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THE GERMAN RESEARCH NETWORK "SEEPAGE WATER PROGNOSIS"

RAINER FREYAS, SIEGFRIED H. EBERLE & SUSANNE HUCKELE(*)

(*) Heinrich-Sontheimer-Laboratorium, Technologiezentrum Wasser, Karlsruher Straße 84, 76139 Karlsruhe, Germany

INTRODUCTION

THE NETWORK'S CONCEPTION

As a consequence of recent German environmental legislation, the German Life Cycle Management Act (1994) gives priority to waste reuse. The German economy produces more than 360 million tons of refuse per year (Federal Statistical Office Germany, 2005). While part of this amount is produced by households and industry, the major share consists of demolition waste, excavated soil, harbour sediments etc. Most of these bulk materials can be used for road construction, landscape design, recultivation purposes etc. due to their properties. However, pollutants contained in these materials pose a threat to the subsurface environment as soon as seepage water carries them from upper layers of the soil down to the aquifer. The Federal Soil Protection Ordinance (1999) defines the transition zone between the unsaturated and saturated layer as the point of compliance for the concentration threshold. The "seepage water prognosis" is introduced as a new instrument for the evaluation of potential hazards caused by existing contaminations as well as waste reuse deposits (BANNICK *et alii*, 2001).

When the Federal Soil Protection Ordinance came into operation, the implementation of the seepage water prognosis was regarded unsatisfactory. In particular, material tests with a low reproducibility were vulnerable from a juridical point of view. As a consequence, an amendment of the ordinance is planned for the year 2007, in order to give accurate instructions to approving institutions. Authorities concerned with the protection of the subsurface environment, as well as consultants in this field and the recycling industry, expressed their need for easy-to-handle but reliable techniques. The German Federal Ministry of Education and Research decided to meet these demands by starting an interdisciplinary research project, dividing the seepage water prognosis into two steps. In the first step, pollutant mobilisation from contaminated materials due to infiltrating rainwater is investigated. Parameter values, particularly substance concentrations, occurring at the bottom of a contaminated layer are then called "source term" or "source strength". In the second step, the transport of pollutants from their origin to the point of compliance is regarded. Hence, the two main issues of the research network conform to a characteristic material reuse scenario with a contaminated layer covering unsaturated subsoil.

The call for proposals was published in the Federal Gazette (FEDERAL MINISTRY OF EDUCATION AND RESEARCH, 1999), attracting remarkable attention among experts and leading to nearly one hundred applications. The network started with 41 selected projects run by scientists from different disciplines such as hydrogeology, geochemistry, waste management, physics and mathematics. Besides many university departments, engineering offices were also taking part. While most of these initial projects were finished from 2004 on, a few of them have been prolonged. Further projects have been added in order to focus on particular aspects. In 2006, the network comprises 57 projects, which are now partially financed by the Federal Ministry for the Environment. The total budget is 11 million Euros.

ORGANISATIONAL STRUCTURE

In regard to the scale of the network, a distinct organisational structure had to be established (figure 1).

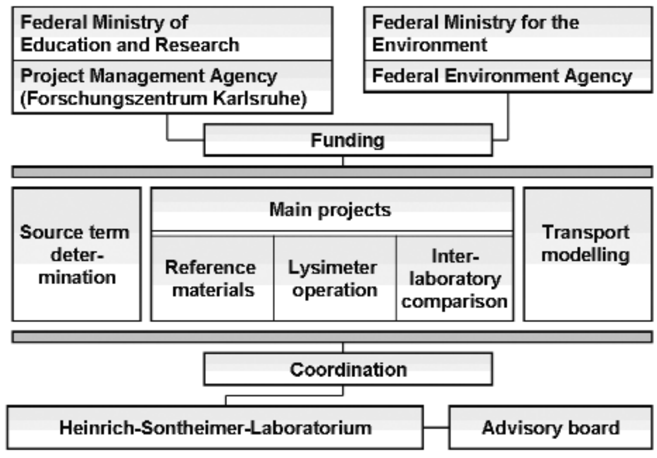

Fig. 1 - Organisational structure of the project network

The two federal ministries fund the network through their executive agencies, the Project Management Agency for Water Technology and the Federal Environmental Agency. In terms of scientific guidance and advice, the Heinrich-Sontheimer-Laboratorium (HSL) in Karlsruhe coordinates the network in consultation with these agencies. Particularly, network conferences that secure information exchange among workgroups are organised and moderated by the HSL. An advisory board was established in order to make sure that scientific results are converted into feasible suggestions for the scheduled amendment. Its members are delegates from the industry, different environmental authorities, state agencies, universities and other scientific institutions.

