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## Use of Chemical Speciation and Reactive Transport to Predict Leaching from Coal Combustion Residues

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### ABSTRACT

The characterization of release from materials, such as coal combustion residues, is entering a new phase with the development and use of standardized characterization leaching tests and associated geochemical modeling capabilities coupled to a leaching database. Geochemical speciation and chemical reaction/transport modeling of coal fly ash leaching test data have been carried out using LeachXS-ORCHESTRA. A chemical speciation fingerprint (CSF) is provided consisting of a mineral selection and parameters for binding to Fe-oxide, Al-oxide, dissolved organic carbon and particulate organic matter, which provides detailed insight in the factors controlling release of >25 substances.

### INTRODUCTION

Coal fly ash has since long been identified as a by-product that can be used beneficially [Malhotra & Metha, 2002; Malhotra, 2002]. Large differences exist in the extent and practices of beneficial use between different countries around the world [Manz, 1996; ACA, 1996]. When coal fly ash is used in beneficial applications it is important to guarantee that no unacceptable risk to health and environment occurs [Essential Requirement 3 on Health and Environment of the CPD, 1988]. The common regulatory tools based on single step extraction tests are clearly inadequate for that purpose [TCLP, 1986; EN12457, 2000]. With the implementation of new characterization leaching tests [CEN/TS14429, 2005; CEN/TS14405, 2004; CEN/TS15683, 2009; draft EPA SW-846 methods 1313 to 1316, 2009] and the associated geochemical modeling capabilities [van der Sloot et al, 2008; LeachXS, [www.leachXs.org](http://www.leachXs.org)], the assessment of the release from materials, such as coal combustion residues, is entering a new phase.

The increased insight in contaminant release controlling processes requires the integrated evaluation of a material as a whole, since the release behavior of individual components is affected by other major and minor constituents in the material. Geochemical speciation modeling and reactive transport forms the basis for prediction of long term release behavior [Kosson et al, 2002]. Inclusion of chemical partitioning of elements between different chemical binding forms and species in solution allows assessment of release under a range of

chemical (e.g., redox, pH) and flow (e.g., percolation, diffusion) conditions. Better understanding of simultaneous interactions of contaminants with minerals and with reactive surfaces in coal fly ash (e.g. minerals, sorption on iron/aluminum oxides, and interaction with clay and natural organic matter) is of crucial importance for environmental risk assessment.

Over recent years this understanding has significantly improved, resulting in multi-component interaction models for ion adsorption onto iron/aluminum oxides, based on work by Dzombak and Morel [1990], and natural organic matter [Kinneburgh et al, 1990; Milne et al, 2003]. These models have been successfully applied to describe the speciation of metals in soils [Dijkstra et al. 2004, Carter et al, 2009] and waste materials [van Zomeren and Comans, 2004; Guyonet et al, 2008] and stabilized waste [van der Sloot et al, 2007; De Windt et al, 2007]. These models have been implemented in the modeling framework ORCHESTRA [Meeussen 2003], which is embedded in the database/expert system LeachXS for easy data handling [van der Sloot et al, 2008; LeachXS, [www.LeachXS.org](http://www.LeachXS.org)].

In this paper, geochemical speciation modeling is used to identify the chemical processes that control the release of substances from selected coal fly ash samples and to address the consequences for the use of coal fly ash in embankments and road stabilization.

## **MATERIALS AND METHODS**

### **Materials**

Coal fly ash samples were obtained from Dutch coal fired power plants operating on coal from worldwide origin. Part of the work reported here is based on alkaline coal ash samples studied back in 1982 [van der Sloot et al, 1982], which can now be placed in perspective to pH dependence test data on alkaline coal fly ash, which was not yet developed at that time. In addition, data from the lysimeter studies with coal ash by Hjelmar [1990] are used to illustrate the consistency of data from different scales of testing and from different sources.

### **Leaching tests**

*pH dependence test [CEN /TS 14429, 2005]* - This test provides information of the pH sensitivity on the leaching behavior of the material. 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 was adjusted at the start of the experiment with HNO<sub>3</sub> or NaOH. After 48 hours of equilibration by end over end rotation in PE containers, the suspensions were filtered (0.45 μm) and analysed. The test provides the response of a material to imposed pH changes and an acid-base titration to understand the response of the material to acid or base reactions under environmental scenarios (e.g., carbonation, infiltration, soil interfaces).

