Table 4. Heavy medium separation tests on the coarse and medium fractions of the Viikinmäki soil.

	Percentage of the soil	Pb concentration (mg/kg)	Percentage of total Pb	Cu concentration (mg/kg)	Percentage of total Cu
Sink product					
coarse soil fraction	12.3	720 000	73.3	180 000	87.6
medium soil fraction	1.6	720 000	9.3	180 000	11.1
both fractions combined	13.9	720 000	82.6	180 000	98.7
Float product					
coarse soil fraction	25.0	300	0.1	0	0.0
medium soil fraction	6.7	200	0.0	0	0.0
both fractions combined	31.7	300	0.1	0	0.0

Table 5. Heavy medium separation tests on the coarse and medium fractions of the Utti soil.

	Percentage of the soil	Pb concentration (mg/kg)	Percentage of total Pb	Cu concentration (mg/kg)	Percentage of total Cu
Sink product					
coarse soil fraction	10.5	720 000	66.7	180 000	80.5
medium soil fraction	1.0	720 000	6.4	180 000	7.7
both fractions combined	11.5	720 000	73.1	180 000	88.2
Float product					
coarse soil fraction	2.5	36 000	0.8	1 100	0.2
medium soil fraction	14.2	98 000	12.2	14 000	8.3
both fractions combined	16.7	89 000	13.0	12 000	8.5

product of the medium soil fraction was especially problematic containing 12.2% of the total lead and 8.3% of the total copper at grades of 98 000 mg Pb / kg and 14 000 mg Cu / kg.

The reason for the different behavior is assumed to be the different ways of screening the samples. The Utti sample was easily dry-screened, while the Viikinmäki sample had to be wet-screened due to the high amount of vegetation-oriented material. While screening the Utti sample, clods of fine-grained soil probably remained on the screen but decomposed later in the heavy medium apparatus. This increased the viscosity of the suspension and, consequently, decreased the selectivity of the separation.

Shaking table concentration

The float products of the heavy medium separation and the original fine soil fractions were mixed in mass-weighed ratios (25.0:6.7:53.0 in the Viikinmäki case and 2.5:14.2:71.8 in the Utti case) and introduced to the shaking table concentration. In the Utti case, shaking table concentration proved to be successful in recovering the bullet weathering products. 12% of the total lead could be recovered in shaking table concentrate and middling product at a grade of 345 000 mg/kg in the high-grade concentrate C_1, with the corresponding figures for copper being 2.9% and 16 000 mg/kg. The grade of concentrate C_1 was high enough so that this product could be combined with the sink product of the heavy medium separation. Hence, 85.1% of the total lead could be recovered in the combined gravity separation process at a grade of 620 000 mg/kg. The corresponding figures for copper were 91.1% and 140 000 mg/kg.

In the Viikinmäki case, the concentrations of lead and copper in the float products of the heavy medium separation were negligible and, consequently, no significant recoveries could be achieved by employing the shaking table. In conclusion, we can note that, when needed, shaking table concentration should be used to complement the performance of the heavy medium separation.

Complex-forming leaching

Complex-forming leaching was applied to the middling product, C_2, and the tailings from the shaking table separation, separately, to remove the lead and copper from the fine disintegrated bullet weathering products. The leaching experiments were carried out the same as in our earlier study (Hintikka *et al.* 1999) and the results on leaching the lead from the Viikinmäki samples are illustrated in Fig. 1. The kinetic data given in this figure indicate that a majority of the lead dissolves immediately and that leaching times longer than one hour do not contribute significantly to the performance of the process. In the case of the shaking table middling product of the Viikinmäki sample, 98% of the lead could be recovered into the solution resulting in a residual lead concentration of 350 mg/kg in the soil. High lead recovery, 96.7%, was also observed when leaching shaking table tailings, but due to the high initial lead concentration of 69 000 mg/kg, the residual concentration in this soil fraction was still over 2000 mg/kg. However, shaking table tailings form only a minor part of the total soil and, in any case, better results will be obtained after further optimization of the leaching conditions.

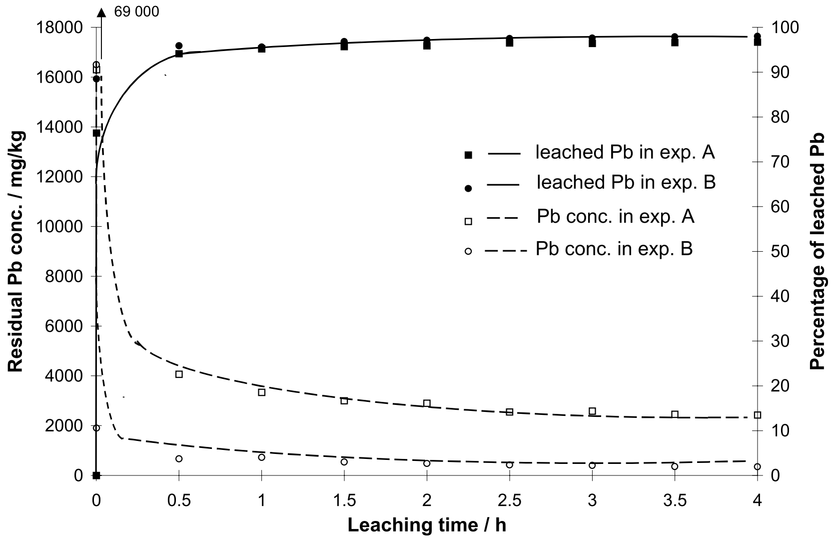

Figure 1. The recovery of lead into solution and the residual lead concentration in the soil as a function of time in the course of the leaching experiments A (■, □, shaking table tailings of the Viikinmäki sample) and B (•, o, shaking table middling product of the Viikinmäki sample). The solid lines and filled symbols refer to the right y axis, while broken lines and open symbols refer to the concentration axis (left y axis). The initial Pb concentration in experiment A is off the scale, 69 000 mg/kg.

CONCLUSIONS

Bench-scale tests on the soils from the Viikinmäki and Utti rifle ranges show that mineral processing techniques can be successfully used for their environmental remediation. Most of the lead and copper in the soil samples is in the form of bullets and their coarser parts, which can be easily recovered mechanically by combining screening with a consecutive gravity concentration stage. In the Viikinmäki case, 82.6% of the total lead and 99% of the total copper could be recovered as a 90% pure bullet product by wet-sieving followed by heavy medium separation. In the Utti case, the sample was dry-sieved making the performance of the heavy medium separation worse. This could, however, be compensated for by a subsequent shaking table concentration stage resulting in a lead recovery of 85.1% at a grade of 620 000 mg/kg, the corresponding figures for copper being 91.1% and 140 000 mg/kg.

The metals present as disintegrated weathering products in the fine soil fraction (particle size below 1 mm) represent the greatest risk to the environment due to the higher solubility of these products. As in our earlier study dealing with shotgun shooting ranges (Hintikka *et al*. 1999), complex-forming leaching proved to be efficient in removing the lead and copper from the fine-grained soil fraction.

By considering the composite process – consisting of wet-sieving, heavy medium separation, an eventual shaking table concentration, and a leaching stage – a total lead recovery of 99.4% can be achieved resulting in an average residual lead concentration of 700 mg/kg in the remediated soil. However, the experimental conditions have not yet been fully optimized and, most probably, a lower residual lead concentration can be achieved allowing the remediated soil to pass the limit value of 300 mg/kg set by the Finnish authorities (Puolanne *et al*. 1994).

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REMEDIATION CENTRE FOR CONTAMINATED SOILS

by
Lauri Kivekäs

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A versatile remediation centre for contaminated soils has operated at Virkkala close to Helsinki since 1998. It is the first regional soil remediation centre in Finland and serves a large area in south-western Finland. Contaminated soils are collected from tens of small and large sites each year and transported to Virkkala for a very high class and environmentally safe treatment under controlled conditions. The centre consists of a 2-ha large hall for storage and treatment of the soils, a 1 ha outside bio-remediation field and a service & truck washing hall. Three treatment technologies are available at the centre: Stabilisation/ solidification, washing and bio-remediation. With these methods all the most common types of contamination can be treated: Heavy metals, oils, PAHs, creosotes and chlorophenols. Special care has been taken with prevention of environmental emissions, because the centre is located close to a housing area and on a lake-front. All the storage and treatment areas are covered with a double or triple bottom liner system and all environmental emissions are being monitored constantly. The EU's Life Environment fund has supported the project.

Keywords: soils, pollution, remediation, methods, environmental protection, Virkkala, Finland

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INTRODUCTION

Remediation of contaminated soils was started in the early 1990's in Finland. In the national survey published in 1995 about 10 000 contaminated sites were identified, most of them in south-western Finland. In the beginning the remediation was only done on-site with mobile equipment, but it was soon realised that several regional remediation centres are necessary for more effectively taking care of the problem. Lohja Envirotec started to plan the Virkkala soil remediation centre in 1994 and after a four-year long environmental permit process (in which one of the first mandatory Environmental Effects Assessment studies, EEA, was done as part of the process) the centre was opened in 1998 as the first regional soil remediation centre in Finland. The centre has been built in three phases, the last one of which was finished in the summer of 2000.

VIRKKALA SOIL REMEDIATION CENTRE

The Virkkala soil remediation centre is a regional centre, where contaminated soils from several sites are being collected and treated in large volumes utilising three technologies.

The centre is located 65 km west of Helsinki in the middle of the triangle formed by the three largest Finnish cities and with a third of the country's population living in the area. Contaminated soils are normally transported from a distance of up to 200 km, but in some cases even from 500 km away. The centre utilises the premises and facilities of a former cement factory, which was closed in 1993. This has provided a splendid opportunity to utilise the available equipment, buildings and infrastructure with much lower costs than in the case of a green-field operation.

The main facility of the centre is a 20 000-m^2 large, 15-m high concrete hall (a former limestone homogenisation hall), inside of which most of the storage and treatment of the soils takes place. Truck washing, service works, social facilities and office facilities are located in the adjacent former truck service hall. A 10 000-m^2 large bio-remediation area and the rain-water collection system are located outside the hall.

In the environmental permit the maximum allowed treatment capacity is limited to 80 000 tons/a, but in the beginning the volumes are 20–30 000 tons/a. The permit allows treatment of soils contaminated with heavy metals, oils, PAHs, creosotes and chlorophenols.

The construction and operation of the centre have also served as a demonstration project in the European Union's Life Environment programme, which has also funded the project considerably.

TREATMENT METHODS

The founding idea of the centre has been to combine several of the best available treatment methods in the same place. The treatment methods have been chosen so that all the most common types of soil pollution can be treated (Fig. 1): The solidification/stabilisation method can be used for heavy metals, soil washing method for organic substances and heavy metals and bio-remediation for organic substances. The availability of all the three methods also allows for effective treatment combinations, so-called "treatment trains"; for example, washing+solidification or washing+bio-remediation.

Solidification/stabilisation

In Finland there are plenty of sites contaminated with heavy metals originating from wood preservation chemicals, many of which contain heavy metals such as arsenic, chrome and copper. Lead is also a common contaminant in such sites. These heavy metals are difficult to treat, because they cannot be destroyed. The only way is to immobilise them by binding them in insoluble form, with chemicals and binders, such as cement. The method is also called eco-concreting, because the soil is actually turned into concrete. This end product, eco-concrete, is normally utilised as a valuable earth construction material as the base layer in fields or roads. After hardening, the leaching of harmful substances becomes so low that the material fulfils the strict Dutch limit values.

At the Virkkala centre, eco-concreting is done with mobile high-capacity mixers, which are taken into the hall twice a year. These Australian continuously operating Aran mixers can treat 1000 m^3 of soil per day.

Eco-concreting took place for the first time in the autumn of 1999. Altogether about 6700 tons of contaminated soil have so far been used in the production of eco-concrete. Contaminated soils directly as well as the dirty fraction of soil washing have been used in the production of eco-concrete. These treated soils were contaminated with copper (Cu), chrome (Cr), arsenic (As), lead (Pb), zinc (Zn), nickel (Ni) or cadmium (Cd). Water that was separated from the soils was utilised in eco-concrete.

Eco-concrete has been utilised in the base layers of waste management fields at two landfill sites. Pre-laboratory analyses were carried out and test samples were taken for a series of analyses to make sure of the suitability of the eco-concrete for these areas and to confirm that the environmental requirements were met. The test results and the requirements are summarised in Table 1.

The leaching of harmful substances from eco-concrete is so low that they have fulfilled the Dutch limit values and the authorities have therefore allowed the material to be used as a construction material in earth construction.

Soil washing

Soil washing is a method consisting of a long process where the contaminated soil is separated into several grain sizes. The idea is that, because the contaminants are bound onto the finest particles, the finer and finer materials are separated in each step from the coarser materials, which are practically free of contaminants. The washing plants normally consist of several (10–30 pieces) components, including sieves, scrubbers, floating, spirals, etc. The process produces clean material, which is normally 70–90% of the original volume, and the end slurry, which normally constitutes 10–30% of the original volume and contains practically all the contaminants. This end slurry must then be treated by solidification or bio-remediation, depending on the type of the contaminants.

At the Virkkala centre there is a Swedish washing plant, which can treat 5–10 tons/h. The method is suitable for treating sandy soil contaminated with organic or inorganic contaminants.

The plant is located inside the hall where it can also operate under mild winter conditions.

Soil washing started in 1999. About 1400 tons of soil have been treated with soil washing so far, containing copper (Cu), chrome (Cr), arsenic (As), lead (Pb), zinc (Zn) or cadmium (Cd). In one lot of soil the feed concentration of lead in the contaminated soil was about 1000 mg/kg and the final concentration in the treated soil ready for disposal was about 100 mg/kg. Soil with a lead concentration of below 300 mg/kg can be disposed of at a landfill in Finland.

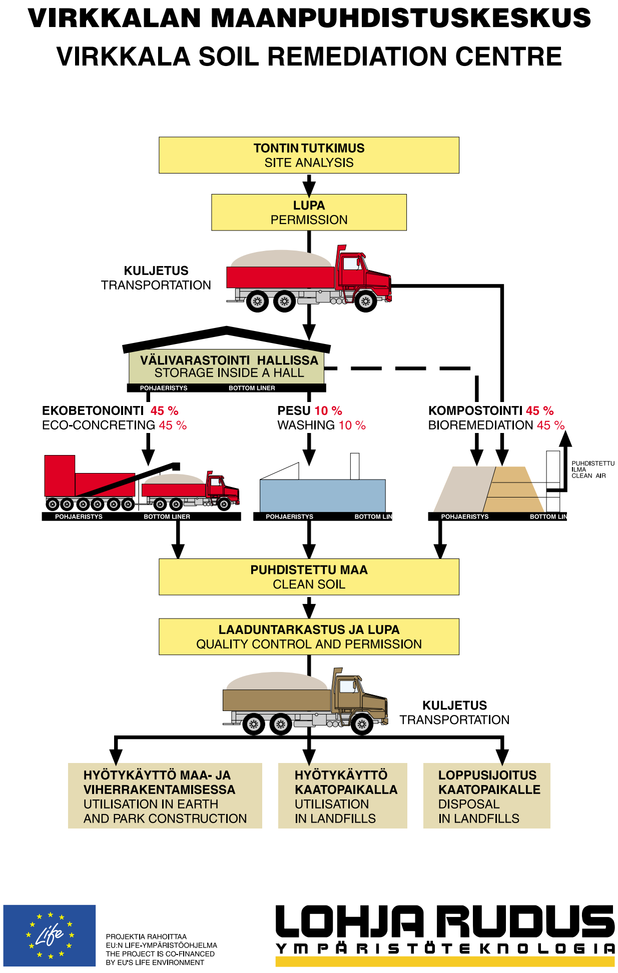

Figure 1. Treatment process of the Virkkala soil remediation centre.

Table 1. Results of laboratory tests on eco-concrete

	Laboratory result	Requirement
Strength	4.3 MPa	>3.5 MPa
Water permeability	4.2 x 10^{-10} m/s	<1x 10^{-9} m/s
Frost durability	3.8%	<10%
Solubility		
As	<6.8 mg/m^2/64d	1B <140 mg/m^2/64d
Cr	<3.4 mg/m^2/64d	1B <480 mg/m^2/64d
Cu	31 mg/m^2/64d	1B <170 mg/m^2/64d
Pb	<20 mg/m^2/64d	1B <400 mg/m^2/64d
Ni	<13 mg/m^2/64d	1B <170 mg/m^2/64d
Zn	<7 mg/m^2/64d	1B <670 mg/m^2/64d

Bio-remediation

Bio-remediation is a very common method used for treatment of organic contaminants, for example, in the case of gas station remediation projects. In this method the soil's own bacteria deal with the contaminants, if the right conditions (air, temperature) are provided inside the soil heaps. So far in Finland bio-remediation has been used only as the traditional method, where the uncovered soil beds are being turned now and then. However, this traditional method is not environmentally sound, because the contaminants are mostly evaporated into the air and only a small proportion of them is treated by the bacteria.

At the Virkkala centre, a so-called bio-pile method is being used for the first time in Finland. In this method the soil beds are covered by plastic sheets and air is pumped through a set of pipes into the soil and then sucked out through another set of pipes. The sucked-out air is then cleaned either by active coal or by catalytic burning or by biological methods, before returning into the bed again. If necessary, the air is heated under winter conditions. The bio-pile method is more effective and environmentally safe than the traditional one. The bio-remediation process is operated by the largest Danish bio-remediation contractor Bioteknisk Jordrens Soilrem A/S.

Bio-remediation with soil vapour extraction (SVE) treatment was taken into operation in January 2000, and so far about 6800 tons of contaminated soil have been treated. All of it was contaminated with creosote. The feed concentration in the soil was up to 1500 mg/kg and the final concentration in the treated soil ready for disposal corresponded to the target limit values (sum of PAH compounds <200 mg/kg). The vapour collected in the treatment was burnt catalytically.

END-PLACEMENT OF THE TREATED SOILS

After remediation has been successfully done and controlled, all the soil is taken out of the Virkkala centre, because there is no end-placement capacity there. So far all the end-placement has been provided by the regional waste management company Rosk'n Roll, owned by the 12 communities and some industrial enterprises in the region. At Rosk'n Roll's two landfill sites, the treated materials have been 100% utilised as earth construction materials in the construction of new roads and fields, and also as the daily covering material. In the future, utilisation will also be considered in other earth construction structures outside the landfill sites.

ENVIRONMENTAL PROTECTION AT THE CENTRE

Since the start of the planning of the Virkkala centre, environmental protection of the centre's site and the surroundings has been one of the major issues, because it is located only 100–200 m from the nearest houses and on the lakeside. In the very thorough EEA process it was found out that with certain environmental protection measures the centre can be located on the planned site. The major environmental protection actions were prevention of leaching of contaminated water into the ground and prevention of dusting. One of the important actions against emissions was the decision to locate most of the activities inside the hall under controlled conditions and protected from rain and wind. On top of this, the following special actions have been taken:

Bottom liner system

During construction of the Virkkala centre all the storage and treatment areas for contaminated soils were covered with a special watertight liner system, which was developed and greatly improved during the construction period. In 1998 in the 1st phase of the construction the bottom liner construction consisted of a sand bentonite mineral liner and two layers of a special, and at that time considered as watertight, asphalt (so-called "Lemdense"). However, soon after the first soil deliveries were received, it was found out that the Lemdense asphalt was leaking. This was noticed because there is a drainage system between the asphalt layers and the sand bentonite layer. Actually, this structure has acted as a giant lysimeter providing valuable information on the effectiveness of asphalt liners. The real water-tightness (k-value) of the asphalt liner was found to be about 100 times higher (i.e. worse) than the design value given by the asphalt producer and required by the structure designer. These experiences from the Virkkala centre have also lead to changes in the design of landfill liners: If the "Lemdense" type of asphalt liners is being used instead of the plastic liner, there should be a layer of rubber-bitumen asphalt (so-called "Lemproof") on top in order to reach the required water-tightness. A more detailed description of this case is given in Kari Ruohonen's presentation at this same Ecogeo 2000 congress.

Based on these experiences, a greatly improved bottom liner system was used in the construction of the 2nd phase in 1999 (Fig. 2). This was a triple liner system with a polymer modified sand bentonite mineral liner (Trisoplast), a 2 mm HDPE plastic liner and two layers of the special asphalt. The polymer modified sand bentonite (Trisoplast) is a Dutch product, which has several greatly improved properties compared to normal sand bentonite. It is more watertight, flexible, resistant to chemicals, etc. Due to these properties the thickness of the mineral layer can be diminished by more than 80% compared to normal sand bentonite without compromising in any of the required properties. This results in considerable savings in landfills and all other types of liner constructions. The HDPE liner was added to the structure, because the asphalt layers could not be considered as a second liner.

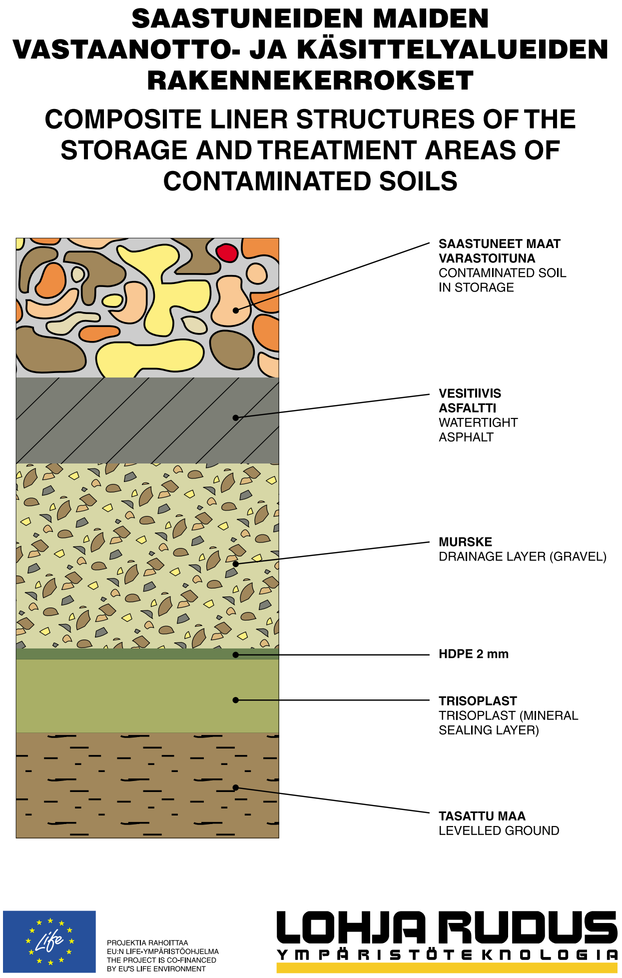

Figure 2. The improved bottom liner system of the Virkkala soil remediation centre.

Emission control system

A special environmental emissions control program was taken into use already before the construction of the Virkkala centre was begun. Ground water quality is being monitored from several wells inside the centre area, soil samples are analysed both from the centre area and from the nearby housing area; air emissions are being monitored inside the hall, outside in the centre area and in the nearby housing areas. A special "odour team" with representatives from the neighbourhood was nominated and all the people living in the neighbourhood have been given a chance to keep "odour diaries". An annual environmental report is given to the authorities and a shorter version to the inhabitants. No significant emission levels have been detected so far.

One issue to be considered was the possibility of contaminated dust spreading into the vicinity by truck tyres. To prevent this a special truck washing line has been built and the driveway leading from the hall to the washing line has also been covered with a liner and water collection system.

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REMEDIATION OF SITES WITH COAL TAR CONTAMINATION – A CASE STUDY

by
Reidar Zapf-Gilje[1], Guy C. Patrick[1] and Pekka Lindroos[2]

Zapf-Gilje, Reidar, Patrick, Guy C. & Lindroos, Pekka 2001. Remediation of Sites with Coal Tar Contamination – a Case Study. *Geological Survey of Finland, Special Paper 32, 165–174,* two figures.