Two project groups of about 25 projects each were created to work on the main issues. The project group engaged in source term determination was separated into projects dealing either with static batch tests or dynamic leaching procedures. The other group applied experimental work as well as computer simulations for investigations on transport processes.

Three main projects form the core of the research programme. The Federal Institute for Materials Research and Testing (BAM) was assigned the production of reference materials, in order to obtain comparable results from the entire network. Municipal refuse incineration bottom ash and demolition waste containing gypsum and asphalt were selected as characteristic mineral waste materials. Soil from a former gasworks site as a third reference material represented existing contaminations. For various experimental activities within the research network, an amount of about 6 tons of each reference material had to be homogenised and distributed to the participating institutions (BERGER *et alii*, 2005).

Basic knowledge about the interaction of source strength and contaminant transport is obtained in three outdoor lysimeter settings. These are run by the National Research Centre for Environment and Health (GSF) near Munich, the North Rhine-Westphalia State Environment Agency (LUA NRW) and the Research Centre in Jülich (FZJ) near Cologne. The so-called transport lysimeters consist of a natural soil column (sand or monolithic loess) covered by reference material. The entire arrangement is 2.5-3 m high; the diameter ranges from 1-1.5 m (KLOTZ, 2006). Pollutant concentrations are determined in pore water directly underneath the covering layer, as well as in the outlet at the bottom of the soil column. This permits the comparison of source terms and concentrations after passage through the subsoil.

Reproducibility of material tests was the topic of an inter-laboratory comparison, which was supervised by the BAM and dealt with static as well as dynamic tests (BERGER *et alii*, 2006). Experiences from previous source strength projects were taken into account to work on precise manuals for the participating institutions. Once more, the use of the standardised reference materials was fundamental for the validation of these instructions, and their acceptance in a future amendment of the Federal Soil Protection Ordinance.

KEY-WORDS: *seepage water, point of compliance, source term, contaminant transport, reference material, lysimeter, transport modelling*

CURRENT STATE OF THE PROJECT

The analytical schedule for the project network comprises inorganic as well as organic pollutant species, primarily heavy metals and PAHs. It is completed by ions that are essential for water analysis, e.g. sulphate and chloride, as well as parameters such as conductivity and pH value. Many workgroups also investigated supplementary parameters, but due to the scope of the entire network, evaluation is mainly focused on the parameters mentioned above. In addition to the reference materials, several participants integrated other slightly contaminated refuse materials into their experimental programme.

In view of a sustainable protection of soil and groundwater, the evaluation of potential hazards aims at a long term perspective for the release of dangerous substances, thus focusing on the progress of concentrations with time. However, the project network adopted a modified time scale, using the amount of water that has seeped through a defined quantity of contaminated material, by a certain time. The quotient of the amount of water and the quantity of contaminated material is called the “liquid/solid ratio” (L/S ratio). This takes into account different dimensions of experimental settings, ranging from percolation columns and batch tests using less than 100 g of material, to outdoor lysimeters filled with nearly 2 tons.

Due to the characteristic release curves starting out on high concentrations rapidly decreasing to a levelled tailing, a logarithmic scale is chosen for the ordinate in many cases. Experimental results from demolition waste are shown as representative examples in the following synopsis. This reference material contains both organic and inorganic pollutants, moreover its properties have been found to be similar to those of incineration ash.

