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INTEGRATED DECISION TOOL FOR ASSESSMENT OF ENVIRONMENTAL COMPATIBILITY OF BIOMASS AND BIOMASS ASH.

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SUMMARY: For biomass ash resulting from energy generation suitable methods of disposal or preferably beneficial use must be found. A proper evaluation of their environmental compatibility is needed, which cannot be judged by total content or a single step leaching test, as specified in current regulations if a regulation actually exists. Proper leaching tests have been standardized in CEN and by US EPA. An integrated decision tool (LeachXS) has been developed to predict release of substances under relevant exposure conditions in service life (beneficial use applications), in recycling scenarios and End of Life (disposal) conditions; thus supporting sustainable development of alternative energy sources. The application of biomass ash as a constituent in a cementious product is used as illustration of the new possibilities.

1. INTRODUCTION

Biomass is used extensively in recent years as an alternative source of energy, either as a single source or in the form of co-combustion with a natural energy source (Venice 2010; Dam-Johansen et al 2012; Savolainen 2003). The residues of such processes are considered to be used beneficially. However, as with many other residues from thermal processes the environmental aspects of beneficial use are a matter of concern (Vamvuka and Kakaras 2011; Tsuchiya et al 2011). Possible uses for biomass ash are use as fertilizer, utilization as a building materials or constituent in a building material and use as a fuel in case the level of unburnt material is still substantial (Pels et al 2005).

In case of its use as a fertilizer, the mix proportion with soil and the mode of application are important. Currently, total content is practically the only legally required parameter for fertilizers. Using total content of the soil improver or soil additive as a measure for impact is not a suitable means of assessing this option as was shown for another waste derived soil additive (Carter et al 2009).

The utilization as a building material as such is a less likely option due to the nature of the material. Its use as a constituent in a mix design for a building material is a possible route, which has been explored (Cheah and Ramli 2011; Corinaldesi et al 2009; Fava et al 2007). However, the environmental compatibility is assessed in a manner that is often inadequate due to insufficiently discriminating methods (such as EN 12457 2002 and TCLP 1986). A good exemption is the work by Rocca et al (2010), who applied the same methods as recommended in this paper.

Currently, CEN/TC351 (Construction products: Assessment of release of dangerous substances) is working on the development of methods to assess release from construction products to soil and groundwater (CPD 1988; Dijkstra et al 2005). Robustness work is almost completed for TS-2 (monolith test) and TS-3 (percolation test). Similar methods have been adopted and validated by US EPA (Garrabrants et al 2012 a and b).

Criteria for construction products are available in the Netherlands (Soil Quality Decree 2007) and are close to being formalised in Germany (BMU, 2011). This allows evaluations of environmental impact to be made.

2. MATERIALS AND METHODS

2.1 Leaching tools

A short description of the characterisation leaching test is given with an indication how simplified testing can be carried out, when a material's release behaviour has been established sufficiently.

2.1.1 pH dependence test

The test provides information of the pH sensitivity on the leaching behaviour of a material (CEN/TS 14429 2005, CEN/TS 14997 2005, ISO TS 21268-4 2007 or EPA method 1313 2009). The listed methods lead to very comparable results (Garrabrants et al. 2012 a and b). The test consists of a number of parallel extractions of a material at L/S = 10 during 48 hours at a series of pre-set pH values. The pH is adjusted at the start of the experiment with HNO_3 or NaOH (or KOH). After 48 hours of equilibration by end over end rotation in PE containers, the suspensions are filtered (0.45 µm) and analysed. In CEN/TS 14997 the pH control is automated. The test provides the response of a material to imposed pH changes under different environmental exposure scenarios (e.g. carbonation, infiltration, sulphur oxidation, soil interfaces).

2.1.2 Percolation test

This test provides information on the leaching behaviour of the material as a function of the liquid to solid ratio (L/S in litre/kg) (NEN 7373 2003; CEN/TS 14405 2004, ISO TS 21268-3 2007, EPA method 1314 2009 and CEN/TS-3 2009). The listed methods lead to very comparable results (Garrabrants et al. 2012 a and b). Seven eluate fractions are collected over the L/S range 0.2 – 10 litres/kg, with the total test duration being approx. 21 days. The leachant is demineralized water. The test material is applied as received and up-flow (15 ml/hour) is applied through a column with a waste height of about 25 cm and a diameter of 5 or 10 cm. L/S can be related to a time-scale through the infiltration rate, density and height of the application (Hjelmar 1990).