The production and use of coal tar was tied to the industrial revolution and its dependence on coal for energy and as chemical feedstock for a large range of organic and inorganic products. Coal tar was produced, often as a byproduct, by coal gasification plants. The North American coal tar production in the mid 1950s was in the order of 25 billion litres. The production, handling, storage and use of coal tar and its derivatives generated a legacy of soil and groundwater contamination that today requires remediation at high costs. At one such site, coal tar was manufactured into a variety of roofing and tar-based products, as well as the production of creosote, oil stains, solvents and anhydrous ammonia. Over its 60 years of operation, a number of chemicals were leaked, dumped or released to the soil and groundwater on the site, of which the most significant was a brown dense non-aqueous phase liquid (DNAPL) with an oil-like viscosity. This DNAPL migrated from the fill, through a pre-development floodplain silt layer and into an underlying sand aquifer. Portions of the DNAPL moved along preferential pathways associated material with the coarser material in the aquifer and reached the nearby river sediments, resulting in elevated PAH concentrations of polyaromatic hydrocarbons (PAH). Site remediation was conducted mitigate risks posed by the coal tar. Remediation has included: in-place management of deep land-based soil contamination, found at depth, removal of shallow soil with high PAH concentrations (*i.e.* 10 times the provincial concentration standards for commercial land use), control of dissolved contamination in groundwater, and recovery of free-phase creosote. The remediation also provided long-term protection of the adjacent aquatic habitat through a combination of groundwater and DNAPL control and recovery, removal of near-shore contaminated sediments, and containment and natural attenuation of far-shore contaminated sediments through the use of a layer of crushed rock placed as a protective cap over the sediments.

Keywords: environmental geology, soils, ground water, pollution, coal tar, polycyclic aromatic hydrocarbons, remediation, case studies

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INTRODUCTION

The production and use of coal tar was tied to the industrial revolution and its dependence on coal for energy and as chemical feedstock for a large range of organic and inorganic products. Coal tar was produced, often as a byproduct, by coal gasification plants. The production, handling, storage and use of coal tar and its derivatives generated a legacy of soil and groundwater contamination that today requires remediation at high costs. Often these facilities were located at or near shorelines for receiving of coal and shipping of products. This led to contamination also of adjacent water bodies and sediments.

Coal tar was produced by coal gasification facilities that produced gas for city lighting and power usage, and the production of coke for the steel industry. The ready availability of coal tar provided ample supply for distillation into creosote for the wood preserving industry. Most major cities in the North America and Europe had coal gasification and coking plants. In the US, 1,150 city gasworks were registered (Lotimer 1992), and total coal tar production in the mid 1950s was in the order of 20 billion litres (Austin 1984). The number of registered gasworks in Canada was approximately 150, and in Denmark 125 (Johansen 1997).

Coal tar production led to the development of a large secondary industry for processing of the tar, including: roof shingle and tar paper for construction; road tars for paving, wood treatment (both tar and creosote derived from tar), and coal tar derivatives used in paints and enamels. These secondary industries switched over to oil-derived tars and solvents in the 1950s, as the manufactured gas plants were being phased out in favour of electricity and petroleum fuels.

Coal tar from coal gasification plants was used by several industries, including the manufacturing plant formerly located at the site discussed in this paper. This plant shut down about 20 years ago and has recently been remediated for redevelopment for commercial use. This paper provides an overview of the contamination found at the site and the remediation approach selected.

COAL TAR AND ITS BYPRODUCTS

Coal tar was commonly produced by high-temperature carbonization of bituminous coals (Austin 1984). The coal tar was further processed to manufacture chemical feedstock for a variety of products including solvents, creosote, roofing materials, and road tars, etc. The main chemical components found in these products include:

paraffins and cycloparaffins
benzene, ethylbenzene, toluene, xylene
low molecular weight PAH (naphthalene, 2-methylnaphthalene, phenanthrene, anthracene, 1-methylnaphthalene, biphenyl and fluorene)
high molecular weight PAH
heterocyclic aromatic compounds (HAC), which may have included quinoline, isoquinoline, carbazole, 2,4-diethylpyridien, acridine, 2-methyquinoline, 4-methylquinoline, pyrrole, pyrrolidine, benzo(b)thiophene, dibenzothiophene and dibenzofuran
phenolics (phenol, cresols, pentachlorophenol, xylenols and 2,3,5-trimethylphenol)
ammonia solutions
sulfate, sulfide, thiosulfate
cyanide, ferro-cyanide, thiocyanide
carbonates and chlorides
acids and bases
metals including sodium, calsium, iron, and others

Physical properties of coal tar that largely determine its behaviour in the subsurface include its specific gravity, which is near that of water, its high viscosity, and low interfacial tension. The specific gravity of coal tar is typically around 1.05. Consequently, it is a DNAPL that will slowly sink when placed in water, although separation into both floating and sinking phases has been reported. Viscosity will vary widely depending on coal tar composition and temperature, but typically is much higher than water, ranging from that of a crude oil (about 20 centipoise) to that of thick molasses (perhaps 50 to 80 centipoises). Such viscosities reduce the velocity at which the coal tar can migrate through soil, although they will have little effect on the ability of the coal tar to flow. Coal tar migration through a porous medium is determined to a much greater extent by the interfacial tension between the coal tar and the soil porewater. Where coal tar has been recovered from soil, interfacial tensions are typically low, on the order of 5 to 10 dynes per centimetre. This suggests that coal tar may travel further and leave behind much lower residual concentrations in soil than would be expected for comparable petroleum-based heavy oil.

MAIN ENVIRONMENTAL ISSUES

The bulk of the compounds found in coal tar are PAH. PAHs are a diverse group of organic compounds composed of hydrogen and carbon atoms arranged in two or more fused aromatic (benzene) rings. PAHs are grouped into two categories based on their molecular structure: 1) low molecular weight (LMW) compounds with fewer than four rings; and 2) high molecular weight (HMW) compounds with four or more rings. LMW-PAH are generally more soluble in water than HMW-PAH. At the site discussed in this paper, the coal tar DNAPL was composed of about 75% of the PAH, which were mainly LMW category-PAH. PAH are also formed by a variety of other natural and anthropogenic processes including biosynthesis and incomplete combustion of organic materials.

Several studies have demonstrated that PAH associated with coal tar can pose risks to aquatic organisms. At sufficiently high concentrations, PAH can cause reduced survival, reduced growth, tumors, developmental effects, enzyme induction, behavioral abnormalities, habitat degradation, fish tainting, and ultraviolet-induced skin lesions (Sved *et al.* 1997, EAG 1988; Eisler 1987, Neff 1979).

Terrestrial activities are much less affected by PAH contamination found at these sites, as it is generally located at depth, and the hydrocarbons present in creosote are not sufficiently volatile to create soil vapour concentrations that would pose a potential threat to people, plants or animals.

SITE CHARACTERISTICS

History and Use

The site was used primarily for the manufacture of roofing, paving and other building materials. Manufacturing operations started near the turn of the century and continued for a period of about 60 years. Major products included:

Roofing shingles, paving and waterproofing materials;
Building papers, roofing felts and roofing pitch;
Creosote, oil shingle stains and enamels;
Coal and tar pitch for road construction, repairs and maintenance;
Asphalt shingles and slate surface roofing;
Chemicals and solvents; and,
Anhydrous ammonia

Coal tar feedstock was provided from at least one manufactured gas plant. The coal tar was brought in by train and barge, and stored in above-ground tanks for processing and resale. The coal tar was distilled into a number of different grades: heavy distillates, used for paving materials (road pitch) on roads, and lighter grades of distillate (mixed with additives) to produce creosote oil, shingle stains, roofing pitch, and paints and enamels. Steam lines from a boiler heated the coal tar, which was stored in one of several large storage tanks, to liquefying temperatures so that it could be pumped. During the latter years, petroleum-based asphalts (oil-based) replaced coal tar as the principal feedstock. Asphalt was typically trucked in from local refineries and pumped directly into the asphalt storage tanks.

Over its period of operation, soil and groundwater contamination resulted from several mechanisms such as:

Spills, leaks, and drips during unloading, transfer, handling and storage of raw materials and distilled products;
Placement of materials from the property along

the foreshore during foreshore filling;
Random on-site filling; and,
Releases of chemicals caused during the demolition and decommissioning of structures on the property.

The potential contaminants of concern identified from the historical reviews of site operations included a) coal- tar derivative oils that were produced at various stages of refining and distillation, b) asphalt and road pitch materials, c) bunker C and other heating fuels, d) some liquid thinners, and e) metals from the granules used in the manufacturing of shingles. Of these, coal-tar derivative oil is the most significant. Coal-tar derivative oil is typically brown in colour and can have a consistency of a moderately light oil such as (creosote and other oils). Coal-tar derivative oils contain mostly of both (mainly PAH) and some monocyclic aromatic hydrocarbons (MAH). The MAHs include the benzene, toluene, ethylbenzene and xylene compounds. When present in a mixture with PAHs, the MAHs in coal-tar derivative oils are generally less soluble in water than would be observed in typical gasoline mixtures. Coal-tar derivative oil commonly has a specific gravity at typical soil and groundwater temperatures that is higher than but very near that of water. Consequently, and, when introduced into the environment, it behaves as a DNAPL.

Regional Geology

Regionally, the site lies within a geological bedrock basin that has been infilled with glacio-marine, alluvial (deltaic), swamp, and flood plain deposits (Fig. 1). The infilled deposits are a few hundred metres thick near the river, and decrease in thickness to the north. Following the last glacial episode, sedimentation at the mouth of the river resulted in a thick sequence of fine-grained deltaic sand deposits that now comprise the river delta. The deltaic sands, which are laterally extensive, are typically between 20 m and 35 m in thickness. On the delta, natural sand levees that formed adjacent to the river channels deposited finer-grained silty sands and silts (overbank or floodplain silts) in the quieter areas upland of the levees. Consequently, the present topography at each of the sites is characterized as relatively flat, with native silt and/or peat overlying a thick, interbedded sequences of sands which, in turn, overlie thick Pleistocene deposits.

Sand Aquifer

The deltaic sands comprise a regional aquifer. The aquifer is hydraulically connected to the river, and responds to diurnal fluctuations in water levels at each of the sites caused by tides. Average groundwater flow is typically horizontal toward the river, and then upward into the river where it discharges. However, during periods in late spring when river flows are high and the water table is low, hydraulic gradients may reverse inland for periods of up to one or two months.

River Sediment Mobility

Sediments along the riverbed are eroded during large flow events and are replaced with sediment eroded from upstream areas as the flow event wanes. Contaminated sediments may be transported away from the site during the large flow events leaving behind freshly exposed contaminated sediments that will be subsequently reburied by newly arriving uncontaminated material from upstream. Fine sediments such as silts may accumulate over the site during the low-flow periods. These fine sediments form a surficial layer (veneer) that is re-mobilized during the next large flow event.

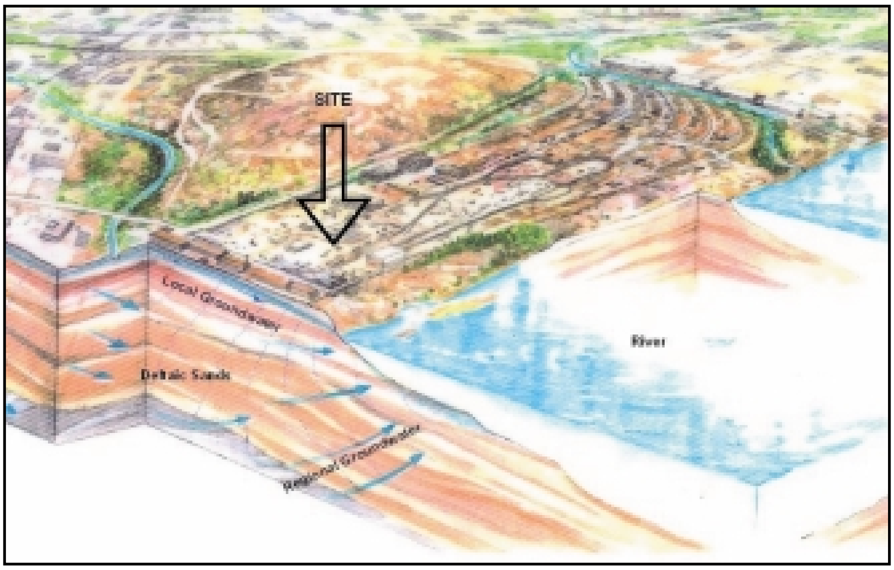

Figure 1. Regional hydrogeological diagram.

SITE INVESTIGATIONS

Scope and Methods

Detailed investigations have been conducted at the site to characterize the physical, chemical and biological subsurface conditions, including an assessment of potential exposure pathways. This information has been used to facilitate the subsequent evaluation of a range of remedial alternatives including in-place management of the contamination. Approaches that have been used in characterizing of the site are described elsewhere (Patrick and Anthony 1998), and include conventional shallow excavations (test pits), hollow stem drilling and sampling, and the use of push technologies (cone penetrometer testing) and continuous soil coring to define stratigraphy, soil chemistry, and the presence and extent of NAPL. Groundwater assessments have included conventional nested well installations as well as vertical chemistry profiling using, for example, the Waterloo Profiler^{TM}. Continuous water-level monitoring of tidal-response and damping in the aquifer, as well as conventional pumping tests have been conducted to evaluate aquifer properties, hydraulic gradients, and groundwater flow velocities and flux.

Continuous coring using the rotary sonic drilling method as well as Macro-Core^{TM} methods (Geoprobe Systems) has been used extensively at eachthe site to allow detailed stratigraphic description and direct observation of NAPL, where present. Typical coring depths have ranged from 5 m to 30 m, providing information through the full thickness of the Sand aquifer. Characterizing river sediment has been largely conducted from a barge using a conventional hollow-stem drill rig and shallow coring and dredging devices.

Where obvious NAPL is observed in soil or sediment cores, it is found almost exclusively in medium to coarse sand, and appears as viscous dark-brown to black globules or oily coatings. However, extensive areas of much lower NAPL concentrations are often revealed in fine sands by weak sheens, soil staining and strong odours. Occasionally, root holes stained by NAPL may be observed in cores of overbank silts located within source zones.

Contamination Sources

The known and inferred extent of the zones of NAPL is illustrated in plan and in section in Fig. 2. Each NAPL zone contains a heterogeneous distribution of coal tar, where concentrations may range from absent to several percent. Lateral spreading has occurred through relatively coarser grained strata bounded on the bottom by apparent capillary barriers of finer-grained material. The deepest zones of vertical penetration appear to have been limited to the areas where chemicals were released (*i.e.*, within those areas where chemical use and handling were greatest). NAPL has reached depths of about 12 to 15 m.

Dissolved PAH

Dissolved PAH emanating from the NAPL source zones are found in both shallow fill onshore, and within the Sand Aquifer both onshore and beneath the bed of the river. Concentrations of dissolved PAH within the source areas approach the expected solubility limits and, in some cases, appear to have exceeded solubility limits where sampling using conventional practice (*e.g.*, purging using foot-valve samplers or bailers) may have entrained NAPL micelles and/or PAH sorbed to sediment in the water samples. PAH concentrations in sediment a few metres beneath the discharge zones at the river are commonly one to two orders of magnitude lower than concentrations measured at the source zones. As discussed below, further reductions are observed within the riverbed prior to discharge, likely as a consequence of tidal pumping and natural attenuation.

DNAPL Mobility

A concern at the site was the possibility that NAPL was continuing to migrate from source zones toward and into river. For the most part, the soil chemistry data and stratigraphy suggested that the potential for NAPL migration under natural hydraulic gradients was low, and that the majority of NAPL represents residual, immobile product. However, NAPL has been recovered from wells at each of the sites. Mobility of DNAPL would be difficult to demonstrate, mainly because the NAPL velocity would probably be very slow (*e.g.*, perhaps on the order of a metre per year). Such low velocities are to be expected because of the high NAPL viscosity, weak stratigraphic slopes along which the NAPL moves, and the NAPL density near that of water.

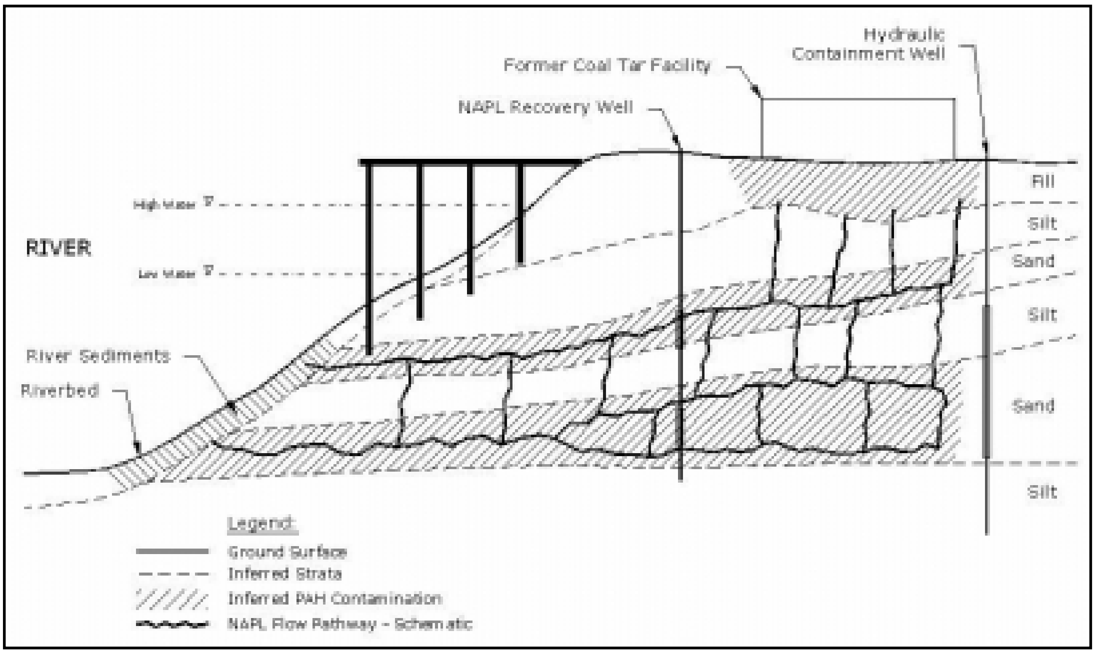

Figure 2. Schematic cross section of typical site conditions

Natural Attenuation

Natural attenuation processes are likely occurring within dissolved plume. The plume extends from the NAPL source onshore to zones of groundwater discharge within the river, within 20 m of the shoreline, (Fig. 2). With the exception of a thin mixing zone in the bed of the river, the plume is anaerobic, and composed mainly of naphthalene. At the river, a groundwater-river water mixing zone is created by tidal pumping, which effectively dilutes the plume and introduces dissolved oxygen and river water. The thickness of this zone is variable, depending on the permeability of the riverbed, and has been measured to be about 0.5 m thick. As a consequence of mixing, PAH degradation is promoted and the sediment porewater often has low PAH concentrations.

Sediment

NAPL occurs randomly in river sediment over an area, measuring about 20 m in width, and extending about 130 m to 160 m in length. Within these NAPL areas, the surficial sediments (to 0.6 m depth) have mean total PAH concentrations ranging between 100 mg/kg and 200 mg/kg, although much higher concentrations are occasionally reported. Total PAH concentrations in deeper sediments can be much higher, probably reflecting less weathering and higher NAPL mass, and higher concentrations at depth related to preferred NAPL migration through deep, gravelly lenses in the sand aquifer.

Outside of the primary PAH zone, a “halo” of surficial PAH-contaminated sediments exists. This secondary surficial contamination is the result of ongoing natural river processes of erosion, deposition, and tidal influences, which have mobilized and redistributed PAH-contaminated sediment from the primary zone. The mean total PAH concentrations within these secondary zones are typically around 10 mg/kg or less.

The riverbed along the foreshore is generally dominated by silt and sand, with little overhanging or instream cover (*e.g.*, riparian vegetation, boulders). Fish spawning and rearing habitat is, therefore, limited. Although the river is a migration corridor for salmonids, it is species such as starry flounder and peamouth chub that use the area for rearing and migration. Common benthic invertebrates found in these sediments include oligochaetes, chironomids, nematodes and copepods.

SITE REMEDIATION

Remediation Constraints

Site conditions posed a number of significant constraints on possible remediation options. These included considerations of minimizing disturbance to ongoing site operations, and ensuring the stability of site structures. The adjacent shipping channel with heavy commercial (tugboats with barges and log booms) and recreational traffic also posed some restrictions on potential sediment remediation options.

Constraints were also imposed by a number of natural conditions including: riverbank geotechnical stability concerns for the implementation of related to dredging or cut-off wall construction; presence of cobble layers beneath the sediments at depth, which could impede the installation of sheet pile cut-off walls; and the erosion and mobility of the river sediments caused by annual freshet flooding.

Remediation Strategy and Implementation

The unique aspects of the environmental contamination from facilities using coal tar related material has led to a remedial strategy of partial source removal, containment and interception. These strategies reflect the knowledge of where the leaks and spills occurred such as tanks and pipes and the science of coal tar migration. These locations are easily identified based on historical information and targeted for investigation, remediation and/or containment. The remaining contamination is significantly more diffuse as a result of random dumping of waste materials as fill or natural migration.

Based on the considerations discussed above, the remedial strategy involved a series of steps to achieve source control and risk reduction. This strategy reflected the knowledge of where the leaks and spills likely occurred, such as the vicinity of storage tanks and piping, and the science of coal tar migration. The source locations were easily identifiable based on historical information, and then targeted for investigation, remediation and/or containment. The remaining contamination beyond the main sources was significantly more diffuse as a result of random dumping of waste materials as fill or natural migration.

Source control involved groundwater pumping and treatment systems to hydraulically control dissolved chemicals and to recover DNAPL from the upland source zones. With these controls in place, the approach to final remediation wais based on defining actual risk to human health and the environment posed by the contamination, and on developing and implementing remedial works that a) reduce the risks to acceptable levels and b) meet the regulatory requirements for permanency and contaminant mass reduction. A large part of the remedial effort was, therefore, focussing on identifying areas of potential mobility of DNAPL, and implementing DNAPL control and recovery measures, and defining ecological risks posed by DNAPL and dissolved components to benthic communities in the river bed.

Interim Contamination Controls

Several interim contaminant controls were implemented prior to the completion of the site investigation. These were aimed at reducing potential environmental impacts, particularly those associated with discharges of dissolved contaminants and DNAPL to the Fraser River. The temporary abatement remediation works included:

Installation of a groundwater collection well and pump-and-treat system to control the offsite discharge of dissolved constituents in groundwater entering the river; and

Excavation of shallow contaminated soil and sediment from the foreshore and beach area.

Removal of Shallow Contaminated Soil

About three metres of fill were placed over native soil when the site was first developed for industrial use. As discussed earlier, part of this fill became contaminated with coal tar over the 60 years of operations at the site. Soil with high levels of PAH contamination (typically ten times higher than the standard for commercial land use) was removed. The remaining soil was safely managed in-place by preventing surface exposures through construction of concrete building slabs, placement of low-permeability caps, and restrictions on land use, as well as control of local groundwater using shallow interceptor trenches.

Hydraulic Control of Dissolved PAH and DNAPL

Dissolved PAH contamination in the deep Sand Aquifer has been controlled by pumping of groundwater from wells, and treating the water onsite using either activated carbon with pre-treatment to remove naturally occurring iron and manganese. Treated water is discharged to the sanitary sewer.

Hydraulic control was also effective in arresting potential DNAPL migration, and in recovering DNAPLrecovery. DNAPL recovery from the deep aquifer is enhanced through cyclic stressing of the aquifer by pumping. Groundwater modelling and on-going monitoring indicate that the hydraulic containment and control system in place provides an effective control with a reverse groundwater gradient away from the river ranging from 1% at low tide to 2% at mean tide. This reverse gradient is sufficient to arrest or reverse the flow of NAPL and capture the entire dissolved-phase plume.