Results from transport lysimeters

In figure 2, a simplified sketch of an outdoor transport lysimeter is presented together with essential results obtained in three years of operation. The scenario-based lysimeter set-up allows observation of its component’s behaviour under most natural conditions. Precipitations infiltrate the column through a covering layer of inert material (quartz gravel) in order to achieve uniform permeation into the reference material. At the bottom of the contaminated layer, suction cups are mounted, which extract pore water by vacuum for the determination of the source term. Additional suction cups at different depths of the transport zone can be added for the observation of concentration fronts progressing to the bottom (PÜTZ *et alii*, 2004). An inert sand filter with graduated grain sizes helps to avoid negative pressure that is otherwise generated by a capillary fringe. Trickling off the sand filter, the seepage water is collected by a tank at the bottom before it is analysed.

Meteorological data is recorded to determine the amount of infiltrating water and to compare it to the amount measured at the outlet, thus acquiring information on the flux regime inside the lysimeter. Additionally, lysimeters are weighable for the same purpose. Any cover of vegetation on top of the lysimeter is continuously removed. Due to a lower evaporation, the determined percolation has been three times higher than German average groundwater regeneration. Hence, one year’s results from the lysimeters forecast observations that would have been made during three years in nature.

On the left side, the progress of the copper source strength can be compared to the outlet concentration. The diagram represents an instructive example for characteristic release curves of many substances belonging to the analytical schedule. The source strength declines below the test value that has been defined by the Federal Soil Protection Ordinance but rises twice, highlighting two summer periods. Concentrations at the outlet are more levelled and differ about ten times from those observed underneath the contaminated layer. Due to the outdoor conditions with their wide range of influencing factors, concentrations vary broadly. Several projects within the network specifically dedicated themselves to investigate side effects such as preferential flow, displacement of particles, biological degradation or effects of heavy precipitations.

In contrast to inorganic pollutants, the release of most PAHs (not represented in this figure) shows little progress as long as biological degradation is prevented, i.e. due to the high alkalinity of demolition waste containing different calcium phases. However, the difference between source term concentrations and those observed in the outlet is even more significant than for heavy metals. It results from both retardation effects and from microbiological activity in the transport layer materials that were not sterilised but feature a high pH buffer capacity.

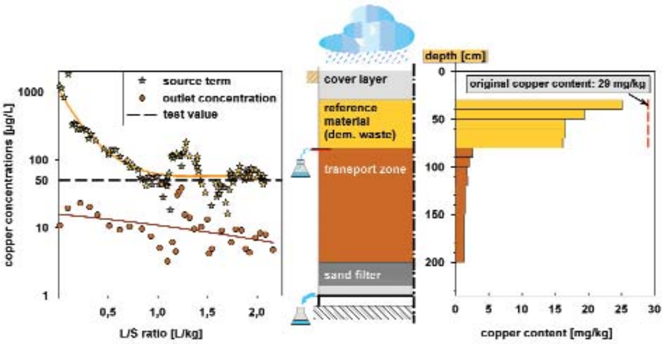

Fig. 2 - Simplified sketch of a transport lysimeter (middle), progress of copper concentrations with time measured in seepage water below the contaminated layer and after passage of the transport zone (left), remaining copper contents after three years of operation as a function of depth (right)
Data sources: KLOTZ D., GSF; BERGER W. & KALBE U., BAM

At the end of the operating time, the lysimeters that had been run by the GSF were dismantled gradually in order to determine remaining pollutant contents. In this manner, values for the total amount of each pollutant before and after the experiment were compiled. On the right side of figure 2, copper contents after three years of operation are presented as a function of depth. Less than 1% of the initial copper content of 29 mg/kg of demolition waste was detected in the outlet. One third of the contaminant remains adsorbed in the transport matrix while two thirds have not left the reference material. Similar observations have been made for PAHs (not represented here) if biological degradation was excluded. This confirms conclusions drawn from many characteristic release curves that a long term emission of contaminants is to be expected even if it takes place at low level.

Laboratory percolation tests

Outdoor lysimeter experiments are the most realistic methods to monitor contamination release, but their application is both too expensive and too time-consuming for approval procedures of contaminated materials. Dynamic leaching tests in laboratory columns have been found to be a good compromise between in-situ source term determination and the requirements of bench-scale work. Due to controlled conditions, these experiments provide release curves with small deviations. However, low concentration levels observed in long term experiments may cause greater variance if the analysis of the samples is not sensitive enough.

