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Technical principles underlying limit values for release of substances for the percolation test TS3: comparison DE and NL

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Final Report

for reconciliation in CEN TC 351 WG 1

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1. Introduction and inducement

Within CEN TC 351 WG 1, standardized, horizontal test methods are developed to assess the release (leaching) of dangerous substances from construction as defined in ER3 of the CPD. For granular materials TS 3, a horizontal up-flow percolation test, was further developed by CEN TC 351 WG1 and will enter the validation phase in 2013. In CEN TC 351 WG1, there are still discussions regarding the sample preparation and some test conditions. Currently, **two options for sample preparation and test conditions** are specified:

Sample preparation

Option A: For granular construction products to be tested under option A, at least 90 % by mass of the test sample shall have a particle size < 4 mm, and 100 % by mass shall have a particle size < 10 mm. Any oversize fraction shall be size reduced by crushing and recombined with the non-crushed fraction to fulfill the particle size requirements. Under this option, the test shall be carried out in columns with an inner diameter d from 50 mm to 100 mm with a packing height h of $300 \text{ mm} \pm 50 \text{ mm}$.

Option B: For granular construction products to be tested under option B, sieve the product applying a sieve with a mesh width of 22,4 mm. Size reduce the fraction $\geq 22,4$ mm to < 22,4 mm by mildly crushing and sieve out the 16/22,4 mm fraction. Add the 16/22,4 mm fraction to the sieved 0/22,4 mm fraction in the amount in which the fraction > 22,4 mm existed in the original construction product. Test the combined fractions in columns with an inner diameter d of at least 3 times the maximum diameter of the grain size of the product ($d_{\text{max}} = 100 \text{ mm}$) with a packing height h of $300 \text{ mm} \pm 50 \text{ mm}$.

Test conditions

Furthermore after the equilibration period, the flow rate is such that the linear velocity is $150 \pm 20 \text{ mm/d}$ for option A and $450 \pm 50 \text{ mm/d}$ for option B.

Controversy between DE and NL regarding sample preparation and test conditions and the need for two separate options in the TS 3 percolation test are possibly for a part caused by different approaches for risk assessment in the Netherlands and in Germany and the resulting regulatory concepts. One important reason for this is, that in soil and groundwater regulations the test method and the impact assessment method are systematically linked together but the impact assessment methods and lab methods in both countries are different. The discussions in CEN TC 351 WG 1 show that there is a need to have a better mutual understanding of the relation between test method and impact assessment on the one hand and a clear overview of differences in impact assessment approaches of each country on the other hand.

The aim of this project is to explain and compare the assumptions, boundary conditions and conventions of the impact assessment approach that are implemented in the upcoming

German Recycling Degree and in the Soil Quality Degree of the Netherlands. Ultimately, a better understanding of the impact assessment approaches provides a basis for further discussion in WG1 to agree on only one option for the percolation test conditions. The following report is prepared by the contractors of Germany and Netherlands together to compare the two country-specific concepts.

This report summarizes and explains the presentation given on the 25th of April at TC 351 WG1 in Berlin, and the extensive discussions afterwards. Detailed information about the German impact assessment are given in Grathwohl & Susset, 2011)

Some important notes

TC 351 does not cover “impact assessment” which is up to MS regulators. However, sometimes we have to look at what “impact assessment” asks from us, i.e. in relation to boundary conditions to the test methods.

The issue is studied exemplary for the risk assessment concepts in the Netherlands and in Germany and makes no claim to be complete regarding requirements from other member states.

Although the authors have been involved in the actual calculations that underlie the “impact assessments” and resulting release limits for percolation tests for the German recycling decree (B. Susset) and the Dutch Soil Quality decree (J. Dijkstra, A. van Zomeren), views and conclusions in this report are not necessarily shared by the Dutch and/or German Government.

2. Working steps and procedure

Main objective of this project, is to explain the basic principles and assumptions of the German and the Dutch impact assessment method. The main part focusses on the scenario description that was used for the modeling and criteria development. This information is necessary to be able to understand the consequences of the results of the robustness validation regarding the two different options A and B. Some preliminary scenario based model calculations for several usual and comparable construction works in Germany and the Netherlands were done to provide a quantitative evaluation of the differences in the limit values in NL and DE. The work can be subdivided into following main working steps.

