A UNIFIED APPROACH FOR THE JUDGEMENT OF ENVIRONMENTAL PROPERTIES OF CONSTRUCTION MATERIALS (CEMENT-BASED, ASPHALTIC, UNBOUND AGGREGATES, SOIL) IN DIFFERENT STAGES OF THEIR LIFE CYCLE

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Abstract

In this paper experiences on leaching behaviour of a wide range of construction materials have been combined to provide an integrated evaluation system for judgement of environmental properties for all construction materials (cement-based, asphaltic, unbound aggregates and soil-like materials). In this system, a consistent approach in test use and development is presented that provides a sufficient level of understanding of the long-term risks from a regulatory point of view and recognizes industry's need for quick and straightforward quality control methods. The system is highlighted by means of examples for leaching behaviour of different materials in the same application and the same materials in different applications, leading to different exposure scenarios and possible tests for different stages in the life cycle of materials. The use of characterization tests for the evaluation of use scenarios and the relationship between characterisation and compliance tests are addressed.

Introduction

In recent years the pressure on primary materials for construction has increased. As a consequence more and more alternative materials are used in Europe in different construction applications. Although alternative materials may prove to be suitable technically, the long-term environmental implications of their use are still uncertain. Several construction applications with alternative materials may perform well in the primary application. However, uncertainty exists about potential environmental impacts from subsequent cycles of use (recycling, reuse in other applications and "end-of-life"). This has been caused partly by the lack of proper assessment tools. Several tools are now available (PrEn14429, 2003; PrEN14405, 2003; NEN 7345,1995; Kosson et al, 2002) and have proven their potential suitability to assess different stages of the materials life cycle. In the CPD (Construction Products Directive, 1988, 1993) the recycling, reuse and end-of-life aspect of a construction material use is not covered, which may prove a serious limitation for some materials.

Full characterisation of all alternative materials used in construction will be economically impossible. Indeed, this is not necessary because many materials behave similarly in many situations. Judgement based on leaching is more appropriate to assess long-term impact from alternative materials in construction than evaluations based on total composition. For release of inorganic constituents (major, minor and trace elements) from alternative materials used in construction it is important to identify a few key issues. These relate to:

- the nature of the constituents of concern, which may have very different release behaviour based on their chemistry and the local conditions (i.e., the conditions of use or recycling); and,
- the major release mechanism in alternative materials used in construction, which will be usually percolation dominated for granular materials and dominated by surface or matrix diffusion controlled release for monolithic materials.

These factors indicate the need for combined use of batch equilibrium tests as a function of pH with either percolation based testing protocols or dynamic monolith leach tests for assessing release. For some cases, equilibrium batch testing as a function of pH (Kosson et al, 2002) can be sufficient for simplified evaluation, avoiding the need for dynamic testing (percolation or monolith). When using results from laboratory testing to evaluate potential impacts during the lifecycle of a material, the following conditions should be considered:

- the hydrology of the situation under consideration (i.e., how much infiltration will occur and what are the relevant/preferential flow paths);
- the nature of the source in terms of release controlling parameters (i.e., pH, EC, redox and dissolved organic carbon (DOC));
- the changes in release conditions with time due to processes such as depletion of source material, changes in main release controlling parameters (e.g., resulting from influence of adjacent materials, biological activity, oxygen exclusion) and changes in permeability due to pore clogging;
- the behaviour of released constituents in the subsoil and in groundwater; and,
- the impacted targets to be evaluated (e.g., surficial soil, underlying soil, groundwater, runoff) in the consideration of acceptance or rejection and subsequent requirements for mitigating measures.

Several questions may need to be answered in an evaluation of alternative materials used in construction. Preferably such questions can be answered with the same type of characterization. In ENV 12920 (1999), a number of steps have been identified, which help address the information needed to answer a specific question. In figure 1 a schematic representation is given of an application of alternative materials in construction, where factors influencing release and targets are illustrated. A major distinction can be made between constituent release from the source under the influence of controlling factors that may vary with time, and the transport of contaminants from the boundary of the site to subsoil or groundwater as a potentially impacted target. Modelling is a key aspect in both describing a time-dependent source term, as well as in quantifying the transport of contaminants through soil and groundwater. The latter part of the evaluation can be made site specific by taking into account different flow regimes and retention characteristics of the subsoil or it can be made in a more generic manner using default values with relatively conservative estimates for flow and retention that are representative of a geographic region.

There is an important difference between the source term description and transport behaviour at a chosen site. For determining the acceptability of a specific use, the source term for constituent release derived from leaching tests is important in conjuction with understanding attenuation processes from the point of release to the point of compliance. Finally, a relationship between laboratory and field verification is crucial to be able to lend credibility to the testing and modelling efforts.