The project network studied effects of different experimental set-ups that are available in the participating institutions. Usual column sizes range from 10 – 100 cm of length and 5 – 15 cm in diameter. In view of practical advantages, most column experiments are run in a saturated state with an upward water flow. In order to limit the duration of the tests, chosen flow rates are between 100 and more than 1000 times higher compared to the average groundwater regeneration in Germany. Column tubes are filled according to national and international standards (DIN V 19736, 1998; ODENSASS & SCHROERS, 2000; prCEN/TS 14405, 2003), which propose layer-by-layer emplacement of the tested material that can be accompanied by simultaneous water saturation. Usually, the columns are operated at ambient room temperature, but some projects also investigated effects of temperatures that are closer to those observed in the subsoil. Different eluent compositions are employed that alter e.g. pH values, dissolved organic carbon, ionic strength or redox potential. Most of these characteristic water parameters play a minor role as they are influenced by the permeated matrix to a large extent. The same effect has been surveyed in transport lysimeters where e.g. pH values are controlled by the rather alkaline materials although they are actually exposed to acidic rainwater. The poisoning of eluent water allows the understanding of how micro-organisms perform in view of the biological degradation of PAHs.

Figure 3 shows characteristic release curves for heavy metals (left) and PAHs (right) in comparison to the test values defined in the Federal Soil Protection Ordinance. High L/S ratios were achieved in both experiments, representing a time of several hundreds of years in nature.

While nickel concentrations decrease after a relatively short time, the emission of chromium and copper lasts until the end of the experiment, even if all three heavy metal concentrations fall quickly below the test value. PAHs are released at much lower concentration levels with different desorption kinetics (GRATHWOHL *et alii*, 2005). Naphthalene displays a release curve with a small gradient, while the pyrene concentration remains constant during the entire operating time and does not decline to the test value for the total of 15 PAHs (including pyrene). In many cases, the characteristic behaviour of contaminants observed in outdoor lysimeters was repeated in laboratory results. Substances of the same chemical category once again showed similar behaviour.

The results of dynamic leaching tests within the individual projects encouraged an inter-laboratory comparison. Eight institutions from the network voluntarily took part in the programme. Its working instructions benefit from experiences with the standards mentioned above and specify most of the experimental conditions. Again, it was accepted that available laboratory equipment in different sizes was employed. Consequently, flow rates were adjusted to these circumstances to guarantee identical contact time between material and eluent (BERGER *et alii*, 2006). The inter-laboratory comparison proved good repeatability and validates the working instructions as a base for a projected technical specification.