2.1.3 Monolith leach test

This test provides information on the release per unit surface as a function of time and it is performed on regular shaped product samples according to standardized procedures (NEN 7375

2003, CEN/TS 15863 2009, EPA method 1315 2009 and CEN/TS-2 2009). The listed methods lead to very comparable results (Garrabrants et al. 2012 a and b). The specimen is subjected to leaching in a closed tank. Demineralised water is used as the leaching solution at a leachant-to-product volume ratio (L/A in cm^3/cm^2) of approximately 8. The leaching solution is renewed 7 times up to 64 days. The pH, electrical conductivity (EC) and, optionally, Eh are measured in all eluates before filtration (0.45 µm) and chemical analysis. From the combination of pH dependence and monolith leach test the predominant release controlling mechanism (diffusion, wash-off or solubility control) can be obtained.

2.1.3 Additional assessment tools

For modelling purposes additional parameters are needed such as the quantities of "reactive" surfaces in the solid phase. Organic carbon partitioning in the solid and liquid phase and the amount of amorphous and crystalline iron and aluminium (hydr) oxides in a material can be quantified by methods are now standardized in ISO/TC 190 (Soil) under series ISO/DIS 12782 parts 1 – 5 (2010). The redox behaviour and redox capacity can be important (NEN 7348 2006). Since the release behaviour under reducing conditions can be very different from the materials behaviour under oxidised conditions, it is important to be aware of the oxidation state.

2.1.3 Tiered approach in testing

As indicated in Kosson et al. (2002) a tiered approach allows limited testing when sufficient knowledge on materials leaching characteristics is gained. An efficient way of achieving this is by selecting one optimal step from the pH dependence test or a combination of the first few leaching steps from either the percolation or the monolith leach test. By presenting the results in combination with prior characterisation data, the value of judgement of the single step increases considerable, as the data is now placed in proper context and the sensitivity of the single step result to slight variations in, for instance, pH can be judged.

2.2 Decision support tool

In recent years a new decision support/modeling tool - LeachXS/Orchestra - has been developed (LeachXS 2010; van der Sloot et al 2008b; Meeussen 2003), which has many different functions embedded to act as a tool for a variety of questions to be answered in relation to environmental acceptability of materials and products for specific applications (van der Sloot et al., 2008). The system is applicable to soil, contaminated soil, sludge, compost, sediments, municipal waste, industrial and hazardous waste, mining waste, preserved wood, treated waste, stabilised waste, and construction products like cement mortars, concrete, aggregates, masonry, asphalt, metal plates, etc. The materials leaching database contains results of laboratory leaching tests, various lysimeter test results and field data from more than 600 materials and wastes. The potential uses of the system for questions around biomass and biomass ash are addressed below.

2.3 Analysis of eluates

The eluates from laboratory tests are preferably analyzed for major, minor and trace elements by ICP (Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Se, Si, Sn, Sr, TI, V, Zn). DOC (dissolved organic carbon) and TIC (total inorganic carbon) can be analyzed by a Shimadzu TOC 5000a analyzer or a similar instrument. Cl, F, ammonium and sulfate can be analyzed by ion-chromatography. The multi element methods are highly preferred, as the major elements dictate the chemical environment that controls release of many substances.

2.4 Materials

Biomass and biomass ash from several studies on fluidized bed combustion, gasification and co-combustion at ECN have been tested using the test methods specified in section 2.1. In Table 1 the nature and the origin of the materials used in this study are indicated. The biomass and biomass ash samples have been used to illustrate the range of available data and their meaning in relation to key factors controlling release and exposure conditions.

Table 1. Materials used in the study.