A: Qualitative comparison of scenario descriptions

B: Scenario based calculations for several comparable construction works in Germany and the Netherlands

C: Comparison and interpretation of results

D: Conclusions for standardization work regarding the two options in TS 3

E: Integrated executive summary.

The work focusses on following issues:

- What is the common goal of protecting soil and groundwater environment?
- Comparison of soil and groundwater criteria in DE and NL
- Conceptual approach in calculating limit values in NL and DE
- Differences of limit values for release between DE and NL
- How are limit values / test results judged and implemented in quality control systems (extensive column test/short test, which LS)?
- What kind of information is needed from the test?
- How to proceed?

3. Results

3.1. Common goals in protecting soil and groundwater

European requirements to soil and groundwater protection (Water Framework Directive, Groundwater Directive, etc.) set boundaries for soil and groundwater protection but no concrete protection values. European directives lead to national values for soil- and groundwater protection which may differ from EU MS to EU MS.

The following comparisons are only given for substances which are regulated in both countries. In DE, there are 30 additional groundwater parameters (mainly organics) published in Draft MantelV (2012) for which such a comparison cannot be made.

Figure 3.1 compares the NL and DE groundwater protection values (ug/l in the solution phase). In NL, the “MTT-gw” value is a maximal permissible addition (MPA), which is derived from the HC5 value by ecotox-testing. This MTT-gw value is the protection level that was used as “target” concentration in the groundwater during the calculation of release criteria for construction products, i.e. irrespective of the background concentrations in groundwater.

In Germany for a given substance, the concentration level avoiding any significant alteration of the chemical status of groundwater is defined as the “Insignificance Threshold Concentration” (German: Geringfügigkeitsschwelle, GFS; given by the Working Group of the Federal States on Water, Länderarbeitsgemeinschaft Wasser, LAWA, 2004, www.lawa.de) comparable to a “no-effect level”. GFS is derived from eco- and humantox. - tests. According to the ‘precautionary principle in the German groundwater policy’ the limit values are not quality targets (or target concentrations) for the groundwater body. Therefore the limits have to be met already above the groundwater in the seepage water drop before it gets groundwater to avoid any alteration of the groundwater. However, any exceedance of GFS within the groundwater is considered to be a harmful change of groundwater in a legal sense.