In the framework of the Standards, Measurement & Testing Programme (DGXII) of the European Community through the Network for the Harmonisation of Leaching/Extraction Tests (1997) the use of widely different leaching test methods in different areas (e.g., for waste treatment and disposal, incineration residues, fuel combustion residues, soil clean-up and reuse of cleaned soil, sludge treatment, use of compost from different sources, use of alternative raw materials in construction) has led to the conclusion that there are too many leaching tests largely addressing the same question, and, that there are insufficient relationships between the test conditions for most regulatory tests and the actual release under field conditions. This observation calls for more harmonisation of leaching test procedures, along with greater verification of field performance, across different fields.

There is no uniform test method or test procedure that could be applied for all construction materials, since there is a wide range of materials and applications. Therefore, different test methods have to be selected from a robust set of leaching methods. The leaching test chosen should be based on the predominant leaching mechanism, which may be different for monolithic and granular building materials. Generally, the leaching behaviour of building materials needs to be considered during their overall life cycle: from production, via the period of use, until demolition and reuse or disposal. Therefore different scenarios and different leaching tests may be necessary for one material. This information is highly relevant for standards to be developed under the European Construction Products Directive (CPD, 1989). This Directive gives the framework for development of Technical Specifications for construction products. The process of development of this directive and the Technical Specifications are guided by the Standing Committee on the Construction Products Directive (SCC). The SCC has decided that for ‘dangerous substances’ a horizontal approach will be followed to avoid development of inconsistent leaching tests (SSC, 7 November 2000 in Vienna). In the contribution by Eikelboom (Recent developments in European legislation; challenges for recycling and horizontal standardisation of test methods, this conference), these aspects are addressed in detail.

SCENARIO: ALTERNATIVE MATERIALS IN CONSTRUCTION

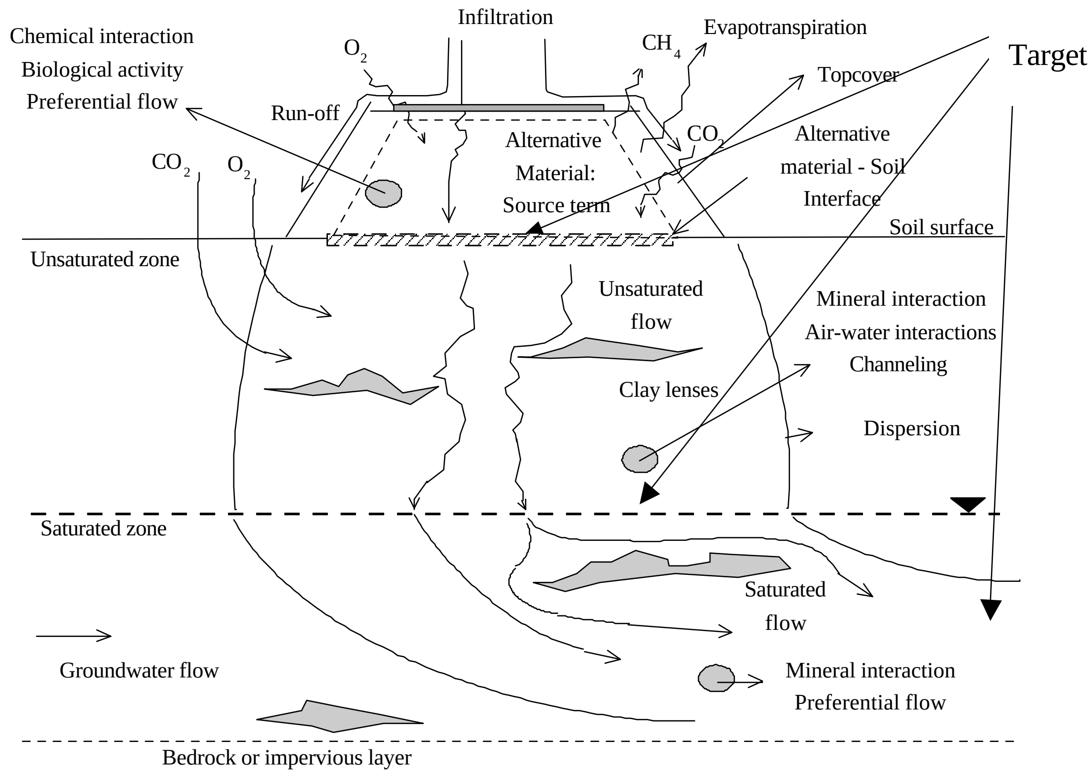

Figure 1. Schematic representation of release from "alternative" construction materials with the possible targets (surficial soil, subsoils, groundwater) for evaluation of environmental impact.