Material type	Samples	Origin
Biomass	Beech wood, cacao, poultry manure, forest residue, paper, preserved wood, sawdust, railway sleeper, straw, wood bark, wood chips spruce, wood pellets, mixed wood.	ECN biomass studies since 1997
Biomass ash (combustion)	Grate ash, coarse and fine ash from fluidized bed combustion	ECN biomass studies since 1997
Biomass ash (gasification)	Gassification bottom ash and fly ash from gasification of straw, grass, wood, cacao, poultry manure, sewage sludge	ECN biomass studies since 1997
Cement mortar CEM I	Cement mortar N1 Norway	Van der Sloot et al 2011

3. RESULTS AND DISCUSSION

3.1 Biomass and biomass ash comparison

A proper comparison of biomass and biomass ash release behaviour can not be made based on composition or single step leach test data. Total content is a meaningless property in relation to environmental impact assessment, as for many substances a very significant portion of the total content will under no foreseen exposure condition be released at short or long term.

3.1.1 Different biomass types

A comparison of different types of biomass using the pH dependence leaching test as shown in Figure 1 reveals that several substances can exceed release limits set for free application of construction products. The upper limit of the box corresponds with the regulatory criterion; the lower limit with the limit of determination of the analytical method employed and the vertical bounds denote the upper and lower pH boundaries for the biomass samples. For As, Cr and Cu this is too a large extent related to biocide treated wood. Poultry manure and railway sleeper exceed Cr, Cu and Zn limits. Several of the materials shown feature rather high dissolved organic carbon (DOC) levels in leachate, which implies substantial mobilization of metal in DOC – complexed form (van Zomeren and Comans 2007).

3.1.2 Biomass ash from combustion

A comparison of different biomass ashes obtained by fluidized bed combustion of various biomass sources is shown in Figure 2. The figure shows results obtained by the pH dependence leaching test since the results from that test allow multiple conclusions to be drawn on the release behaviour. The release behaviour for several substances defined as the pattern of release as a function of pH proves to be the same for different source materials, although the level at low pH for metals and mildly alkaline pH for oxyanions may vary. The L/S=10 condition corresponds with the regulatory conditions, which implies that not only the release from the fresh ash, but also the release under aged/ carbonated conditions can be assessed.

A comparison with regulatory criteria reveals that several substances can exceed release limits set for free application of construction products. RDF derived ashes show significantly higher release levels for Cr, Cu, Pb and Zn. This effect is particularly noticeable at pH below 7 – 8 for Cu, Pb and Zn between straw and RDF derived ash. A single point measurement at pH 9 or 10 may reveal no difference between the two, but characterisation testing as shown here will. For Mo the differences are smaller, but here the ash resulting from straw shows the higher release levels.