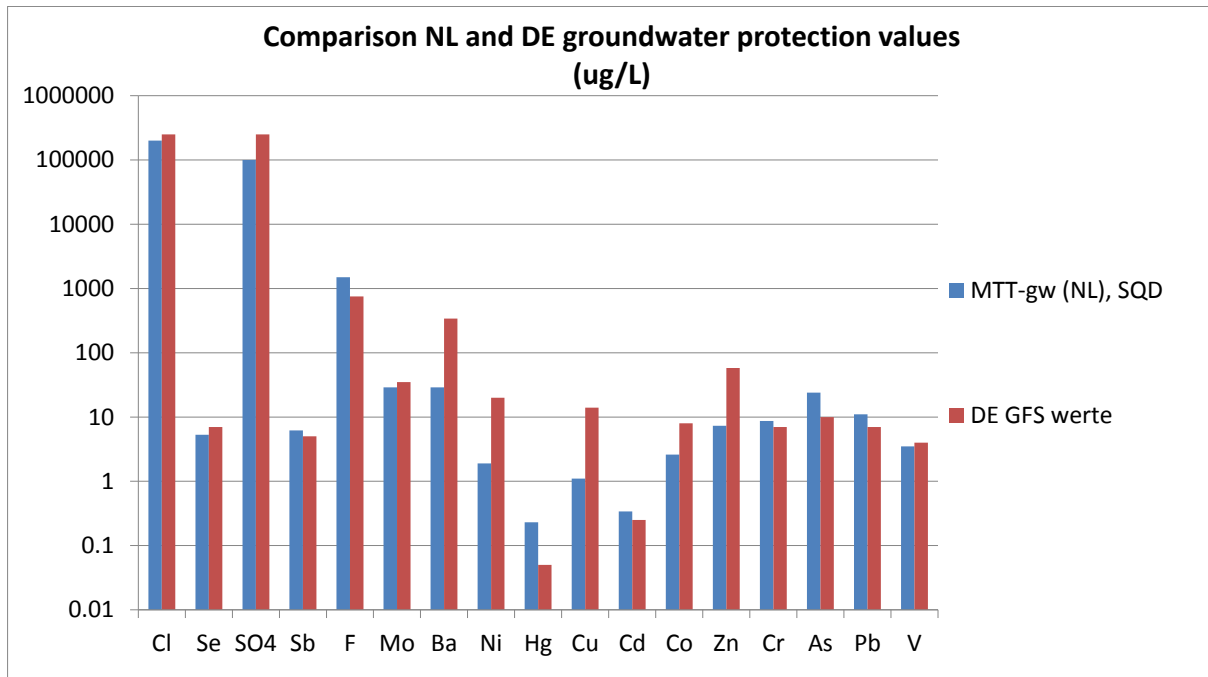


Fig. 3.1: Comparison of NL and DE groundwater protection values [$\mu\text{g/L}$]. MTT-gw is a maximum permissible addition (MPA) to groundwater. GFS is an “Insignificance Threshold Concentration” within the groundwater (German: Geringfügigkeitsschwelle).

Figure 3.2 compares the NL and DE soil protection limits (mg/kg in the solid phase). In NL, the MTT-soil value is a maximal permissible addition (MPA) which is again derived from MTT+Background (MPC-value derives from HC5 value by ecotox-testing). In Germany “soil-protection policy” is based on content concentrations, so called precautionary values in the soil solids, given by the Federal Soil Protection and Contaminated Sites Ordinance (Länderarbeitsgemeinschaft Boden, LABO, www.labo.de), which again according to the “precautionary principle” are not the quality targets (or target concentrations) for soils. They are not directly comparable to the MTT-soil values in NL as they are not permissible addition values.

For the comparison, the so called accumulation values have to be taken into account, which are 50 % of the filter capacity given by the precautionary values minus background values of different soils. In the German risk assessment approach the accumulation of contaminants in the solids is averaged over 1 m transport zone and has to be limited to 50% of the filter capacity of soils within an appraisal period of 200 years. As the background value and precaution values differ for sand, loam and clay, for the risk assessment, two different soil categories were distinguished: the standard sand and standard loam (or silt or clay) category. To make these values comparable with the NL values (which are based on an appraisal period of 100 years) the German values which rely on a more conservative appraisal period of 200 years (versus 100 years in the Netherlands) have to be subdivided by

factor 2. The resulting maximum allowed accumulation for sand and loam is given in Figure 3.2.

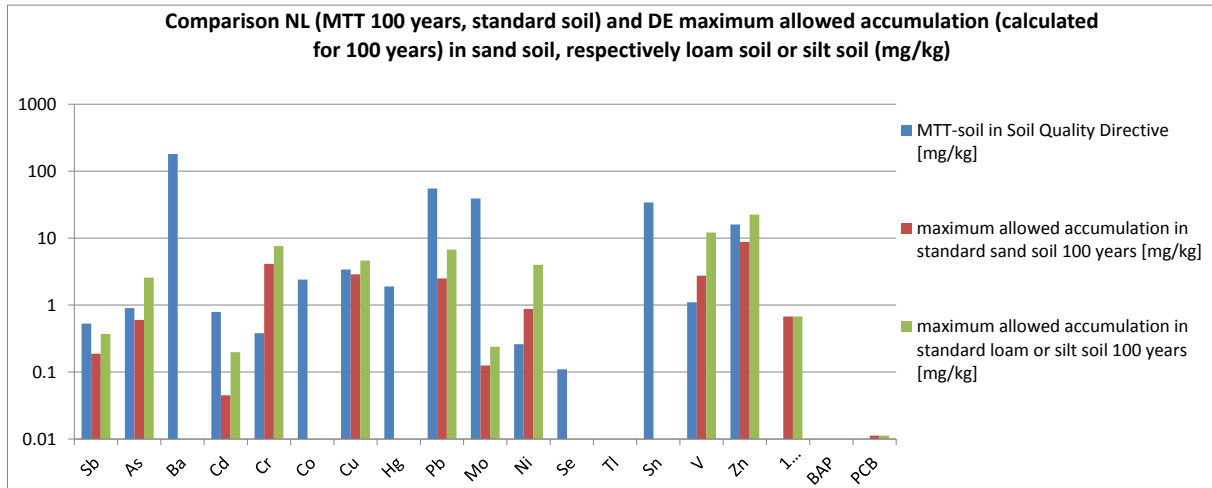


Fig. 3.2: Comparison of NL and DE soil limits [mg/kg]. The Dutch MTT-soil is a maximal permissible addition (MPA) to soil averaged over 1 meter soil over an appraisal period of 100 years. In Germany the soil limits are calculated from the filter capacity given by the precautionary value minus background value. In Germany the maximum allowable accumulation is limited to 50 % of filter capacity averaged over 1 meter soil within an appraisal period of 200 years. As the appraisal period is 2 times more conservative than in NL, the DE values were subdivided by factor 2 for direct comparability with the Dutch MPA

