REVISION OF THE DUTCH BUILDING MATERIALS DECREE: ALTERNATIVE EMISSION LIMIT VALUES FOR INORGANIC COMPONENTS IN GRANULAR BUILDING MATERIALS

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ABSTRACT: This presentation shows how new emission limit values for inorganic components in stony and earthy building materials were derived for the revision of the Building Materials Decree in the Netherlands. Three key factors determine the emission limit value: (1) the environmental compartment under consideration with its dimensions, (2) the desired level of environmental protection and (3) the scenario applying to the intended use of the construction material. Emission limit values have been derived here from modeled concentrations in soil, groundwater and surface water. Two models were used for the vertical reactive transport through the soil profile and for retardation of components in the soil profile. The PEARL model was used for binding described by a linear distribution coefficient approach (Kd). The ORCHESTRA modeling framework was used for modeling binding that takes speciation and surface complexation into account. For surface water, a dilution factor was computed assuming a certain mixing zone, dependent on the size of the surface water. The consequences of the proposed alternative emission limit values have been estimated on the basis of information in a database containing results of leaching tests for construction materials. The alternatives proposed form the basis for a political decision-making process, where environmental protection was balanced with other socio-economic concerns.

INTRODUCTION

Since 1995 the Netherlands has a Building Decree based on the potential impact of materials on the environment. The decree gives quality criteria for the application and re-use of stony materials and earth used as building materials. No difference is made between primary materials, secondary materials and waste materials. The decree is applicable in case these materials are used in constructions where they are in contact with rain, surface water and groundwater (e.g. in embankments, road buildings, outside walls of buildings, foundations and roofs). The backgrounds of this regulation are described by Eikelboom et al. (2001). After 10 years of experience with the regulation of the environmental quality of building materials several bottlenecks/drawbacks were encountered in the enforcement of the regulation. The revision was necessary because the regulation of 1995 has led to a complicated administration that involves high costs. Due to several amendments with exemptions the regulation has become quite untransparent. The aim of the revision is to offer a simplified regulation, containing a consistent set of emission limit values. Conditions of the emission limit values are protection of soil and groundwater quality with minimal restrictions for the re-use of secondary materials. Many of the building materials involved are secondary building materials or waste materials such as recycled asphalt, MSWI-bottom ash, blast

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furnace slag etcetera. Re-cycling of these materials decreases the amount of waste and also prevents unnecessary use of primary materials such as sand and gravel.

Part of the simplification in the new regulation is achieved by the introduction of emission limit values that can be directly compared with compliance test results. The standard compliance test for granular building materials is the column test NEN 7343 (NEN, 1995), which is almost similar to CEN/TS 14405 (CEN, 2004). In this test, the cumulative leaching and the leaching rate of inorganic components in granular materials are determined. The total amount of percolated water is 10 times the mass of the solids in the column (L/S ratio = 10). A lot of monitoring data obtained with this method is available and could be used to assess the potential economic consequences of proposed emission limit values for individual building materials. The new regulations are included in the so called Soil Quality Decree and will come into force as from July 1st 2008.

Emission limit values have been derived in order to comply with soil, groundwater and surface water quality criteria. In this paper we focus on the derivation for the soil-groundwater system, which also appears to be protective for the surface water also. Computations for surface water as well as the method and results for monolithic building materials can be found in the full report of Verschoor *et al.* (2006) and are not presented in this paper.

The inorganic components involved in this study are antimony (Sb), arsenic (As), barium (Ba), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), mercury (Hg), lead (Pb), molybdenum (Mo), nickel (Ni), selenium (Se), tin (Sn), vanadium (V), zinc (Zn), bromide (Br), chloride (Cl), fluoride (F) and sulphate (SO₄).

OUTLINE OF THE METHOD FOR DERIVATION OF EMISSION LIMIT VALUES

The emission limit values are calculated in six steps, which are visualized in figure 1.