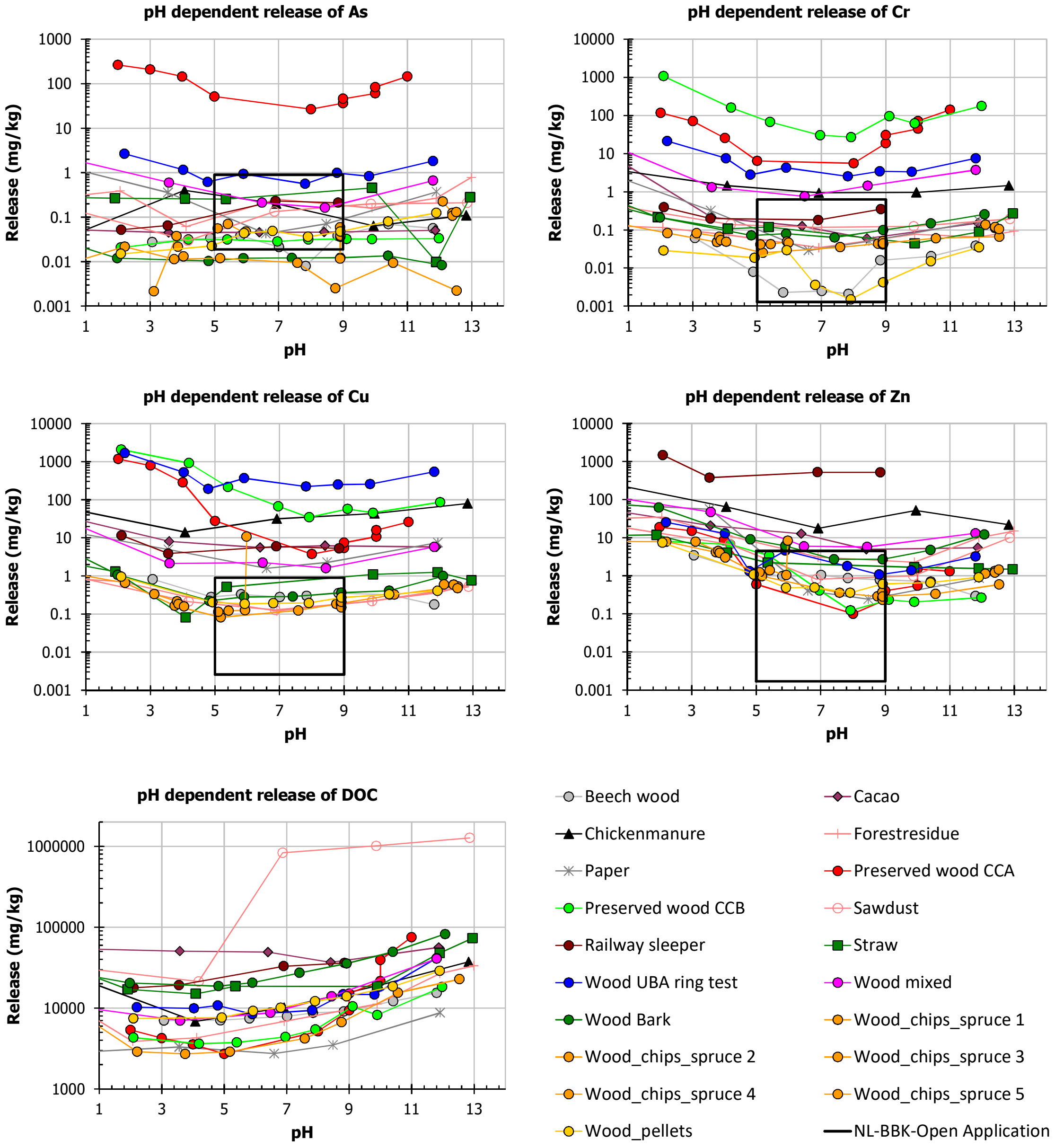

Figure 1. Comparison of pH dependent leaching behaviour of selected substances from different biomass types in comparison with release limiets as specified by the Dutch Soil Quality Decree (2007).