Conclusions soil & groundwater limit values

Groundwater: In general, the groundwater limit values are quite comparable for most of the substances (except for Ba, Ni, Hg, Cu and Zn). German GFS are in 10 cases more tolerant (Cl, SO₄, Se, Mo, Ba, Ni, Cu, Co, Zn, V), Dutch MTT are in 7 cases more tolerant (Sb, F, Hg, Cd, Cr, As, Pb); In Germany exist 30 additional parameters (mainly organics) published in Draft MantelV (BMU, 2012)

Soil (accumulation values): For antimony, arsenic, copper, zinc comparable values. Extreme differences for cadmium, lead and molybdenum, which are in Germany 10 (Cd, Pb) to 26 (Mo) times lower, driven by very low precautionary soil protection values (these accumulation criteria lead to a stringent construction limit value of GFS * 1,5 in most scenarios). For chromium, nickel and vanadium the Dutch values are 5 to 10 times lower.

3.2. Impact assessment in Germany and the Netherlands

The fundamental principles of impact assessment in Germany and the Netherlands are similar (see scheme in figure 3.3). Groundwater risk assessment is subdivided into the assessment of the source term and the transport term. The seepage water concentration can be estimated most reliable by aqueous leaching tests. Extended column up-flow percolation tests are used for basic characterization of the leaching behavior. From the laboratory release test results (concentration versus testing time or liquid to solid ratio, L/S) the leaching in practice is assumed by an L/S-time conversion (time t is expressed based on the dynamic liquid to solid ratio (LS), which is the amount of water percolated through the column after a certain time relative to the dry weight of the solids in the column), using typical seepage water rates in the field.

A time-dependent leaching behavior (NL: metals and salts, DE: salts) or a constant averaged source term concentration (DE: metals and organics), act as input for the assessment of the transport term (see also 3.2.1). Below the source zone, attenuation may lead to a decrease of concentrations within the seepage water (organics), whereas sorption leads to a retarded breakthrough of the concentrations at the point of compliance. Adequate numerical and analytical tools to describe the transport term are available. From the transport modeling the breakthrough of concentration versus time at the point of compliance can be estimated. Release limits for constructions are back-calculated in that way, that the maximum concentrations within soil, into the seepage water drop or into the groundwater at the point of compliance meet the soil and groundwater limits and/or to ensure, that there is no exceeding of the soil and groundwater limits within a certain time period (the “appraisal times”, which is 200 years in DE and 100 years in NL).