There are many different types of construction materials with widely ranging properties from the perspective of leaching of"regulated"substances. Cement-based materials, bituminous materials, filling materials and alternative construction materials derived from materials formerly considered wastes obviously have widely different properties such as pH, porosity, wetting behaviour and morphology. Besides the widely different nature of the various construction materials, the application scenarios differ also and may range from indoor construction to underwater applications. These differences call for identification of the relevant exposure conditions, because they will lead to different release rate and extent for the same material.

Product families

Below a large number of relevant construction materials are listed for which an evaluation will be needed:

Aggregate used in concrete (bound), road base (unbound), and as structural fill (unbound); cements, building limes and other hydraulic binders; concrete, mortar, grout and related products; gypsum products; masonry and related products; road construction products, fill materials and cover materials such as asphaltic products; drinking water pipes (concrete); coatings and finishes;

roof coverings, gutters, sheet plating; preserved wood; etc. In general, to provide consistency in evaluation amongst the material categories described above, materials can be classified according to the matrix in which they are incorporated (e.g., unincorporated aggregate, Portland cement, asphalt cement), the physical form of the product or material in use (e.g., monolithic, including compacted granular materials, or granular), and the expected water contact regime (e.g., continuously saturated, intermittently saturated, unsaturated). For many cases, the matrix in which the material of concern is incorporated will control the release chemistry. For example, the release behavior of most materials incorporated in Portland cement matrices will be controlled by the Portland cement chemistry and physical properties.

Different scenarios and different leaching tests may be necessary for different materials, but also for one material in different applications (figure 2). In the previous evaluation of harmonisation (Van der Sloot et al, 1997) this scheme was presented illustrating the raw materials, products derived from such raw materials, the different life cycle stages and the possible impacts.

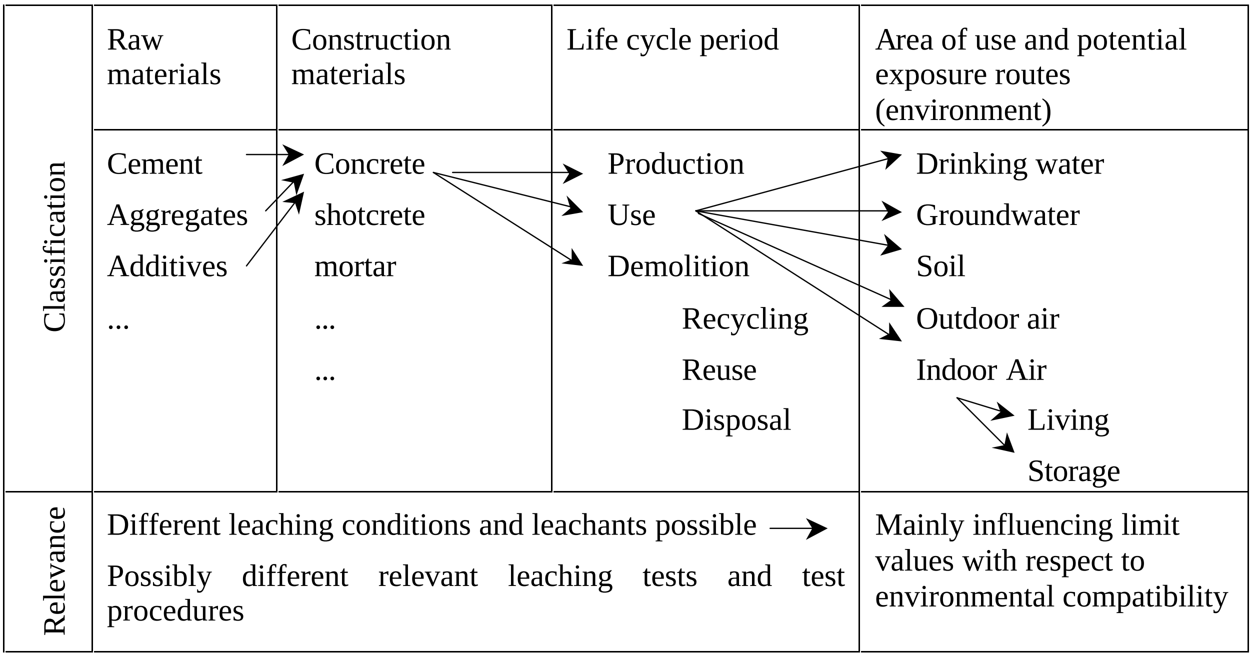

Figure 2. Environmental compatibility of cement-based materials (basic scheme)(Schiessl and Hohberg 1995, Van der Sloot et al, 1997).