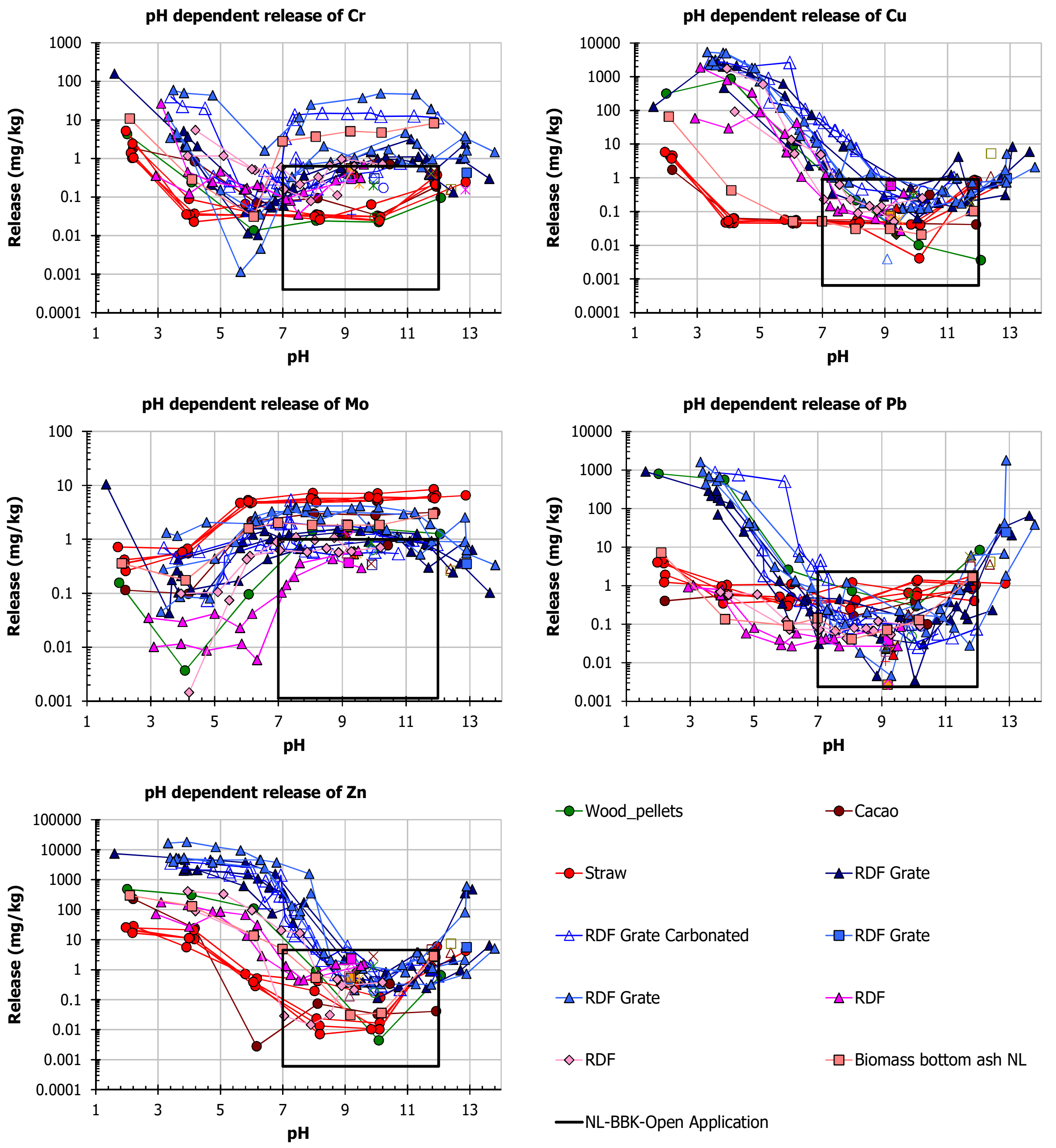

Figure 2. Comparison of pH dependent leaching behaviour of selected substances from different biomass ash types (combustion) in comparison with release limiets as specified by the Dutch Soil Quality Decree (2007).

3.1.1 Biomass ash from gasification

In figure 3 the results of leaching by pH dependence and percolation test are shown for Zn from gasification residues. The results obtained are shown in different units (mg/L and mg/kg dm) to highlight different aspects of the leaching behaviour of Zn from these residues. Depending on the nature of the source material relatively large variation in release levels can be found. The typical behaviour of Zn with high release below pH 8 - 9 and low release between pH 9 and 11points at solubility control in the latter pH domain. Through geochemical modelling (van der

Sloot et al, 2008a) the minerals and/or sorptive phases controlling release can be identified. The concentration level at low liquid to solid ratio (L/S in L/kg) is relevant for assessing the possible leachate composition in the initial stages of storage and disposal. This level can be considerably higher than the level found at L/S=10 to 20 (common testing condition in regulatory tests) and may prove not to be acceptable in spite of compliance with regulatory criteria (e.g. TCLP). Large differences can be noted between fly ashes, as some may be neutral, while others are very alkaline. Since this has significant influences on release at short and longterm, this aspect deserves more attention than given in regulation until now. From the cumulative release curve, there can be two cases identified leading to more or less the same release pattern. One relates to wash out of a soluble species, which will then be depleted by the time L/S=1 is reached. The other relates to a pH change, which can imply either pH increasing from neutral to 10 – 11 or pH decreasing from pH > 12 to around 9. In both cases, the initial release is high in case of Zn followed by a decreasing concentration. The cumulative release curve reaches a plateau. Further change in pH may lead to an increase again and hence understanding of the relationship between release and pH change is crucial for long term behaviour.

3.2 Biomass ash use in construction

In the prior sections the residues resulting from combustion or gasification of biomass have been assessed as obtained from the process. However, in many instances the material may be used as a constituent in a product or, as in the case of soil, mixed with soil to neutralize acidity or provide more structure to the soil. The release behaviour of the biomass ashes in usch circumstances will not be the same any more and thus the release under the new exposure conditions must be assessed to be able to judge acceptability. The case of biomass ash as a soil amendment is not addressed here further. Since total content is unsuitable, an alternative approach based on the tests described in this paper has been presented for alkaloam, a product derived from red mud (Carter et al, 2009). A similar testing approach would be suitable for assessing environmental acceptance as soil improver or fertilizer. Here the use of biomass ash as a constituent in cement based formulations is addressed. To address this question testing of a product made with biomass ash will be necessary, but in the process of developing possible utilisation options, it is costly to test all possible combinations. An alternative approach, it to test the constituting ingredients and then through geochemical modelling assess the possible effects, which then can be verified by a much more limited set of experiments. The geochemical modelling of materials using LeachXS is described elsewhere (van der Sloot et al 2008a). The main aspect is to cover all major, minor and trace elements to btain a good match between model description and measurement. The Chemical Speciation Fingerprint (CSF) thus obtained is used to describe the release behaviour from any mix proportion of the constituting parts (van der Sloot et al 2007).