Sediment Removal or Control

Contaminated sediment in the intertidal zone was removed by excavation, and treated off-site by thermal desorption. For contaminated sediment in the subtidal zone, it was concluded that capping/containment by capping would provide an effective level of protection. The selection of this option was based on regulatory considerations, the results of the aquatic risk assessment, the already existing source control measures that were already in place (*i.e.*, mass reduction and hydraulic control of NAPL and dissolved PAH), and specific site constraints such as: mobility of river sediments, river slope stability concerns if disturbed, and increasing PAH contamination to a depth of 4 m to 5 m below the surface of the sediments of 4 to 5 m.

The designed sediment cap consists of an engineered layer of crushed rock barrier. The cap will prevent erosion and downstream transport of contaminated sediments, provide a layer of protection-barrier between the contaminated sediments and the aquatic community, and provide a dispersion and attenuation zone for groundwater that may contact sediments if groundwater pumping containment by pumping was terminated.

CONCLUSIONS

The site has been successfully rehabilitated for continued industrial and/or commercial use through the partial removal of contaminant sources removal and in-place controls, especially in the shallow soil. Long-term site improvements are expected through recovery of mobile DNAPL from NAPL recovery wells. Protection of the aquatic habitat has been achieved through removal of intertidal sediments, capping of contaminated subtidal sediments, and control of dissolved PAH and DNAPL releases by groundwater pumping. Over the long term, as the mobile DNAPL is removed, it is predicted that pumping of groundwater will no longert be required. Natural attenuation and dispersion of PAH during migration through the ground and through the crushed rock barrier cap are expected to reduce dissolved PAH concentrations to levels that do not pose risk to the aquatic habitat.

Complete removal or reduction of PAH contamination to applicable clean-up standards was not ractical at the site with today’s technologies. Even if the removal or reduction of all accessible contamination had been pursued, this would have resulted in a lengthy and very costly process. The partial removal of sources and containment of the majority and less accessible contamination in the ground and the sediment have provided for an effective remediation at reasonable cost. The remediation includes long- term operation and maintenance of the hydraulic control system and the sediment cap, under an environmental management system for the site, and financial security that is required by the provincial government to ensure adequate funding for its continued implementation.

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PERMEABLE REACTIVE BARRIER SYSTEMS FOR GROUND WATER CLEAN-UP

by
R. Hermanns Stengele and S. Köhler

Stengele, R. Hermanns & Köhler, R. 2001. Permeable reactive barrier systems for ground water clean-up. *Geological Survey of Finland, Special Paper 32, 175–182,* four figures.

Permeable Reactive Barriers (PRBs) represent a new *in situ* method for remediation of contaminated ground water. The paper comprises a short specification of variant designs as well as the description of a current research project at ETH Zurich, concerning the development of novel filling media to apply with this promising technology.

Keywords: environmental geology, ground water, pollution, remediation, in situ, methods

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INTRODUCTION

Increasingly it is realised that, under economical and ecological aspects, conventional pump-and-treat systems for remediation of contaminant plumes do not lead to satisfying results. This is particularly true if remediation periods of several decades are anticipated, which means a continuous and, ultimately, high input of energy for pumping water from extraction wells and operating water treatment systems, as well as periodic maintenance and monitoring.

In the early 90s the method of "Permeable Reactive Barriers" (PRBs) was suggested in Canada and USA. PRBs are a passive *in situ* technique that runs without any need of permanent and cost-intensive operations. Once installed in the subsurface, geochemical or geophysical reactions between the material in the PRB and ground water contaminants take place without any further external interference.

Research efforts at the Institute of Geotechnical Engineering (IGT) include the development of an adsorptive barrier material containing different reactive media to treat a wide range of contaminants and contaminant classes. The research programme will comprise both laboratory and full scale geochemical and geotechnical tests.

SPECIFICATION OF THE PERMEABLE REACTIVE BARRIER (PRB)-PROCEEDINGS

Permeable Reactive Barriers are installed in the aquifer across the flow path of a contaminant plume. As the contaminated groundwater moves through the PRB due to the natural gradient, the contaminants are removed by physical, chemical and/or biological processes. Depending on what processes take place, the reactive barrier material can remain permanently in the subsurface, or replaceable units can be provided. As the reactions that occur in such systems are affected by many parameters, successful application of this technology requires a sufficient characterisation of contaminants, ground water flow regime and subsurface geology. In case of instability of the subsoil or if great depths are to be reached, techniques of specialised heavy construction are essential for the implementation (i.e. sheetpiling, contiguous bored cased piles).

Important for the decision on the feasibility of a PRB is, apart from a financial point of view, the examination of:

- Installation facility and permeability ($k_{f,\ PRB} > k_{f,\ Aquifer}$)
- Efficiency and clean-up performance
- Reliability and potentiality to monitor the performance of the geochemical / geophysical processes
- Hydraulic and mineralogical long-term stability
- Environmental compatibility

Currently, two basic designs are being used in full-scale implementations of PRBs. A continuous trench, filled with the reactive media, is called "*reactive wall*" or "*treatment wall*". The combination of cut-off walls and permeable *in situ* reactor(s) is known as the *"funnel-and-gate" system* (EPA 1998, Gavaskar *et al*. 1998, NATO/CCMS 1998, Starr & Cherry 1994, Teutsch *et al*. 1996).

Reactive Walls

Ground water remediation with continuous reactive walls represent an especially environmentally friendly way. The disturbance of ground water flow by the constructional interference is minimal. The permeability of the filling material must at least be as high as that of the natural aquifer. By this, the ground water flow is prevented from changing its direction and passing around instead of through the wall. The horizontal thickness must be designed in a way that, concerning the chemical and physical processes, a sufficient residence time of the polluted ground water is provided within the wall to achieve the required effluent concentration.

Funnel-and-Gate Systems

To remedy polluted ground water *in situ* successfully, a continuous reactive wall must be big enough to cover the whole range of the contaminant plume. If the latter is wide and reaches great depths, the extent of the reactive wall would become very large. To avoid this problem and to increase the efficiency of the flow-through, “funnel-and-gate” systems can be conceived alternatively. Herewith, the contaminated ground water flow is enclosed and directed towards the permeable reactive zone (gate) by cut-off walls (funnel). The cut-off walls typically consist of interlocking sheet pilings or slurry walls. It is important to note that, by this method, backwater upstream of the system emerges which causes an increased gradient and hence higher flow velocity. This must be taken into account with respect to the contact time of the contaminants with the reactive material and the thickness of the gate, respectively.

Some variant designs of PRBs are shown in Fig. 1. The capture zone of a funnel-and-gate system is related directly with the flow through the gate.

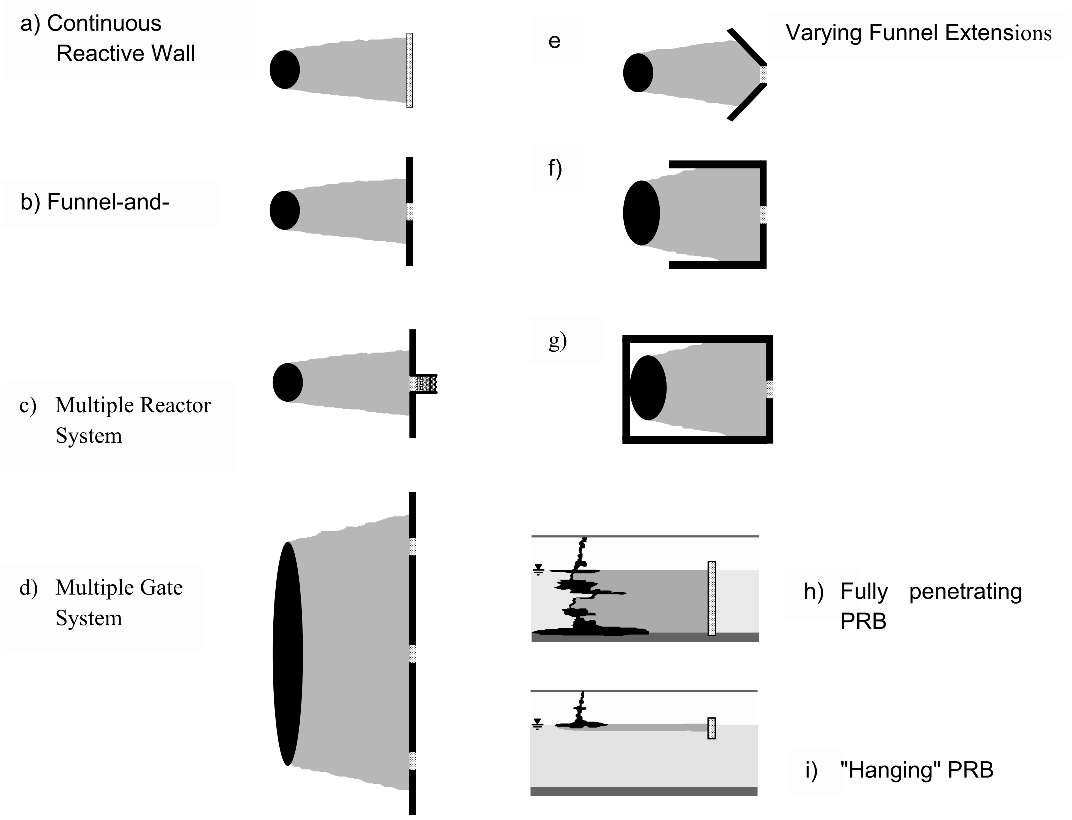


Figure 1.Variant configurations of Permeable Reactive Barriers (PRBs) (after Starr & Cherry 1994).

Plumes with a mixture of contaminants that require more than one type of *in situ* reactor can be controlled using multiple gates *in series* (Fig. 1c). On the other hand, multiple gates *in parallel* can be used for controlling wide plumes (Fig. 1 d). Generally speaking, the capture zone of a funnel-and-gate system with several (small) gates is larger than that of a system with the same overall extent and only one single reactive zone. This is due to the fact, that the water stream flows around each half of a funnel-segment (Fig. 2).

An *in situ* PRB should function until either the reactive capacity is consumed or it is clogged by precipitants or micro-organisms. As a consideration, a reactor enclosed in a relatively small gate offers better possibility to remove and replace the reactive material than a continuous reactive wall. If the extent of a plume is relatively large, the latter should be designed for a longevity exceeding the calculated remediation durance.

Another advantage of funnel-and-gate systems is the feature, that they offer a better and uniform exploitation of the reactive filling material in heterogeneous underground, particularly in combination with drainage trenches

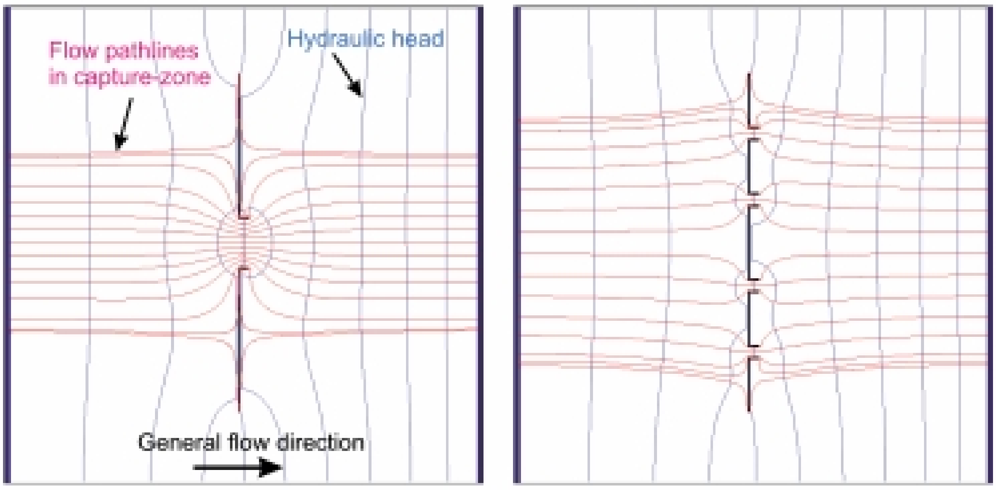


Figure 2. Capture zone of different funnel-and-gate configurations (exemplary demonstration of a numerical flow model). a) Single funnel-and-gate system b) Multiple funnel-and-gate system.

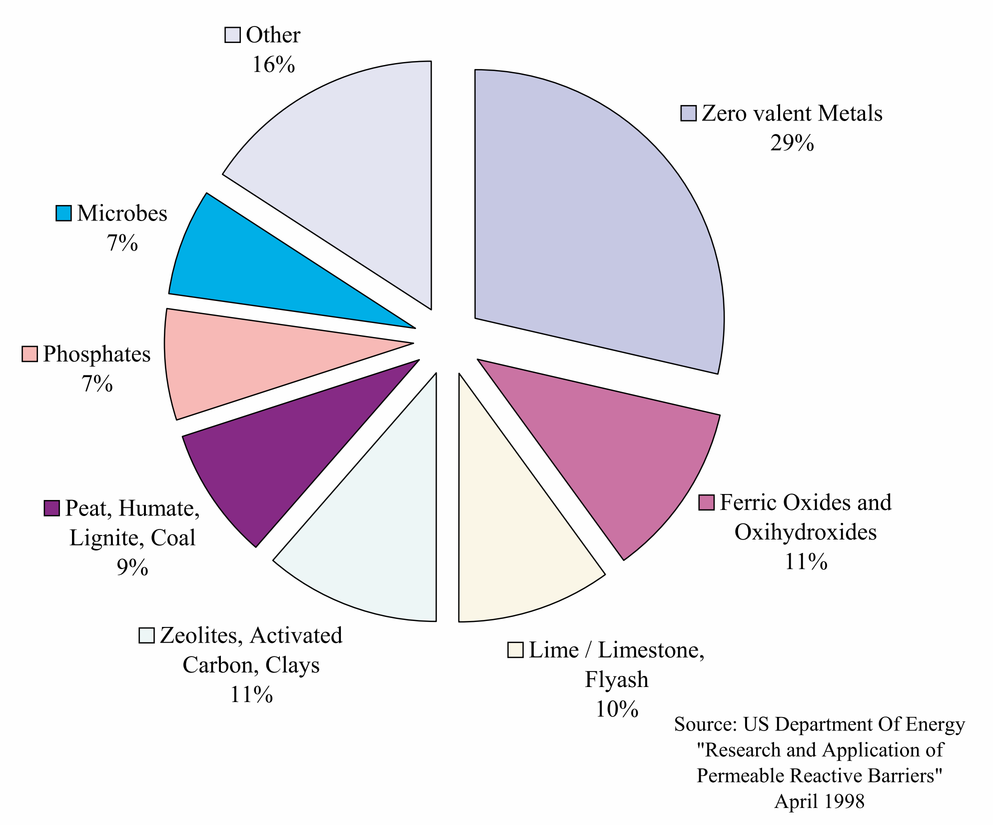
REACTIVE MATERIALS

The interest in PRBs has increased eminently over the past years, whereas implementations of full-scale projects have been realised especially in North America. So far, the predominant filling material has been zero-valent iron (Fe^0) in form of chips, jet blasting media, ironfoam or Fe-filler material for concrete etc. These materials can often be found as by-products in metalworking industry. Additionally, some highly reactive metals and metal alloys are being developed and modified with complex and expensive procedures. Fe^0 is very suitable for degrading chlorinated organic compounds (Dahmke (1997), Dahmke *et al*. (1999), Gillham *et al*. (1994), Gillham & O'Hannesin (1994), Johnson and Tratnyek (1994)). Hereby, an abiotic, heterogeneous surface process occurs, which involves adsorption of the contaminant and hereafter a progressing, normally complete reductive dehalogenation of the chlorinated compound. In the end, essential degradation products are ethene or ethane, respectively. In small quantities, also some problematic low-grade chlorinated compounds, such as cis-dichloroethylene (cDCE) and vinylchloride (VC) occur as metabolic products. Generally speaking, Fe^0-reactive barriers represent an important tool for remediation of ground water contaminated with halogenated compounds. Nevertheless, from scientific point of view, there are still several unsettled questions (e.g. degradation processes; influence of further substances in the ground water; performance of contaminant mixtures with diverse degradation- or sorption-characteristics; processes occurring downgradient of the PRB, concerning subsequent reactors and, not to forget, water protection rights). Recent research studies show that using this degradation method, serious problems, such as clogging effects and/or inhibition of the chemical processes might occur in presence of HCO_3^-, SO_4^{2-}, PO_4^{3-} or NO_3^- or other competing oxidants (Dahmke *et al*. 1999). Furthermore, the influence of microbiological activities on the degradation process is widely unknown.

Zero-valent metals are suitable for a series of heavy metals as well (Cr^{VI}, As^{VI}, As^{III}, Se^{VI} and TC). Soluble forms of these metals are transferred into insoluble forms by the reduction process, which then precipitate as hydroxide, for example (Blowes *et al*. 1996).

Significantly less distributed is the application of *adsorptive PRBs*. That is to achieve an immobilisation of the pollutants by chemically attaching them to mineral surfaces. With drinking water, treatment by adsorption of organic components on solid surfaces is established. For this, activated carbon is used predominantly. Activated carbon has also been used as filling material in first applications of adsorptive PRBs (Grathwohl & Peschik 1997, Teutsch *et al*. 1996). The tendency of a solvent to leave the fluid phase and to adsorb on a certain solid surface, is quantified by sorptionisothermes. This process provides just a retardation of the contaminant transport. With the reactive material showing a finite capacity, in most cases frequent replacement of the adsorptive medium must be considered.

Beside these commercially applied technologies, further ones are being developed, in order to optimise known processes as well as to develop novel methods. Figure 3 gives an overview of diverse research activities.



Source: US Department Of Energy "Research and Application of Permeable Reactive Barriers" April 1998

Figure 3. Present laboratory research projects on the development of reactive materials.

DEVELOPMENT OF ADSORPTIVE FILLING MATERIALS FOR PERMEABLE REACTIVE BARRIERS

Development of novel adsorptive media to apply with Permeable Reactive Barriers (PRBs) is the object of a present research project at the Institute of Geotechnical Engineering at ETH Zurich. Materials and material mixtures are being generated that can be utilised for adsorption of a wide range of contaminants, as typically occur at hazardous waste disposal sites. One goal is in particular, to achieve the immobilisation of pollutants which have proven to be difficult to manage with other remediation methods. Exceedingly, environmental impact is heeded, and economical aspects are given prime importance. It is hoped to find suitable media among those which typically occur as by-products in industry.

Adsorptive processes are based on physical-chemical properties for separation of certain components from mixtures. Hereby, the detached substances affiliate on the interface between solid and fluid. Adsorption is more intensive the larger the area surface per unit mass is (specific surface). It is increasing rapidly with decreasing grain size. Furthermore, an effective detachment of contaminants from water can only be expected if specific affinities between the compounds and the adsorptive medium exist. Additionally, surface tension of the fluid is relevant, since it affects the wetting capability of the solid.

For application in PRBs, relatively coarse grained materials are eligible, categorically, which guarantee high hydraulic permeability. To be able to use the adsorptive properties of clay minerals, for example, they must show several soil mechanical and mineralogical characteristics that are unusual and even undesirable in many conventional geotechnical fields which deal with clays. Primarily only such minerals are suitable, which are available in fractured form without any fine grained material or in granulated form (so called pellets). Moreover, the following characteristics have to be fulfilled:

- Hydraulic and mineralogical long term stability
- Small deformations
- Erosion stability of the pellets
- Uniformly graded grain sizes

With certain materials, like zeolites, clays / bentonites, silicon oxides, alumina clays, hydrotalcite etc., geotechnical tests are carried out with respect to the feasibility in PRBs on one hand. First of all, oedometer tests are performed on the reactive materials to relate permeability to *in situ* stress conditions. Additionally, swelling tests can be performed to check out if effective pore volume remains stable. On the other hand, one-dimensional flow tests in columns are carried out to quantify the adsorptive kinetics. The design of a typical column set-up is shown in Fig. 4.

For volatile compounds, a collapsible Tedlar bag (a material similar to Teflon) is used as repository, so that no head space emerges while outpouring. The column flow is from bottom to top with a pump providing a constant rate. The flow velocity should comply with that of a typical ground water flow. Sampling ports are equipped with gastight and watertight fittings. Sampling syringe cannulae are left permanently inserted into the column, with the tip at the centre of the column. Concentration profiles can be generated periodically for the distribution of the solute compounds by collecting and analysing samples systematically.

Column tests are not only used for basic research to find out about eligible filling materials, but they are indispensable in view of definite applications and designs of PRBs with selected reactive materials. To comply with all relevant boundary conditions on laboratory scale, it is essential to use original site ground water. The flowrate through the column should be set to approximate site conditions to adjust the relevant reaction kinetics. Therefore, it is desirable to have reliable ground water velocity data from the site at the location of the proposed barrier. Especially with designing a funnel-and-gate system, considerably higher ground water velocities than in the surrounding aquifer must be taken into account. Furthermore, the significant influence of temperature has to be considered. Sorption processes (exothermal) normally run faster in cool underground than at normal room temperature in the laboratory.

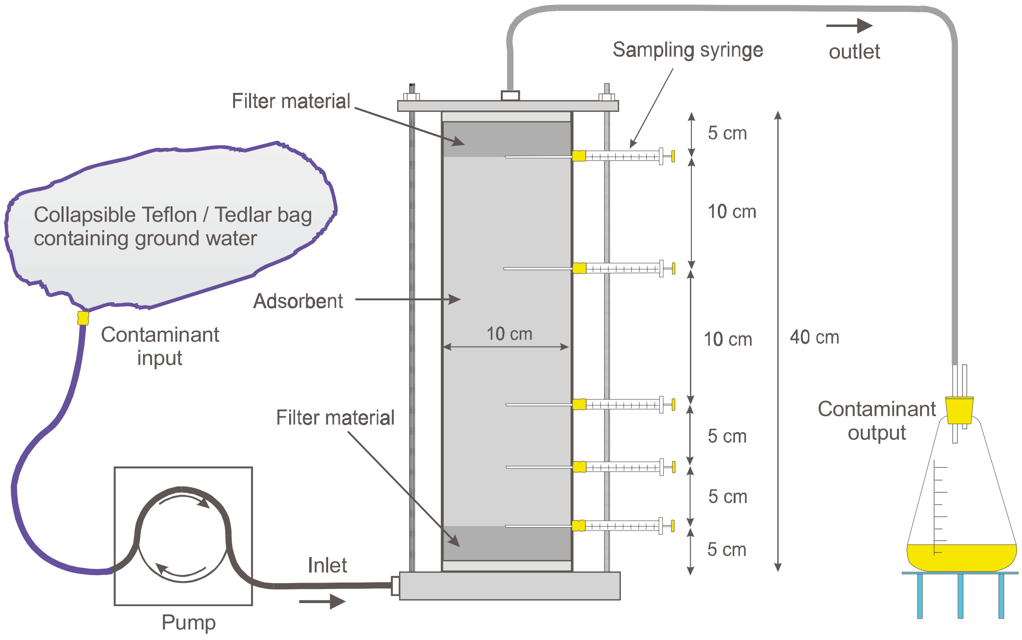


Figure 4. Schematic of a column test used to quantify the adsorptive properties of the reactive materials.

OUTLOOK

Column testing for basic research is currently being undertaken at the Institute of Geotechnical Engineering of ETH Zurich. Simultaneously, data from different hazardous sites are collected. As soon as suitable reactive materials can be confirmed, it is aspired to apply and verify the results gained on laboratory scale by implementing a field test or even rendering a contribution within a full scale remediation project.