*Column leaching test [CEN/TS 14405, 2004]* - The column leaching test provides information on the leaching behavior of the material as a function of the liquid to solid ratio (L/S in L/kg). Seven eluate fractions are collected over the L/S range 0.1 to 10 L/kg, with the total test duration being approximately 21 days. The leachant is demineralized water. The test material is applied as received and up-flow (14mL/h) is applied through a column with a waste height of 28 cm and a diameter of 5 cm. L/S can be related to a time-scale through the infiltration rate, density and height of the application.

*Preparation for analysis* - For each eluate, the pH and conductivity are measured and analytical samples are conserved for chemical analysis.

## **Estimation of model parameters**

The quantities of “reactive” organic carbon in the solid phase (i.e. HA and FA) were estimated by a batch procedure [van Zomeren and Comans, 2007]. In short, the procedure is based on the solubility behavior of HA (flocculation at  $\text{pH} < 1$ ) and the adsorption of FA to a polymer resin (DAX-8). The amount of amorphous and crystalline iron (hydr)oxides in the waste mixture was estimated by a dithionite extraction [Kostka and Luther III, 1994]. The amount of amorphous aluminum (hydr)oxides were estimated by an oxalate extraction [Blakemore et al., 1987]. The extracted amounts of Fe and Al were summed and used as a surrogate for hydrous ferric oxides (HFO) in the model [Meima and Comans 1998].

## **Chemical analysis**

The leachates and extracts from laboratory tests were 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, Tl, V, Zn). DOC (dissolved organic carbon) and TIC (total inorganic carbon) were analyzed by a Shimadzu TOC 5000a analyzer. Cl, F, ammonium and sulfate were analyzed by ion-chromatography.

## **Geochemical speciation and release modeling**

Chemical speciation of the solutions was calculated with the ORCHESTRA modeling framework [Meeussen 2003] embedded in LeachXS [van der Sloot et al, 2007; van der Sloot, 2008]. The input to the model consists of metal availabilities, selected possible solubility controlling minerals, active Fe-and Al-oxide sites, particulate organic matter and a description of the DOC concentration as a function of pH or L/S (polynomial curve fitting procedure). The mineral phases that were allowed to precipitate were selected after calculation of their respective saturation indices (SI) in the original pH dependence leaching test eluates. Saturation indices were calculated for all > 650 minerals in the thermodynamic database and a selection of the most likely and relevant phases was made based on the degree of fit over a wider pH range, the closeness of the SI value to 0 and expert judgment on suitability of possible minerals for the waste mixture (e.g. exclusion of high temperature minerals). Generally, minerals were selected if the SI was in the range of -0.2 to 0.2 for more than two pH data points. For ettringite, solid solution parameters have been derived from Portland cement mortars [Meeussen et al, 2009].

## **RESULTS AND DISCUSSION**

### **Interpretation of leaching test results**

In this work we are building on the experience gained in leaching studies on a wide range of materials [van der Sloot et al, 1997, Kosson et al, 2002], which has shown that there is no correlation between leaching and total content with the exception of a few cases [van der Sloot et al, 1997] and that single step extraction tests like TCLP [1986] are definitely insufficient to properly address environmental aspects of any material. This has recently been demonstrated for a wide range of coal combustion residues, including fly ash [Kosson et al, 2009; Kosson et al, 2010]. The recent adoption of the same characterization leaching tests (pH dependence, percolation test, monolith leach test and compacted granular leach test) for waste, soil, and soil like materials (sludge, compost) and construction products (wide spectrum of products covered in CEN TC 351) illustrates that these methods have very widespread applicability. Here we address the leaching behavior of pulverized coal fly ash, of

