

## **Verification of laboratory - field leaching behavior of coal fly ash and MSWI bottom ash as a roadbase material**

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### **ABSTRACT**

For an environmentally acceptable utilization of primary and secondary construction materials regulations have been developed in the framework of the Soil Protection Act (WBB) and the Clean Water Act (WVO). This has resulted in the Building Materials Decree.

An extensive field study has been carried out to determine the relation between predictions based on laboratory leaching tests and the release as established by measurements at applications of secondary materials in roadbase constructions. The study consisted of an extensive sampling program based on previous release estimates, an estimate of actual water management, leaching of the underlying sand layer as well as calculation and modelling of release based on these profiles. For many relevant elements a quantifiable release has been established after about 10 years of field exposure and for most elements the predictions based on laboratory tests agree with the field data fairly well.

### **Introduction**

For an environmentally acceptable utilization of primary and secondary construction materials regulations have been developed in the framework of the Soil Protection Act (WBB) and the Clean Water Act (WVO). This has resulted in the Building Materials Decree [1]. Based on leaching tests in the laboratory and their translation to practical situations, a theoretical relation has been established between the desired level of protection of soil and groundwater and the results of laboratory leaching tests. Since results of laboratory leaching tests cannot be translated directly to field conditions, RIVM developed translation factors and formulas, which are described in the Guidance to the Building Materials Decree. This concerns corrections for the temperature difference between lab and field, the degree of contact with water under field conditions, and the extrapolation to longer time scales. For instance to the reference point of 100 year as used in the concept of marginal burdening of soil. For road construction applications the question is here to

what extent laboratory leaching data can be correlated with field observations taking into account the correction factors specified above. The CROW Coordination Committee Construction Materials and Environment, originally the Steering Committee Environmental Hygiene and Road Construction, supported in 1993 a research plan prepared by INTRON and ECN to provide a relation between predictions of release based on laboratory test data using the calculations specified in the Building Materials Decree and measured impact in applications in practice. Four characteristic situations in road construction have been selected to verify the degree of agreement between predicted emissions using the prescribed calculations and the field situation after about 10 years of field exposure. In these road base applications coal fly ash and MSWI bottom ash have been used as stabilization layer.

The aim of the study was the verification of the predicted release from secondary materials with measured immission in practice in the four selected road construction applications using results from laboratory leaching tests.

The following approach was followed to study the relation between the predicted immission on the basis of laboratory data and formula's specified in the Building Materials Decree with the measured impact under field conditions:

- extensive sampling at the four selected road construction applications,
- determination of release under field conditions,
- prediction of release according to the Regulation of Construction Materials,
- comparison of predicted release with measured release under field conditions,
- evaluation of observations.

The determination of release in practice has been based on:

- I. The concentration decrease in the construction material relative to the starting situation.
- II. The net concentration increase in the underlying soil and collected leachate.
- III. The difference in leaching between fresh and „field aged“ construction material.

The release in practice as derived from these different complementary methods is compared with the calculated release based on predominantly percolation controlled system or a predominantly diffusion controlled system. To provide a sufficient statistical basis for the comparison 10 cores were sampled for each of the four road base constructions in the study. The prediction of release was carried out in accordance with the formulas specified in the Building Materials Decree. Geochemical reaction / transport modelling has been carried out to support the interpretation of the field measurements. Figure 1 gives a view of the approach described above.

#### Selection of sites

Four conditions in road base application were selected. In all cases, secondary materials have been applied as stabilization layer on a drainage layer of sand. As cover materials, asphalt concrete, sand and road construction bricks have been utilized.

The selected applications (code) are:

**CA**, coal fly ash cement stabilization under asphalt.

**CZ**, coal fly ash cement stabilization under sand.

at the Coloradoweg (Province of South Holland, Municipality of Rotterdam, year of construction: fall 1983).

**VF**, Feniks under road construction bricks.

**VA**, MSWI Bottom ash under road construction bricks.

and the Vondelingenweg (Municipality of Rotterdam) (year of construction: mid 1986)

The coal ash cement stabilization at the Coloradoweg is a stabilization of pulverized coal ash with blast-furnace slag cement (9 %). Feniks is a stabilization of MSWI bottom ash (from Roteb, Rotterdam), with sand and blast-furnace slag cement in the following proportions, 60 %% MSWI bottom ash, 40% sand and 3 % cement The MSWI bottom ash consists of sieved, aged MSWI bottom ash from ROTEB and applied as granular material.