Obviously, the description is not suitable for all trace substances, as not for all substances adequate thermodynamic data are available. In spite of that limitation, very interesting observations can be made. For instance, the adding a new material in a mix will generally not alter the release behaviour of the orginal product very much. There seems to be some tolerance before significant changes in release behaviour are noted. The percolation test data match generally well with the pH dependence test data, which indicates that solubility rather than percolation behaviour drives the concentrations. This is also reflected by the fact that the release prediction for L/S=0.3 agrees well with the description of release at L/S=10 over the pH range 9 – 12 for the gasification ash.

The release of Ni between the three conditions (biomass ash, cement mortar and mix) is very similar. In the mix, the Ni release behaviour is hardly changed. In case of V, the description for V release in gasification ash is described by hydrated ironoxide sorption and Pbvanadate solubility. For the cement mortar, hydrated ironoxide and ettringite substitution are controlling.

In the mix, the Pb vanadate phases become more prominent than originally in the mortar. In the gasification ash $BaCrO_4SO_4$ proves to be an important controlling phase besides solid organic matter. In the cement mortar, at high pH ettringite substitution is controlling release of Cr. In the mix, the ettringite substitution remains important, but at neutral pH organic matter, $Cr(OH)_3$ and $BaCrO_4SO_4$ appear as controlling phases. In the gasification ash, Pb is controlled by $Pb(OH)_2$, hydrated ironoxide and organic matter. The description for Pb in cement mortar is off, but the Pb concentration is very low. In the mix, $Pb(OH)_2$, hydrated ironoxide and organic matter are controlling release, which is to a large extent related to the significant increase in the Pb content of the mix due to addition of biomass ash.