Table 1 shows examples for relevant scenarios for building materials. If one takes for instance mortar or concrete, there are three life cycle periods: service life, recycling and "end of life", which have to be considered. During the service life a possible application of the concrete is the construction of foundations, which could be in contact with groundwater or soil. A suitable characterisation test would be the tank test. Mortar could be used in drinking water pipes in contact with drinking water. The suitable characterisation method is also the tank test, but the admissible emissions could be much lower than for foundations in contact to soil. After demolition, the construction debris could be used as aggregate in concrete, than the same conditions are used as above. Since recycled concrete may be applied also in road construction as a granular material with more direct exposure to soil or rainwater, a column test or compacted granular test could be the suitable characterisation test. If demolition waste has to be disposed, the exposure conditions could be rather extreme, and the pH-dependence test would be a more suitable method to assess long-term leaching characterisation.

In the process of evaluating environmental properties of raw materials, construction products, the recycling of construction debris and the use of alternative raw materials, different stages can also be identified with different testing requirements. For an initial proper understanding of material properties characterisation leaching tests are needed. Once a base of knowledge is gained, simpler testing will suffice to ensure compliance is achieved with previously determined characterisation information. This approach can be compared with common practice in concrete technology, where for a proper understanding of the strength development the compressive strength is measured over a period of 56 or 90 days. For compliance purposes, only the 28-day strength is commonly used for reference purposes. In figure 3 this is illustrated for monolithic construction materials. The knowledge gained in the process of utilization can be used to derive acceptability of materials for use in specific construction products and applications.

Table 1. Relevant Scenarios for Building Materials – Examples.

Material	**Life cycle period**	**Area of use**	**Exposure condition**	**Suitable characterisation test method***
Cement-based materials	Service life	Foundation	Flowing/stagna nt ground water	Tank test
			soil	
		Drinking water pipe, container	Drinking water	
		Facades	Rain water, splash water	Tank test with modified conditions, special test methods
	After demolition	as aggregate in mortar/concrete	see above	see above
		Road construction	Soil/rain	Column test, compacted granular test
		Disposal	Low pH; extreme conditions water	pH-static leach test
MSWI bottom ash	Used as recycled waste	Road construction	Soil/rain	pH dependence, olumn test, compacted granular test

* Details of the test methods listed in the table can be found in the CEN standards referenced as well as in the paper by Kosson et al. (2002).

Release

The controlling release mechanisms most often can be described in terms of either equilibrium controlled or mass-transfer rate controlled. Equilibrium controlled release occurs for slow percolation through porous or granular materials. Mass transfer rate controlled release occurs when flow is predominantly at the exterior boundary of monolithic materials or percolation is very rapid relative to mass transfer rate of constituent release to the percolating water. Intrinsic leaching parameters that are to be measured using laboratory testing are: constituent availability, constituent partitioning at equilibrium between aqueous and solid phases as a function of pH and liquid-to-solid (LS) ratio, acid and base neutralization capacities (ANC and BNC), and constituent mass transfer rates. Definition of management scenarios and application of intrinsic parameters, release models and decision criteria are discussed in later sections.