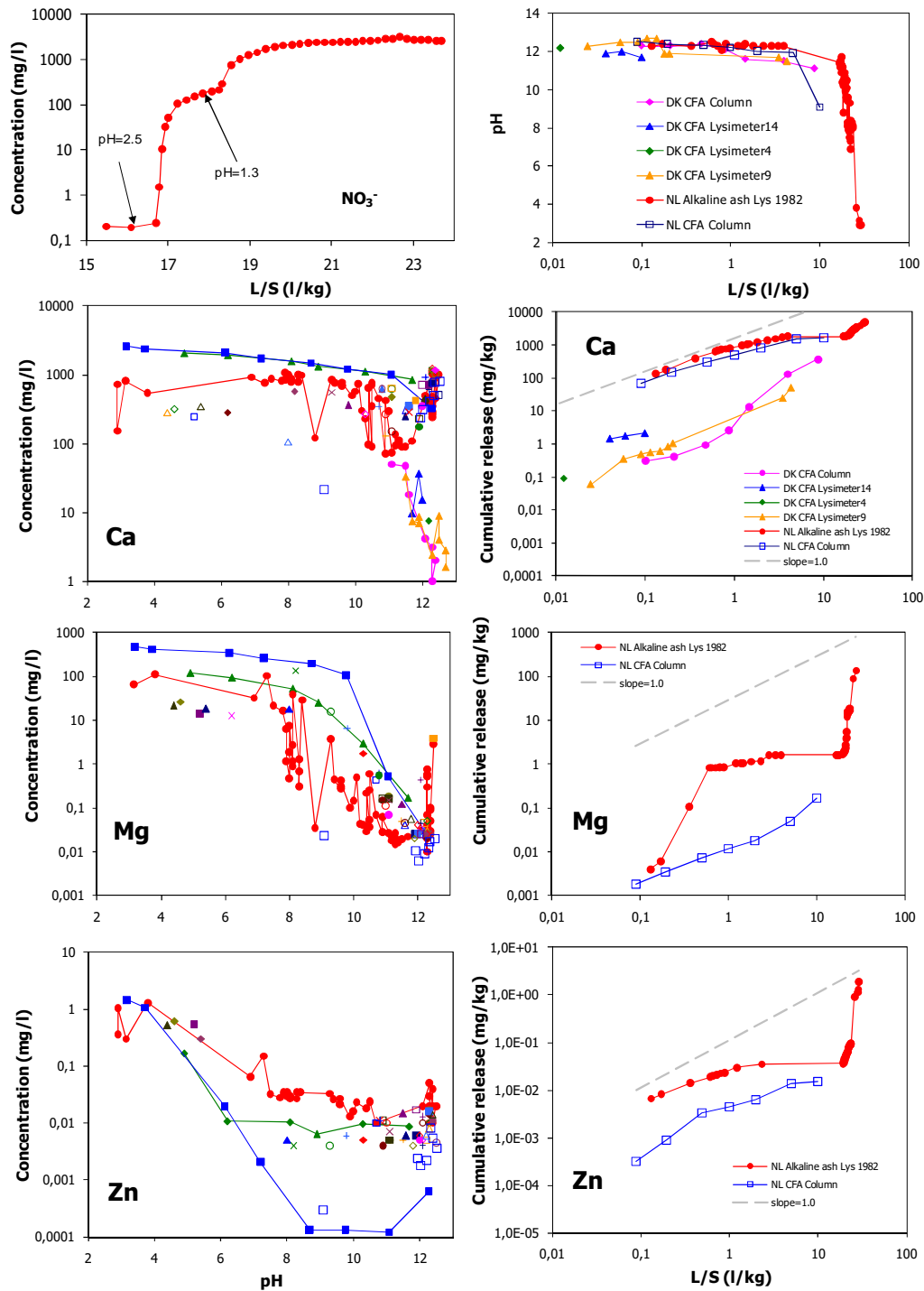
which the test results obtained with these methods are given in Figure 1. In the graphs pH dependence test data for coal fly ashes from different sources are given in the left graph. In this graph the data from the column test on alkaline ash carried out in 1982 [van der Sloot et al, 1982] are shown, as well as L/S=10 batch test data on 50 coal fly ashes from widely different coal origin (single data points). Data for Ca from large scale lysimeters operated in Denmark [Hjelmar, 1990] are given to illustrate the level of carbonation occurring in the collection tanks and not necessarily reflecting the condition in the lysimeter itself. This comparison illustrates that the leaching response to elution with an acidic solution largely corresponds with the release behavior as observed in a pH dependence leaching test on alkaline coal fly ash, also indicating local equilibrium conditions in the release from a column test.

### **Geochemical modeling**

In Table 1 the input parameters (forming the chemical speciation fingerprint, CSF) for the speciation modeling are given. These comprise the element availabilities (maximum concentration that was obtained in the pH dependence leaching test in the range pH 2-12), the mineral selection, the clay content, the quantity of reactive Fe- and Al- oxide surfaces and the reactive part of particulate and dissolved organic matter. In Figures 2 and 3 the model results for the alkaline coal fly ash is given in comparison with the original pH dependence test data. The percolation test data on the same ash are given for comparison. The model run is carried out both at L/S=10 L/kg and at L/S 0.2 L/kg (all other parameters remaining the same) to assess the validity of the mineral and sorption parameter selection for both a wide pH range as well as a wide L/S range. The L/S of 0.2 L/kg reflects pore water conditions in the column. The starting point for the modeling was the L/S=10 L/kg pH dependence test data. The optimization of the mineral selection is based on obtaining a prediction that provides the closest fit between model and actual test results. The low L/S modeling (0.2 L/kg) using the first fraction of the percolation test is to evaluate whether the same selection of minerals or a slight modification can simultaneously predict the release behavior at both low and high L/S under the assumptions that local equilibrium prevails and a representative and robust CSF has been selected. The selection of relevant minerals is reduced by selecting only those that represent at least one promille of the available amount of the element in the system (marked with \* in Table 1). A total of 24 elements are included in the chemical speciation modeling of the coal fly ash. The selection of minerals for the calculation run is wider than the actual minerals found to be of relevance.