### Sampling

The release of constituents from the construction materials containing secondary materials has been established by measuring the decrease in concentration in the construction material itself as well as from the concentration increase in the underlying sand layer. By taking cores through the stabilization layer well into the underlying soil using a core-catcher (PVC inter tube in the stainless steel drilling core) an undisturbed profile of construction material and adjacent soil can be obtained. The PVC cores were capped, transported and stored.

In the laboratory, the cores have been sliced in the full length and subsequently sliced in segments for chemical analysis. For each core (10 per application) this has resulted in concentration profiles in the construction material and the adjacent soil. These data have been used to determine the release from the secondary construction material in the exposure period of about 10 years.

For each application samples have been taken from the field exposed construction material to assess its residual leaching behavior in a column test (NEN 7343) [2] after aging/weathering.

For the prediction of release, the predominant transport mechanism of constituents is a crucial property that needs to be addressed to allow a comparison of lab test data with field measurements.

The leaching mechanism - percolation or diffusion - can be identified for practical applications based on water management, verification of water content in the core profiles and on transport modelling. With the exception of application CA - coal fly ash under an asphalt top cover, the leaching behavior is controlled by percolation. Under asphalt, the release is largely diffusion controlled.

A study of the water management of the four applications has provided information on the liquid to solid ratio (L/S) under field conditions and information on the occurrence of interferences such as temporary contact of the stabilization layer with ground water as a result of a high water table. The calculated liquid/solid ratio based on formulas in the Building Materials Decree in agreement with the study of water management and projections from (collected) leachate production.

### **Determination of release from field data**

For the determination of release, three methods have been applied:

- I. Concentration decrease in the secondary construction material.
- II. Concentration increase in the underlying soil and/or the collected percolate.
- III. The difference in leaching between „fresh” and „aged” material.

In the figure 2 the determination of  $E_i$  and  $E_{ii}$  is shown.

Release E<sub>i</sub>

The determination of release ( $E_i$ ), which is based on the concentration decrease of the construction material, can be carried out when a proper reference can be found or when a concentration decrease is observed in a part of the construction material. For instance, at the interface of construction material and soil. In the latter case, the part of the construction material that is not or not appreciably leached can be used as reference. Based on a statistical evaluation of all cores sampled, it is possible to identify whether a statistically significant decrease relative to the reference level has occurred.

Release E<sub>ii</sub>

The determination of release  $E_{ii}$ , which is based on the concentration increase in the underlying soil directly in contact with the construction material, can be carried out when a proper reference for the original background concentration can be found or when a concentration increase is observed in a part of the sampled profile. In the latter case, the concentration in the section further away from the construction material/sand interface can be used as reference. Based on the results of all the cores, the statistical significance of a measured increase in the underlying sand layer can be identified.

The concentration measurement in samples from the sand layer are based on a mild acid extraction as the elements causing the increase in soil are surface bound.

Elements that are not sorbed in the soil are transported to deeper soil layers or to ground water.

From a concentration increase in collected percolate in combination with the fraction that is retained in the sand layer, a better estimate of release for mobile elements (e.g. anions) can be given. This implies that only in case of leachate collection as applied in demonstration projects this aspect can be covered adequately. For the road base applications at the „Coloradoweg” and the „Vondelingenweg” a leachate collection system has been placed for monitoring purposes thus allowing to close the mass balance for mobile species.

Release E<sub>iii</sub>

The release of constituents from the construction material can also be assessed from the difference in leaching of the fresh material and the leaching of field-exposed material. The underlying assumption is that no appreciable change in leaching behavior has occurred. For aged and weathered materials this cannot be fully excluded. This manner of determining release is further identified as  $E_{iii}$ .

**Release calculation according to the Building Materials Decree.**

In the Building Materials Decree a distinction is made between granular materials (N) and monolithic construction materials (V). In general, the leaching of the former will be governed by percolation, while leaching of the latter is dominated by diffusion. For both types of materials leaching test protocols and models are available (Guidance document Building Materials Decree) that allow a prediction of the release under field conditions. For the four applications predictions of

release have been made for the respective exposure time to determine whether measurable release can be expected in practice.

