DEVELOPMENTS IN THE CHARACTERISATION OF CONSTRUCTION MATERIALS AND WASTE MATERIALS FOR ENVIRONMENTAL IMPACT ASSESSMENT PURPOSES

With special emphasis on beneficial use of coal fly ash

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MAIN CONCERNS IN RELATION TO LEACHING TEST USE AND DEVELOPMENT

- Too many leaching tests addressing the same question
- Too many ways of data representation
- Too limited relation of test conditions with the actual problem (TCLP used far outside its scope of development, unsuitable for evaluation of construction materials)
- Too limited use and relevance of the vast amount of leaching test data generated annually in the industry and research (missing parameters)
- Key information relevant to the outcome and possible interpretation of a leaching test often not reported (pH, EC, Eh, DOC)

STRONG NEED FOR HARMONISATION OF LEACHING TEST METHODS AND DATA EVALUATION!
A robust and scientifically sound, while practical, framework for characterisation of environmental behaviour of soils, sludges, sediments, wastes and constructions materials in a range of applications and exposure scenarios is in development in EU and US.

The framework is a tiered approach, allowing the user to select the level of testing and evaluation required based on the degree of conservatism needed, prior information available, and balancing costs of testing against benefits from more detailed information.

ROLE OF CHARACTERISATION LEACHING TESTS IN ENVIRONMENTAL JUDGEMENT

Judgement of the application of materials

Limit values
Relation lab-practice (Scenarios)
Modelling

Characterisation leaching tests
(identification of mechanisms and processes)

Assessibility of data: data base/expert system LeachXS

Quality control of products
Efficient measurements
Precision measurement data
Compliance leaching tests

Product improvement
Product modification
Measurement for verification

Development of criteria for regulation

Regulation
SCENARIO APPROACH IN JUDGING IMPACT

Problem definition and test selection

Lab, lysimeter, field data collection, data management, data formatting, storage and retrieval

Management Scenario Description - configuration, design specifications, infiltration, climate

Physical, chemical, biological properties

pH, L/S & time dependence - redox, DOC, EC, ANC

Release with time

Granular
Percolation related

Monolithic
Surface area

Source term description

Impact evaluation subsoil and groundwater

Judgement and decision making
QC; Regulatory aspects
Treatment, Disposal, Utilization, Remediation endpoints, long-term stewardship requirements

APPLICABLE TO:
CONSTRUCTION MATERIALS,
SOIL, SLUDGE,
SEDIMENT,
WASTE,
MINING WASTE,
PRESERVED WOOD,
AIR DUST

Data integration between fields and tests, modeling and verification against field data

Expert system/database

EN 12920
STANDARDS ARE NEEDED FOR EXISTING OR UPCOMING EU REGULATIONS

- Sewage Sludge Directive 86/278/EEC
- Landfill Directive 1999/31/EC
- Mining waste Directive
- Construction Products Directive (ER3)
- Water Framework Directive
- Groundwater Directive
- .............
BASIC CHARACTERISATION TESTS

GRANULAR MATERIALS

pH DEPENDENCE TEST : BATCH MODE ANC prEn 14429 or COMPUTER CONTROLLED

MONOLITHIC MATERIALS

pH DEPENDENCE TEST : BATCH MODE ANC prEn 14429 or COMPUTER CONTROLLED

PERCOLATION LEACHING TEST (PrEN 14405)

TANK LEACH TEST (MONOLITH) and COMPACTED GRANULAR LEACH TEST.

Chemical speciation aspects

Time dependent aspects of release
RELEVANT pH DOMAINS FOR DIFFERENT FIELDS

- MATRIX MINERALOGY
- CHANGES DUE TO REMINERALIZATION
- POTENTIALLY LEACHABLE

Leached quantity (mg/kg)

pH

- WASTE SLUDGE
- SOLUTION CHEMISTRY
- WOOD
- NATURAL SOIL
- COMPOST
- SEDIMENT
- CONTAMINATED SOIL
- STABILIZED WASTE AND CONCRETE

ACTUAL LEACHING (Metal)
COMPARABILITY OF TESTS AND TEST CONDITIONS RELATED TO DIFFERENT EXPOSURE CONDITIONS

Relevant pH domains for assessing different questions in relation to different types of impact.