In Figure 3 the model prediction is further detailed by the partitioning between dissolved and particulate phases for Zn and Cr. In general, the model describes the leaching behavior of the coal fly ash quite well, especially when it is realized that model is highly constrained because changes in input parameters may simultaneously affect the predicted behavior of several elements. It should also be noted that coal fly ash, which is generally considered devoid of organic matter, does contain small amounts of particulate and dissolved organic matter, which is apparently relevant for describing release behavior. For the low concentrations of Zn and Cr observed at pH 9 (0.06 µg/l) and pH 6 (5 µg/l), respectively, the small amount of DOC (around 1mg/l) is significant.

The geochemical model results for several major, minor and trace elements match quite well with the measured concentrations in the pH dependence test (Figure 2.)



**Fig 1. Comparison of pH Dependence (connected solid squares) and Column Test Data (connected open squares) on Alkaline Coal Fly Ash. For Comparison Purposes, the Data from a Column Test Using an Acidic Eluent [connected solid dots; van der Sloot et al, 1982] and Leachate Collected from Large Scale Lysimeters [Hjelmar, 1990] are Included in the Graphs**

**Table 1. Input Used in Geochemical Modeling of Coal Fly Ash.**

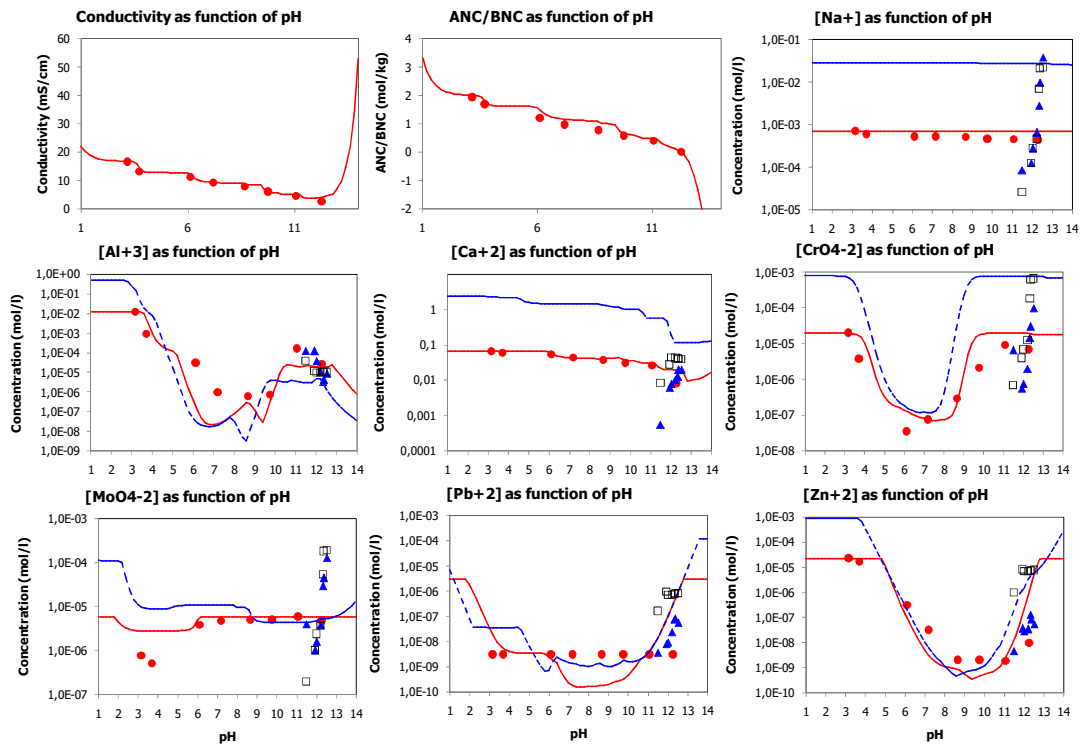
Material	Coal_fly_ash_NL	DOC/DHA data	pH	[DOC] (kg/l)	JHA fraction	[DHA] (kg/l)	Polynomial coefficients	
Solved fraction DOC		0.2	1.00	1.507E-06	0.20	3.014E-07	C0	-6.30E+00
Sum of pH and pe		14.00	3.17	1.400E-06	0.20	2.800E-07	C1	-3.036E-01
L/S		10.0784 l/kg	3.72	1.000E-06	0.20	2.000E-07	C2	9.460E-02
Clay		1.000E-02 kg/kg	6.12	1.400E-06	0.20	2.800E-07	C3	-1.106E-02
HFO		8.000E-03 kg/kg	7.20	1.200E-06	0.20	2.400E-07	C4	4.125E-04
SHA		2.000E-04 kg/kg	8.68	1.500E-06	0.20	3.000E-07	C5	0.000E+00
Percolation material	Coal_fly_ash_NL		9.77	5.000E-07	0.20	1.000E-07		
Avg L/S first perc. fractions		0.1420 l/kg	11.08	7.000E-07	0.20	1.400E-07		