Research and development of Permeable Reactive Barriers and of appropriate filling materials in particular, as well as on-site applications, are being promoted continuously at many places, but especially in North America (EPA 1998, EPA 1999). It would be desirable if this innovative and comparatively economical long-term remediation method could obtain global acceptance.

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LANDFILL LINERS AND COVERS – DEVELOPMENT AND PRACTICAL APPLICATION OF SEALING SYSTEMS IN GERMANY

by
E. Gartung

Gartung, E. 2001. Landfill liners and covers – development and practical application of sealing systems in Germany. *Geological Survey of Finland, Special Paper 32, 183–190.*

Liners at the base of solid waste landfills prevent leachate from migrating towards the ground water. Covers keep snow and rainwater away from the waste deposit and help to minimize the generation of leachate in the long term. The sealing layers consist of soils of low hydraulic conductivity and of geomembranes or asphalt. Extensive research and development work aiming at an appropriate assessment of the effectiveness of different sealing systems has been carried out in Germany during the past three decades. The practice of constructing composite basal liners is well established. But the standard solutions for cover systems are being questioned at the present time and presumably will be updated. The paper presents a brief review of several liner and cover systems.

Keywords: engineering geology, landfills, sealing, disposal barriers, clay, geomembranes, Germany

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INTRODUCTION

In Germany and in some other European countries strategies to reduce, reuse and recycle residues have been effective in minimising the amount of solid waste to be disposed of. The demand for storage space for solid waste has declined dramatically as compared to the conditions 10 to 15 years ago, the time when the development of technical rules for safe waste disposal was a major issue for the geotechnical profession. The political commitment of the German Federal Ministry of the Environment aims at further reducing the amount of solid waste by promoting the development of waste processing techniques. However, the need to design and construct new and to rehabilitate old landfills for solid waste still exists and calls for the attention of geotechnical engineers.

In order to achieve a comparably high level of safety with respect to the protection of the environment at all waste disposal sites, technical instructions for hazardous and domestic waste landfills have been issued by the German Federal Government (TA-Abfall 1991 and TA-Siedlungsabfall 1993). These instructions are supplemented by the regulations of the German States in order to account for regional particularities. Recently, a group of experts from all German State Administrations (LAGA-Arbeitsgruppe 2000) reviewed the Federal instructions for cover systems and prepared an update accounting for recent technical and scientific developments.

Independent of the Federal Government regulations but in accordance with them, since 1985 the German Geotechnical Society (Deutsche Gesellschaft für Geotechnik, DGGT) has edited “Geotechnical Recommendations” for the design and construction of landfills annually in the journal Bautechnik. They have been elaborated by the technical committee on Geotechnics of Landfills (Arbeitskreis Geotechnik der Deponiebauwerke, AK 6.1). The present state of the art in this area was compiled and presented in the third edition of GDA-Empfehlungen, (DGGT 1997). The series of recommendations is updated and extended by new issues every year (Gartung & Neff 1997, 1998, 1999, 2000).

SOLID WASTE LANDFILLS – ENGINEERED STRUCTURES

Solid waste may contain substances which are harmful to the environment. Thus the engineering problem to be solved consists of designing and constructing a containment which safely prevents hazardous and harmful substances from migrating into the surroundings and endangering the natural environment. The philosophy to achieve this follows the concept of the multibarrier system (Stief 1986). The first barrier is the waste itself. It should be deposited in such a condition that no substantial chemical and biological reactions will take place and that harmful components are not present or at least are not mobile. The second barrier is the basal lining system for the prevention of leachate from contaminating the ground and the ground water. The next barrier is the natural ground on which the landfill rests. It should possess a low permeability and a high adsorption capacity and thus act as an efficient geological barrier. Finally, the landfill body will be provided with a cover when the landfilling operation ceases. The capping barrier acts in two ways, it retains dust, odours and gas developed within the waste and it keeps precipitation water outside the waste body, thus minimising the generation of contaminated leachate.

The natural and the technical barriers of solid waste landfills interact. They are influenced by the geology of the site, the climatic conditions, the type of waste and the technical means by which they are constructed. So each landfill project requires careful studies of the natural site conditions and must be designed individually in such a way that all relevant aspects are encompassed.

BASAL LINERS

General aspects

The technical barrier at the base of a landfill has to prevent seepage out of the waste body. Since any repair of the basal liner of an operating or of a closed landfill would be practically impossible, the basal liner must serve its function reliably and permanently. The basal liner system consists of a mineral liner of low hydraulic conductivity, in most cases a synthetic geomembrane, a protector for the sensitive geomembrane and leachate collection facilities.

These consist of a coarse-grained drainage blanket inclined at an angle of at least 3 to 5 degrees, sufficient to facilitate gravitational flow of the leachate towards perforated pipes which conduct it to maintenance shafts and further out of the landfill containment for treatment in a purification plant.

The basal liner system rests directly on the natural ground, that is at the surface of the geological barrier. There should be no drainage layer or capillary block below the mineral liner, in order to prevent the loss of moisture. The hydraulic conductivity of mineral liners is lowest when they are well compacted at a water content slightly above the Proctor-optimum and should keep this moisture content. If the mineral liner lost water, it might develop desiccation cracks and the sealing function could be impeded.

Compacted clay liners

A major component of the standard basal seal consists of a well-compacted layer of natural clay. By the selection of a suitable clayey soil, thorough homogenisation, adequate compaction at the correct water content, it is possible to obtain hydraulic conductivity values of the mineral liner of less than 5×10^{-10} m/s. This is the highest accepted permeability of mineral basal liners, according to German technical standards. For the preparation of each individual job, suitability testing in a soil mechanics laboratory and at site compaction tests in a test field for the optimisation of the applied construction technology and equipment are required. It is of further great importance to have well-trained and quality-minded personnel at the construction site who perform the earthwork reliably under a rigorously exercised quality management. Finally, it is relevant for the quality of the mineral liner to protect the finished surface against desiccation, rain, snow and frost action. The achievement of the desired low hydraulic conductivity and efficient permanent sealing function in the field depends largely on the execution of the job. So great emphasis is placed on quality management and testing.

The scientific basis for the assessment of the efficiency of mineral liners to act as seals with respect to seepage and diffusion is well established by soil mechanics fundamentals and by specific research. If alternative mineral liners are proposed, their equivalency with the standard compacted clay liner is demonstrated in accordance with the rules of the German Institute of Construction Technique (DIBt 1995). They include experiments for the determination of the properties of the liner material, in particular the hydraulic conductivity and the diffusion coefficient under specified boundary conditions and the calculation of leakage and diffusion rates for specified assumed actions.

Gartung *et al.* (1999) exhumed a compacted clay liner at the base of a domestic waste landfill after more than 12 years of operation. Samples were taken and analysed by scientists from the disciplines: soil mechanics, mineralogy, geochemistry and microbiology. In summary, the results lead to the conclusion that under the pertinent favourable conditions the sealing function of the compacted clay liner improved with time and that the barrier performed better than expected on the basis of permeation calculations. It seems that the phenomena of coupled flow due to hydraulic, electrical, chemical concentration and thermal gradients as described by Mitchell (1976) and microbial life at the surface of the compacted clay liner contribute considerably to the barrier function and almost eliminate the hydraulic and diffusive migration of pollutants. This result is of great interest for the evaluation of the contamination potential of many older landfills which are equipped with compacted clay liners only without geomembranes. It is also noticeable for predictions of the long-term behaviour of landfills with composite basal liners for time periods beyond degradation of the polyolefine geomembranes.

Alternative mineral liners

In areas where natural clays, clayey sands or silts are not available as construction materials for basal liners, coarser soils are blended with a sufficient amount of imported clay powder (between 3 and 15 %), such as bentonite, kaolinitic or illitic clays. Well-composed gradations of this type mixed at the plant and compacted to high values of dry density can reach a lower hydraulic conductivity than typical natural clays. Systematic research into the behaviour and performance of the blended mineral liner materials has lead to products called Bentokies, Dywidag Trockengemisch and Chemoton, the last of which displays improved performance and high resistivity against aggressive chemicals due to additives, silicates, and finally Dynagrout (Lauf & Müllner 1993).

The thicknesses of compacted clay liners, which comply with the hydraulic conductivity requirements mentioned above, are 50 cm for landfills of category I, containing harmless waste such as inert construction residues; 75 cm for landfills of category II, containing domestic waste and its incineration residues; and 150 cm for landfills of category III, containing hazardous waste. If mineral liners with superior sealing performance such as Chemoton are used, the thickness of the basal liner can be reduced according to their improved lower hydraulic conductivity as compared to standard requirements.

Composite liners

Landfills of categories II (domestic) and III (hazardous) are provided with composite liners at the base. Composite liners consist of a low permeability mineral layer, either compacted clay or blended minerals as described in previous chapters and a 2.5-mm thick geomembrane on top of it. This combination is an especially effective barrier when both components are placed in direct contact. Geomembranes of high-density polyethylene PEHD consist of nonpolar molecules. They completely retain harmful polar substances such as heavy metal ions, but they are somewhat pervious to nonpolar liquids such as chlorinated hydrocarbons. If hydrocarbons permeate through the PEHD geomembrane, they encounter the surface of the mineral liner which is composed of polar molecules restricting further migration. When there is no lateral flow at the interface of the geomembrane and the compacted clay liner, because both components are in direct contact, there will be an increase of the concentration of hydrocarbons at the lower side of and within the geomembrane causing a reduction of the concentration gradient and thus the diffusion of hydrocarbons through the geomembrane ceases (August *et al.* 1992).

To achieve this beneficial performance of the composite liner, direct contact between the geomembrane and the mineral liner must be reached in construction. This means that the spread geomembrane is not allowed to have any waves or wrinkles. Very careful planning and sequential execution of the construction work is asked for, taking the physical properties, in particular the thermal expansion of the geomembrane and the daily temperature cycle at the site, into account.

In order to guarantee the high quality of the geomembranes, German standards require that geomembranes used in landfill construction are approved by the Federal Institution for Material Research and Testing (BAM). The criteria which geomembranes have to meet for the application in landfill construction with respect to manufacturing as well as with respect to installation at the construction site are very strict, well established and during the past 10 years have proved to be reliable and efficient (BAM 1992). Great emphasis is placed on quality management in the production, transportation, placement and fusion welding of the geomembrane sheets.

Geomembranes are sensitive to damage. To prevent puncturing caused by coarse-grained drainage gravel or crushed rock, special protection layers are placed between the geomembrane and the drainage blanket. These protectors formerly consisted of 10-cm thick sand layers. Nowadays, geosynthetics or geocomposites like sand filled geotextile mats are used. Cushioning geosynthetic products without mineral filling have also been applied successfully (Zanzinger & Gartung 1999).

Asphalt is sometimes used instead of polyolefine geomembranes. The sealing performance and the resistance of asphalt to aggressive chemical substances have been demonstrated by long-term testing (Arand *et al.* 1992). Rules for the design and execution of asphalt liners at the base of solid waste landfills were laid down by the German Institute of Construction Technique and the German Geotechnical Society, GDA (1997).

COVER SYSTEMS

General aspects

When the filling process of a solid waste landfill or of a large portion of it is completed, the surface of the waste body is covered with an upper barrier to close the waste confinement to the atmosphere. Many older waste dumps, deposited before the time of engineered landfills and thus neither equipped with a basal liner nor with an impervious cap, have to be retrofitted with cover systems which are essentially designed along the same lines as covers for modern engineered landfills. The cover system prevents the infiltration of rainwater, the emission of odours, dust and gas, and it facilitates the growth of vegetation and the development of landscaping. The cover should be impervious or of low hydraulic conductivity; it ties the waste landfill into the environment, into the nature and thus has to be designed for the pertinent local climatic conditions.

The main components of the cover are: a regulating soil layer immediately above the waste body, a gas venting system, sealing layers, a drainage system and the restoration profile. Depending on the requirements for different landfill categories and different stages of landfill age, these layers may vary to some extent. The properties and the behaviour of the waste influence the performance of the cap. They are evaluated according to methods of waste mechanics and taken into account in stability analyses and settlement predictions (Jessberger 1993). For waste bodies which contain mineral solids that do not undergo chemical or biological reactions, no major long-term settlements are expected.

Most solid waste is not suitable for profile completion. So in general a regulating soil layer of about 30 to 50-cm thickness is needed for this purpose. If gas is generated in the landfill body, a gas venting system has to be installed below the seal. The functions of the regulating layer and the gas venting layer are most often served in combination by a single sandy soil layer, typically recycled material. It must be sufficiently pervious and has to be compacted to a degree that facilitates qualified earthwork, in particular adequate compaction of the overlying mineral sealing layers. The gas is collected by pipes and either used for energy production or is processed and released in a harmless state.

Mineral sealing layers

For landfills without long-term differential settlements, the placement of the final cover can be carried out as soon as the design height is reached. However, common municipal waste bodies are very compressible and in addition to that are essentially bioreactors, where degradation processes take place in the waste body, associated with significant gas production and volume changes. The surfaces of domestic waste landfills experience large settlements for quite some time; substantial settlement differences may occur locally, impeding the integrity of mineral seals and straining geomembranes. So it is advisable to provide municipal landfills with an interim cover and to install the final impervious capping system after the ending of major settlements. To determine the right time for this action, the deformation of the landfill surface is monitored.

Since bioreactors need a certain amount of water for the degradation processes, some leakage of the interim cover is probably of no concern. In a few German landfills which were provided with impervious geomembranes at an early stage, gas production slowed down to such a degree that technical means had to be provided for the supply of water below the seal in order to keep the microorganisms alive and the degradation of organic waste ongoing. Therefore, the former practice of covering every landfill with a mineral sealing layer and a geomembrane as soon as the waste body had reached the design height, is presently changing towards the application of interim mineral sealing layers which are not absolutely water tight and which can be integrated into the final cover systems at a later stage.

The Federal instructions of TA-Siedlungsabfall (1993) specify a compacted clay layer placed in two lifts of 0.25 m thickness, each with a coefficient of hydraulic conductivity of no more than 5 x 10^{-9} m/s. This type of mineral sealing layer has been constructed at many landfill sites. However, often the side slopes of waste bodies are steeper than the long-term shear strength of natural clays permit. Blended mineral sealing materials with a high content of coarse-grained soils such as the Bentokies or Chemoton mentioned earlier, have large angles of internal friction and provide sufficient stability. They can be used on slopes which could not be covered with natural clays. The coarse-grained mixed in plant mineral seals further have the advantage that lower coefficients of hydraulic conductivity can be achieved, so the thickness of the mineral seal can be reduced accordingly.

The use of capillary barriers in landfill cover systems is gaining the recognition of supervising authorities (LAGA 2000). The system is based on the physical phenomenon that water flows laterally in an inclined, partially saturated layer of fine sand placed above a layer of coarse sand/gravel, provided the difference between the two layers in void size and in the associated degree of saturation is large enough to prevent the water from breaking through the interface into the coarser soil. Research and pilot applications have led to rules for the design, construction and quality management of capillary barrier systems (*e.g.* Von der Hude *et al.* 1999, Gartung & Neff 2000). To date, this method has reached a degree of maturity to be regarded as competitive with conventional mineral sealing layers.

Another alternative mineral seal is the geosynthetic clay liner (GCL). The criteria of equivalency of GCLs with compacted clay liners have been discussed in the international literature. In Germany they were elaborated on by a technical committee on the basis of the guidelines of DIBt (1995). The advantages and shortcomings of GCLs as members of landfill cover systems have been presented by Gartung & Zanzinger (1998), among others.

Practical experience shows that GCLs have some advantages over compacted mineral liners. Handling and installation are easier, less time is needed for placement and waste storage space can be saved due to the smaller thickness. However, GCLs are thin and sensitive to mechanical damage. Most of the GCLs on the market use sodium bentonite with a very high swelling capacity and extremely low hydraulic conductivity. Observations at many sites indicate, that the hydrated bentonite undergoes cation exchange with time. By the replacement of sodium with calcium ions which takes place in nature, the swelling capacity of the bentonite is reduced by one half and the hydraulic conductivity increases by one order of magnitude. GCLs may lose their sealing function completely if the protection against desiccation is insufficient. Once desiccation cracks have occurred, it is unlikely that they will fully self heal, because of the simultaneous effects of drying and wetting cycles and cation exchange (Melchior 1999).

As briefly discussed for thin GCLs, the thicker mineral liners in capping systems are also exposed to fluctuations in their water content. Their larger thickness and the greater amount of water stored makes them less prone to complete failure due to desiccation than GCLs, but under Central European climatic conditions they may also experience desiccation cracking after a few years of seasonal drying – wetting cycles, if the soil cover above does not prevent them from drying out.

The evapo-transpiration rate is high during the growing season from about April until late September, whereas at the same time precipitation may be low. During this time the water content of the mineral landfill cover layers is reduced. In autumn and winter, evapo-transpiration decreases, precipitation may be high and the cover is re-wetted. Observations at large test fields indicate that desiccation may cause the formation of micro-fissures and cracks in the cohesive cover soils in summer. These defects are preferably utilised by the plants for root paths, and the detrimental effect of the desiccation due to thermal gradients is further increased by suction of the roots. As a result, within a few climatic cycles the mineral liner may experience fissuring to a considerable extent, the overall hydraulic conductivity may increase, and the sealing function may be impeded. Blended mineral seals with a coarse grained skeleton such as Chemoton are less sensitive to desiccation cracking than compacted clay liners and GCLs. Among the clays the more active species such as the bentonites are more prone to cracking than the less active clay minerals.

Nowadays it is well understood among geotechnical engineers that the performance of the soil cover above the sealing layer and the vegetation combined are crucial with respect to water storage and release in the cover system and thus control the moisture content and the sealing function of the mineral sealing layer. The thickness of cover soils of 1 m as specified in the Federal instructions TA-Siedlungsabfall, is insufficient in most cases; 1.5 to 3 meters are considered more appropriate for German climatic conditions. Guidelines have been developed for the design, placement and quality management of cover soils by a team of geotechnical engineers and soil scientists of the German Geotechnical Society (DGGT; Gartung & Neff 2000).

Geomembranes as members of cover systems

Fissuring and growth of roots in mineral seals of landfill capping systems is prevented by the placement of a geomembrane. A geomembrane intercepts the migration of moisture from the underlying mineral seal upwards into the cover soil and vice versa. Final cover systems of landfills commonly consist of the combination of compacted mineral liners or GCLs with geomembranes.

Since geomembranes of covers are not exposed to a corrosive chemical environment, they must not be made of HDPE and their thickness must not be 2.5 mm as for the BAM-approved basal liner geomembranes and as often used in German cover systems until now. The installation of thinner geomembranes or of softer polymers such as VLDPE are more favourable with respect to the anticipated deformations of the landfill surface and are preferred nowadays.

The construction requirements and installation techniques are essentially the same for geomembranes of the cover as of the basal liner. The seaming technique and all details of manufacturing and construction quality assurance apply to covers as well. The surface of the landfill or of the regulating layer has to be modelled to a shape which allows plane geomembranes to be spread without distortions. This design requirement is especially important, when HDPE geomembranes of 2.5-mm thickness are used, because it is impossible to place them on three dimensionally curved surfaces with small diameters of curvature.

Usually landfills are hills with sloping surfaces. Slope stability is an important issue. In order to mobilise enough shear resistance, geomembranes with rough structured surfaces are used in cover construction. Particular attention is paid to their long-term tensile strength and stress cracking resistance. In order to avoid tensile forces in the geomembrane, the mobilised friction at the lower surface of the geomembrane should be greater than at the upper surface.

Summary and conclusions

During the past 10 to 15 years, the activities in design and construction of solid waste landfills have seen a steady development. Based on observations in the field and on research into the performance of the components of the landfill structure, technical instructions have been issued. They specify the minimum requirements for sealing and dewatering systems on a high technical level. Great emphasis is placed on quality assurance in manufacturing and construction in order to achieve the efficient performance of the sealing and the dewatering elements that have been established theoretically and experimentally by numerical and physical modelling. The components of liners and covers consist of mineral materials and of geosynthetic products, in most cases in combination. The design, manufacturing and construction practice for composite basal liners are well established in Germany. The technical rules for cover systems are being revised at the present time, taking positive and negative experience and recent scientific research results into account.

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POTENTIAL FOR *in situ* REDUCTIVE DEHALOGENATION OF CHLOROETHYLENES

by
Päivi H-M. Kauppi and Jaakko A. Puhakka

Kauppi, H-M. & Puhakka, Jaakko A. 2001. Potential for *in situ* reductive dehalogenation of chloroethylenes. *Geological Survey of Finland, Special Paper 32, 191–196,* three figures and 1 table.

Tetrachloroethylene (PCE) and trichloroethylene (TCE) from a former dry cleanery have contaminated the groundwater in Oitti, Finland. The AOX-analyses of this and former studies suggest a DNAPL-source in the aquifer. The DNAPL is a continuous source of groundwater contamination. The aquifer was investigated for redox conditions. The objective of the study was to estimate if the groundwater conditions would be suitable for the reductive dehalogenation of PCE and TCE. Transformations of PCE were studied in anaerobic microcosms, which were fed with revised anaerobic mineral medium, PCE and either lactate, ethanol, glucose, H_2 or their mixture as the electron donor. The control bottles were made without added electron donor or they were injected with NaN_3 as killed controls. The bottles were incubated at 8 and 25°C. The results suggest reductive dechlorination of chlorinated ethylenes in contaminated groundwater in Oitti if a suitable electron donor is provided. Pilot-scale experimentations are being started to study the design criteria of *in situ* bioremediation.

Keywords: environmental geology, ground water, pollution, tetrachloroethylene, trichloroethylene, dechlorination, in situ, Oitti, Finland

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INTRODUCTION

Tetrachloroethylene (PCE) and trichloroethylene (TCE) are common groundwater contaminants. PCE and TCE have been widely employed as dry-cleaning and degreasing solvents (Mroueh 1993). PCE and TCE from a former dry cleanery have contaminated the groundwater in Oitti, Finland. Approximately 8–10 million m^3 of groundwater has been contaminated with the average concentration of the solvents being 100 µg/l (Silvennoinen 1999, pers. communication). The concentrations in Oitti exceed the limit values given by the World Health Organisation (Savolainen 1998).

For PCE, the reductive dechlorination is the only known biodegradation mechanism. By reductive dehalogenation a halogene substituent is removed from a molecule with concurrent addition of electrons to the molecule. The reductive dehalogenation requires an electron donor. (Mohn & Tiedje 1992)

The electron donor may affect the rate and extent of dechlorination (Holliger 1995). In many cases dechlorinators use H_2 as the electron donor, even though it is not always the only electron donor (Ballapragada *et al*. 1997). Several studies have demonstrated that no single substrate can be recommended for all sites (Holliger 1995). However, various studies indicate that dechlorination can only be sustained supplying an electron donor (Holliger 1995).

The bioremediation of the sites contaminated with chlorinated ethylenes is a young technology. Many field applications are still unaccomplished. The environmental conditions at the contaminated sites vary. In Finland a low temperature may be a limiting factor for the use of bioremediation. However, many field scale studies demonstrate the reductive dehalogenation under anaerobic conditions (Tschistowskaja & de Kreuk 1999, Dooley *et al*. 1999, Litherland *et al*. 1999).

The objective of the study was to estimate whether the groundwater conditions in Oitti are suitable for the reductive dehalogenation of PCE to TCE, DCE, VC and ethene (Fig. 1).