- 1. a source term is computed which describes the release pattern of substances from the building materials. The release pattern is a result of properties of the substance in combination with properties of the material and dimensions of the construction. In our approach an average release pattern for each substance is used, based on measured data in many building materials.
- 2. two dynamical models are applied which compute substance concentrations variable with time and depth of the soil profile. A transport model using linear distribution coefficients (PEARL 2.2.2) and a model that includes speciation and surface complexation (ORCHESTRA) are applied. Both model codes are freely available through the internet (see references).
- 3. the resulting concentrations in groundwater and are compared with corresponding compliance values (quality criteria).
- 4. the source term is adjusted in such a way that computed soil and groundwater concentrations exactly equal the compliance



Figure 1 Outline of the calculation of emission limit values (CV=compliance value)

values in soil and groundwater. The adjusted source term represents a critical release. Two different source terms can be the result; one derived from groundwater compliance values and one from soil compliance values.

- 5. the adjusted source terms are transformed into emission limit values.
- 6. the most stringent emission limit value is protective for the whole environment.

ENVIRONMENTAL QUALITY CRITERIA

The compliance values used for the derivation of emission limit values are set at the Maximum Permissible Concentration (MPC). At this level ecosystems are not significantly affected by chemical exposure. For the inorganic substances occurring at natural background concentration, the MPC is transformed to a maximum permissible addition (MPA) using the following equation MPC = MPA + background. The method is explained by Struijs *et al.* (1997). In table 1 the MPC values for the substances involved are listed. No MPA values are available for Cl, Br, F and SO₄ in soil. As a consequence emission limit values for Cl, Br, F and SO₄ are determined by the effect on groundwater only. Other criteria have been also elaborated, such as target values, serious risk values and drinking water limits. The results of it are not presented here, but can be found in the full report of Verschoor *et al.* (2006).

Component		MPA _{soil} (mg/kg)	MPA _{groundwater} (μg/L)						
Antimony	Sb	0.53	6.2						
Arsenic	As	0.9	24						
Barium	Ba	180	29						
Cadmium	Cd	0.79	0.34						
Chromium	Cr	0.38	8.7						
Cobalt	Co	2.4	2.6						
Copper	Cu	3.4	1.1						
Mercury	Hg	1.9	0.23						
Lead	Pb	55	11						
Molybdenum	Мо	39	29						
Nickel	Ni	0.26	1.9						
Selenium	Se	0.11	5.3						
Tin	Sn	34	20						
Vanadium	V	1.1	3.5						
Zink	Zn	16	7.3						
Bromide	Br	n.a.	8,000						
Chloride	CI	n.a.	200,000						
Fluoride	F	n.a.	1,500						
Sulphate	SO ₄	n.a.	100,000						

 Table 1. Quality criteria for soil and groundwater in The Netherlands

n.a. = not available

Release Pattern – Source term

The release pattern of substances from granular materials in constructions can be described by equation 1.

$$I_{soil} = E_{construction} = \frac{E_{material} * d_b * h * (1 - e^{-\kappa * \frac{N_i * Y}{d_b * h}})}{(1 - e^{-\kappa * 10})} \quad (\text{mg.m}^{-2} \text{ in Y years})$$
(1)

in which:

I _{soil}	source term (mg/m ² per Y years);
Econstruction	emission from a construction (mg/m ² per Y years);
E _{material}	measured emission of material in a column test at $L/S = 10 (mg/kg)$;

db	bulk density of a material (default 1550 kg/m ³)
h	height of material in a construction (m);
κ	kappa, rate constant for release ;
Ni	effective infiltration of rain (mm/years);.
Y	time (years)

For granular building materials constructions with heights of 0.2, 0.5, 0.7 and 2 meters are simulated with and without isolation on top. For isolated constructions an infiltration of 6 mm/year was assumed and for open constructions an infiltration of 300 mm/year. The results for building materials under isolation are not presented in this paper, but can be found in Verschoor *et al.* (2006). Kappa is a rate constant for the release of substances and is obtained from column experiments. Average kappa values have been obtained from a large database with all different types of granular building materials. Kappa values are listed in table 2, together with some other important model input parameters. As an example, two figures are presented to illustrate the effect of kappa and construction height.



Figure 2. Effect of release rate constant kappa and construction height on the cumulative release of a substance to the soil.

Figure 2 shows that substances or materials with higher kappa values tend to be released faster and as a consequence the material is depleted at an earlier stage. Materials used in thicker layers exhibit a prolonged release of substances, resulting in a higher cumulative release. A description of all simulated scenarios for application of building materials has been presented at Consoil 2005 (Lijzen *et al.*, 2005).