			12.28	8.000E-07	0.20	1.600E-07		
Reactant concentrations	Reactant	mg/kg	Reactant	mg/kg	Reactant	mg/kg	Reactant	mg/kg
	Al+3	3.331E+03	CrO4-2	1.045E+01	Mn+2	9.731E+01	Sb[OH]6-	3.362E-01
	H3AsO4	9.233E+00	Cu+2	6.360E+00	MoO4-2	5.648E+00	SeO4-2	2.770E+00
	H3BO3	3.324E+02	F-	5.000E+01	Na+	1.659E+02	H4SiO4	6.112E+03
	Ba+2	1.613E+01	Fe+3	7.115E+01	Ni+2	6.684E+00	Sr+2	3.207E+02
	Br-	5.000E-01	H2CO3	1.000E+04	NO3-	5.000E+01	VO2+	3.917E+01
	Ca+2	2.623E+04	Hg+2	1.000E-02	PO4-3	7.283E+02	Zn+2	1.474E+01
	Cd+2	2.500E-01	K+	1.403E+02	Pb+2	6.376E+00		
	Cl-	1.050E+02	Mg+2	4.730E+03	SO4-2	1.678E+03		
Selected Minerals								
	* AA_2CaO_Al2O3_SiO2_8H2O[s]	AA_Magnesite	CaZincate	* Pb2V2O7				
	* AA_3CaO_Al2O3_6H	* AA_Tobermorite-I	Cd[OH]2[C]	Pb3[VO4]2				
	* AA_3CaO_Fe2O3_6H2O[s]	AA_Tricarboaluminate	Cerrusite	* PbMoO4[c]				
	* AA_Brucite	* Ba[SCr]O4[77%SO4]	Cr[OH]3[C]	Rhodochrosite				
	* AA_Calcite	* BaSrSO4[50%Ba]	Fluorite	Sb[OH]3[s]				
	* AA_CO3-hydroxalcalite	* Ca2Cd[PO4]2	* Manganite	Strontianite				
	AA_Fe[OH]3[am]	Ca2Cu[PO4]2	* Ni[OH]2[s]	* Tenorite				
	* AA_Fe[OH]3[microcr]	* Ca4Cd[PO4]3OH	* OCP	* Wairakite				
	* AA_Gibbsite	* Ca4Zn[PO4]3OH	Otavite	Willemite				
	AA_Gypsum	CaMoO4[c]	* Pb[OH]2[C]	Zincite				

For some elements (Ba, P and Si at pH 11) the rate of mineral dissolution may result in an under-saturated solution at test conditions [Dijkstra et al, 2006]. In many cases (Al, Ba, Ca, Fe, Si, Cu, Sr, Mg, P, Cl, K, Na), the modeled L/S=0.2 L/kg results agree well with the first fraction(s) from the column test.

The material properties listed in Table 1 in terms of element availabilities, Fe- and Al-oxide quantity, clay content, relevant minerals and reactive particulate (designated as solid humic acid - SHA) and dissolved organic matter (designated as dissolved humic acid DHA) forms a chemical speciation fingerprint (CSF) for the coal fly ash. This CSF is a good starting point for pH dependence test data and transport modeling for other coal fly ash samples since most minerals and reactive surfaces identified here are likely to be relevant.