For the calculation of the immission ( $E_p$ ) in case of a percolating system the following formula is used [3]:

$$E_p = d_b \cdot h \cdot [E_{(10)} - E_g] \cdot [1 - e^{-\kappa \cdot LS_{\text{practice}}}] / [1 - e^{-\kappa \cdot 10}]$$

with:

$d_b$	dry density of soil kg/m <sup>3</sup>
$h$	height of the applied secondary construction material (m)
$E_{(10)}$	leached quantity at LS=10 in the standardized column test (mg/kg).
$E_g$	average leached quantity for clean soil at LS=10 (mg/kg)
$\kappa$	constant related to the degree of interaction of the constituent with matrix
LS	liquid/solid ratio (l/kg)
$LS_{\text{practice}}$	liquid/solid ratio reached in practice (l/kg)
$j$	period of field exposure (y)
$N$	net infiltration rate (mm/y)

The correction for the leaching of soil ( $E_g$ ) implemented by the regulator to account for the difference in leaching between lab and field is not applied in this study. It would lead to negative release predictions.

In view of the fact that leaching data for the precise materials applied in the four works are not available, data from other sources obtained or very similar materials have been used instead.

For the studied applications only in case CA diffusion is relevant mechanism of transport. The release for diffusion controlled systems as specified in the Building Materials Decree is given by:

$$E_d = E(64d) \cdot f_{\text{tem}} \cdot f_{\text{ext,v}}$$

with:

$f_{\text{tem}}$	correction for temperature difference between lab and field (0.7 here)
$f_{\text{ext,v}}$	correction for the degree of water contact and the correction for a different exposure time (64 days to 11 years, here 7.81).

The coal ash stabilization under asphalt concrete is assumed to be permanently wet, which implies that the water contact correction factor becomes unity. From earlier studies leaching information on similar material as applied in this work has been used as the relevant data for the actual material applied in these is not available.

### Comparison of predicted and measured release in practice

From the field samples and the various calculations many release data with their uncertainties have been obtained. In view of the relatively large variations in measured and predicted release values a qualitative assessment of the comparison has been adopted. A difference between predicted and measured release at a time scale of about 10 years or less than a factor 2 is considered good to

very good. A difference of a factor between 2 and 5 is considered of comparable order. In case of differences by more than a factor of 5 the agreement is poor or not existing. In table 1 and 2 the results of the different methods to determine the leaching in the field are presented, for detailed information see [4].

From a comparison of predicted and measured release a large part of the measured constituent (65%) shows a good to very good agreement between field measurements ( $E_i$ ,  $E_{ii}$  or  $E_{iii}$ ) and the calculated release ( $E_d$  or  $E_p$ ) based on laboratory test data. It should be realized that prediction of no release and observing no measurable release in 10 years is also a good prediction.

The level of agreement is even better, when in case of mobile elements (e.g. B, Cr, Mo, V and  $SO_4$ ) the release  $E_{ii}$  is corrected for the release to ground water. For these mobile elements, the agreement between the concentration increase in the underlying soil and the predicted release is often poor. The high mobility of these elements is related to the fact that all of these elements occur as oxyanionic species (e.g. borate, molybdate), and as such feature limited interaction with soil. For these elements a good agreement can be obtained, when the mass balance is closed by calculating release from concentration and volume measurements of collected percolate. This has been possible for Mo and Cr in coal ash application CZ and for Cu in MSWI bottom ash application VF and VA.

The agreement between  $E_{iii}$  and  $E_p$  is generally good. The application CA, however, the discrepancy between  $E_{ii}$  and  $E_d$  can be explained by the occurrence of a diffusion resistance at the interface between construction material and unsaturated sand layer. Based on geochemical modelling, the water management and the measured concentration profiles in the construction material as well as in the underlying sand layer it can be concluded that transport of mobile species to ground water is highly unlikely. This implies that the  $E_{ii}$  measurement in practice is in this case the most reliable determination of actual release. The poor agreement between prediction and measurement necessitates a adjustment of the prediction to account for this diffusion resistance.

In all cases, the release determination based on a concentration decrease in the construction material is the least accurate.

## Conclusions

For the first time an extensive field study has been carried out to determine the relation between predictions based on laboratory leaching tests and the release as established by measurements at applications of secondary materials as road base materials. The study consisted of an extensive sampling program based on previous release estimates, an estimate of actual water management, leaching of the field-exposed material, concentration profile analysis of the construction material and the underlying sand layer as well as calculation and modelling of release based on these profiles.