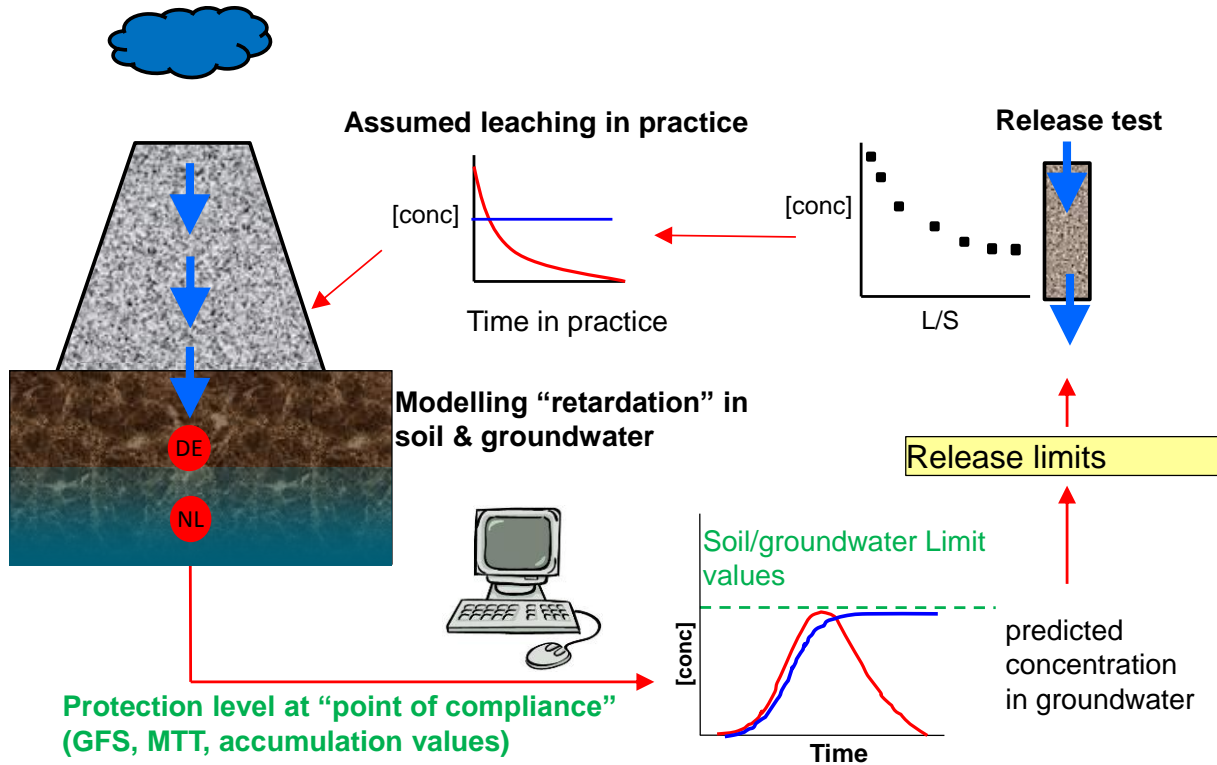


Fig. 3.3: Schematic overview about impact assessment in Germany and the Netherlands. The fundamental principles of impact assessment in Germany and the Netherlands are similar.

3.2.1 Differences in source term definition DE and NL

In the NL impact assessment a time dependent source term is assumed for all substances (salts and metals), described by a substance-specific function. In this first order decay function, a “kappa” parameter describes the concentration decrease as a function of liquid to solid ratio (L/S, convertible to time in a scenario in practice):

$$C_{L/S} = C_0 \cdot e^{-\text{kappa} \cdot L/S}$$

The kappa value is substance-specific, but product-generic. The kappa value is for each substance derived from a database with 360 historical percolation tests on different construction products. Although the standard deviations of the “average” kappa values is of course very large (due to material-specific differences, outliers, low concentrations etc. etc.), the general outcome is quite logic. For instance, the average kappa for chloride (a soluble salt) results in a sharp decreasing concentration curve (consistent with the average behavior of chloride in a percolation test) and a metal such as arsenic is represented by an almost constant concentration, which is also a common observation.

Because the kappa value is generic for products and specific for substances, the “source term” in the impact assessment modeling only depends on the value of C_0 , the initial concentration. During calculation of release limits, the C_0 is the only parameter that is iteratively varied, and fitted in such a way that the concentrations of substances at the POC complies with the soil and groundwater criteria.

The above formula allows extrapolation of column test data to scenario's in practice, because the L/S ratio converts easily to time scales (years) given the bulk density (kg/m^3), annual net precipitation (mm/year) and the height of the application (m). For open granular scenarios in NL, the generic choice is an application height of 0.5 m (defined as the single representative scenario for all application heights), precipitation of 300 mm/year (open application, isolated applications 6 mm/years) and a generic dry bulk density of 1550 kg/m^3 .

It is important to understand that the NL limit values for release to which construction products should comply, are expressed in cumulative leached amounts (mg/kg) after L/S 10 L/kg, as measured with a percolation test in 7 fractions for basic characterization. Nevertheless, the impact assessment that underlies these cumulative leached amounts in mg/kg relate through the kappa value directly to a value of C_0 . The cumulative release, expressed in mg/kg at L/S 10, is found by integrating the above formula between L/S 0 and 10.

The DE limit values for release to which construction products should comply, are expressed in cumulative leached concentrations (mg/L) after L/S 2 L/kg, as measured with a percolation test in 4 fractions for basic characterization and 1 single fraction for compliance.