### Percolation test results

Starting from the chemical speciation fingerprints (CSF) of coal fly ash as described above, model predictions have been carried out using a dual porosity chemical reaction/transport model [Dijkstra, 2007; Grathwohl and van der Sloot, 2007] to describe the release from a column test carried out with the same coal fly ash. The following additional parameters are needed for column modeling: the initial pH, the porosity of the packed column, the density of the material and the height of the packed column. A stagnant fraction of the total column content of 20 % is assumed. A diffusion distance between stagnant and mobile phase is assumed, which is calibrated on the release of non-interacting elements (usually salts like Na, Cl).

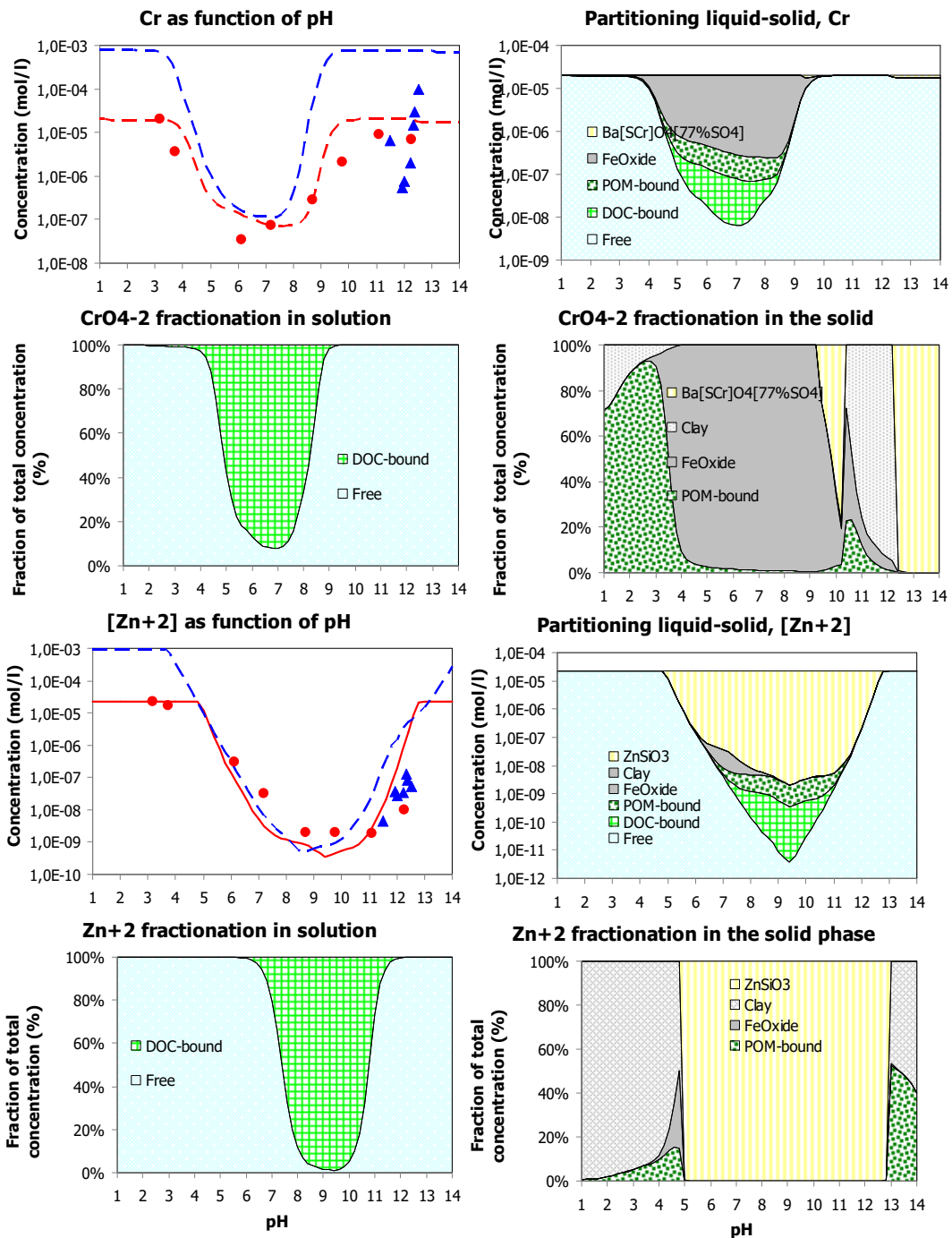


**Fig. 2. Measured and Predicted Leaching Behavior of Major, Minor and Trace Elements as a Function of pH in the Coal Fly Ash. Solid circles: pH dependence test; triangles: percolation test data; solid line: prediction at L/S=10; dotted line: prediction at L/S=0.2; open squares: prediction of percolation test data.**

Once this parameter is fixed the initial pH is adjusted to be able to predict the pH response as measured. In the model approach local equilibrium dictated by the mineral and sorptive phases as determined in the modeling of pH dependence test (Table 1) is assumed. Initial aqueous concentrations throughout the column are assumed to be the same and at equilibrium with the solid phase. In most cases, the leachant is demineralized water, which is assumed to contain negligible amounts of the constituents of interest. A constant eluant flow rate is assumed for the duration of the test.