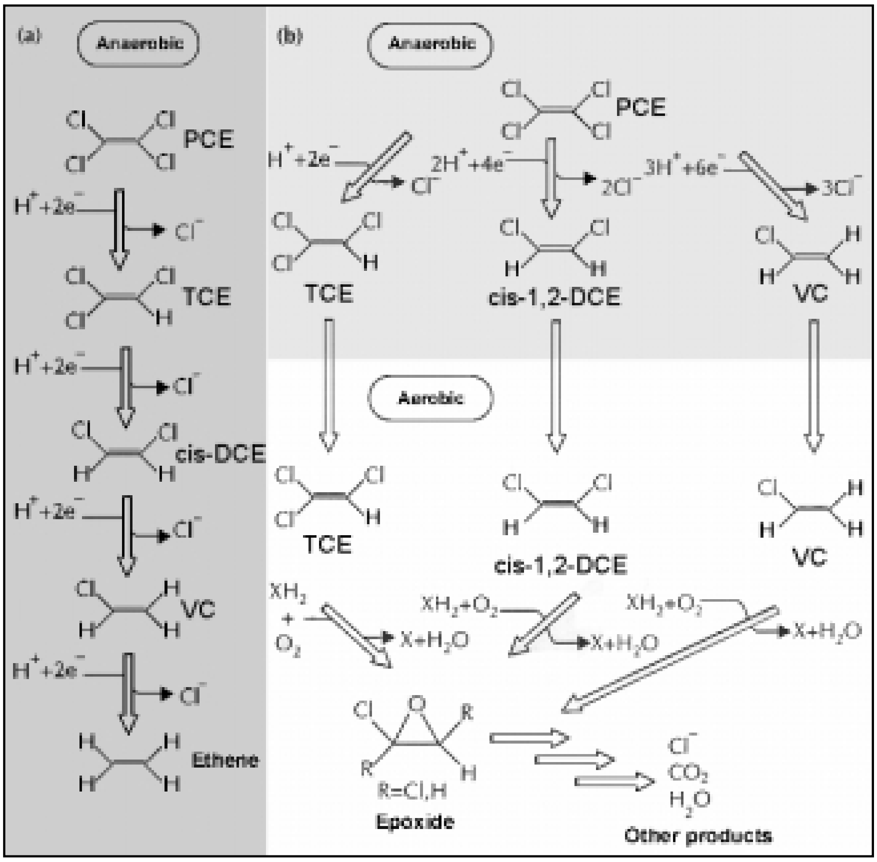

Figure 1. Two possible approaches for the biotreatment of PCE. (a) Anaerobic reductivedechlorination to ethene. (b) Anaerobic dechlorination to less-chlorinated ethenes, followed by aerobic cometabolic ar metabolic (*e.g.* via VC) oxidation to harmless products. (Holliger 1995)

MATERIALS AND METHODS

AOX was determined using ECS 1000 Euroglas BV-equipment. Nitrate was measured using nitrate specific electrode and reference electrode. Sulfate concentration was obtained by precipitating sulfate with $BaCl_2$ and determining Ba concentration with AAS. Ferrous-iron was determined using phenantroline-method 3500-Fe D and Shimadzu UV 1601 –spectrophotometer (Standard Methods 1992). DOC (Dissolved Organic Carbon) was analyzed using Shimadzu TOC-5000 analyzer. Calibrated WTW pH 96 –meter was used to determine pH in the groundwater.

Transformations of PCE were studied in anaerobic microcosms, which were constructed from samples of the contaminated site in Oitti. 10 grams of contaminated soil was added in every bottle. Bottles (120 ml) were fed with revised anaerobic mineral medium, PCE (aqueous concentration of approximately 5 mg/l) and either lactate, ethanol, glucose, H_2 or their mixture as the electron donors in two concentrations (100 or 200 mg COD/l). The control bottles were made without added electron donor or they were injected with NaN_3 as killed controls. Reducing conditions were maintained by the presence of Na_2S. Resazurin served as the redox indicator. The color of the indicator changed from blue to pink to colorless indicating anaerobic conditions in the bottles. The bottles were incubated at 8 and 25 °C. Methane was analyzed using gas chromatograph (Perkin-Elmer Sigma 300) equipped with a hot wire detector. PCE and TCE were measured on a gas chromatograph (HP 5890 series II) equipped with an electron capture detector. All chlorinated ethenes were analyzed at the end of the experiments on a gas chromatograph (HP 6890) equipped with a mass selective detector (HP 5972 A) using head space -method.

RESULTS AND DISCUSSION

Groundwater conditions

The aquifer was investigated for redox conditions. Concentrations of ferrous-iron were high indicating anaerobic groundwater. Nitrate and sulfate can serve as electron acceptors and in high concentrations inhibit the reductive dehalogenation of chloroethylenes (Doong & Wu 1992, Mohn & Tiedje 1992). Nitrate and sulfate concentrations varied between 3,5–5,8 mg/l and 19–26 mg/l, respectively. Nitrate and sulfate at these concentrations do not inhibit reductive dehalogenation. DOC (Dissolved Organic Carbon) demonstrates the presence of organic material in the groundwater. DOC may serve as an electron donor for the reductive dehalogenation (Stiber *et al.* 1999). DOC levels were in most cases lower than 1 mg/l indicating that the suitable electron donor for dehalogenation is not naturally available in the Oitti groundwater. pH-values varied between 6,4...6,6. Neutral pH favors biodegradation (Norris *et al.* 1994). Natural pH in Oitti is near neutral and does not inhibit biodegradation. AOX-level varied between the detection limit and 159 µg/l. In many sampling points of Oitti site the AOX-levels have not significantly decreased since the year 1992, when the monitoring was started (Öhberg 1997).

Dechlorination experiments

Microbial production of gas and methane was determined in order to indicate reductive conditions in the bottles. The theoretical production of gas was 2,3 ml for one bottle when 100 mg COD/l was added and 4,6 ml for the addition of 200 mg COD/l. The production of gas was low. The concentration of methane remained below the detection limit in all bottles at 8 °C and at 25 °C in the bottle supplemented with NaN_3. In all other bottles at 25°C methane was formed indicating very reducing conditions.

At 25°C the dehalogenation started in all bottles except for that supplemented with NaN_3 (killed controls) (Fig. 2). These results show that the degradation of PCE occurred biologically. At 25°C, it made very little difference what electron donor was

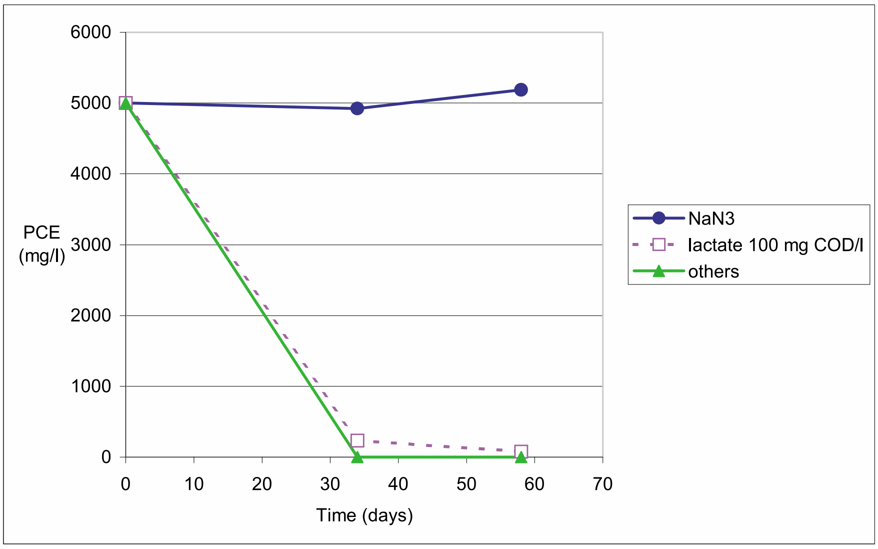

Figure 2. Anaerobic tetrachloroethylene biodegradation at 25°C. Others(Δ)=no donor, glucose, ethanol, H2 and mixture 100 and 200 mg COD/l, lactate 200 mg COD/l.

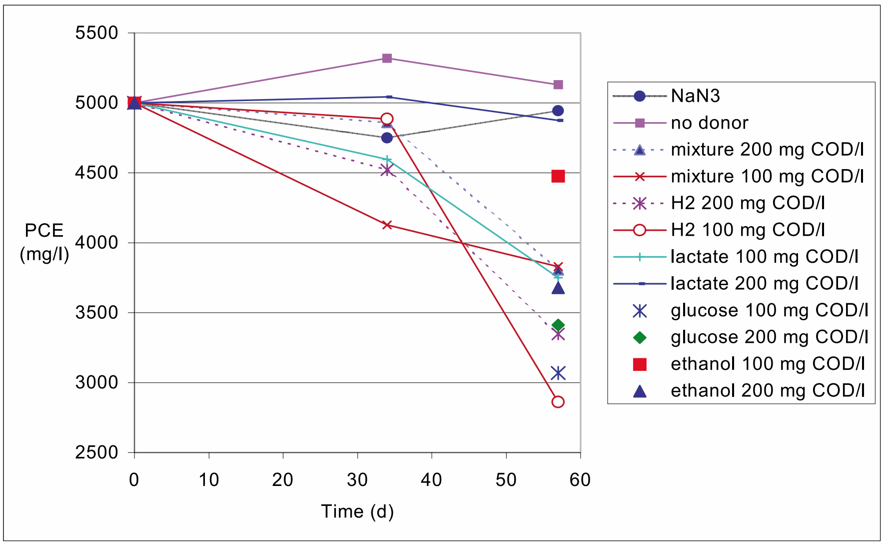

Figure 3. Effect of electron donor on tetrachloroethylene anaerobic biodegradation at 8°C.

added. PCE degraded even without any added electron donor.

At 8°C, the rate of the dehalogenation was signicantly lower than at 25°C. At 8°C glucose and H_2 were the most efficient electron donors, but resulted in very slow dehalogenation (Fig. 3).

At the end of the experiments (120…125 d) all the chlorinated ethylenes were qualitatively analyzed in selected bottles and the results are shown in Table 1.

VC was found only in some bottles indicating that VC does not accumulate. The degradation of VC is the rate limiting step in reductive dechlorination of chlorinated ethylenes (Ballapragada *et al*. 1997). VC is undesired end product since it is more toxic than the parent compounds.

Table 1. Chlorinated ethylenes in anaerobic microcosms after 120...125 days incubation time. (+) = detected, (–) = not detected, (±) = detected in one of the duplicates. Mixture = ethanol, glucose, H2 and lactate.

Compound	mg COD/l	T (°C)	PCE	TCE	DCE	VC
No donor	–	25	+	–	–	±
NaN_3	–	25	+	+	+	–
Ethanol	200	25	+	–	–	–
Glucose	100	25	+	–	±	–
Glucose	200	25	+	–	–	–
H_2	200	25	+	–	–	–
Lactate	200	25	+	–	±	–
Mixture	200	25	+	–	–	–
No donor	–	10	+	+	+	–
NaN_3	–	10	+	+	+	–
Ethanol	200	10	+	+	+	±
Glucose	200	10	+	+	+	±
H_2	200	10	+	+	+	–
Lactate	200	10	+	+	+	–
Mixture	200	10	+	+	+	–

CONCLUSION

The AOX-analyses of this and former studies suggest a DNAPL-source in the aquifer in Oitti. The DNAPL is a continuous source of groundwater contamination. The results suggest reductive dechlorination of chlorinated ethylenes in contaminated groundwater in Oitti if a suitable electron donor is provided. However, the complete dehalogenation of chlorinated ethylenes will take many years in a boreal groundwater aquifer. Pilot-scale experimentations are being started to study the design criteria of *in situ* bioremediation.

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POSSIBILITIES FOR BIOTECHNICAL *in situ*-REMEDIATION OF GROUNDWATER CONTAMINATED BY SULPHITE PULP MILL EFFLUENTS

by
H. M. Anttila and J. A. Puhakka

Anttila, H. M. & Puhakka, J. A. 2001. Possibilities for biotechnical *in situ*-remediation of groundwater contaminated by sulphite pulp mill effluents. *Geological Survey of Finland, Special Paper 32, 197–201,* four figures.

Biotechnical *in situ*-remediation of groundwater contaminated by sulphite pulp mill effluents was examined at laboratory scale. The contaminated groundwater and soil consisted mainly of organic substances dissolved from wood and formed during sulphite pulping. The contaminants consume oxygen in the groundwater. *In situ*-simulation with air-lift percolators and a semi-continuous reactor were performed in darkness at 9 °C. The highest DOC concentration was 8400 mg/l and the average DOC reduction rate was 56.7 mg/d. The DOC reduction achieved was 13–17%. The effects of pH and the presence of the original soil material were studied. There was no significant difference in the performance of the percolators at the ambient (4.3–4.5) or adjusted (6.5–7.5) pH. Neither was the DOC reduction of the percolators containing the original soil material higher than the one containing only burned and washed sand. The possibility of maintaining a stable DOC reduction was studied with a semi-continuous reactor test. Average DOC reduction of 20 and 7.8 % with hydraulic retention times of 7.6 and 1.6 days, respectively, were achieved.

Keywords: environmental geology, ground water, pollution, paper industry, waste water, bioremediation, *in situ,* Central Finland

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INTRODUCTION

A lot of organic contamination of the soil and groundwater occurred in earlier years as a result of industrial activity. Because of carelessness and the lack of knowledge, large amounts of industrial waste has been released into in the environment without any treatment. Under today's environmental regulations, a restoration of contaminated environment is often demanded by the authorities.

The restoration can be based on physical, chemical or biological processes. The use of biological remediation technologies has been growing in popularity as an increasing number of micro-organisms has been found with the ability to degrade organic compounds earlier classified as non-biodegradable.

The aim of the work was to study the possibilities for biological *in situ*-stabilisation of polluted groundwater.

MATERIALS AND METHODS

Site description and sampling

The study area is located in Central Finland about 20 kilometres east of Jyväskylä. The groundwater and the soil of the area are contaminated by old sulphite pulp mill effluents. The effluents consist mainly of organic substances dissolved from wood. A large part of the organic substances are lignin derivatives (probably lignosulphonates) formed during the pulping process (Vilén 1993). The contaminants increase the consumption of oxygen when released into the water system. The contamination occurred in the period extending from 1935–1967. The total amount of waste water released in the area is approximately 2.9 millions m^3 (Suvilampi & Rintala 1999). The estimated total area of contaminated soil is about 225 000 m^2 of which the area of the pond where the effluents have been released has been about 95 000m^2 (Mäkelä 1994). The water used in the experiments was taken from a well, situated about 200 meters south-east from the pond. Water samples were stored at 9°C in darkness.

Air-lift percolators

The *in situ*-bioremediation simulation was performed in glass air-lift percolators in the dark at 9°C. (Puhakka & Tuovinen 1986, Langwaldt *et al.* 1998) (Fig. 1). The percolator was filled with two layers of sand. The sand was dried at 180°C for 3 hours. Dry sand was passed through a sieve with a mesh size of 0.5 mm (fine-grained sand) and 1.68 mm (coarse-grained sand). The fractionated sand was heated at 550°C for 1 h and then boiled for 1 h in 5mM HCl to remove organic matter. 200 g of coarse-grained sand was added to the bottom of the percolator and 1000g fine-grained part to the top of it.

The liquid phase of the percolators was replaced with a new waste water sample and nutrient solution when the degradation was getting slower (three times) or at the end of the experiment every two weeks to see if the velocity of degradation is still increasing. The liquid level in the percolator was held above soil level to simulate saturated subsurface conditions. The amount of water added at the time was 1000 ml. The total time of the experiment was 149 days.

The effects of pH and presence of the original soil material were studied simultaneously in parallel percolators. The oxygen concentration in the liquid was maintained at 5 mg/l by aeration of the recycle flow. Samples were taken from the sampling port at the bottom of the percolators. The parameter monitored was dissolved organic carbon (DOC). At the end of the experiment, NaN_3 (200 mg/l) was added to the water to show non-biological removal.

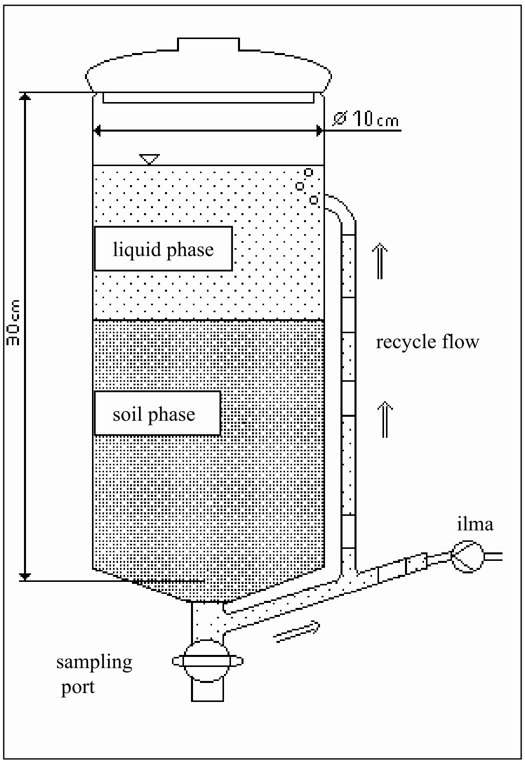

Figure 1. A schematic diagram of an air-lift percolator.

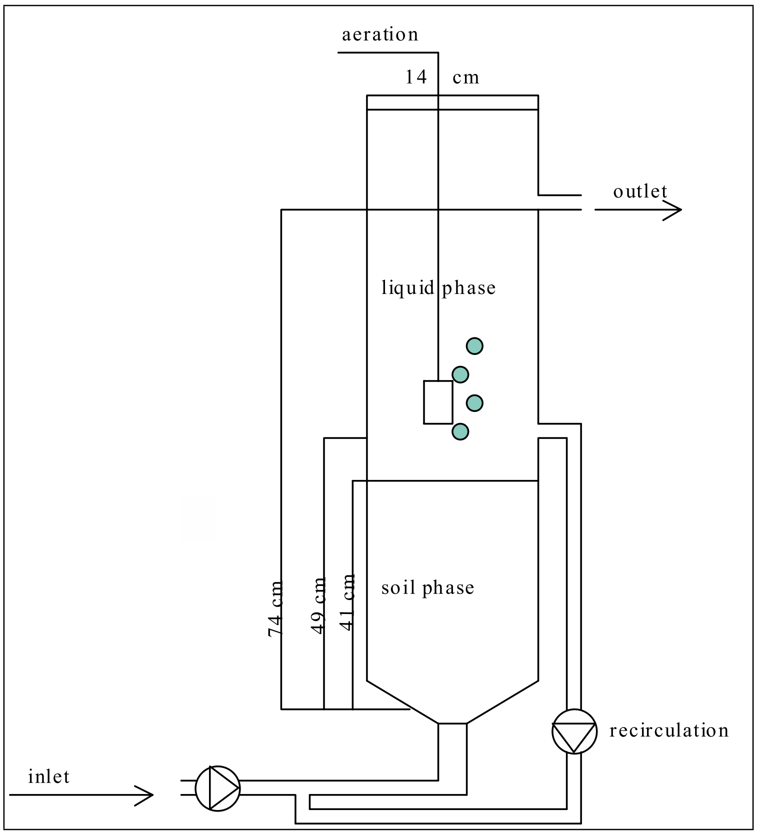

Figure 2. A schematic diagram of the semi-continuos flow reactor.

Semi-continuous flow reactor

The principle of the semi-continuous reactor is shown in Fig. 2. The total liquid volume of the reactor was 7.4 l. 1000 g of gravel was added to the bottom of reactor. On the top of this layer were added 5000 g of gravel (mesh size > 5 mm) mixed with 3000 g of the soil from the polluted area. The gravel was heated at 550 ^{0}C (1 h) before the experiment. The water was neutralised with 10% NaOH and a sufficient amount of nutrients were added. The oxygen concentration in the reactor was kept at 10 mg/l.

The water was first circulated in the rector over 18 days to enrich a microbial population. The circulation flow was about 6 l per day. After the enrichment period a flow of 970 ml/l of water was passed through the reactor for 35 days. After that period the flow was increased to 4500 ml/d for 9 days. The hydraulic retention times in the reactor were 7.6 and 1.6 days, respectively. The DOC concentration and pH of the inflow and the outflow were monitored. The experiment was performed at 7.5 ^{o}C in darkness.

Analyses and the nutrient media

The pH of the water was determined with the SenTix 97T electrode (WTW Ltd., Weilheim, Germany). The oxygen concentration was determined with an oxygen sensitive electrode EO96 (WTW Ltd., Weilheim, Germany). The dissolved organic carbon (DOC) concentration was analysed with Shimadzu TOC-5000 analyser connected to a Shimadzu ASI-5000 autosampler. Diluted (1:100) samples were filtered before analysis with 0.45 µm Whatman Puradisc™ 25 AS filters. In the experiments, a buffered nutrient solution based on Ortega-Calvo *et al.* (1995) was used. The nutrient solution was added to the water to achieve DOC:N:P -relation 100:5:1. The initial BOD_7/DOC -ratio of the water was 0.86.

RESULTS AND DISCUSSION

Air-lift percolators

The aim of the air-lift percolator experiment was to simulate the bioremediation of polluted groundwater. The liquid phase of the percolators was replaced with a new ground water sample and nutrient solution when the degradation slowed down or at the end of the experiment every two weeks to see if the velocity of degradation was still increasing. The liquid level in the percolator was held above soil level to simulate saturated subsurface conditions. The amount of water added was 1000 ml. The total time of the experiment was 149 days. The results are shown in Fig. 3.

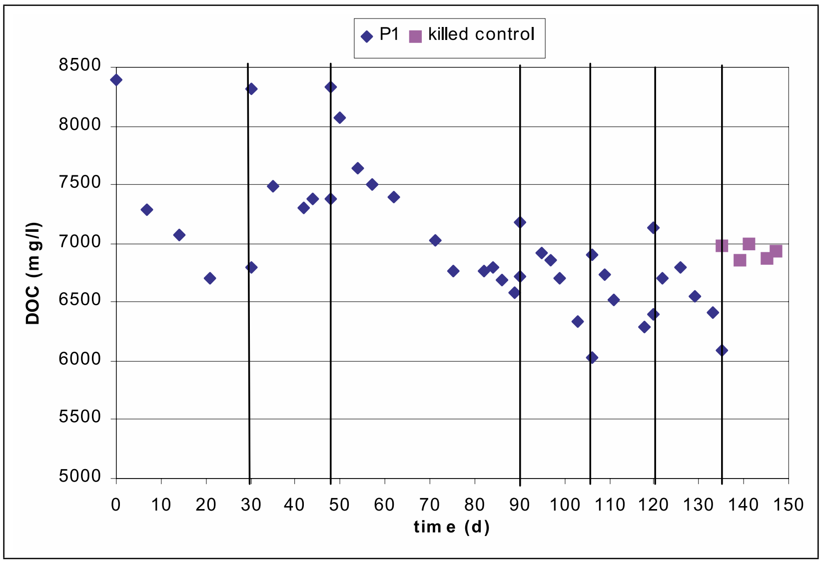

Figure 3. Groundwater DOC removal in air-lift percolator. The vertical lines show the times of groundwater replacements. The percolator was operated at ambient groundwater pH.

The highest initial DOC concentration was 8400 mg/l and the average DOC reduction was

56.7 mg/d. The DOC reduction was between 13–17 %. At the end of the experiment 200 mg/l of NaN_3 was added to the water which stopped biodegradation (Fig. 3). The effects of pH and presence of the original soil material were also studied. There was no significant difference in the performance of the percolators of the original (4.3–4.5) or regulated (6.5–7.5) pH. Neither was the DOC reduction of the percolators containing the original soil material higher than the one containing only burned and washed sand. The biodegradation rate did not increase during the experiment. This may be due the strong biomass accumulation in the percolator, which decreased the recirculation rate of the water.

Semi-continuous flow reactor

The aim of the semi-continuous flow reactor experiment was to find out the possibility of maintaining stable DOC-reduction. The effect of retention time was also studied.

The water was first circulated in the reactor for 18 days to enrich a microbial population. The circulation flow was about 6 l/d. After the enrichment period a flow of 970 ml/l of was passed through the reactor for 35 days. After that period, the flow was increased to 4500 ml/d for 9 days. The hydraulic retention times in the reactor were 7.6 and 1.6 days, respectively. The DOC concentration and pH of the inflow and outflow were monitored. The results are shown in Fig. 4.