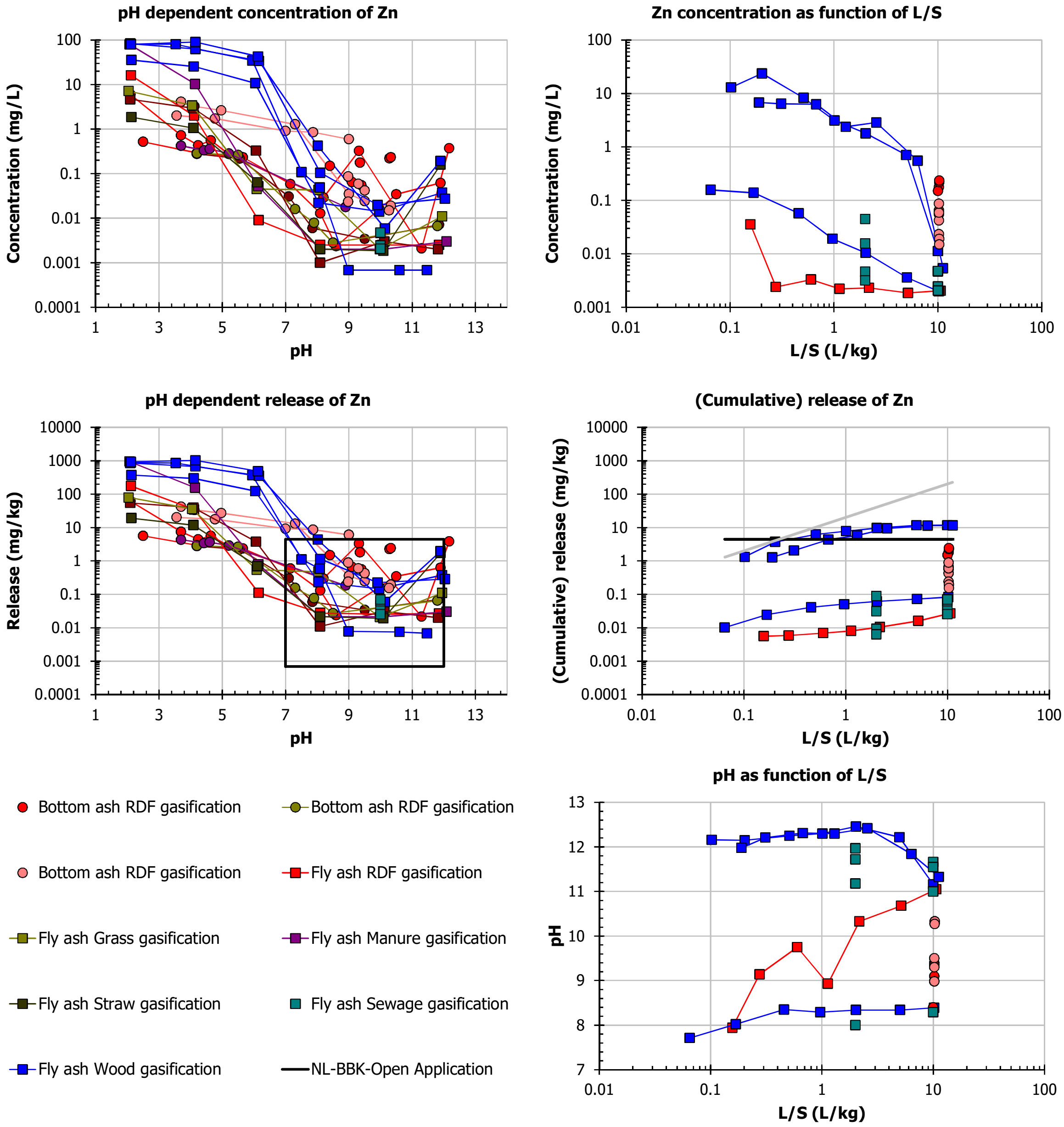

Figure 3. Release behaviour of Zn from gasification residues as obtained by pH dependence (CEN/TS 14429) and percolation test (CEN/TS 14405) expressed in concentration as well as release units.