Heavily Sewage Sludge Amended Soil

Leached at L/S=10 (mg/kg)
LeachXS Structure

Materials (Leaching data, Composition, Physical characteristics)

Scenarios (e.g., fill characteristics, geometry, infiltration, hydrology)

Regulatory (Regulatory thresholds and criteria from different jurisdictions)

Materials Leaching Database

Scenario Database

Regulatory Database

Thermodynamic Databases

LeachXS (Materials and Scenarios Evaluation)

Excel Spreadsheets (Data, Figures)

Reports (Figures, Tables, Scenario and Material Descriptions)

Orchestra (Geochemical Speciation and Reactive Transport Simulator)

Other Models (Source Term and Parameters for Fate, Transport, and Risk Models)
Comparison of Coal fly ash data with regulatory criteria - DUTCH Building Materials Decree (BMD)

For granular materials info obtained from pH dependence and percolation is complementary. In this case, Zn proves not to be critical under any normal exposure condition.
COMPARISON BETWEEN LAB, LYSIMETER AND FIELD DATA

Percolation test equipment (0.0005 m³)

Lysimeters (1.5 m³), NL

Pilot Nauerna (12,000 m³), NL
INDIVIDUAL WASTES VERSUS INTEGRAL WASTE MIX

- **Ba**
- **Cu**
- **DOC**
- **Ni**
- **Mg**
- **Zn**

Individual wastes test data
INDIVIDUAL WASTES VERSUS INTEGRAL WASTE MIX

A waste mixture behaves quite systematic

Integral waste mix enriched with organic rich waste

Individual wastes

Ba

Cu

DOC

Ni

Mg

Zn
Consistent behaviour between different scales of testing. Predominantly inorganic waste concept meets inert landfill criteria for Pb (and others, except Cl and SO4)
Increased organic matter level leads to increased PAH release
Batch test leads to higher release than a percolation test.
PAH behaviour very consistent at different scales of testing.
Largely same approach in testing and judgement possible as for inorganics.
Solubility control and wash-out offer possibilities to predict release with reasonable certainty.

Mobile non-interacting constituents (e.g. Cl) can provide insight in role of preferential flow aspects at different scales of testing.
## Calculated Cumulative Wash-out in % Relative to the Column Experiment at Lab Scale
(Upflow, same L/S condition)

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Na %</th>
<th>K %</th>
<th>Cl %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nauerna Pilot</td>
<td>46</td>
<td>36</td>
<td>28</td>
</tr>
<tr>
<td>LYSIMETER</td>
<td>23</td>
<td>16</td>
<td>22</td>
</tr>
<tr>
<td>LYS + Shredder and Sludge</td>
<td>33</td>
<td>23</td>
<td>21</td>
</tr>
</tbody>
</table>

Rather consistent wash-out behaviour at the lysimeter and field scale. After correction for the lab test (85 % leached in upflow) preferential flow leaves 70 - 80 % of the material unaffected.
1. pH dependence leaching test on samples for chemical speciation purposes (monolithic materials after size reduction)

2. measurement of release from granular material (column) or monolithic specimen (type of tank test)

3. speciation modelling using LeachXS a database-coupled version of the modelling environment ORCHESTRA to identify relevant mineral phases (SI-indices)

4. refined prediction of leaching behaviour in a pH dependence test based on the selected minerals (from SI units) and other relevant phases (e.g. Fe, clay, DOC, POM)

5. this resulting speciation is used as input for the chemical reaction/transport modelling to describe the release from a column or monolithic specimen

6. model the field scenario with external factors (e.g. carbonation, oxidation) and realistic estimates of infiltration.
### INPUT PARAMETERS FOR GEOCHEMICAL MODELLING OF COAL FLY ASH

**Coal fly ash (anonymous source)**

**Input specification**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solved fraction DOC</td>
<td>0</td>
</tr>
<tr>
<td>Sum of pH and pe</td>
<td>15.00</td>
</tr>
<tr>
<td>L/S</td>
<td>10.000</td>
</tr>
<tr>
<td>Clay (Al oxide)</td>
<td>0.000E+00</td>
</tr>
<tr>
<td>Fe oxide (HFO)</td>
<td>2.000E-03</td>
</tr>
<tr>
<td>Organic matter - solid (SHA)</td>
<td>0.000E+00</td>
</tr>
<tr>
<td>Organic matter - liquid (DHA)</td>
<td>0.000E+00</td>
</tr>
</tbody>
</table>