In Table 2 the model parameters for the column prediction of coal fly ash leaching are given. As a prediction of DOC from the organic matter content of the solid is not (yet) possible, we have used the DOC data as measured in the percolation test. These data have been corrected to obtain the reactive fraction of DOC (DHA) relevant for metal interaction. A power function fit is used based on the equation:  $DHA_{L/S} = q_2 + q_0 \cdot e^{-q_1 \cdot L/S}$ , which gives at present the best possible description for DHA at intermediate L/S values necessary for modeling.

Figure 4 provides representative results of column modeling for a highly soluble salt (Na) and an element with highly pH dependent leaching (Mg). The result of salt wash-out is described well as indicated by Na. The prediction of pH is quite acceptable, however, at L/S=10 L/kg the pH is much lower than predicted.



**Fig. 3. Partitioning for Zn and Cr from Chemical Speciation Modeling of Coal Fly Ash Using pH Dependent (TS 14429; solid dots) and Column (TS 14405; triangles) data. Line and dotted line: model predictions at L/S=10 L/kg and L/S=0.2 L/kg respectively. Partitioning in the solution and in the solid is given.**



This is most likely related to the test conditions, where the eluate is in contact with the atmosphere for a long time during the last step of the test<sup>1</sup>. This pH reduction does not reflect the behavior of the ash in the column and stresses the need to avoid CO<sub>2</sub> uptake in the collection vessel. The release of Mg further indicates that this is a side effect of the test, as Mg does not increase in leachability as would be expected based on a significant pH change. Also important is the pattern of release as that reflects the release mechanism (wash-out, solubility limitations, etc).

**Table 2. Input for Chemical Reaction Transport Modeling of Coal Fly Ash.**

Case	Coal fly ash NL					
Solved fraction DOC	0.2	[DOC/DHA data]				
Sum of pH and pe	14.00	L/S	[DOC] (kg/l)	DHA fraction	[DHA] (kg/l)	
Clay	1.000E-02 kg/kg	0.09	1.520E-05	0.20	3.040E-06	
HFO	8.000E-03 kg/kg	0.19	7.700E-06	0.20	1.540E-06	
SHA	2.000E-04 kg/kg	0.50	3.000E-06	0.20	6.000E-07	
Porosity Fraction	0.35	0.99	1.400E-06	0.20	2.800E-07	
Density	1.8 kg/l	1.99	6.000E-07	0.20	1.200E-07	
Initial ph (solid)	12.5	5.02	6.000E-07	0.20	1.200E-07	
Initial ph (liquid)	1.5	10.03	1.000E-06	0.20	2.000E-07	
Column length	25 cm	Curve fitting coefficients				
Rel. stagnant volume	20 %		Q0	2.625E-06		
Eff. diffusion dist.	3 cm		Q1	2.500E+00		
Initial water concentrations			Q2	1.000E-07		
Reactant	mol/l					
All	1.000E-09					
H2CO3	3.000E-06					

SHA = humic acid fraction in the solid phase; DHA = humic acid fraction in dissolved organic matter; mineral selection is the same as in Table 1.