The main goal of the study was to verify the predicted release based on laboratory leaching tests with measured release under field conditions. For many relevant elements a quantifiable release has been established after about 10 years of field exposure. As expected, the variations in the measured concentrations in the soil samples were large, for some elements up to 200 %. This can be due to the small increases relatively to the background level or variability in composition between the different cores. Based on the results of validation studies of leaching tests the calculated release from laboratory data also have relatively large uncertainties (20 - 50 %). In spite of the sometimes large uncertainties in measured release most elements show a reasonable to good agreement between predicted release and measured release in the field.

From this first large scale lab-field verification of leaching behavior, the following specific conclusions can be drawn:

### General

The method of sampling and analysis applied in this study provides the possibility to assess actual release after about 10 years of field exposure of construction materials. Future demonstration projects can make valuable use of these observations. Recommendations have been made in the Handbook for leaching characteristics [5].

### *Methodology for release measurement in practice*

- Based on the water management in practice and geochemical modelling it proved possible to distinguish between predominantly diffusion-controlled release (coal ash under asphalt cover, CA) and percolation dominated release (other applications, coal ash and MSWI bottom ash under respectively sand and bricks). Under a closed top cover such as asphalt concrete diffusion proves to be the controlling release mechanism.
- The choice of the most useful methodology to determine the release ( $E_i$ ,  $E_{ii}$  or  $E_{iii}$ ) in practice depends on the specific situation under consideration.

Determination of release from the concentration increase in the underlying soil ( $E_{ii}$ ) is most appropriate for elements that sorb strongly to soil (e.g. Co, Cu, Pb, Zn). In view of the relatively low background concentrations in soil (after mild acid extraction) this approach is usually the most sensitive. In case of release by diffusion release from soil profile analysis ( $E_{ii}$ ) is appropriate for all constituents.

Elements which do not or weakly interact with soil (e.g. B, Cr, Mo and  $SO_4$ ) will be transported to deeper layers and ultimately to ground water. In this case the release based on the release between fresh and aged material can give a good estimate. Another option is the combination of a soil increase and measurements of collected percolate (only relevant for demonstration

projects).

The release from concentration decrease of the construction material ( $E_c$ ) is only applicable to homogeneous materials. Due to the subtraction of two large numbers the uncertainty is generally high in this method.

*Comparison of measured release in practice and predicted release according to Building Materials Decree.*

- For several elements (**CA**: Cd, Cu, Ni, V and  $SO_4$ ; **CZ**: Cr, Cu, Mo, V, Zn and  $SO_4$ ; **VA/VF**: Cu, Pb, Ni) the predicted release according to the formulas specified in the Building Materials Decree agree well with measured release under field conditions after about 10 years exposure.
- In case of diffusion controlled release, the occurrence of a diffusion resistance as a result of a large difference in the level of water saturation between the construction material and the underlying soil needs to be taken into account.
- In all applications the release of Zn measured in practice is significantly higher than predicted based on release calculations of the Building Materials Decree. A possible explanation can be the initially higher pH in the construction material shortly after placement compared to the pH in the leaching tests that form the basis for the release prediction.

*Formulas in the Building Materials Decree*

- For the evaluation of field data through a comparison of measured release in practice with predictions based on formulas in the Regulations of Construction Materials, the correction factor for soil ( $E_g$ ) is omitted as it leads to negative release predictions.
- Based on the release predictions some elements (Cd, Co, Ni, and Cu) are hardly leached from coal fly ash stabilization leaching to a hardly measurable release in practice. These predictions are in agreement with the different field release measurements.
- Modelling of transport using the code ECOSAT on measured profiles in the construction material and the adjacent soil has provided indications that a diffusion is substantially less than predicted on the basis of saturated conditions. In practice the underlying sand layer is unsaturated. This leads to a diffusion resistance at the stabilization/soil interface. By introducing a correction factor in the diffusion coefficient, the results may be translated to varying degrees of saturation.
- For the coal ash stabilization under sand and the MSWI bottom ash under bricks, the pH of the construction material after about 10 years exposure proves to be significantly less than shortly after placement. This is caused by release of mobile pH controlling species and/or carbonation. This change in pH can lead to a significantly different leaching behavior in subsequent time intervals. The prediction of release over long periods of time (e.g. 100 year) on the basis of laboratory experiments (higher pH) can deviate significantly from the true behavior in practice.
- Anions such as sulphate, but also oxyanions such as molybdate, borate and chromate are poorly retained in the underlying soil. After a relatively short time (<10 y), the leachable fraction of these components can be washed out completely to the ground water in a percolation dominated scenario.