MODELING BOUNDARIES

The simulation time was set at 100 years. The soil profile consists of 1 m unsaturated soil and 1 m saturated soil. The average groundwater level is 1 m below soil surface (bss), being an average for the Dutch situation.

Daily groundwater concentrations have been computed at several depths. These data are elaborated to annual average concentrations over the upper 1 m layer of the groundwater. The maximum annual concentration within a period of 100 years is used as an endpoint for derivation of the emission limit value in the groundwater.

Daily soil concentrations have been computed at several depths. The annual average concentration over 1 m soil at the end of the simulation (100 years) is used as an endpoint for derivation of the emission limit value in the soil.

MODELING BY A LINEAR DISTRIBUTION COEFFICIENT APPROACH (KD)

The Kd approach has been applied using PEARL 2.2.2 (Leistra *et al.*, 2000). The major input parameter is the adsorption coefficient (K_d). This parameter assumes a constant

ratio between sorbed and dissolved substance. The K_d is dependent of soil type - caused by differences in pH, organic matter, clay content and ferrous and aluminium(hydr)oxides - and can vary orders of magnitude. For Cu, Cd, Pb, Ni and Zn relations of Kd with these soil parameters are amongst other described by Römkens et al. (2004), Buchter et al. (1989) and Sauvé et al. (2000). However for the other substances, the K_d cannot be predicted by soil properties. The Netherlands has a great variety of soil types, ranging from poor sandy soils, to peat and marine clays. The choice of the value of the K_d is crucial for the computed soil and groundwater concentrations, and for the ultimate emission limit values. In order to guarantee a safe use of materials on all soil in the Netherlands, realistic worst-case K_d -values are selected from a collection of literature data. This implies that relatively low K_d-values have been selected for groundwater protection. In that way relatively high groundwater concentrations are predicted and as a consequence protective emission limit values are computed. For soil protection, relatively high K_d-values have been selected, resulting in relatively high soil concentrations and relatively low emission limit values. The 10- and 90-percentile of the collected K_d-values are assumed to be representative for Dutch soils and to generate realistic worst case concentrations in respectively groundwater and soil. The resulting set of K_d-values is listed in table 2. The adsorption is assumed to be reduced deeper in the soil profile. For the soil layer at 50-70 cm bss the K_d has been reduced by a factor 3 and deeper than 70 cm bss the K_d has been reduced by a factor 10.

Component		Карра	10-p K _{ads}	90-p K _{ads}
Antimony	Sb	0.04	10	550
Arsenic	As	0.01	280	17000
Barium	Ba	0.17	530	14000
Cadmium	Cd	0.32	15	1700
Chromium	Cr	0.25	1200	50000
Cobalt	Со	0.13	30	1100
Copper	Cu	0.27	30	830
Mercury	Hg	0.14	350	1100
Lead	Pb	0.18	400	47000
Molybdenum	Mo	0.38	140	1400
Nickel	Ni	0.26	46	1500
Selenium	Se	0.16	200	2000
Tin	Sn	0.1	130	10000
Vanadium	V	0.04	70	2200
Zink	Zn	0.28	11	550
Bromide	Br	0.508	0.3	15
Chloride	CI	0.65	0.125	0.5
Fluoride	F	0.26	75	300
Sulphate	SO ₄	0.33	3.75	15

 Table 2
 Major input parameters for release and transport modeling

MODELING SPECIATION AND SURFACE COMPLEXATION USING ORCHESTRA

A speciation and surface complexation model is applied for verification and validation of modeling with linear distribution coefficients. The ORCHESTRA model approach is able to compute adsorption of substances to several organic matter fractions (solid as well as dissolved organic matter fractions), clay and ferrous-oxides. The uncertainty of the intrinsic thermodynamic constants for these specific interactions is considerably less than the variability of K_d -values in soils. An advantage is that the adsorption can be computed for soils with different binding properties. In this approach, non-linearity of adsorption, competition between substances for adsorption sites and the influence of pH is accounted for. Adsorption constants for specific and non-specific binding to organic matter have been taken from Milne et al. (2003). For adsorption to ferrous and aluminium(hydr)oxides the 'Generic Two Layer Model' of Dzombak and Morel (1990) was implemented. Adsorption to clay is simulated by a simple Donnan-model, assuming a CEC of 0.25 eq/kg clay. For more detail on the Orchestra modeling approach is referred to Dijkstra *et al.* (2004) and Meeussen (2003).