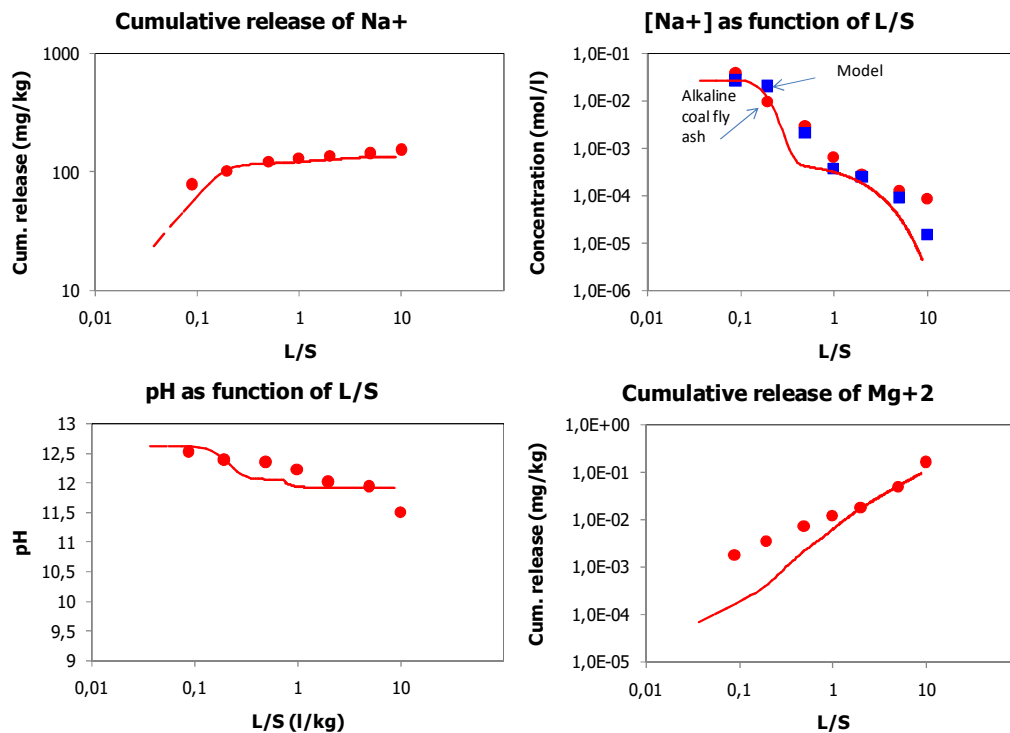
The prediction of trace element release from a percolation test is not easily accomplished when all elements (>25) need to be matched simultaneously. In most cases elements are modeled one by one, but this may lead to conflicting descriptions for the same sample. When the pH dependence test data are not adequately matched, then the prediction of release under the low L/S conditions in the field is a big challenge. As leaching of trace elements often represents only a minor fraction of the total elemental content, a deviation between modeling and measurement of a factor of 3 to 5 is often acceptable and the resulting model is useful for evaluating potential changes in elution of specific elements under different chemical conditions (e.g., pH, redox). However, the magnitudes of these uncertainties must be recognized as part of any decision making process. The modeling also provides insight in the concentration profiles along the column in solid and liquid phase. This type of modeling can be very useful in case pore water samples have been taken along the length of the column.

## CONCLUSIONS

Understanding the mechanisms controlling release and identifying the mineral and sorptive phases likely to control release allows drawing conclusions on possible long term leaching behavior. In this context it needs to be stressed that the result from any one of the leaching tests applied here cannot be compared directly to surface water and groundwater criteria, as

<sup>1</sup> In more recent testing, flushing with nitrogen or a water lock (i.e., bubbler) has been used to minimize absorption of atmospheric carbon dioxide during testing.

the attenuation/dilution relevant for the specific scenario is not factored in. Separate models have been developed to describe impact on soil and groundwater starting from a decreasing source term. A model to describe release in multilayer systems is currently tested.



**Fig. 4. Measured (solid dots) and Predicted Release (line: continuous concentration change; open circles: calculated concentrations corresponding with the measurement points) in a Column Test (TS 14405) on the Coal Fly Ash.**

Multi-element, multi-phases geochemical modeling has led to improved insight in possible release controlling phases. There are gaps between data and model predictions for a number of constituents, which are either due to missing thermodynamic data, as yet unidentified mineral phases or kinetics of dissolution and precipitation. Dissolution kinetics have been shown to be of relevance for evaluating the test data used in this work. Modeling uncertainty must be considered when deciding what level of agreement between measurement and prediction is sufficiently accurate for subsequent decision processes.

The advantage of the integral approach applied here is that more guidance can be derived as to which factors require further investigation. The potential to predict behavior under conditions that have not yet been tested before (e.g. low L/S, imposed redox condition, increased contamination) can provide important insight into how to design verification experiments.

From the modeling it has become very clear that the interactions between major, minor and trace elements forms a very significant limitation for studies in which only a limited set of elements is evaluated. Evaluation of a complete set of major elements is essential because they dictate the leaching conditions that are imposed on the trace constituents of concern. The

parameter settings for the coal fly ash used here forms a good starting point for modeling other ashes as well as a basis for more detailed chemical reaction transport modeling by LeachXS™ - Orchestra.

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