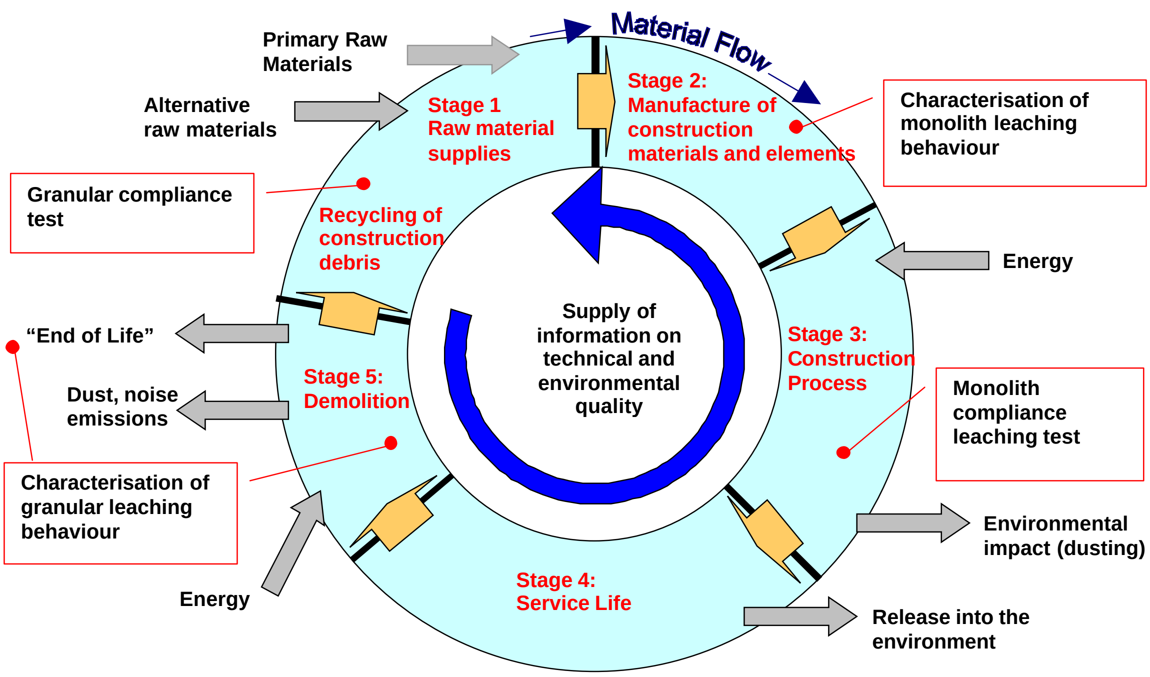

Figure 3. Life cycle stages of alternative materials in construction with characterisation and compliance leaching tests allocated to the different stages. (Modified after Rilem 22, 2000).

A special case are very fine grained materials such as clays, which may behave as monolithic materials, provided they are occurring in a situation surrounded with more porous materials. Then water will flow around the material, instead of though the material as represented by a percolation test generally prescribed for granular materials. The data handling of the compacted granular leach tests (NVN 7347, 1997) is applicable for this case and very similar to the tank leach test. The category of coarse granular materials is intermediate between percolation and diffusion type of behaviour. The larger particles are certainly dominated by diffusion or wash-off effects. Assessing the release properties of this intermediate class of materials is still subject of discussion as a current test for this class of materials (EN 1744-3, 2002) has some flaws that make data interpretation cumbersome. Experimental work is needed to identify the role of individual particle size fractions on the overall leaching result. This type of experiments will provide guidance on choices to be made, which are less arbitrary than the current approach.

To evaluate environmental impact resulting from utilisation of construction materials a scenario approach is needed, in which the question to be answered must be properly specified. In ENV

12920 (1997), worked out by CEN TC 292, a framework to describe such a scenario approach is provided for waste, which is sufficiently general to be equally applicable to construction materials after minor modifications. In general, defining a scenario will include defining (a) the range of anticipated field conditions, (b) the leaching tests to determine the applicable intrinsic release characteristics of the material, and (c) mass transfer models to provide estimates of long-term release under the range of conditions for the material and scenario(s).

For scenarios, in which percolation dominates the release, such as in embankments, road base materials and structural fill, a column leaching test in combination with pH stat or ANC forms the most suitable method. Depending on the nature of the material, additional information may be required to assess redox changes and pH changes in time. The tank leach test in combination with pH stat or ANC is the most relevant method to address mass-transfer constraints. The method can be modified to impose dominating exposure conditions such as a neutral pH environment, seawater exposure, etc.

Hydrology plays a significant role in release from granular percolation dominated materials and from monolithic materials with mass-transfer constraints. In the case of percolation, the L/S is related to a time-scale. In this relationship infiltration rate, height and density are linked (Kosson et al, 1996). For monolithic materials, diffusion or dissolution from the surface only takes places during wet periods. So summing wet periods to relate release to permanently wet conditions to actual conditions can provide an initial estimate. Models that account for limited water contact and internal gradient relaxation during periods with drying or without water contact have been proposed (Garrabrants et al., in press).

It is crucial for the evaluation of construction products to use a scenario type of approach, in which a generic basis provides the relevant background to select appropriate conditions for testing materials in specific applications. A hierarchy in testing is very useful, as the characterization provides the relevant background information to make decisions on quick tests suitable for compliance purposes (PrEN 14405, 2002; PrEN14429, 2002; Kosson et al 2002). However, it is of major importance that the quick tests / compliance tests are linked to the generic characterisation tests. For a proper comparison, it is important to take the performance characteristics of the tests into account.

Cement-based monolithic products

The characterisation test methods have by now been applied to some 50 different types of materials providing a good insight in the relationships in leaching behaviour of specific contaminants in different matrices. Among others the use of alternative raw materials in cement production have been assessed by studying detailed leaching behaviour of now more than 20 cement mortars from world wide origin (van der Sloot et al, 2002). The environmental properties of the cement mortars in their service life as well as in the secondary life (construction debris) and their end-of-life condition are considered (van der sloot, 2000). For cement mortars and concrete for that matter, the recycling stage and the end-of-life scenarios are most important from an environmental point of view (van der Sloot et al, 2002). Evaluations for the use stage of the life-cycle are most appropriately based tank leach testing, but evaluations during recycling and end-of-life conditions are most appropriately based on pH stat testing (van der Sloot, 2000; van der Sloot et al, 2002). Both for cement-based products as well as aggregates in concrete, the oxyanions such as molybdate, chromate and vanadate are more important than cationic metals, because the cationic metals feature a rather low mobility in the relevant pH domain for common applications. For quality control purposes, compliance leaching tests have been developed, which provide more valuable insights then most current regulations when they are presented in conjunction with prior characterisation information.