During the recirculation period, the DOC concentration decreased by 25 %. Average DOC-reductions of 20 and 7.8 % with hydraulic retention times of 7.6 and 1.6 days, respectively, were achieved. In previous tests at room temperature (Anttila 1999), 46 % of the DOC was biodegrad-

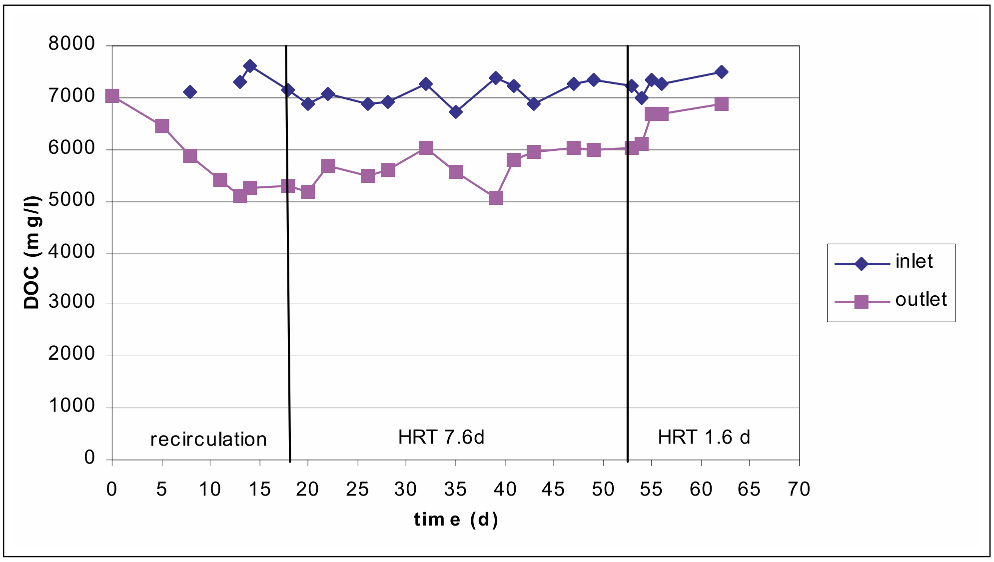

Figure 4. Degradation of DOC in continuous-flow reactor.

able. Rintala and Suvilampi (1999) reported similar results when testing the biodegradability of the water of the bottom layers of the pond. With 4 days aerobic biodegradation tests in a lab-scale batch reactor they measured soluble chemical oxygen demand (SCOD) reductions of about 46±2 % at 20°C.

CONCLUSIONS

Over 20 % reduction of groundwater DOC was achieved in a lab-scale in-situ simulation system at ambient groundwater temperature in darkness when nutrients and air are added to the system. A stable DOC reduction of 20% was achieved at a hydraulic retention time of 7.6 days.

The better results derived from the semi-continuous test than with the percolator tests were mainly due to better recirculation of water in the reactor. That was because of higher flow rate and bigger soil particle size in the reactor.

In field conditions, the biomass accumulation is a limiting factor for biotechnical *in situ* remediation. On the other hand, the biomass accumulation could be used for controlling the water flow in the aquifer.

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Edited by Reijo Salminen.
Geological Survey of Finland, Special Paper 32, 203–209, 2001.

FULL-SCALE LANDFILL BOTTOM LINER TEST STRUCTURES AT THE ÄMMÄSSUO LANDFILL, ESPOO

by
Minna Leppänen[1)], Antti Kaartokallio[2)], Erkki Loukola[3)]

Leppänen, Minna, Kaartokallio, Antti & Loukola, Erkki 2001. Full-scale landfill bottom liner test structures at the Ämmässuo landfill, Espoo. *Geological Survey of Finland, Special Paper 32, 203–209,* eight figures and 1 table.

Full scale tests structures were constructed in summer 1996 in the Ämmässuo landfill, Espoo, Finland, to gather experience on quality control during the construction and long-term behaviour of mineral liners and combination liners. Actual leachate was used to create the chemical loading and a hydraulic pressure of one meter. The structures were monitored for two years. The leachate seeping through the liner structures was collected in lysimeter basins and additionally in lysimeter wells, in which the amount of the water was measured automatically by pressure sensors and water samples taken for chemical analyses. The structures were dismantled under controlled conditions in November 1998, and material samples were taken to evaluate the effects of leachate.

Keywords: engineering geology, landfills, disposal barriers, seepage water, construction materials, chemical properties, experimental studies, Ämmässuo, Espoo, Finland

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INTRODUCTION

In The summer of 1996, full-scale landfill bottom liner test structures were constructed in the Ämmässuo landfill. The purpose of the project was to:

- verify the importance of new, strict requirements and necessity of the artificial liner
- gather practical experience during the construction
- test available field quality measuring equipments
- monitor the behaviour of the liners, and especially combined liners, under chemical loading caused by the landfill leachate
- collect information about the effects of frost.

The project was funded by the Ministry of Environment, Finnish Environment Institute, Helsinki Metropolitan Area Council, Technology Development Centre and private companies.

STRUCTURE AND MATERIALS

Materials

Three different mineral liners and two artificial liners were used: a natural clay from the vicinity of the landfill, a mixture of bentonite and crushed stone from the landfill area and fly ash with desulphurating agent were used as a mineral liner; and HDPE geomembrane and dense asphalt concrete as a artificial liner. Actually seven different combinations were investigated (the number and letter refer to the specific part; see Fig. 1):

1) natural clay (1B),
2) natural clay with geomembrane (1A),
3) bentonite soil mixture (2B),
4) bentonite soil mixture with geomembrane (2A),
5) fly ash and desulphurating agent 50 %:50 % (3B),
6) fly ash and desulphurating agent with dense asphalt concrete on the top (3A), and
7) dense asphalt concrete alone (4A).

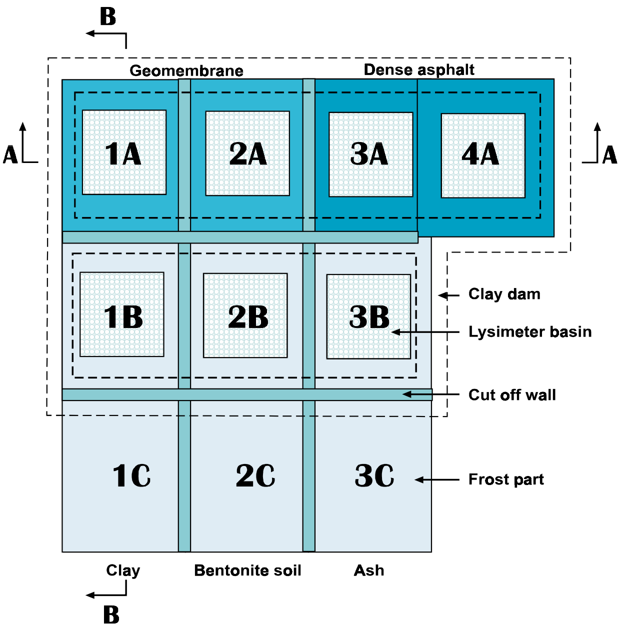

Figure 1. Schematic layout of the test structure.

Structural principles

The test structure was based on 0.5 m layer of crushed rock compacted on the bedrock, thus no settlement was to occur. Lysimeter basins, which collected the leachate seeping through the liners into the collection wells, were constructed under the liners (see Fig. 2). The average area of the lysimeter basin was 80 m^2.

Liner materials were separated by vertical cut-off walls made from a special bentonite soil mixture containing 10% bentonite. The walls were extended below the liners to prevent horizontal water movements between the materials.

The surface of all the liners were at the same level. The landfill conditions were simulated by a 1-m layer of actual leachate from the Ämmässuo landfill pumped into the drainage layer on the liners. The level of the water was monitored from pipes and wells, and additional leachate was pumped automatically to sustain this level. The whole structure was covered by a thick clay layer to prevent infiltration of rainwater.

One third of the mineral liner was covered by the artificial sealing line (A-part) and one third was left unprotected, directly exposed to the leachate (B-part). The third part of the mineral liner was left without the leachate layer and soil cover, and exposed to frost and other climate effects (the so-called frost parts or C-parts).

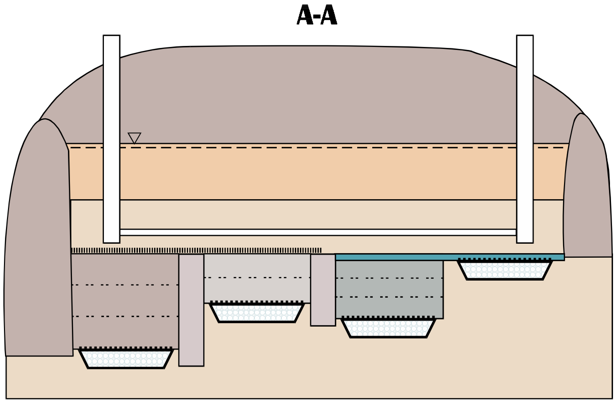

Figure 2. Schematic cross section of the test structure.

RESULTS AND DISCUSSION

The results of laboratory and field permeability, frost and temperature measurements are discussed in the paper published in Proceedings Sardinia 99, 7th International Waste Management and Landfill Symposium. In this paper, the emphasis is on the results of chemical analyses of water and material samples.

The quality of the leachate seeping through the liner structures was frequently followed by pH and conductivity measurements, and water samples were taken from lysimeter wells and leachate storage for laboratory analyses.

Quality of the leachate

The loading water used in test structures was pumped from the settling pond of the Ämmässuo landfill. The quality of the leachate in the pond is regularly monitored as a part of landfill observation.

During the monitoring period from March 1995 to August 1997, the pH of the leachate was between 6.7 – 7.7 and the solid contents between 60 – 550 mg/l, with the corresponding average of 185 mg/l. The electrical conductivity and chloride content of the leachate have a good correlation, and is shown in Fig. 3.

The leachate quality monitoring includes, in addition to wastewater parameters, the monitoring of the following substances: Ca, Mg, Al, Co, Ni, Fe, Mn, Cd, Cu, Pb, Zn, Cr, Hg and As.

The pond gathers leachate from all the filling areas, both old and new. Therefore, the quality of the leachate has a large range of variability. In the comparison, the maximum and minimum values of the leachate have been plotted to describe the variability of the leachate.

pH and electrical conductivity

The pH and electrical conductivity of the water samples were measured immediately after sampling in the laboratory of the Finnish Environment Institute using glass electrodes. The conductivity was also measured in the laboratory of Vesihydro Ltd. The results are presented in Figs. 4 and 5.

The electrical conductivity of the water seeping through the ash structure (3B) differs clearly from

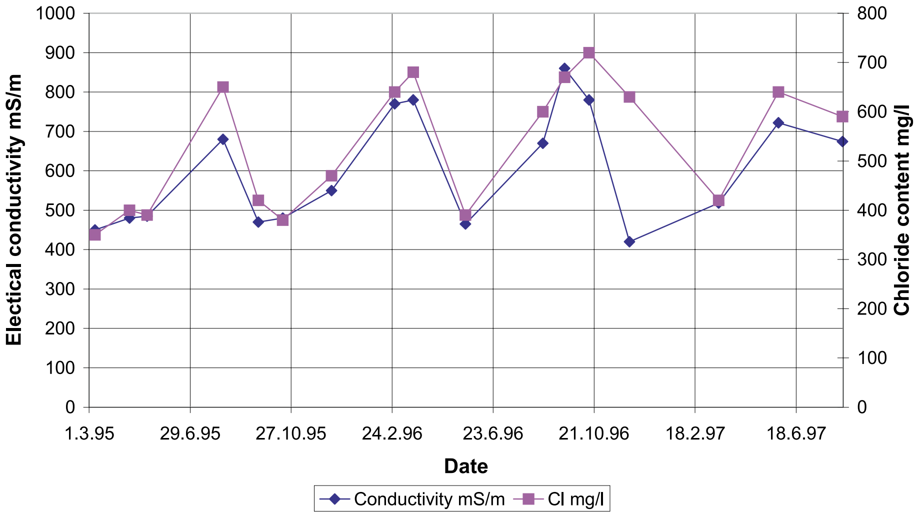

Figure 3. Electrical conductivity and chlorine content of leachate samples and seeping water samples from lysimeter wells.

that of the other two. At first, it increases rather quickly to the maximum value, about 3000 mS/m. Then it slowly decreases to the same level as the leachate. Thus, the chloride is being leached from the material.

The electrical conductivity of the water seeping through the bentonite structure (2B) reaches the level of leachate soon after the implementation of the leachate. A small peak can be noticed at the moment of increasing flow rate, in January 1997.

In the samples taken from the lysimeter well of clay structure (1B), the level of conductivity remains almost the same throughout the monitoring period. Values are clearly below the conductivity of leachate. For this reason, the clay liner provides the best ground water protection out of these three structures.

In contrast to the conductivity, there are no clear trends in the pH. The results from the ash structure display a slight decrease. In the soil bentonite structure, there is a minimum value at the moment in an increasing flow rate. The pH of clay remain close to neutral.

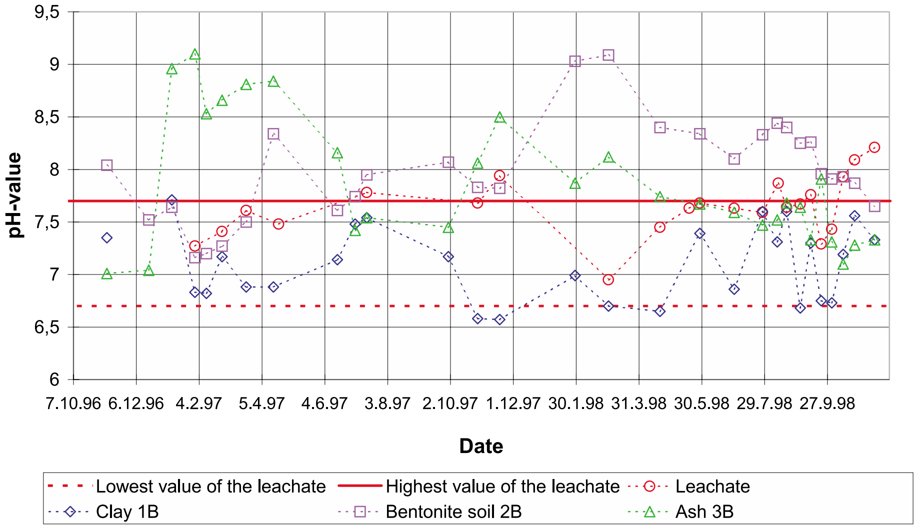

Figure 4. pH of leachate samples and seeping water samples from lysimeter wells.

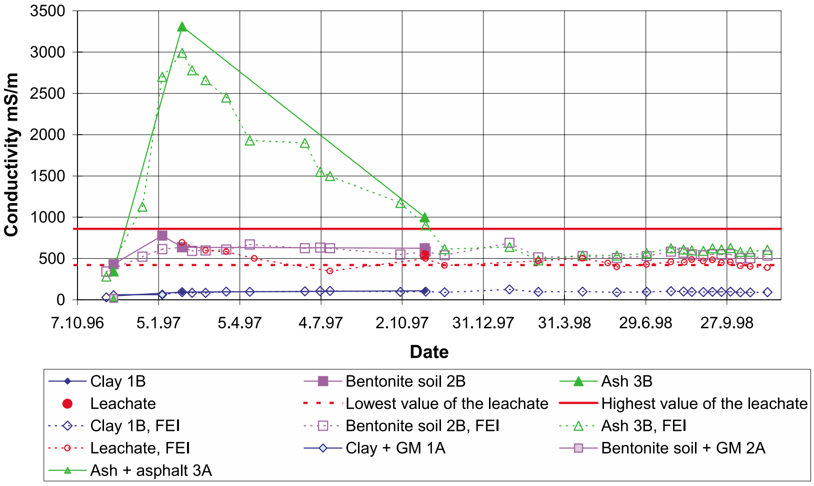

Figure 5. Electrical conductivity values of leachate samples and seeping water samples from lysimeter wells. Results are from two laboratories, Vesihydro Ltd and Finnish Environment Institute (FEI). There is only one set of samples from the A-parts.

Results of chemical analyses

In order to evaluate the propagation of the contaminants through the liner, the effects of sorption and ion exchange and the possible occurrence of chemical reactions, the chemical analyses were done on water samples taken from the lysimeter wells and leachate storage. The content of Ca, Na, Cl, Cr, Mg, Al, Ba and SO_4^{2-} were measured. The analyses were done in the laboratory of Vesihydro, Ltd and at the Geological Survey of Finland (GSF).

Figs. 6, 7 and 8 illustrate the trends of three different materials. The first results describe the water used in lysimeter well calibration, and therefore should be ignored or considered with caution. The assessment of results should be done taking into the consideration the amount of water seeping through the liner.

In the clay structure, the consistent results of chemical analyses support the conclusion that the clay acts as a filter and provides a good protection.

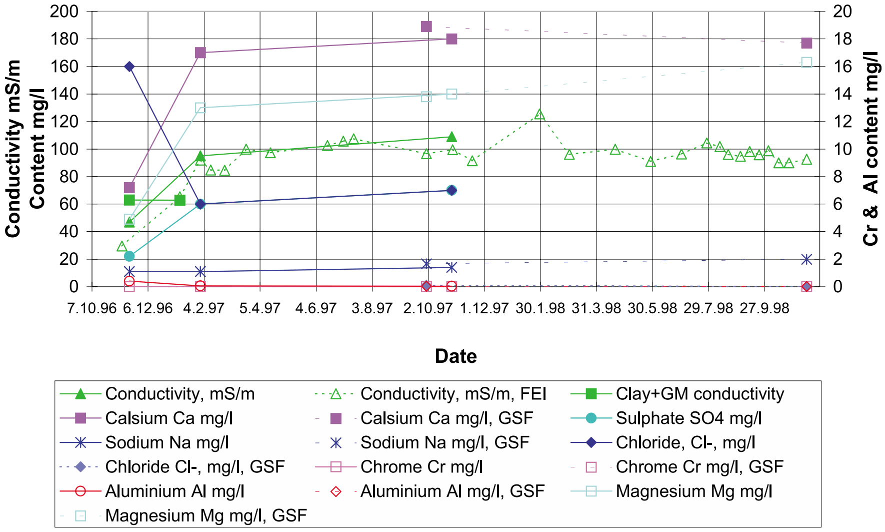

Figure 6. Results of chemical analyses of seeping water collected from the lysimeter of the clay liner.

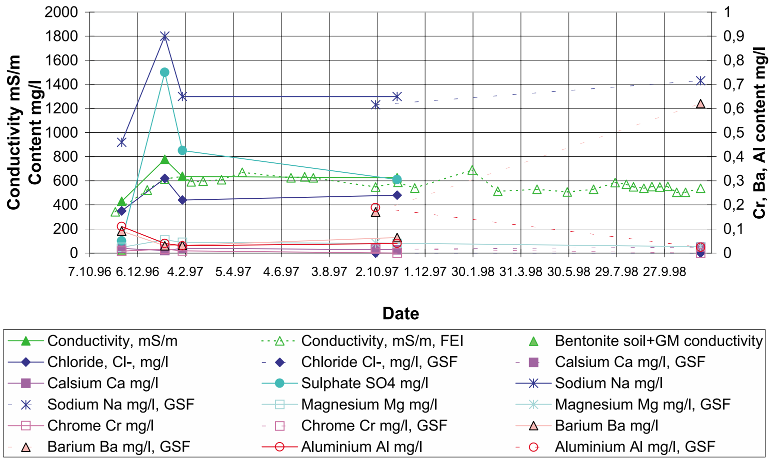

Figure 7. Results of chemical analyses of seeping water collected from the lysimeter of the soil bentonite liner.

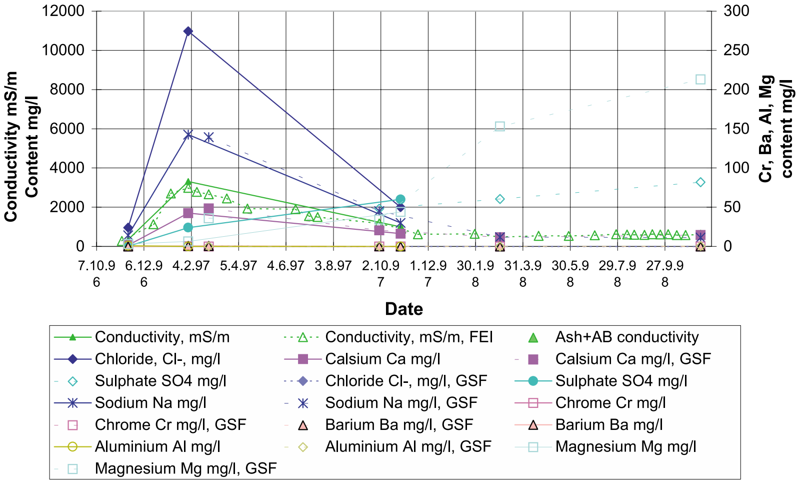

Figure 8. Results of chemical analyses of seeping water collected from the lysimeter of the ash liner.

Chemical analyses of the materials

The test structures were demolished in November 1998 and material samples were taken for laboratory analyses.

In order to evaluate the propagation of the contaminants through the liner, effects of sorption and ion exchange, and the possible occurrence of chemical reactions, chemical analyses were done on material samples. The concentrations of 25 substances, including heavy metals, were measured. The analyses were made in the laboratory of Geological Survey of Finland (GSF). A selected part of the results are presented in Table 1. The concentrations are compared to the Finnish guiding and limiting values used in assessing soil contamination. Similar to the water analyses, the assessment of these results should be done taking into the consideration the amount of water seeping through the liner.

In clay, the natural concentrations of some contaminants, such as vanadium and chrome, exceed the guideline values. No clear sign of effects of leachate can be noticed.

The guideline values are exceeded in fly ash and desulphurating agent, too. Typically the concentrations decreases due to the presence of leachate. It follows that the sorption processes aren't significant, whereas diluting is.

The chloride content of ash is high at the A-part, where the structure was protected by the asphalt layer and was not in a contact with the leachate. At the B-part, where the leachate can infiltrate to the ash layer and seep through it, the chloride content is significantly lower, only a few percent of the original values. The smallest values are from the surface of the liner structure, where the effect of the water is largest. By contrast, the concentration of arsenic is rather even at all ash parts, due to the poor solubility of arsenic in water.

In bentonite soil the amount of sodium diminishes due to the presence of leachate. The effect can be noticed from the sample taken from the surface of the B-part and from the sample taken under a geomembrane failure point found at the A-part during the demolition. The results do not show signs of accumulation of contaminants.

Based on these results, the leachate has a detrimental effect on both of the ash and bentonite soil liners investigated in this study. The effects on clay liner are undetectable.

Table 1. The concentration (mg/l) of selected substances in liner material samples. Samples taken before the leachate implementation are marked with text “before”. n.d. – not detected, DSA – desulpurating agent, BE-soil – soil bentonite.