**DOC concentrations**

<table>
<thead>
<tr>
<th>DOC</th>
<th>pH</th>
<th>kg/l</th>
<th>Polynomial coefficients</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>4.90</td>
<td>0.0E+00 C0 0.0E+00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6.20</td>
<td>0.0E+00 C1 0.0E+00</td>
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<tr>
<td></td>
<td></td>
<td>8.10</td>
<td>0.0E+00 C2 0.0E+00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>8.90</td>
<td>0.0E+00 C3 0.0E+00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10.30</td>
<td>0.0E+00 C4 0.0E+00</td>
</tr>
<tr>
<td></td>
<td></td>
<td>11.70</td>
<td>0.0E+00 C5 0.0E+00</td>
</tr>
</tbody>
</table>

**Reactant concentrations**

<table>
<thead>
<tr>
<th>Reactant</th>
<th>mg/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag+</td>
<td>n.a.</td>
</tr>
<tr>
<td>Al+3</td>
<td>1770.00</td>
</tr>
<tr>
<td>H3AsO4</td>
<td>0.59</td>
</tr>
<tr>
<td>H3BO3</td>
<td>84.13</td>
</tr>
<tr>
<td>Ba+2</td>
<td>10.77</td>
</tr>
<tr>
<td>Br-</td>
<td>n.a.</td>
</tr>
<tr>
<td>Ca+2</td>
<td>20730.00</td>
</tr>
<tr>
<td>Cd+2</td>
<td>n.a.</td>
</tr>
<tr>
<td>Cl-</td>
<td>62.60</td>
</tr>
<tr>
<td>H2CO3</td>
<td>9070.00</td>
</tr>
<tr>
<td>CrO4-2</td>
<td>23.06</td>
</tr>
<tr>
<td>Cu+2</td>
<td>n.a.</td>
</tr>
<tr>
<td>F-</td>
<td>6.50</td>
</tr>
<tr>
<td>Fe+3</td>
<td>0.13</td>
</tr>
<tr>
<td>H+</td>
<td>n.a.</td>
</tr>
<tr>
<td>I-</td>
<td>n.a.</td>
</tr>
<tr>
<td>Li+</td>
<td>n.a.</td>
</tr>
<tr>
<td>Mg+2</td>
<td>1201.00</td>
</tr>
<tr>
<td>Mn+2</td>
<td>7.47</td>
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<tr>
<td>Na+</td>
<td>317.30</td>
</tr>
<tr>
<td>NH4+</td>
<td>n.a.</td>
</tr>
<tr>
<td>Ni+2</td>
<td>n.a.</td>
</tr>
<tr>
<td>NO3-</td>
<td>n.a.</td>
</tr>
<tr>
<td>PO4-3</td>
<td>20.00</td>
</tr>
<tr>
<td>Pb+2</td>
<td>5.00</td>
</tr>
<tr>
<td>SeO4-2</td>
<td>0.90</td>
</tr>
<tr>
<td>SO4-2</td>
<td>1003.73</td>
</tr>
<tr>
<td>Sr+2</td>
<td>n.a.</td>
</tr>
<tr>
<td>Th+4</td>
<td>n.a.</td>
</tr>
<tr>
<td>UO2+</td>
<td>n.a.</td>
</tr>
<tr>
<td>Zn+2</td>
<td>1.67</td>
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</tbody>
</table>

**Selected Minerals**

- Boehmite
- BaCaSO4[50%]
- Ferrihydrite
- Manganite
- Otavite
- Pb[OH]2[C]
- Diaspore
- CaMoO4[c]
- CSH_ECN
- Hilgenstockite
- Calcite
- Wairākite
- monosulfate_EC
- Fluorite
- Brucite
- Strontianite
- Pb2V2O7
- Ba[SCR]O4[77% Portlandite]
- Magnesite
- Willemite
- PbMoO4[c]
One set of conditions for all parameters simultaneously: Element availabilities, selected minerals, Fe-oxide and clay sorption, POM and DOC interaction.
Prediction and partitioning in the pH dependent leaching test for Cr and Mo in Coal Fly Ash

Relevance of mineral formation, interaction with Fe phases for retention of oxyanions in coal fly ash (ettringite type phases)
Geochemical modelling of Cement Mortar

All elements simultaneously, including all relevant minerals, Fe oxide sorption and solid solutions such as ettringite substitution.
Prediction of Na concentration in the tank test NEN 7345

Cumulative release of Na

Cumulative emission of K

Liquid renewal cycles in the test

pH not yet good for this specimen in early cycles.