If the more relevant and useful information initially gained from characterization testing results in more appropriate and cost effective options for material use, the cost of implementing the three levels of testing will not be excessive and provide the possibility to focus on key aspects rather than measure irrelevant parameters. Collective organisation of characterisation testing for groups of materials expected to have similar characteristics will benefit industry by reducing testing costs and at the same time highlighting the key issues to be resolved in a given industry sector. A database/expert system to guide testing, data management and comparison of characterisation data would be very beneficial for all parties. Such a database will prove very useful in view of its use:

- as reference for compliance test data,
- as basis for regulatory development,
- to limit unnecessary duplication of work
- to avoid generation of useless data at great cost,
- in focussing on key parameters for specific materials,
- in identifying controlling factors for guiding the choice of treatment options/ recipes.

Evaluation of constituent release can be approached by a series of steps: (i) define management scenarios and mechanisms occurring in the scenarios (e.g., rainfall infiltration) that control constituent release, (ii) measure intrinsic leaching parameters for the material being evaluated (over a range of leaching conditions), (iii) use release models incorporating measured leaching parameters (corresponding to anticipated management conditions) to estimate release fluxes and long-term cumulative release, and (iv) compare release estimates to acceptance criteria. Management scenarios can either be default scenarios that are designed to be conservative or incorporate site-specific information to provide more-accurate estimates of release (Kosson et al, 2002).

Granular - percolation dominated and monolithic - mass transfer limited release

The integrated approach that forms the basis of evaluating the release from monolithic construction materials in a variety of conditions is given in figure 4. Here mass-transfer constraints (i.e., diffusion or surface dissolution) control the rate of release. The same scheme is applicable for granular materials, however, the monolith leach test should be replaced with a percolation test such as PrEN 14405 or NEN 7343. Other than that, the primary aspects of chemistry and the release controlling factors such as pH, redox and DOC will be the same. For the judgement of impact, the source term description will be different, but the soil groundwater impact evaluation will be very much the same. The integrated evaluation provided by this approach allows for many aspects to be addressed, which are relevant for an evaluation of the long-term release behaviour of materials under the influence of external factors and stresses.

The combination of pH dependence test results on size reduced material and the monolithic leach test results provide a means to distinguish between diffusion controlled release and solubility limitations. In figure 5 this is illustrated for Sr and Zn from a cement-based material tested under imposed neutral pH conditions.

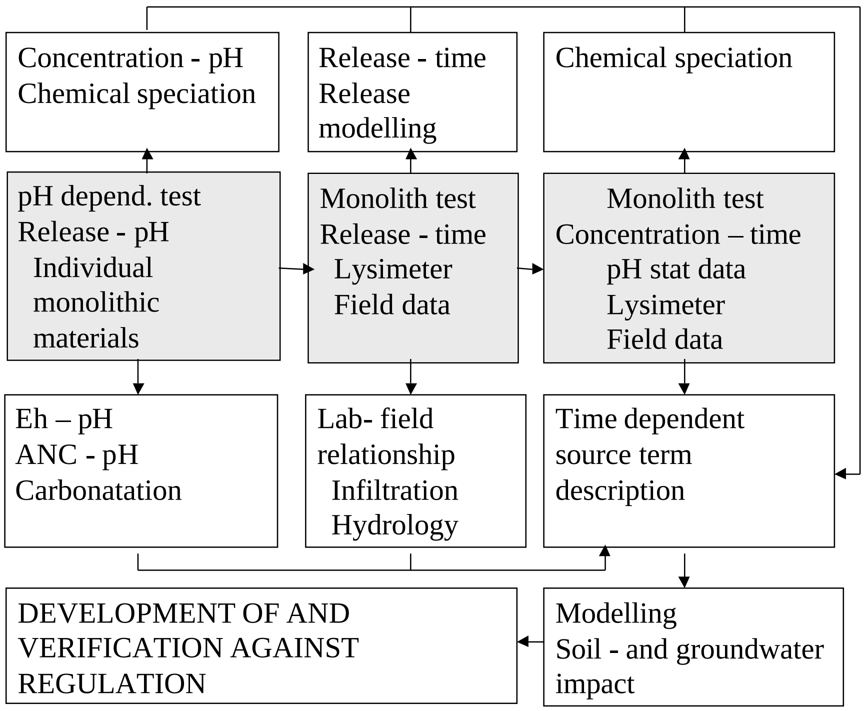

Figure 4. Scheme showing interrelations between test for monolithic materials, data processing and evaluation aspects for monolithic construction materials.