The substances are assumed to be simultaneously present in the source term. Macro-elements such as Na and Ca are added to the source term, because they are the major counter-ions of respectively Cl and SO_4 in many building materials. They are added in the same amounts and with the same kappa values as Cl and SO_4 .

In order to represent a wide range of conditions in The Netherlands, an average sand, clay and peat soil have been selected from a large database with 465 real Dutch soil profiles (Kroon *et al.*, 2001). The properties of these soil types are shown in figure 3.



Figure 3 Properties of selected soil profiles. (DHA=dissolved humic acid, SHA = solid humic acid, SHFO = sum of ferrous- and aluminium (hydr)oxides)

RESULTS

In figure 4-left typical concentration patterns in the soil are shown, dependent of the sorption of a compound using the linear distribution coefficient approach. The endpoint (shown as dots) for the derivation of the emission limit value is the concentration after 100 years.

In figure 4-right typical concentration patterns in the groundwater are shown. The endpoint (shown as dots) for the derivation of the emission limit value is the maximum concentration within a period of 100 years.



Figure 4. Concentration pattern of substances in soil (left) and groundwater (right) depending on K_d (for height 1 m and kappa 0.5). Dots are modeling endpoints.

In figure 5 typical concentration patterns in the groundwater are shown for a variety of application heights and kappa values. It is shown that kappa and application height act together. It is the product of kappa times application height that determines the shape of the concentration curve. The endpoint for the derivation of the emission limit value is the maximum concentration within a period of 100 years.



Figure 5. Concentration pattern of substances in groundwater depending on kappa and application height (for $K_d = 20 l/kg$). Dots are modeling endpoints.

Aiming at a simplification of the regulation for building materials, the government has decided not to distinguish between application heights. The emission limit values derived from an application height of 0.5 m were proposed for the new regulation (Dutch Soil Quality Decree).

In table 3 modeling results of the linear distribution coefficient approach and the speciation and surface complexation approach are compared. It appears that both models are quite comparable when it comes to calculations for very mobile substances, such as Sb, Br, Cl and SO₄ and for very immobile substances such as As, Cr, Pb, Mo, Se and Sn.

For some substances a difference in prediction of the mobility is observed between the two models. That is the case for Ba, Hg and V. This could be explained by the absence of reliable data for K_d and effects of nonlinearity, speciation/complexation and competition. For vanadium there is apparently a good agreement, however, the Kd- model results indicate that groundwater is the most vulnerable compartment, whereas the speciation model predicts that the soil is the most vulnerable compartment. By a coincidental combination of compliance values in both compartments the final emission limit values are almost equal.

The last group of substances, substances with an intermediate mobility - Cd, Co, Cu, Ni, Zn and F - shows considerable disagreements in emission limit values, whereas the agreement in calculated maximum concentrations is quite good. It appears that by cutting off the simulation time at 100 years the maximum concentration in the groundwater within 100 years has become very sensitive to relatively small differences in mobility.

Table 3. Emission limit values (mg/kg) for granular building materials with an application height of 0.5 m. Values printed in bold are determined by soil and values printed in italic are determined by groundwater. Grey shading indicates a good agreement between linear distribution (K_d) and speciation/surface complexation model results. Reasons for agreement or disagreement are given in the last column.

	Kd- approach	Speciation/surface complexation	Reason for (dis-)agreement
Sb	0.08	0.03	High mobility in both models
As	0.5	0.5	Low mobility in both models
Ва	290	1.6	Large difference in predicted mobility
Cd	0.005	0.3	Difference is magnified by cut-off time of 100 years
Cr	0.7	0.6	Low mobility in both models
Co	0.06	0.5	Difference is magnified by cut-off time of 100 years
Cu	0.03	3.4	Difference is magnified by cut-off time of 100 years
Hg	2.9	0.8	Large difference in predicted mobility
Pb	91	77	Low mobility in both models
Мо	75	64	Low mobility in both models
Ni	0.07	0.4	Difference is magnified by cut-off time of 100 years
Se	0.2	0.2	Low mobility in both models
Sn	44	39	Low mobility in both models
v	0.7	0.8	Large difference in predicted mobility. However due to a combination of compliance values in soil, groundwater and the cut off time of 100 years, the emission limit values are coincidently comparable.
Zn	0.08	5.5	Difference is magnified by cut-off time of 100 years
Br	24	24	High mobility in both models
CI	480	480	High mobility in both models
F	370	9.4	Difference is magnified by cut-off time of 100 years
SO ₄	640	640	High mobility in both models