Location	Cl mg/kg	As mg/kg	B mg/kg	Ca mg/kg	Co mg/kg	Cr mg/kg	Mg mg/kg	Mo mg/kg	Na mg/kg	Ni mg/kg	P mg/kg	V mg/kg
Clay BEFORE	n.d.	< 10	n.d.	5400	22.0	**106**	15000	< 5	540	50.0	597	**117**
Clay 1A surface	n.d.	< 10	**5**	3730	18.7	73.9	11300	< 0.3	417	41.1	357	**90.4**
Clay 1A bottom	n.d.	< 10	**7**	6260	28.3	**110**	18200	< 0.3	672	**61.3**	566	**134**
Clay 1B surface	n.d.	< 10	**12**	4660	21.6	87.5	13500	< 0.3	816	48.0	499	**108**
Clay 1B bottom	n.d.	< 10	**8**	5610	24.8	**102**	16500	< 0.3	611	55.8	531	**125**
Clay 1C	n.d.	< 10	**9**	5490	22.3	99.0	15400	< 0.3	580	53.9	553	**116**
Fly ash BEFORE	n.d.	**31**	n.d.	9800	10.0	22.0	2900	**6**	550	26.0	1740	45
Fly ash&DSA BF	n.d.	**14**	n.d.	104000	12.0	17.0	4400	< 5	11000	23.0	1380	37
Ash 3A 0 m	10400	**26**	**113**	10400	9.5	11.9	2650	**7**	4890	22.9	1720	39.2
Ash 3A 0.2 m	13100	**25**	**116**	9460	7.9	11.4	2350	**6**	5170	20.2	1490	35.9
Ash 3A 0.6 m	20900	**16**	**197**	56600	6.6	7.9	3020	4	6700	16.0	1020	22.7
Ash 3B 0.2 m	529	**38**	**128**	82900	9.8	15.4	3900	< 0.3	973	23.8	1480	40.9
Ash 3B 0.6 m	716	**22**	**473**	103000	10.5	12.9	5660	**6**	746	24.8	1390	35.7
Fly ash 3B surface	125	**20**	**42**	10400	10.0	14.4	2530	< 0.3	416	25.3	1890	37.2
Fly ash 3C	2.7	**28**	**61**	8840	9.9	11.6	2700	< 0.3	230	24.0	1790	39.3
BE-soil BEFORE	n.d.	< 10	n.d.	9600	8.0	36.0	6000	< 5	2270	15.0	337	42
BE-soil 2A surface	n.d.	< 10	< 5	7010	6.2	19.1	4390	< 0.3	2100	9.2	240	28.8
BE-soil 2A bottom	n.d.	< 10	< 5	9110	5.6	17.7	4250	**10**	3000	9.2	234	25.4
BE-soil 2A defect 0 m	n.d.	< 10	< 5	6450	5.6	17.5	4240	< 0.3	986	9.6	228	27.8
BE-soil 2A defect 0.2 m	n.d.	< 10	< 5	7170	4.7	13.6	3620	< 0.3	2210	6.0	205	21.7
BE-soil 2B surface	n.d.	< 10	**8**	11100	5.7	17.9	4280	< 0.3	541	9.2	318	27
BE-soil 2B bottom	n.d.	< 10	< 5	9350	5.2	15.6	4260	< 0.3	3000	7.1	252	25.7
BE-soil 2C	n.d.	< 10	< 5	8630	5.8	17.0	4320	< 0.3	2460	8.1	223	25.4
Guiding value		**10**	**5**		**50**	**100**		**5**		**60**		**50**
Limiting value		**50**	**50**		**200**	**400**		**200**		**200**		**500**

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International conference on practical applications in environmental geotechnology ecogeo 2000,
Edited by Reijo Salminen.
Geological Survey of Finland, Special Paper 32, 211–218, 2001.

LONG-TERM INVESTIGATION OF AN ALTERNATIVE SURFACE-SEALING SYSTEM WITH AN ENHANCED MINERAL CLAY LINER AND AN UNDERLYING CAPILLARY BARRIER AT THE KARLSRUHE-WEST SANITARY LANDFILL (GERMANY)

by
W. Breh and H. Hötzl

Breh, W. & Hötzl H. 2001. Long-term investigation of an alternative surface-sealing system with an enhanced mineral clay liner and an underlying capillary barrier at the Karlsruhe-West sanitary landfill (Germany) *Geological Survey of Finland, Special Paper 32, 211–218,* two figures and 2 tables.

At the sanitary landfill Karlsruhe-West an alternative surface-sealing system with an enhanced mineral clay liner and an underlying capillary barrier is employed. A macro-scale lysimeter was used to obtain water balance data over a period of more than six years. The data of the long-term investigation show that the alternative surface-sealing system is an effective and economic way for covering a landfill and so reducing the seepage water infiltration rate to a minimum.

Keywords: engineering geology, landfills, sealing, clay, disposal barriers, water balance, Karlsruhe-West, Germany

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INTRODUCTION

Due to subsurface contamination in the vicinity of the Karlsruhe-West sanitary landfill, the city of Karlsruhe was forced to realize a comprehensive remediation concept for this site. The central concern of this concept was the emplacement of a qualified surface-sealing system covering the entire 22-hectare area of the waste site.

Because of the steep slope angles of 1:2.3 (23.5°), the installation of the regular sealing system involving a combination of a polymeric membrane with high density (PE) on a highly compacted mineral clay liner, as defined in the Technical Instruction for Domestic Waste (Technische Anleitung Siedlungsabfall, TA-Si, Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit 1993) was not feasible. Therefore, an alternative surface-sealing system comprised of an enhanced mineral clay liner with an underlying capillary barrier was utilised as the main sealing unit.

For alternative surface-sealing systems, the TA-Si explicitly requires proof of equivalence. To provide such proof, a test field of 2.2 hectares, including an integrated macro-scale lysimeter of 10 x 40 metres, was built at the landfill Karlsruhe-West in 1993. This lysimeter is used to obtain water-balance data on the alternative sealing system. The processing of the data and the fieldwork is done by the Department of Applied Geology, University of Karlsruhe.

STRUCTURE

In 1993 Lysimeter I was built in the southern part of the landfill (Fig.1). With a length of 40 m and a width of 10 m, it has a base area of 400 m^2. The surface is sloped with a gradient 1:2.3 (23.5°) to the south. In Fig. 2 the schematic structure of Lysimeter I is shown.

Extent of the investigation

The extent of the performed investigations and the necessary measuring equipment at the sanitary landfill resulted from the following objectives:

- Proof of equivalence for the alternative surface-sealing system in comparison to the standard composite liner system postulated by the TA-Si regulation.
- Gaining data for a soil water balance with special emphasis on the vegetation support layer.
- Determination of the effectiveness of the capillary barrier system and of the whole alternative surface-sealing system.
- Long-term investigation of the hydraulic conductivity of the enhanced mineral clay liner.

The main input parameter is the daily precipitation depth. It is measured with three separate standard precipitation gauges. The daily depths of discharge for each different layer of the lysimeter were measured by central measuring equipment and manually fed into the computer for processing.

On the lysimeter test field, four pipes (NS1 – NS4) for the use of a neutron probe were installed. The neutron probe was used for the weekly radio-

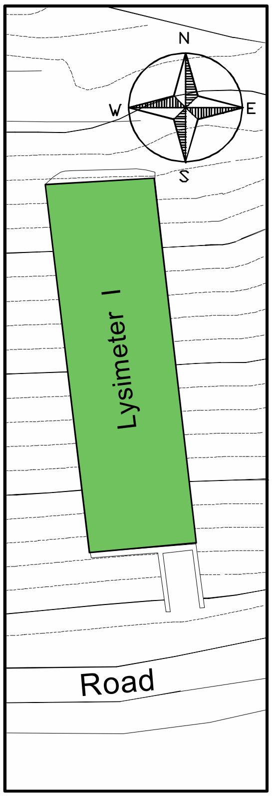

Figure 1. Site plan of Lysimeter I.

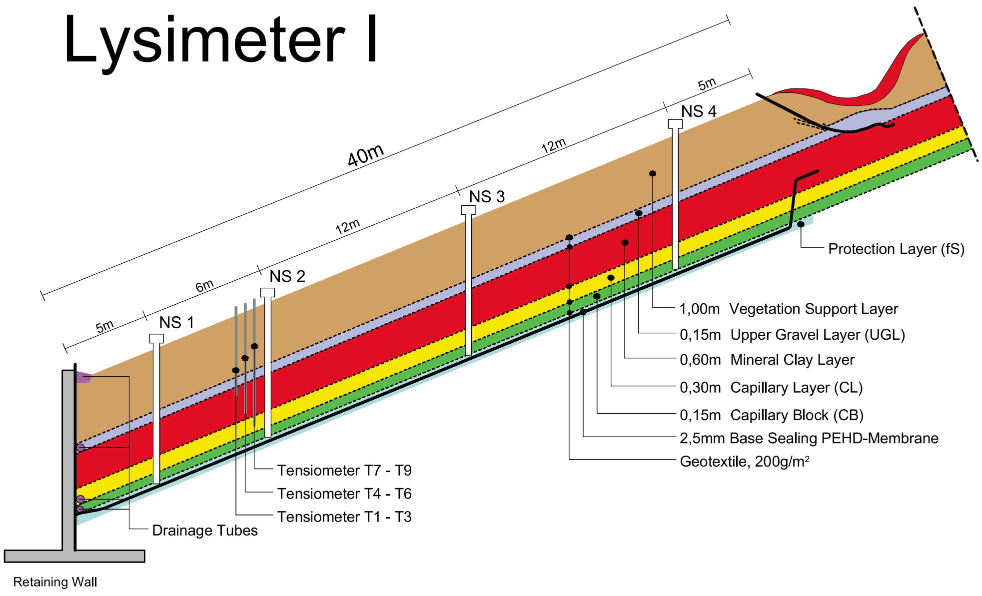

Figure 2. Schematic cross section of Lysimeter I with the permanent measuring equipment.

metric determination of the soil water content. The variation of the soil water content is an important factor for calculating the soil water balance.

Near to test point NS 2 nine tensiometers were installed to measure the soil water potential and the hydraulic gradients in the unsaturated zone. From the data collected conclusions about the saturation conditions and the direction of water movement in the unsaturated zone are possible. The tensiometers measure water tension and soil temperature every four hours at several depths.

RESULTS OF THE LONG-TERM INVESTIGATION

Precipitation (Prec.): Fig. 3 shows the daily precipitation distribution for the entire observation period. The precipitation situation is characterized by a quite uniform and constant precipitation distribution, which only occasionally exceeds a precipitation depth of 30 mm in 24 h. The absolute maximum was observed in February 1997 with a precipitation depth of more than 80 mm a day. The monthly and yearly precipitation sums are given in Table 1. The listed data show that in the past period of measurement four years (1994, 1995, 1997, 1999) were characterised by a higher annual total than the long-term average (739 mm) and in two years (1996 and 1998) a lower annual total was observed. Hence the surface sealing in the period of observation was exposed to rather wetter weather conditions with respect to the long-term average.

Actual evapotranspiration (ETa): The difference of all water balance factors can be used for the computation of the actual evapotranspiration. For the undertaken measurements, data ascertained after Wendling *et al.* (1984) was compared to the data for the actual evapotranspiration derived from the lysimeter measurements. For the individual water balance years this resulted in satisfying to good conformity. For the entire period of observation the actual evapotranspiration comes on average to 83.5% of the precipitation.

Surface runoff (Q_{SURF}): For the surface runoff of the test field its considerable decrease and finally complete absence is remarkable. This phenomenon can be explained by the increasing density of the vegetation on the test field, but was not expected because of the steep slope. However, an intensive check of the measurement unit, which was undertaken several times, confirmed the correctness of the determined data. The long-term water balance (Table2) shows that an average of approximately 0.3% of the precipitation runs off at the surface. In 1998 the surface runoff stopped totally.

Upper gravel layer (Q_{UGL}): The upper gravel layer is normally supposed to drain the main bulk of percolating water in this kind of system. The layer was emplaced with a reduced thickness of 0.15 m compared to the TA-Si standard (0.3 m). It is striking that for the period of observation the discharge in the upper gravel layer was considerably reduced and only small peaks are still observable. The discharge maxima correlate well with stronger precipitation events.

Over the entire period of measurement on average 8.2% of the whole precipitation is drained in the upper gravel layer. For the last three years the upper gravel layer discharge lies below the one in the capillary layer. The development of the data gives a clear indication that the mineral clay layer looses its property of being mostly impermeable with time, because increasingly larger amounts of percolating water pass the mineral layer and are drained in the capillary layer.

Capillary layer (Q_{CL}): Fig. 3 shows the discharge for the entire period of observation. A yearly change is clearly visible in the way that during the main precipitation period (November/December to April) the highest discharge intensities occur. This is characterized by a discontinuous curve due to precipitation and percolating activity of varying intensity. After this high discharge activity a normal discharge behaviour develops, which can be compared to a storage hydrograph (Zischak 1997). In the period of observation about 8.1% of the precipitation was drained in the capillary layer. It is apparent that the relation between the upper gravel layer discharge and the discharge of the capillary layer clearly shifted towards the latter. This indicates again the retention and draining property of the capillary layer.

Capillary block (Q_{CB}): The lateral discharge beneath the capillary block is equivalent to the sealing property of the whole system and in the end gives the amount of the potential percolating water recharge of the landfill. In the entire period of observation, 0.6% of the precipitation runs off in permanent discharge activity. The permanent measurable low discharge beneath the capillary block is a result of the not optimally adjusted grain sizes in the capillary layer and the capillary block (Zischak & Hötzl 1996). With optimally dimensioned materials only the smallest amounts of percolating water pass the boundary layer and these are merely added to the adhesion water. A measurable discharge is supposed to occur exclusively due to a failure during a hydraulic overstrain.

The four pronounced discharge peaks correlate well with the discharge maxima in the capillary layer and occurred only in periods of clearly higher precipitation activity. But even during these breakthrough events the daily discharge depths stayed relatively low. By far most of the time the daily discharge heights were below 0.025 mm. The low discharge depths in the capillary block show clearly that practically the entire percolating water is drained reliably by the capillary layer, which fulfils its function as a sealing system within the capillary barrier.

Soil water storage (Δ_{SW}): The development of the soil humidity for the observation period is characterized by a pronounced decrease of the water contents in the summer months and a subsequent rapid increase. This development is observable in all layers, including the mineral clay layer. The measurements of the water contents in the mineral clay layer indicate a persistent dry-up of this layer. This drying behaviour was documented by neutron probe measurements in 1998 and 1999. All together the soil humidity decreased in the period of observation by 36.6 mm (0.7% of the entire precipitation).

Hydraulic gradients: Tensiometers were used to determine the hydraulic gradients in the unsaturated zone. Calculation of the hydraulic gradients was undertaken with the capillary block as a reference level. This means that positive gradients show a water movement from top to bottom and negative values a motion in the opposite direction. The development of the gradients between the first and the third layer of the mineral sealing confirm the aforementioned statements about the soil water conditions and the dry-up behaviour of the mineral clay layer.

The tensiometer data show that in summer time the mineral clay layer experiences a strong negative hydraulic gradient, which means it loses water. This result, in combination with the discharge data and the data of the neutron probe, is a strong indication for the at least partial dry-up of the mineral clay layer which leads to the observed increase in hydraulic permeability.

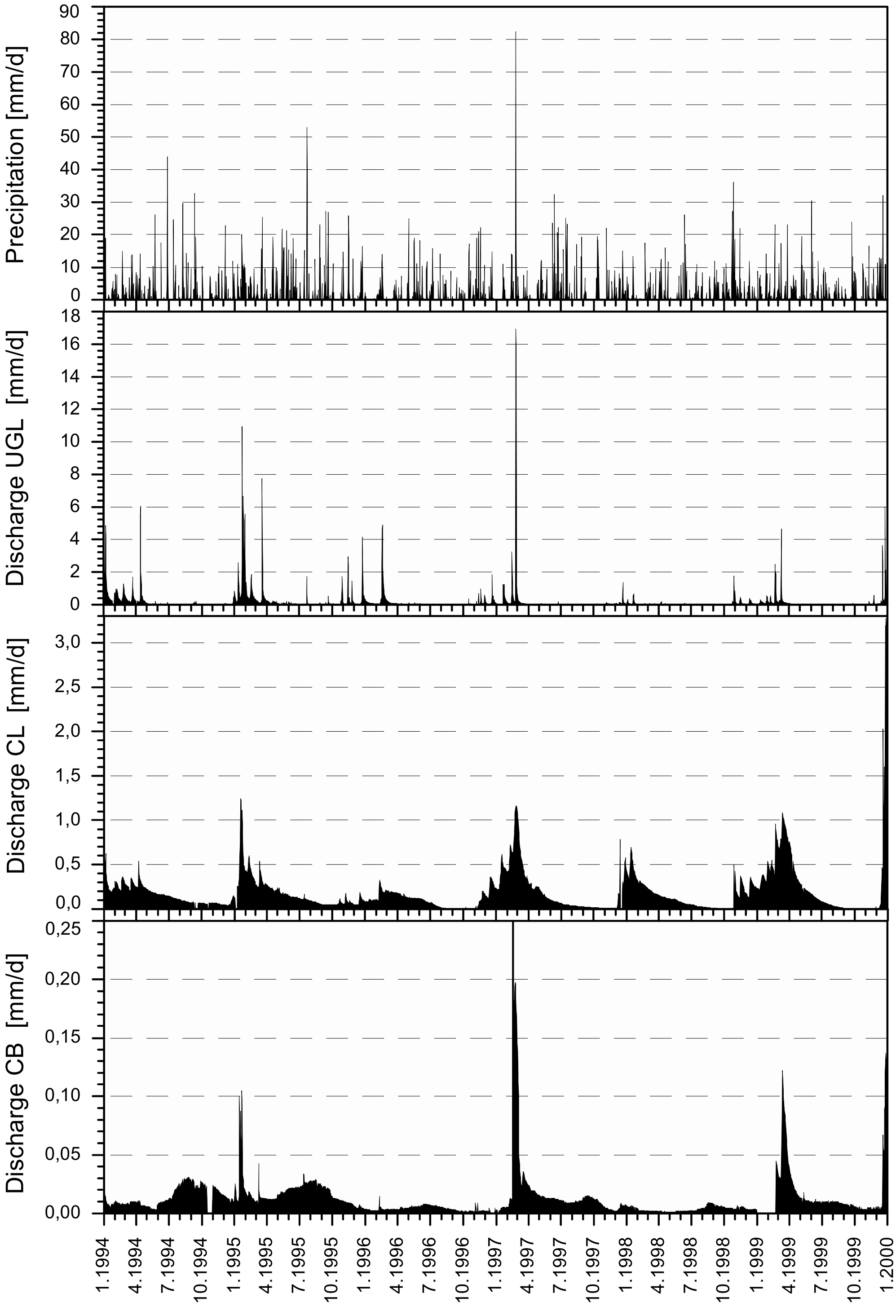

Figure 3. Daily precipitation and discharge depths in [mm/d].

WATER BALANCE OF THE ALTERNATIVE SURFACE-SEALING SYSTEM

Table1 shows the monthly water balances, the annual sums and the percentage of the precipitation (% of Prec.).

For the last period of observation, an over-average hydraulic strain for the surface-sealing system has resulted from the relative abundance of precipitation (four wet years: 1994 with 812 mm, 1995 with 1068 mm, 1997 with 879 mm and 1999 with 846 mm) and two dry years: (1996 with 660 mm and 1998 with 717 mm).

Table 1. Monthly soil water balances (all data in mm).

1994	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	Sum	% of Prec.
Prec.	61.6	49.7	71.2	69.0	79.7	78.5	52.4	83.1	107.5	38.4	42.3	79.2	812.7	
Q_{SURF}	0.2	0.3	0.8	0.3	0.6	0.7	0.3	0.5	0.3	0.0	0.0	0.3	4.2	0.52
Q_{UGL}	32.4	17.1	11.7	21.1	1.3	0.5	0.1	0.0	0.0	0.0	0.2	2.7	87.2	10.73
Q_{CL}	8.9	7.0	8.4	8.4	6.5	4.9	4.2	3.0	1.5	1.8	1.9	1.5	58.1	7.15
Q_{CB}	0.3	0.2	0.2	0.3	0.2	0.2	0.4	0.8	0.8	0.6	0.5	0.4	4.9	0.60
Δ_{SW}	6.0	2.3	–16.3	–17.0	–39.6	–29.0	–25.3	–6.0	17.0	1.9	0.0	41.5	–64.5	–7.94
ETa	13.8	22.9	66.3	55.9	110.8	101.1	72.6	84.8	87.9	34.1	39.8	32.8	722.7	88.94
1995	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	Sum	% of Prec.
Prec.	133.6	63.2	90.3	77.2	123.3	96.6	136.8	71.1	106.4	31.5	79.9	57.5	1067.5	
Q_{SURF}	0.9	0.1	0.2	0.2	0.2	0.2	0.2	0.2	0.1	0.0	0.0	0.0	2.2	0.20
Q_{UGL}	63.3	19.4	21.4	4.7	1.9	1.4	2.8	0.3	1.6	4.1	11.4	13.6	146.0	13.68
Q_{CL}	12.6	13.4	10.1	7.5	6.0	4.4	3.7	2.7	1.6	1.8	2.6	2.5	68.8	6.44
Q_{CB}	1.0	0.5	0.4	0.4	0.5	0.6	0.8	0.8	0.7	0.4	0.3	0.2	6.7	0.63
Δ_{SW}	41.9	–10.8	–1.0	–14.4	1.8	–24.0	–27.6	–17.5	34.3	15.8	13.4	17.0	28.9	2.71
ETa	13.9	40.5	59.2	78.9	112.9	114.0	157.0	84.5	68.1	9.4	52.2	24.3	814.9	76.34
1996	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	Sum	% of Prec.
Prec.	5.1	66.4	21.7	23.1	111.7	74.0	69.3	41.9	19.0	62.7	127.4	37.8	660.1	
Q_{SURF}	0.0	0.0	0.0	0.0	0.4	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.6	0.10
Q_{UGL}	3.7	22.6	3.6	1.0	1.2	0.2	0.1	0.0	0.0	0.5	4.7	7.1	44.7	6.77
Q_{CL}	2.9	4.7	6.0	5.0	3.9	3.2	1.7	0.3	0.1	0.1	1.1	6.9	36.0	5.45
Q_{CB}	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2	0.1	0.1	0.1	0.1	1.5	0.22
Δ_{SW}	–13.0	17.0	–13.8	–9.0	–2.3	–70.0	–39.2	–9.7	–19.3	31.3	91.4	26.5	–10.1	–1.53
ETa	11.4	22.1	25.7	26.0	108.3	140.3	106.4	51.1	38.1	30.7	30.1	–2.8	587.4	88.99
1997	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	Sum	% of Prec.
Prec.	37.2	155.6	36.6	24.8	62.8	165.0	112.1	62.1	13.8	65.7	73.1	70.4	879.2	
Q_{SURF}	0.1	0.2	0.0	0.0	0.0	0.2	0.1	2.8	0.0	0.0	0.0	0.0	3.3	0.37
Q_{UGL}	8.1	45.7	4.4	0.1	0.2	0.5	0.1	0.0	0.0	0.2	1.1	4.5	64.8	7.37
Q_{CL}	10.4	15.5	23.7	8.4	6.0	2.9	1.8	1.1	0.7	0.4	0.3	3.0	74.3	8.45
Q_{CB}	0.1	1.6	2.8	0.7	0.5	0.4	0.3	0.3	0.4	0.4	0.1	0.1	7.6	0.87
Δ_{SW}	6.4	34.9	–45.2	–30.8	–50.9	45.1	–24.5	–26.3	–38.9	37.7	52.2	26.0	–14.3	–1.63
ETa	12.2	57.7	50.9	46.4	107.0	115.9	134.4	84.2	51.6	27.1	19.4	36.7	743.5	84.57
1998	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	Sum	% of Prec.
Prec.	62.5	23.8	39.8	94.0	14.8	96.5	45.5	26.8	71.1	143.1	61.7	37.8	717.1	
Q_{SURF}	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.00
Q_{UGL}	3.9	0.4	0.3	0.8	0.0	0.3	0.1	0.0	0.0	2.9	4.0	2.8	15.5	2.15
Q_{CL}	14.9	8.8	6.6	4.4	3.5	2.1	1.1	0.5	0.2	1.0	7.0	7.3	57.4	8.01
Q_{CB}	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.1	0.1	0.1	1.4	0.20
Δ_{SW}	10.9	–7.0	–15.9	–31.9	–53.1	–25.3	–4.8	–13.4	14.4	100.6	18.0	12.5	4.8	0.66
ETa	32.6	21.5	48.7	120.7	64.3	119.4	49.0	39.5	56.3	38.4	32.6	15.1	638.0	88.97
1999	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	Sum	% of Prec.
Prec.	77.1	67.1	93.0	49.3	66.1	79.7	52.0	36.8	67.0	49.3	48.0	160.1	845.6	
Q_{SURF}	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.00
Q_{UGL}	4.6	11.6	12.1	0.8	0.2	0.0	0.0	0.0	0.3	0.2	1.7	21.2	52.7	6.23
Q_{CL}	9.9	16.0	26.5	13.9	6.7	3.8	1.9	0.7	0.1	0.1	0.1	31.4	111.1	13.14
Q_{CB}	0.2	0.5	2.0	0.7	0.3	0.3	0.3	0.3	0.2	0.1	0.1	1.4	6.4	0.76
Δ_{SW}	27.5	8.6	–5.0	113.7	18.2	–29.3	–27.1	–13.1	–0.3	27.2	40.2	89.2	22.7	2.68
ETa	34.9	30.4	57.4	147.6	40.7	104.9	76.9	48.9	66.7	21.6	6.0	16.9	652.7	77.19

The water balance term ETa results from the difference of all other terms of the water balance with ETa = Precipitation – ΣQ – $\Delta_{\text{soil water}}$ (= variation of soil water content) directly from the lysimeter measurements.