In the calculations according to the Building Materials Decree this aspect is addressed for sulphate and Cl, but not for mobile oxyanions.

**Literature**

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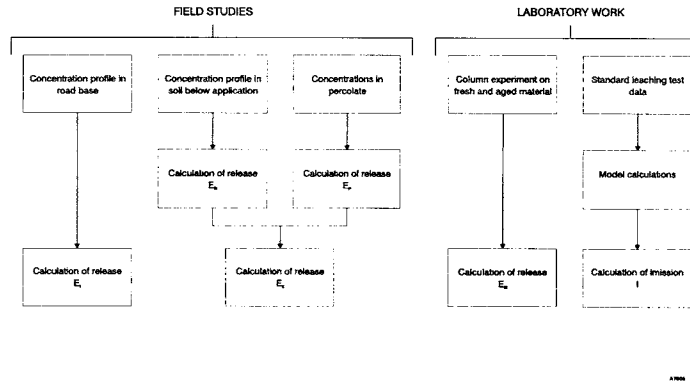


Figure 1. Approach of the verification of laboratory-field leaching behavior

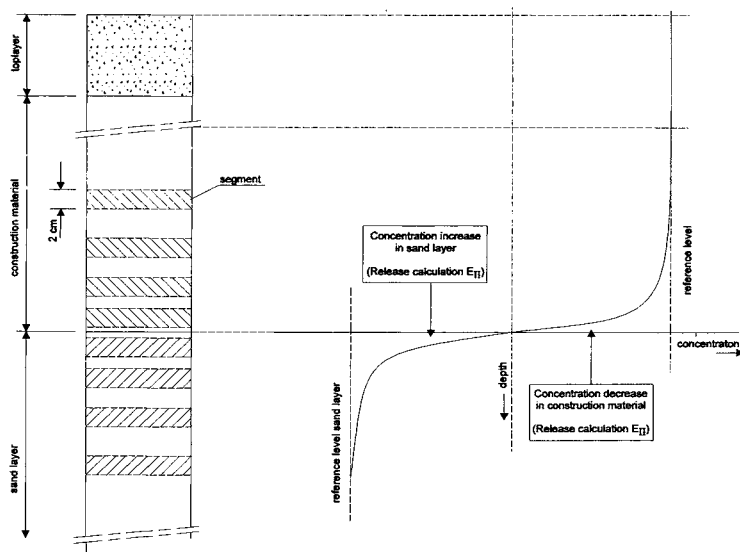


Figure 2. View of the segmentation of the cores and the determination of  $E_I$  and  $E_{II}$  from concentration decrease and increase in the construction respectively the underlying soil.

Table 1. Comparison of predicted ( $E_p$ ) and measured ( $E_i$ ,  $E_{ii}$ ,  $E_{iii}$ ) release ( $\text{mg}/\text{m}^2$ ) for the percolation scenario (shoulder of the road construction), location Colorado road

Component	$E_i$	(sd)	$E_{ii}$	(sd)	$E_{iii}$	(sd)	$E_p$
Cr	-	-	300	(280)	-	-	290
Mo	954	(1980)	1580	(600)	2049	(1945)	1470
V	8130	(6715)	300	(300)	1134	(848)	440
Zn	-	-	135	(140)	-	-	42
$\text{SO}_4$	857.700	(353.000)	-	-	924.500	(528.000)	830.000

Table 2. Comparison of predicted ( $E_d$ ) and measured ( $E_i$ ,  $E_{ii}$ ,  $E_{iii}$ ) release ( $\text{mg}/\text{m}^2$ ) for the diffusion scenario (central part of the road) Project CA, location Colorado road

Component	$E_i$	(sd)	$E_{ii}$	(sd)	$E_{iii}$	(sd)	$E_d$
Cr	-	-	31	(35)	-	-	317
Mo	91	(130)	60	(28)	1767	(718)	443
V	548	(593)	183	(270)	24	(48)	208
Zn	338	(502)	130	(270)	-	-	2.7
$\text{SO}_4$	46.740	(25.422)	15.400	(10.000)	132.150	(34.200)	58.300