In table 3 it is shown that the emission limit value for some substances is determined by the groundwater and for other substances by the soil. It depends on the mobility of the substance and the level of the compliance values in soil and groundwater which compartment is the most vulnerable within the time frame of 100 years. This phenomenon is illustrated in figure 6. The critical release is the endpoint of step 4 (see outline of the method). It is the amount of substance that can be released to the soil in 100 years, without exceeding soil or groundwater compliance values. The figure shows that at lower K_d -values – generally lower that

approximately 100 L/kg – the groundwater is the critical compartment. The final emission limit value will thus be determined by the groundwater. At higher K_d-values the soil is the most critical compartment and as a consequence the emission limit value will be determined by the soil. In general the lowest line (the lowest critical release) determines the emission limit value. It is dependent on the realistic range of adsorption coefficients whether the observed sensitivity is relevant. For example, for Pb the realistic K_d-range is on a level that the emission limit value is always determined by the soil compartment (given a simulation time of 100 years). This value is independent op de K_d-value of Pb. For cadmium the range in K_d is such that in certain soil types the groundwater will be most critical while in other soil types the soil will be more critical. For Br no critical release in soil has been computed while there is no compliance value for Br in soil. Whereas for cadmium the effect on the critical release – and consequently on the emission limit value – is a factor 1000, it is for Sb only a factor 10. This is a result of the level of the compliance values in soil and groundwater, relative to each other. Sensitivity analyses for all the substances are given by Verschoor *et al.*, (2006).



Figure 6. Examples of the effect of mobility (Kd) on the critical release and critical compartment. Straight lines represent critical release in the groundwater and dotted lines represents critical release in the soil. On the x-axis the realistic K_d-range for the substance is shown. Dots represent the 10-percentile, the geometrical mean and the 90-percentile K_d-value.

Consequences

Before the proposed emission limit values were implemented in the Soil Quality Decree, consequences of the values were investigated using 60,000 test data of column experiments with granular building materials (de Wijs and Cleven, 2008). From these test data it is calculated how many batches of building materials exceeded the proposed values. The results were reason for adjustment of emission limit values for most of the substances.

In some cases the modeled values were unnecessarily or undesirably liberal. This was the case for Br and Mo, Hg, Pb and Sn. For these substances the emission limit values were set at a more stringent value than proposed.

On the other hand, some proposed values could potentially cause too many restrictions in the use of secondary building materials, and therefore the emission limit value was set at a higher level than proposed by environmental modeling. This concerns Sb, As, Cl and SO₄.

For a number of substances the twp environmental models did not agree on the level of the emission limit values. In that case an economically feasible value in between is chosen, depending on values for these substances in the old Building Materials Decree and other regulations, for example landfill directives. That is the case for Ba, Cd, Cu, Zn and F.

For Co and Ni the two modeled values also differed significantly. For Co and Ni the highest modeled values, obtained by the speciation/surface complexation model, are included in the new regulation.

For Cr and Se the values proposed by the two models are identical and taken over in the new regulation.

Table 4 Compliance of building materials with new emission limit values, based on test data in 2003-2004. Values represent the percentage of batches that exceeds the emission limit value. Printed in red are exceedings >20%, orange exceedings 5-20% and green exceedings < 5% of the batches.