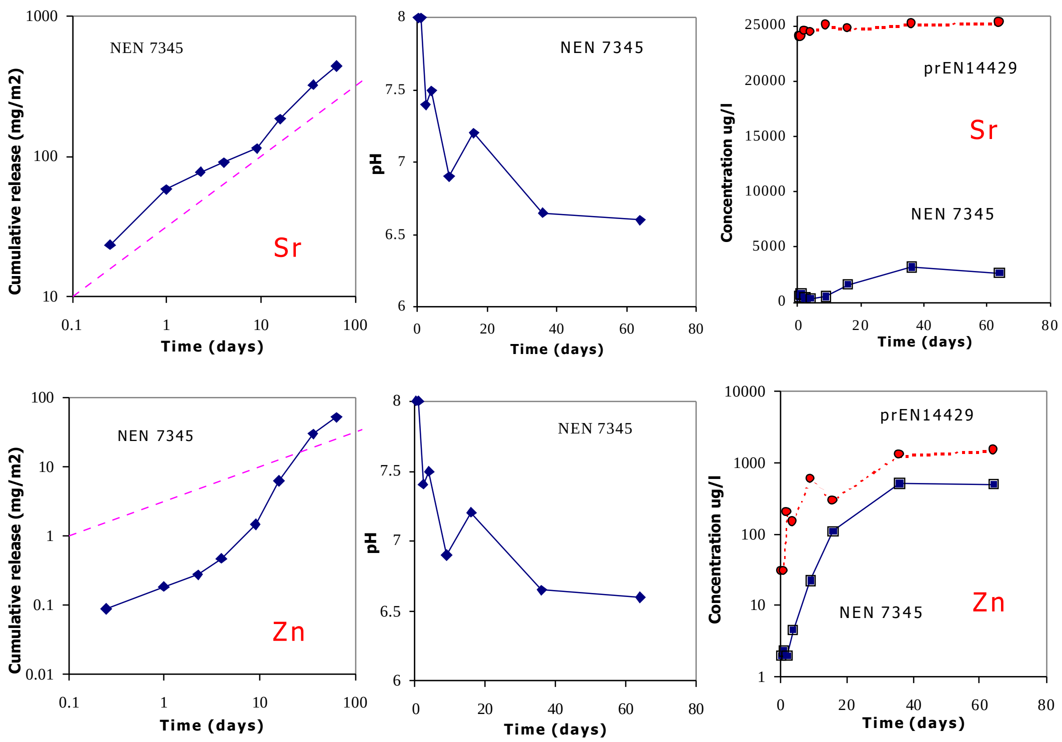

Figure 5. Cumulative release of Sr and Zn from cement-based material, pH variation during the test and concentration in eluate in relation to pH dependence test data at corresponding pH.

If under the experimental conditions release is controlled by a specific mechanism, then it must be possible to recognize the mechanism and use this information in making long-term assessments. In case of Sr, the difference between the solubility limitation and the measured concentration is at least a factor of 5. However in the case of Zn, the solubility curve derived from the pH dependence test is close to the concentrations measured in the eluate and follows it indicating solubility limitations for release from Zn. This implies that for long-term prediction in case of Sr a different description must be applied than for Zn.

The monolith leach test (like NEN 7345) is a tool that is suitable for most monolithic forms that maintain their physical integrity during testing, but the release controlling process, not the material type, should determine the test choice. The applicability of monolith testing is illustrated in figure 6 for a wide range of different materials. As can be seen from the release curves in comparison to the slope 0.5 for diffusion controlled release, tentative conclusions on behaviour can be drawn, which need to be verified. Still this type of test provides one of the best options to resolve the release mechanism, which is so crucial for long-term prediction. Improvements in the test conditions (time sequence of leachant renewal) can further enhance its use. In addition, coupled use of pH dependent testing with monolith testing allow for understanding of the underlying coupled mechanisms that influence the rate of diffusion controlled release. Appropriate representation of these underlying mechanisms in mass transfer models is important for extrapolation of testing results to long-term release estimates (e.g. Sanchez et al, 2002, Sanchez et al, 2003, Garrabrants et al, in press, Garrabrants et al, Meeussen, 2003).