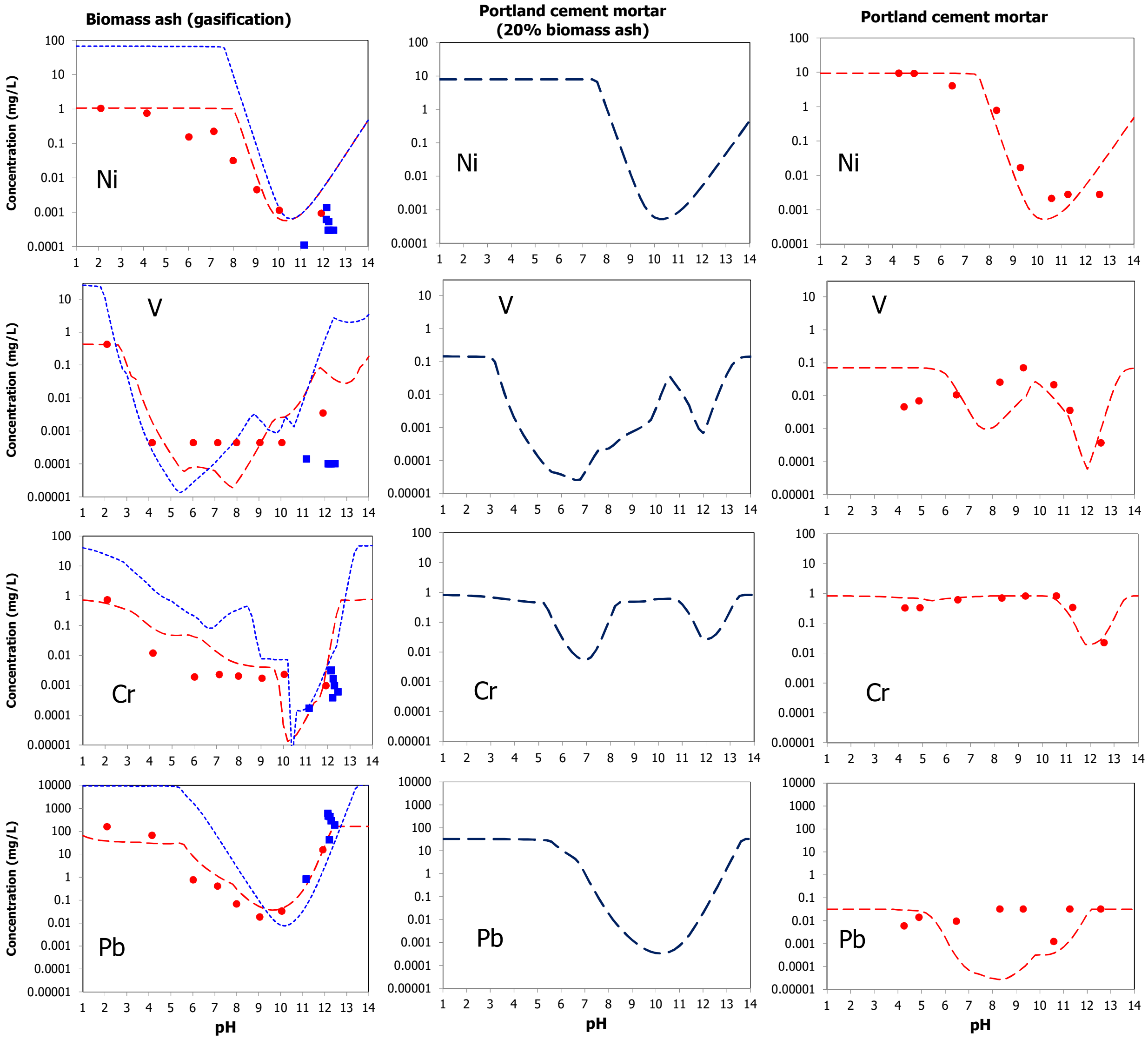

Figure 4. Comparison of measurements and associated model descriptions for Ni, V, Cr and Pb from multi-element and multi-phase geochemical modelling with a prediction of release from a mixture of 20% biomass ash and Portland cement mortar (CEM I). Red dots are pH dependence test results; blue dots are percolation test data plotted as a function of pH; the red broken line is the model description based on CSF; the blue dotted line is the prediction of release at L/S=0.3; the blue broken line I the middle graphs represent the prediction of release from the mixture.

4. CONCLUSIONS

The beneficial use of biomass ash is a very worthwhile endeavour. However, the judgement of environmental properties of biomass conversion residues in view of their possible uses in either agricultural or construction applications is still limited by the often poor testing approaches, that either leave a lot of questions unanswered or give the wrong or an incomplete answer. The now more common characterisation leaching test, which have been validated by US EPA for soil and waste and are in the process of being validated for construction products, are suitable tools to provide the necessary insight in long term behaviour of residues and an understanding of the consequences of incorporation of such residues in construction products and in agricultural applications. Since the methods are applicable to a wide spectrum of material and products, the test result can be used to assess a wide range of regulatory questions related to utilization and disposal covered by the EU Landfill Directive, Construction Products Directive, End of Waste considerations and use in agricultural context. Once the characterisation is done for a given class or group of materials simpler testing (part of the characterisation tests) will suffice for decision making.

Although thermodynamic data are still missing for important substances multi-element geochemical speciation modelling taking multiple interactions between substances into account (mineral precipitation, sorption, ion exchange, solid solutions,..) is already providing valuable insights in release behaviour under conditions that are not easy to simulate in the laboratory. Also in design of recipes for beneficial use modelling will allow identification of potential critial environmental reactions, which can than be verified experimentally, rather than carrying out trial and error type testing approaches.

Biomass and consequently biomass ash is much more variable in composition and leaching behaviour than several traditional energy sources. Also the variability within a source material is likely to be greater. This implies that a good understanding of such variability is needed to ensure environmentally acceptable uses of the residues. The results presented here by bringing together data from multiple test runs provides a means to determine a bandwidth for source materials based on which decisions for suitable uses can be build. LeachXS Lite as a management tool will provide the capability to compare results from different tests in a sound manner and lay the groundwork for impact prediction through reactive transport modelling.

The approach sketched here is not limited to biomass ash, but is equally applicable to a variety of other material and product streams. Observations from other fields prove beneficial in resolving questions relevant for biomass and biomass conversion residues.

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