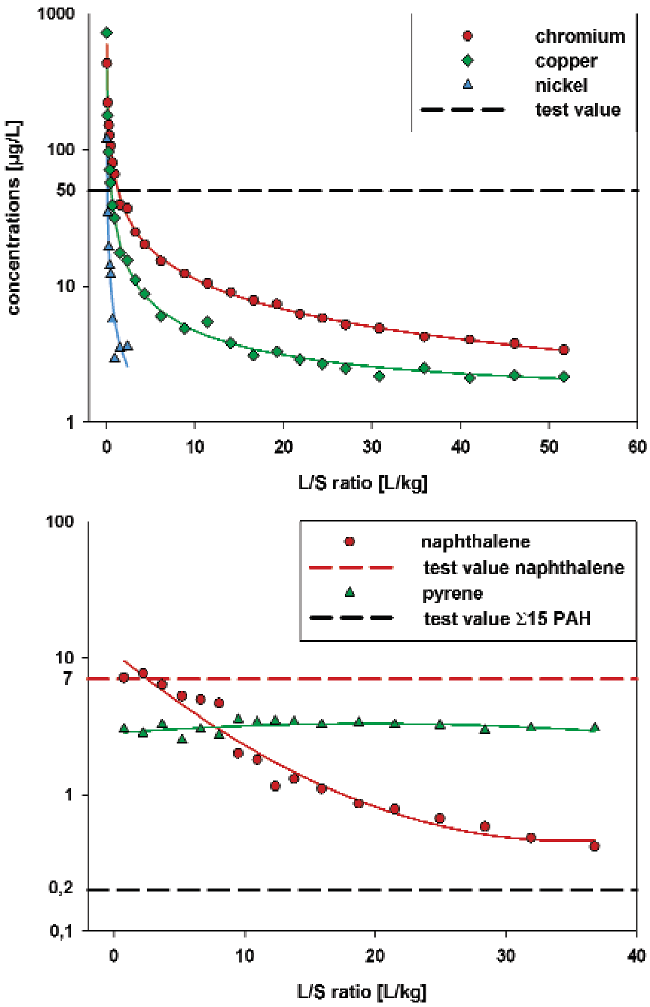

Fig. 3 - Release of contaminants from demolition waste, observed in two different saturated column experiments at room temperature. Left: release of heavy metals (column height 24 cm, flow rate 1.2 m/d). Right: release of PAHs (column height 20 cm, flow rate 0.27 m/d, eluent poisoned with sodium azide).
Data sources: DELAY M., University of Karlsruhe; LANGE F.T., Water Technology Centre, Karlsruhe

RESULTS FROM BATCH EXTRACTION TESTS

Static batch extraction represents the most practical alternative for bench-scale material tests. While few source term projects developed new methods, most of them focused on two procedures. The first is the “S 4” shake test with a L/S ratio of 10:1 that has been part of German standardisation for a long time (DIN 38414 Teil 2, 1984), and its modified form with a L/S ratio of 2:1. The second type is the saturated soil extract at a L/S ratio of about 1:4 according to the Federal Soil Protection Ordinance. It is closer to natural conditions but provides little water for analysis.

Results from the network revealed that batch extraction methods had a low reproducibility. An inter-laboratory comparison of shake tests and saturated soil extract endorsed this conclusion. It was repeated with more precise working instructions, which systematically minimised deviations.

Single static tests cannot depict concentration progression with time. Their results hardly allow secure forecast of long term mobilisation in natural systems. However, they may be suitable for preliminary decisions as to whether a material should undergo further tests on its release potential. A new approach aims at the assessment of a combination of batch tests at different L/S ratios. It takes into account recent knowledge on the mathematical description of contaminant emission. Continuous functions that have been found so far show aberrations that are not larger than deviations observed in the experimental batch tests themselves.

THE NETWORK’S FURTHER RESEARCH ACTIVITIES

Transferring results from bench-scale tests to outdoor conditions has become an essential problem for the seepage water prognosis. In particular, laboratory measurements in a saturated state contrast with observations in the unsaturated subsoil above the point of compliance, according to its definition.

In 2005, the HSL conducted a poll among network participants concerned with transport modelling as a transfer tool. The intention was to find out how the source strength as the main input to computer simulations was implemented in the individual projects. Many transport models still assumed constant emission, despite of network’s results proving most substances to be released at changing concentrations. Discussions during the final network conference in 2005 revealed that functional descriptions of release curves might represent an adequate solution. Based on identical mechanisms, they may be attributed to a fairly small number of characteristic mobilisation patterns. Figure 3 shows two types, a hyperbolic concentration decrease and a constant emission.

In regards to the scheduled amendment of the Federal Soil Protection Ordinance, the HSL decided to form a task force from experts that have already participated in the network. Three of its members are occupied with the derivation of process-based source term functions, mainly using data from completed network projects. For direct comparison of saturated and unsaturated conditions, the fourth project carries out experimental work with two identical sets of columns.

At the moment a central database is being created in order to improve access to the network's results. It is fed from forms which were developed with Excel® and have been distributed to all participants. The database features optional plots to compare different workgroups' measurements and will be part of a future internet presence after completion of the network.

With most of the projects finished, the mission of the HSL has changed to the evaluation of the entire network's outcome. Final reports from the workgroups are collected, moreover contractors have been asked to provide an executive summary to brief the most important conclusions and postulations from their project. The HSL will edit a summary report as an important contribution to the committees concerned with the scheduled amendment of the Federal Soil Protection Ordinance.

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