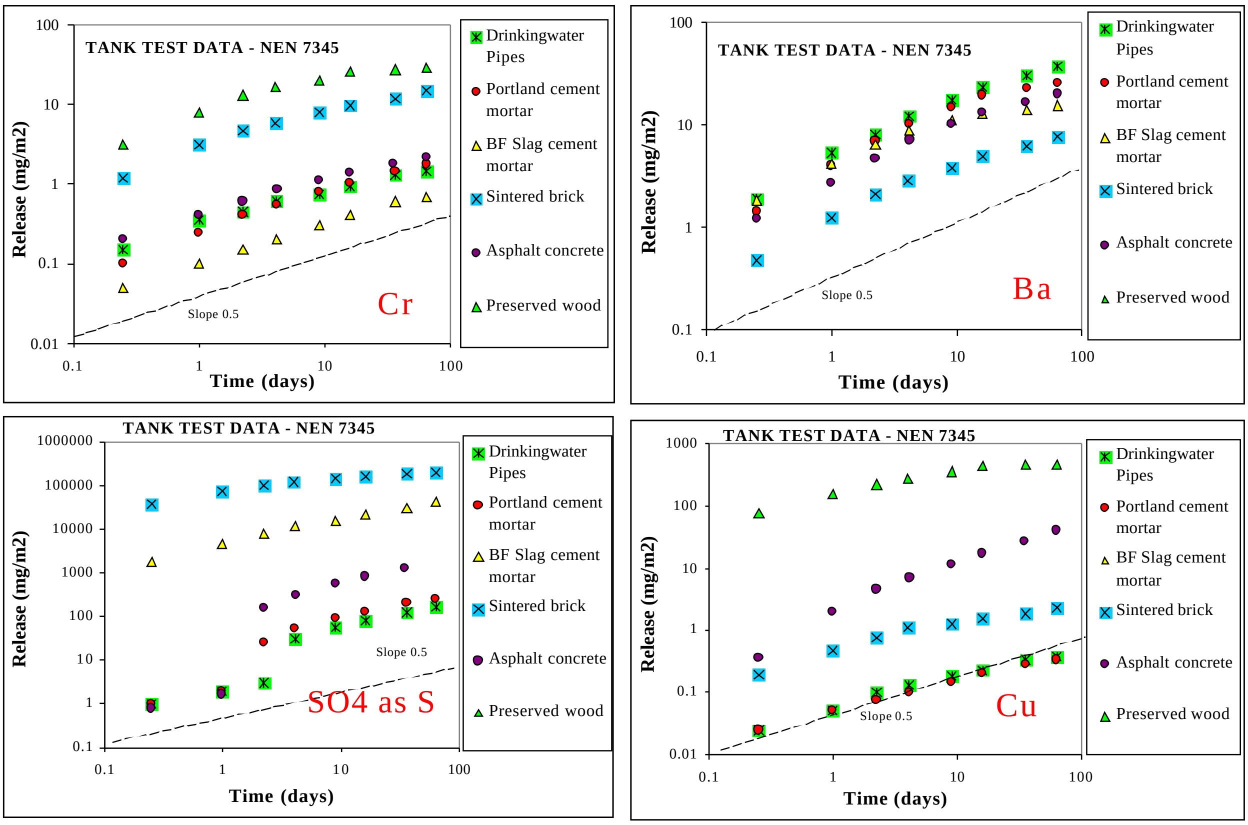

Figure 6. Cumulative release curves as a function of time for a wide range of monolithic materials as obtained by NEN 7345(1995).

Geochemical speciation modeling has been shown to be valuable to identify solubility controlling conditions. Currently, developments are in progress to use ORCHESTRA (Meeussen, 2003) for geochemical solubility calculation using an extended MINTEQA2 database in combination with the NICCA Donnan model for simultaneous organic matter interaction. For this purpose ORCHESTRA is coupled to the database/expert system for quick data retrieval. ORCHESTRA is further developed to couple chemical speciation and transport to describe interface processes, concentration profiles in products at given exposure times and long term release predictions. Finally, the approach followed in the development of acceptance criteria for the landfill of waste (Hjelmar, 2002), provides the basis for developing acceptance criteria for construction materials in applications in and on soil as well as in contact with surface and groundwater.

CONCLUSIONS

A tiered testing approach coupled with a database/expert system provides a sound basis for understanding and quantifying release behaviour from a wide range of different construction materials of both alternative and traditional nature in a wide range of settings and time scales. It can address different life cycle stages of construction materials and forms a basis for quality improvement on sound scientific grounds. Furthermore this approach provides a basis for quality control focused on key parameters against a background of characteristic material behaviour. Finally, it offers the possibility to develop risk based acceptance criteria using a scenario approach with the option of defining different points of compliance with associated target values.

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