Transport for “inert” species apparently well described
Transport for a solubility controlled major element like Ca quite well described.
Depending on the modeled groundwater quality at the POC (which might be chosen anywhere), limits to the release of the construction products might be adjusted such that the product eventually complies with the groundwater limit value at the POC.
Modeling release from construction material by percolation
Construction Products Directive - Essential Requirement No3 on Health & Environment - Release of Dangerous substances

Mandate accepted by Standing Committee Construction on October 26 2004

Mandate has been issued to CEN

Work plan for BT176 is now being drafted

Start of the horizontal standardisation work on:
- Sampling
- Indoor air
- Impact to soil and groundwater
  (ECRI CEM/ECO Serve Cluster 1 input)
CONFORMITY ASSESSMENT IN THE CPD

Assessment of construction products in view of CPD/ER No. 3

WT-Products

Non WT-Products

WFT-Products

Ft-Products

Conformity Evaluation acc. to prescribed Conformity System

Conventionally Approved Materials

WT = Without testing
WFT = Without further testing
FT = Further testing

CE-marking

Main aim: avoid unnecessary testing and focus on the key issues
LIFE CYCLE ISSUES IN SOIL & GROUND AND SURFACE WATER IMPACT

• Service life: different exposure scenarios
  • Constructions in water
  • Constructions on land
  • Drinking water pipes/basins

• Recycling stage (bound): same as service life

• Reuse stage (unbound): different exposure scenarios
  • Road base/embankment
  • Structural fill (dikes, soundbarriers)

• “End of life”: Landfill scenarios
  • Inert landfill
  • Non-hazardous waste landfill

Characterisation testing provides information on the abovementioned life-cycle stages
TEST USE IN RELATION TO THE LIFE CYCLE OF CONSTRUCTION PRODUCTS

Stage 1: Raw material supplies

Stage 2: Manufacture of construction materials and elements

Stage 3: Construction Process

Stage 4: Service Life

Stage 5: Demolition

Primary Raw Materials

Alternative raw materials

Recycling of construction debris

Supply of information on technical and environmental quality: Database/expert system

Characterisation (ITT) of monolith/ granular leaching behaviour

Monolith/granular QC and compliance leaching test

Release into the environment

Energy

Dust, noise emissions

“End of Life”

Characterisation of granular leaching

Granular compliance test

Energy
RELEVANT PAPERS

HOW TO JUDGE RELEASE OF DANGEROUS SUBSTANCES FROM CONSTRUCTION PRODUCTS TO SOIL AND GROUNDWATER

CPD Topic 1 - Soil and groundwater impact

CPD Topic 2 - Hierarchy in testing: Characterisation, initial type testing, further testing and selection of tests in specific stages of material judgement

CPD Topic 3 – Proposal for reference to ER 3 aspects in product standards and in CE marking

J. J. Dijkstra (ECN) H. A. van der Sloot (ECN) G. Spanka (VDZ) G. Thielen (VDZ) ECN-C--05-045

DEVELOPMENT OF HORIZONTALLY STANDARDIZED LEACHING TESTS FOR CONSTRUCTION MATERIALS: A MATERIAL BASED OR RELEASE BASED APPROACH?

Identical leaching mechanisms for different materials

H.A. van der Sloot and J.J. Dijkstra ECN-C--04-060

Presentations at Workshop by EAWAG in Meiringen May 2005
CONCLUSIONS

Characterisation leaching tests, such as developed in CEN/TC 292/WG 6, provide the source term for modelling long term release from coal fly ash, wastes, contaminated sites and construction materials.

A limited number of well selected leaching tests can provide the crucial answers needed to assess impact and evaluate sustainability.

Comparison between laboratory scale leaching tests, lysimeter experiments and full scale verification is needed to be able to develop sustainable utilisation and disposal concepts. First steps are very promising.

Understanding the role of mineral formation, sorption onto clay, Fe-oxides and “residual” organic matter (non-degradable) in relation to mobilisation in DOC-bound form is of importance to minimize porewater concentrations in the material matrix.
CONCLUSIONS

Reduction of DOC levels in leachate by proper acceptance criteria will lead to significantly decreasing concentrations of inorganic and organic contaminants in the leachate from any type of material. This does not imply banning organic matter as residual, non-degradable organic matter is quite acceptable (provides binding).

Transport modeling in its simplest form already provides good prediction potential for long term release for a range of constituents.

The leaching of organic contaminants and the judgement of impact can be addressed in a very similar manner as for inorganic contaminants.

An expert system coupled with a database of characterisation and field data will provide a very useful tool for construction and waste treatment industry, consultants and regulators.
CONCLUSIONS

With the modelling capabilities now presented sequential chemical extraction (SCE) is outdated.

The present model capabilities replace the widely used Kd concept by a more detailed description of the actual chemistry. The main aspect being the mutual interaction between constituents which is ignored in a Kd approach.