The data in Table 1 demonstrate clearly how the discharge behaviour in the individual layers of the alternative surface-sealing system changes within the period of observation:

1. The surface runoff decreases progressively and could not be observed anymore from 1998 at all.
2. The relationship between the discharge in the upper gravel layer and the discharge in the capillary layer shifts progressively towards the capillary layer.
3. The discharge of the capillary block, which resembles the percolating water infiltration rate of the landfill, always stays below 1% with respect to the annual precipitation, in low precipitation years even below 0.25%.

Setting up a water balance for the entire period of observation results in the data given in Table 2.

The calculation of the average efficiency factor E_{cap} of the capillary barrier system from the equation

Table 2. Soil water balances for the entire observation period 01.01.1994–31.12.1999.

	mm	% of Prec.
Precipitation	4982.1	
Q_{SURF}	12.5	0.25
Q_{UGL}	409.6	8.22
Q_{CL}	404.8	8.12
Q_{CB}	28.5	0.57
$\Delta_{Soil\ Water}$	–32.6	–0.65
ETa	4159.3	83.48

$$E_{cap} = 1 - \frac{\sum Q_{CB}}{\sum Q_{CL} + Q_{CB}}$$

gives a value of 0.93 for the period of observation. The average efficiency factor E_{tot} of the entire alternative surface-sealing system can be calculated from the equation (P = Precipitation)

$$E_{tot} = 1 - \frac{\sum Q_{CB}}{\sum P} = 0.994.$$

CONCLUSIONS

On the basis of the presented field data there are several conclusions with regard to the results of the long-term investigations on the alternative surface-sealing system with an enhanced mineral clay liner and an underlying capillary barrier (Breh & Hötzl 1999):

- The presented data prove that the realized alternative surface-sealing system at the Karlsruhe-West sanitary landfill is an effective and economic surface-sealing system, which reduces the amount of leachate in consequence of precipitation to a minimum. The results of the long-term investigation convinced the proper authorities to approve the employment of this alternative surface-sealing system.
- An increase of the thickness of the vegetation support layer from 1 m to 2 m should have positive consequences for the whole surface-sealing system because it increases the transpiration capability of the vegetation, reduces the risk of root penetration and has a positive effect on the retardation of percolating water.
- A thickness of 0.15 m for the upper gravel layer (TA-Si standard is 0.3 m) is sufficient for reliable draining of the percolating water in this layer.
- The development of the discharge distribution between upper gravel layer and capillary layer in favour of the latter, as well as the results of the neutron probe and the tensiometer measurements are a clear indication that the hydraulic sealing properties of the mineral clay layer, even in enhanced form (thickness 0.6 m instead of 0.5 m), are already in the medium-term (within three years) significantly reduced. It is therefore to be further investigated by which technical precaution the required properties of the mineral sealing can be secured in the long-term or in what way the mineral layer is to be substituted by another sealing component, e.g. an asphalt layer, or to be omitted completely.
- A better adjustment of the materials of the capillary layer and the capillary block should result in a further increase of the efficiency of the capillary barrier system.

All these conclusions resulted in a modified alternative surface-sealing system for the Karlsruhe-West landfill site.

For the investigation of the modified structure and for demonstration of the equivalency according to TA-Si, the concept for an additional lysimeter test field (Lysimeter II) was developed together by Ingenieurbüro Roth & Partner and Prof. Dr. Hötzl of the Department of Applied Geology, Karlsruhe. The construction of Lysimeter II was completed in April 2000. The first results of Lysimeter II will be presented at a later date.

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CLEAN-UP CRITERIA FOR CONTAMINATED LAND MANAGEMENT – HOW TO ACHIEVE A COST EFFECTIVE BALANCE.

by
M. Loxham[1] and S. Dack[2]

Loxham, M. & Dack, S. 2001. Clean-up Criteria for Contaminated Land Management – How to achieve a Cost Effective Balance. *Geological Survey of Finland, Special Paper 32, 219–225,* two figures.

It has become quite clear that even in the rich industrialized world there are not enough resources to clean contaminated soils to the desired levels. Furthermore, even if the resource were to be available, the technologies are inadequate to achieve these aims. This has necessitated the development of alternative cost effective contaminated land management strategies. These are based around the twin concepts of Risk Analysis and Cost Benefit Analysis. This paper explores the methodology and some of its' consequences.

Keywords: soils, pollution, risk analysis, remediation, cost

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INTRODUCTION

The contaminated land problem was brought into focus in the late seventies by the incidents at Love Canal in the United States and Lekkerkerk in the Netherlands. Other contaminated sites in the entire world were quickly identified and it soon became apparent that contaminated land was a frequent rather than a rare phenomenon. It also triggered a public response that demanded both clean-up and prevention of future incidents. This in turn prompted politicians and their servants to promulgate rigorous soil quality criteria backed up by some of the most draconian legal sanctions hitherto found in the environmental field. The Dutch, for example, developed the multi-functionality principle that required all soils to be of such a quality that they could be used for all functions, from agriculture to supporting waste sites. It was embedded in the criminal code of Dutch law. The United States addressed the clean-up problem directly through the Super Fund that was based directly on the two pillars of clean-up and that the Polluter Pays.

Unfortunately the costs both in financial terms and in disruption to society terms have proven to be beyond the bearing capacity of even the richest nations. This has lead to a general retreat from the initial position of land quality restoration to one of a more economically feasible contaminated land management philosophy. In fact it has taken to May of this year, that is 20 years, for the Dutch to abandon in principle the Multi-functionality objective.

Whilst this readjustment of our collective expectations from the land quality restoration programs has been going on there has been a second related development that is emerging as a key driver in contaminated land problems. This is the, often only perceived, impact of the land quality parameters on the value of the land in a financial transaction. This development has brought a whole new spectrum of considerations to the fore and is forcing a reappraisal of the meaning and use of land quality standards.

It is against this rapidly changing backcloth that the search for new and more robust assessment methodologies and criteria has to be seen. There are several possible starting points for developing novel procedures for the examination of contaminated land and the design of response strategies. These include the application of the Observational Method and the formulation of Generic Characteristic Values for identified site archetypes.

In recent years the methodologies and technologies of quantitative risk analysis have been applied to the problem. Whilst they can give rational answers to the soil quality problem they in fact push the debate back to the community that then has to agree on what is an acceptable risk from soil pollution. None the less these methodologies are the most promising to date and will be looked at further in what follows. It should be noted that the United Kingdom has based her recent contaminated land legislation entirely on the risk analysis principle.

RISK ANALYSIS

Risk Analysis provides a context for the whole analysis and management of the contaminated site. It provides a common, if sometimes difficult, forum for communication of the problem to all concerned and it can be coupled with cost-benefit techniques to provide a very powerful management tool. Equally the techniques can be used in the field of pollution prevention design to balance of the need for pre-investment in measures to avoid possible problems. In what follows a description of this approach and its' application will be presented. The methodology has several major disadvantages or issues and these will also be presented.

Risks and Risk Based Methodologies

The key distinguishing factor between risk based design methodologies and other more conventional ones is that they are based on a recognition that soil contamination is only relevant because it has the potential to cause harm. Or put another way, the chemical characteristics of the contaminated soil itself are of no importance in themselves as such. This approach is in marked contrast with earlier ones that sought to reduce the contaminant level at the site to a prescribed, and often legislated, value. It is this paradigm shift from site contaminant parameters to receptor (or target) impact values that brings with it the many advantages of the new methodology.

A second important consequence of this approach is that it is no longer possible to use design procedures that are based on prescribing management techniques or the properties of components of remediation engineering. Only the functionality counts.

The harm or risk involved can be of several types. These include;

- Risk to Human Health either in the long or short term.
- Risk to Flora and Fauna including uptake in food chains.
- Risk to the Eco-system as a whole including diversity.
- Risk to the Asset Value of the Site even though realization is not necessarily planned.
- Risk to the Use Value of the Site restricting its economic value.
- Risk of incurring Liabilities to others by cross boundary migration.
- Risk of Legislative non-compliance leading to fines or imprisonment.
- Risk to the Reputation of the owner or user of the site.
- Risk to Ground Resources such as groundwater.

There is a notional risk to the bearing capacity of ground in its’ support function but this has been only rarely documented. Often these risks can be concurrently experienced and many of the categories overlap strongly. It is clear from this list that the span of any suite of Risk Analysis techniques will have to be quite wide for them to be effective.

This simple picture is complicated by the fact that no one person appreciates and deals with risk in an entirely ration manner. A distinction has to be made between risk as such and perceived risks. It has to be recognized that perceived risk is the real driving force behind many calls for remediation. The methodologies can be extended to include risk perception elements but there has been little or no documented study of this.

The Source-Path-Target Methodology

Soil contamination problems give rise to four basic questions:

1. What contamination is present? – The site investigation problem.
2. Where are the contaminants going to? – The migration problem.
3. If the contaminants get there, will there be a problem? – The impact problem.
4. If the impact is significant, what can be done about it? – The remediation problem.

The first three problems can be integrated in the “Source-Path-Target” methodology, a viable subset of risk analysis techniques. The last is concerned with remediation technologies themselves rather than the design methodologies and will not be considered further here.

The methodology starts with identifying three elements of the contamination management problem, the Source, the Path and the Receptor (or Target). The objective of the analysis is to relate events, such an emission of toxic substances from a contaminated site, to its effect at some sensitive point, or “target” in the environment. This relationship is a chain of sub-events, such as leaching followed by transport by groundwater and subsequent abstraction as drinking water, and is referred to as the “Path” connecting the Source of risk to the Target where its’ impact will be felt. In the general methodology the path can refer to effect transfer functions more abstract than the physical flow of toxic substances.

The Source is characterized by the nature and emission strength of the chemicals in the site. The Path is the chain of pollutant migration events as described above and is a vector. In general there are multiple pathways between source and target and the further removed these two are from each other, and the more members in the chain of events forming the path that have to be considered, the more difficult the analysis.

The target is defined by its sensitivity and its’ position in the environment. This is expressed as a maximum allowable impact value and is target specific. In theory the sensitivity of the target could be defined in toxicological terms if the detailed nature of the chemicals arriving at the target from the site contaminants and the required dose-effect relationships were to be known. However this is not always possible as the data are either not available or because the pollutants reaching the target often form a complicated cocktail of individual substances whose cumulative impact is only poorly understood. None the less there has been rapid progress in this area for hydrocarbons and several simple risk analysis models such as RBCA from the ASTM or Risk Human from the van Hall Institute have quite extensive data bases associated with them. These allow very viable estimates of the impact at target to be made.

Many countries have approached this problem of poor toxicology by setting chemical concentration standards rather than impact values. These are the maximum allowable concentration of MAC based standards of which the Dutch A, B, and C values or the new target and intervention levels have received wide application. This can result in a drastic simplification of the problem but sacrifices some of the advantages of the impact analysis route.

The practical problem of using the methodology however turns not only on what the MAC, or impact values are, but where along the impact pathways are they to be applied. The conceptual position around the site where the MAC values are to be applied is called the “target envelope”. Changing the position of this envelope can have profound influence on the range of remedial countermeasures available and the costs involved.

For the risks other than those to health a different set of maximum allowable impact values have to be set. These can be financial or less quantifiable and can reflect elements of perceived risk as well as real risk.

The pathways connect the source with the receptor and are vectors characterized by direction and speed. They may be simple as in convection and dispersion through the groundwater system or complex and multi-membered such as a food chain pathway. Although physical pathways can be easily visualized, some of the risk areas mentioned above require abstract pathway notions that are very difficult to work with. The physical pathways can dilute, delay and disperse toxic material emitting from the site, can provide opportunities for biodegradation or chemical stabilization or conversely can result in re-concentration of toxins by bio-accumulation or adsorption.

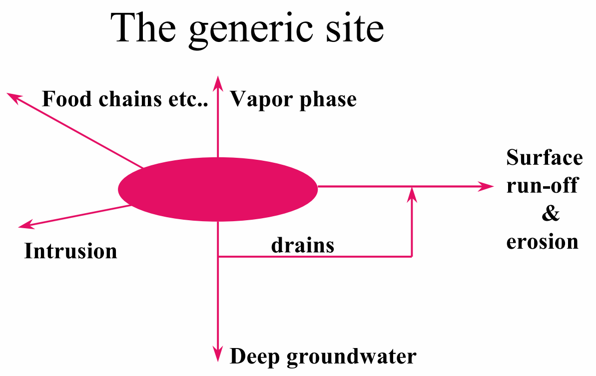

Figure 1. The generic site.

For a typical site at least the following pathways have to be considered,

- Leachate through the shallow groundwater system and local drains.
- Leachate to the deeper groundwater system especially where this is an important aquifer.
- Surface run-off of toxic inventory either by erosion or leaching Vapor phase dispersion into surroundings.
- Dust (particulate) dispersion into the surroundings.
- Uptake in the food chains.
- Intrusion into the site and physical displacement.

All these pathways can be operative at some point in the history of the site, from initial contamination, through the remedial actions to the long-term inevitable loss of institutional control on the site.

Again the pathways by which the fact of contamination at a site can reach a target and impact it for risks other than health risks requires careful study and delineation.

The source is characterized by its’ nature and emission as a function of time. This is given by the sort of chemicals in the site, their mobility and their concentration. The concentration alone says little of the emission strength, for example 500 ppm of cadmium in a site as sulfide is a very different risk than 5 ppm of the same cadmium present as a highly mobile chloride. This is of particular importance as most sites contain a cocktail of pollutants whose behavior furthermore changes in time. This is especially the case as remediation technologies are applied and conditions deliberately changed. A common example is where the lowering the groundwater table prior to excavation can result in a major re-mobilization of heavy metals as they are re-oxidized up into soluble species and their subsequent migration along the convection pathways caused by the dewatering operation. The mobility of a pollutant in a site is also influenced by physical factors such as temperature and pressure.

The source is the point where the risk perception of people involved focuses. This is often one of imagined emission and potential danger on the site itself. The language used to describe the site is telling. Words such as "chemical time bomb" and "disaster waiting to happen" are well known. It is however these notional emission factors that have to be used in the risk analysis.

The three elements of Source, Path and Target are put together to form the so-called "linkage". One way of expressing this is in the following diagram

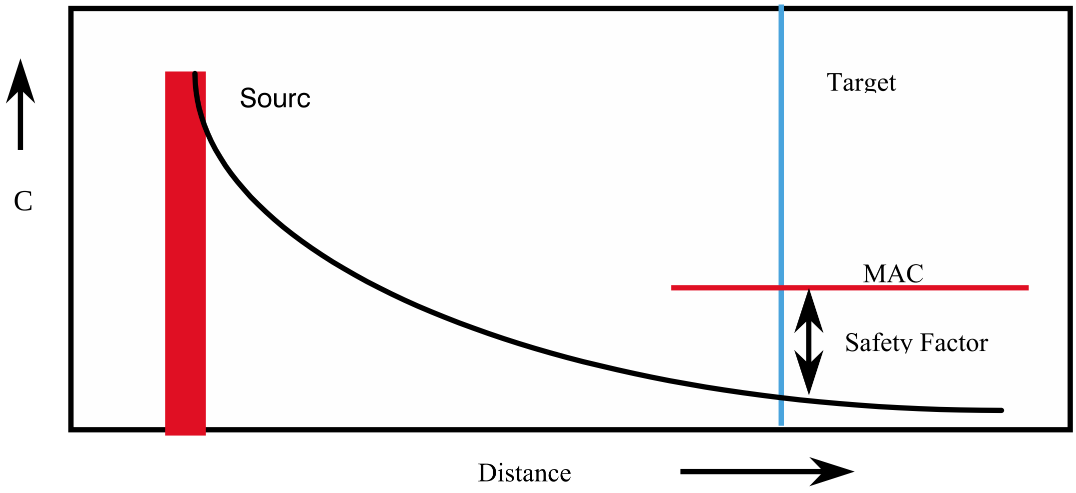

Figure 2. The linkage.

Here the drop in concentration with distance away from the emitting source is shown. Somewhere on the pathway is found the target. If the concentration is below a maximum allowable level then there is, in principle at least, no problem. It is above a remedial management strategy, which might or might not involve clean-up, is called for. The difference between the concentration at the target and the maximum allowable concentration can be expressed as a safety factor. Traditionally these safety factors are huge reflecting our fear of contamination and our uncertainty of understanding the system involved.

Implementation and consequences.

Systematic and quantitative application of the source-path-target methodology makes it possible to translate the environmental problem at the target to an emission characteristic at the source. In turn this makes it possible to approach the soil remediation problem in terms of manipulating the emission characteristics at the source to achieve a desired risk level at the target.

In order to do this, the following steps are necessary;

1. List all the possible source-path-target combinations including the irrational and non-quantifiable ones.
2. Construct a data-set based on available information, common sense, similar sites and incidents or just plain guesswork.
3. Use the data set and an appropriate risk analysis model to assess the impact of the source at the target. At this point many if not most of the potential source– path–target linkages will be

eliminated leaving only realistic combinations over.

4. Conduct a sensitivity analysis on the remaining linkages to identify those where the evaluation is sensitive to parameters that are poorly known. (the "phase 1 sensitivity analysis")
5. Carry out a cost-benefit analysis between the consequences of not knowing a parameter to a higher level of certainty against the cost of re-evaluating the parameter. ("Need to know" principle).
6. Determine the "need to know" parameters and repeat the steps to this point until there is sufficient confidence in the parameter database.
7. Evaluate the source emission characteristics that will just reduce the impact at the target to below those maximally allowed.
8. Design and cost a remedial management scheme that will achieve such an emission characteristic.
9. Evaluate the sensitivity of the impact at target to errors or failures in the design and/or implementation of the management scheme. (the "phase 2 sensitivity analysis")
10. If there is a significant chance of unacceptable failure, go on to perform a cost benefit analysis on reducing that chance. (The "discrimination analysis").
11. Converge, by more than one cycle if necessary, to the final optimum management strategy.
12. It is very advantageous to formalize the above process so that it can be easily integrated into a modern IT based environment and an adequate quality system.

It is important to address the correct problem with the methodology. Many sites contain multiple sources and it is worthwhile considering whether these should be treated together or separately in the analysis.

ISSUES ASSOCIATED WITH A RISK BASED METHODOLOGY

Unfortunately every methodology has its' own limitations and failings. For this methodology there are several issues involved and they can be usefully divided into three categories.

The first category relates to the technology of Risk Analysis itself. There are three major issues

- There is the issue of methodology. Is the methodology secure? What risks are being taken as important? What levels of risk are acceptable, and who determines this and how?
- The second issue has already been touched upon and is related to the inadequacy of the relevant databases. Toxicology is notoriously weak, especially for chemical cocktails. Models of the future characteristics of the emission curve, the failure mechanisms and the migration along the multiple pathways are all open to question and few if any have been verified for the time-scales involved. An especially difficult area is the incorporation of these uncertainties into a statistical assessment of the design performance. Failure to cater for this adequately can lead to massive over-design of the management strategy as large safety factors are built in.
- A third set of technology issues is thrown up by the difficulties caused by the uncertainties in each of the elements that go to make up the problem.

These issues will become clearer as more applications are based on the methodology but at this point in time great care and skill is required to avoid discrediting it. The effect of this can be seen by the massive safety factors that have to be used to meet most stringent legislation such as the old Dutch A, B, C values.

The second category relates to the perception of the rest of the public to such a design proposal, or indeed to contaminated site management in general (as distinct from total contaminant removal).

- The most serious issue here is the generally very poor credibility standing enjoyed by engineers and scientists proposing these measures.
- A difficult issue is the lack of experience of many in the community in dealing with the concepts and jargon of risk analysis. This is compounded by the fact that there are rarely clear-cut black and white answers to simple questions such as "is it safe? ". In fact the problem may be deeper and the very fact of having to confront risk may trigger irrational responses throughout the community.

These issues can only be dealt with by clear and often bespoke communication procedures to ensure that everybody knows what is really going on and can satisfy themselves that their concerns are being treated seriously and adequately. In fact the application of this methodology almost always goes ahead in parallel to a carefully crafted communication program.

A final category involves that of the engineers themselves.

- The first issue here is one of professional skill. Moving away from traditional “look it up in the standard” approaches to a Risk Based approach puts a heavy burden on the skill base of the engineer responsible for the design. This is especially so given the wide range of diverse disciplines involved in the essential core of the process.
- The second issue follows on from the first. When is such an engineer “qualified” and by whom? This is also related to a whole range of professional concerns such as, what is professional liability position of the engineer at the moment if such methodologies were to be applied and who exactly is taking the risk?

These issues require addressing adequately in order to gain the very considerable advantages associated with this methodology. These advantages are a very significant cost reduction for the site management compared to a conventional approach.

THE ADVANTAGES OF USING THE RISK ANALYSIS METHODOLOGY

Notwithstanding the difficulties to be faced in applying the methodology it has already proved successful. The main successes have come from avoiding the prohibitively expensive need for excavation and removal of contaminated soils. On the one hand the costs are much reduced for the same local risk mitigation but it is also often the case that the risks associated with the removal of the contaminated soil and its disposal elsewhere are in fact greater on aggregate that leaving it in place.

A second advantage of the risk-based methodologies is that it makes explicit the residual risks in a contaminated land management strategy. This helps a little way to rationalising the risk perception aspects of the problem that can dominate the public debate on any strategy.

A third advantage is that of the possibility of innovative remedial action designs specifically tailored to controlled emission release based on risk assessments of that release. As an example, recent work at GeoDelft has been considering the relative costs of achieving a satisfactory risk level using new and older techniques. The results show that the new techniques of partial containment behind chemically active barriers with controlled but safe emission levels cost only 2% of the cost of restoring the site to multi-functionality and but 4% of the excavation strategy.

RISK BASED CONTAMINANT MANAGEMENT STRATEGIES – THE FUTURE.

There are several obstacles to be overcome before the risk-based methodology is proven and accepted. None the less the obvious economic advantages of the results of using the methodology make it inevitable that this will take place.

These advantages are gained by only managing the risks that are significant and relevant, by developing site specific criteria for action rather than generic ones and by providing the tools for accurate and credible communication to the problems’ stakeholders.

The future will see a move towards standardisation of methodologies and tools, a drastic reduction in the cost of the analysis process itself, a certification of the consultants working in that area and a recognition by all concerned of the need for a robust quality standard based on transparent and agreed protocols.