		8				0															
	Sb	As	Ва	Cd	Cr	Co	Cu	Hg	Pb	Мо	Ni	Se	Sn	V	Zn	Br	CI	F	SO4		
New emission limit value (mg/kg)	0.16	0.9	22	0.04	0.63	0.54	0.9	0.02	2.3	1	0.44	0.15	0.4	1.8	4.5	20	616	55	1730		
Recycled asphalt														5					11		
Asphalt			50																		
MSWI-bottom ash	77						94			52							96		37		
Concrete CDW aggregate																					
Armour stone																					
Residues from drinkwaterproduction																					
Puverished fly ash			72		68					100		29		10					6		
Furnished bottom ash																					
ELO-slag														20							
Flugsand																					
Phosphorus slag													20				10	10			
Phosphorus slag (hydraulic bond)													17				36				
Blast furnace slag mixture																			17		
Hydraulic mixed CDW aggregate																					
Lava stone														20							
LD-mixture																					
LD-slag																					
Mixed CDW aggregate																					
Masonry CDW aggregate																					
Mining stone		13															50				
Natural stone																					
Plaster board			10		10										10						
Crushed stone																			13		
Cleaned recycled asphalt (TAR)																					

The final set of emission limit values for granular building materials included in the Soil Quality Decree is listed in table 4. In the table is also shown what fraction (percentages) of tested batches of building materials exceeds the emission limit values. MWSI-bottom ash and furnished bottom ash have difficulties to comply with the new emission limit values. It is suggested that these materials should be applied in such a way that infiltration of water is limited (isolated conditions). Computations for a scenario with isolation (6 mm infiltration per year) showed that none of the building materials exceeds criteria then (Verschoor *et al.*, 2006).

CONCLUSIONS AND DISCUSSION

It is shown to be possible to apply realistic, mechanistic models for the derivation of emission limit values for building materials. These models take into account a distribution of substances over liquid and one or more solid phases. The fact that these models have a very detailed output in time and in depth, raised the question on how to choose the modeling end points. After some trials with different depths, cut-off times and averaging windows the emission limit values were at last derived from annual average concentrations in 1 m soil and annual average concentrations in 1 m groundwater. For soil, the endpoint is taken at the end of the simulation time, 100 years. For groundwater, the maximum annual concentration within a 100 year window is taken. On the one hand, the averaging method levels out many differences between the linear distribution model and the speciation/surface complexation model, thereby offering a robustness to the assessment. On the other hand, cutting off the simulation time after 100 years magnified small differences in maximum concentrations to large differences in emission limit values for substances with an intermediate mobility (Cd, Co, Cu, F, Ni, and Zn).

It is concluded that both models - linear distribution as well as speciation/surface complexation - are suitable for modeling fate and behaviour of substances in soil and groundwater. Each method has its advantages and drawbacks. Calculations using the Kd approach are in general easier to perform and to understand. However it is not possible to account for non-linearity of adsorption, specific interactions in mixtures of substances, to account for pH, to account for adsorption to dissolved organic matter and to account for precipitation reactions. Speciation/surface complexation models can account for all these processes, but require more input parameters which are, however, in many cases available (e.g., pH, solid and dissolved organic matter content, clay content).

In case of generic scenarios with many assumptions it is more difficult to qualitatively assess whether the result is representative for a certain level of protection. This can be overcome by performing sensitivity analyses. Because the availability of input parameters is improving by the development of digital databases with soil profile information, a major drawback hampering generic application of speciation models will be overcome. Moreover, computer hardware is improving, so that calculations with complete soil databases and iterative procedures in the calculation routines are no longer a reason to turn down these type of methodology. Still, performance of speciation modeling requires more expert knowledge and skills than of the linear distribution coefficient approach . In this respect, it is essential that user-friendly, non-expert versions of these modeling applications are made available (e.g., through LeachXS (http://www.leachxs.com/lxsdll.html)).

The economic feasibility appeared to be a major and in many cases a decisive reason for the choice of the final emission limit value. The fact that the final emission limit values are higher than the proposed ones does not mean that soil or groundwater are at risk on a large scale. It means that the level of protection is not 90%, as was assumed by the choice of the range in soil types or K_d values, but is less than 90%. As is shown in figure 6, higher levels of critical emission values can, for some substances still be safe in many situations. The emission limit values were derived from ecotoxicological risk levels. However, it should be emphasized that for some substances these risk levels are based on very poor datasets. Data poor substances have higher uncertainty ranges and therefore relatively stringent MPC values. It is recommended that more effort is put in research to improve ecotoxicological risk levels for data-poor substances.

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