Multi element and multi parameter modelling allows identification of missing or inadequate stability data for minerals or sorption parameters.
CONCLUSIONS

The methodology and release controlling processes illustrated here for coal fly ash and cement with coal fly ash are equally relevant for contaminated soil, amended soil, mining waste and alternative materials used in construction.

The emphasis on specific release controlling factors may be stronger for one material than for another, but all aspects may play a role as different parameters are affected by different release controls (e.g. Cu is very sensitive to even very small amounts of DOC).

Understanding leaching behaviour should be exploited more extensively as it holds the key to treat materials in such a way that long term solutions are achieved rather than reaching temporary gains.
FUTURE

Challenges: proper description of pH and DOC generation from various materials for prediction of inorganic and organic contaminant release from construction materials, soil and disposed waste.

Mining of old data sources containing valuable leaching data currently inaccessible because of poor data format, non-electronic data forms.

Prediction of release behaviour of mixture of materials

Developing generic scenario descriptions based on fundamental leaching information for easy application by end-users in construction applications, contaminated soil site evaluation, treatment of waste and development of sustainable landfill concepts.
ACKNOWLEDGEMENT

In this work information has been presented from Sustainable Landfill project that is an initiative of the Dutch Cooperation of Waste Processing Industries, Afvalzorg, Essent, and VBM.

Part of the work presented relates to activities in ECO Serve (EU project) as well as work for the Dutch Ministry of Environment.
INFORMATION ON LEACHING AVAILABLE AT:

LEACHING BACKGROUND
www.leaching.net (Wascon 2003 workshop)

CONSTRUCTION PRODUCTS DIRECTIVE

LEACHING IN PROJECT HORIZONTAL
www.ecn.nl/horizontal

GRACOS EU project on contaminated soil and sediment
www.uni-tuebingen.de/gracos
Additional Background Slides
pH DEPENDENCE TEST TO ASSESS SENSITIVITY TO CHANGES IN pH, E_H AND TEMPERATURE

(PrEn14429 Batch mode test)
ADVANTAGES OF pH DEPENDENCE TEST

- Identification of sensitivity of leaching to small pH changes
- Provides information on pH conditions imposed by external influences
- Basis for comparison of international leaching tests
- Basis for geochemical speciation modelling
- Provides acid neutralization capacity information
- Mutual comparison of widely different materials to assess similarities in leaching behaviour
- Recognition of factors controlling release
- For non-interacting species possible to assess subsampling error

Applicable to almost any material
PERCOLATION TEST TO ASSESS LONG TERM RELEASE FOR GRANULAR MATERIALS PrEn14405

Liquid to solid ratio (L/S) related to a time scale by infiltration rate, density and height of application.

TEST CONDITIONS:
Pre-equilibration after saturation for more than 48 hrs
Up-flow
L/S range 0.1 - 10 (100 - 1000 yrs)
Test data in mg/l or mg/kg cumulative
ADVANTAGES OF PERCOLATION TEST

- Identification of solubility control versus wash out
- Indication of pore water concentrations relevant to field leachate from low L/S data
- Local equilibrium established quite rapidly
- Basis for geochemical speciation modelling
- Allows comparison with lysimeter and field data provided L/S value can be obtained from such measurements
- Projection towards long term behaviour possible
  Solubility controlled release
  Wash-out of non-interacting species

Applicable to many materials. Limited or not applicable to clayey soils and sediments (low permeability).
TANK LEACH TEST OR COMPACTED GRANULAR LEACH TEST (CGLT) FOR MONOLITHIC MATERIALS (modified)

**TEST CONDITIONS:**
First step: pre-equilibration for 48 hrs at liquid to volume ratio: 5
Second step: leaching at low L/V ratio (1) for 24 hrs
Then contact times: 2, 4, 8, 16, 32 and 64 days
Leachant: demineralised water (own pH)

Expression of results in mg/m² (cumulative) against time
EXPERIMENTAL SET-UP

CGLT = Compacted Granular Leach Test
ADVANTAGES OF TANK LEACHING TEST OR COMPACTED GRANULAR LEACH TEST

- Relevant for materials with monolithic character (durable materials) or materials behaving as monolith (low permeability soil and sediments)
- Identification of solubility control versus dynamic leaching possible
- Isolation of surface wash-off effects
- Quantification of intrinsic release parameters
- Basis for reactive/transport modelling
- Projection towards long term behaviour possible

Applicable to sediments, clayey soils, stabilised materials and construction materials produced