In the German approach for salts (chloride and sulphate) a time dependent source term is used, which is described by a substance-specific and product-specific function (derived from percolation tests with substance- and product-specific K_d -values (parameter used to fit column test data) which is the solid/water-distribution coefficient). Like in the NL approach the impact assessment relate through the K_d -values directly to a value of C_0 . The cumulative release, expressed in mg/L at L/S 2, is found by integrating between L/S 0 and 2.

For the risk assessment of metals and organics, Germany uses a constant (time independent) source term. A cumulative constant source term concentration at LS 2 L/kg describes an averaged medium time-scale concentration (LS 2 L/kg \approx 8 - 50 years, depending on construction scenario). It corresponds to the cumulative mass released up to LS 2 L/kg divided by the total volume of water collected up to LS 2 L/kg. Motivation for this choice are results of a huge number of column tests showing, that standard deviations of the mean release rates are much higher for metals and organics than for salts and the release behavior is hardly to describe with a product-generic type curve as the leaching behavior differs from sample to sample.

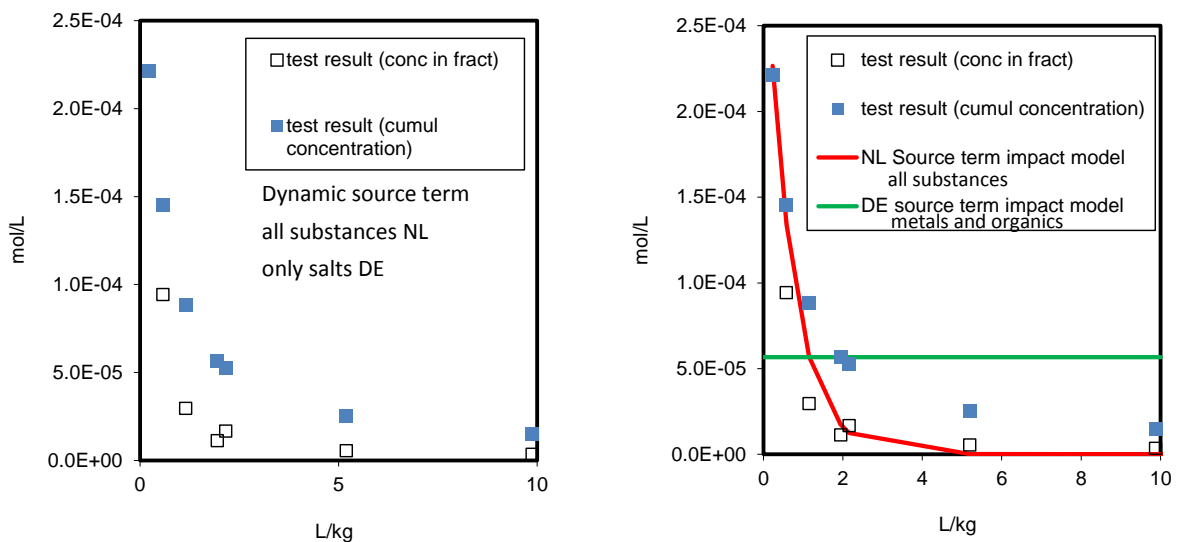


Fig. 3.4: Source term definition in Germany and the Netherlands. In the Dutch impact assessment for all substances (salts and metals) a time dependent source term is used, described by a substance-specific function, which is generic to all products. In the German approach for salts (chloride and sulphate) a time dependent source term is used, which is described by a substance-specific and product-specific function. For metals and organics Germany uses a constant (time independent) source term. A cumulative constant source term concentration at LS 2 describes an averaged medium time-scale concentration.

3.2.2 Differences in scenario definition DE and NL

Figure 3.5 compares the scenario definitions in Germany and the Netherlands. Beside the differences in the source term assessment discussed in chapter 3.2.1, there are further differences regarding the situation-specific distinction of source term assessment: Germany distinguishes between construction-specific averaged seepage water rates and dilution factors of partly open constructions, construction-specific averaged seepage water rates of open constructions (313 mm/y for noise protection wall, 242 mm/year below cobblestone, 377 mm/year below flagging, 583 mm/year below open covers) and isolated constructions (construction materials with statistical derived construction limits are accepted per convention). This leads to more than 120 different construction-specific “media related application values”, which are maximum concentrations allowed in the seepage water and can be interpreted as the “release limit values” in Germany.

Compared to the German approach, the Dutch approach is much more simplified with 3 different generic scenarios (granular open, granular closed and monolith). In the German upcoming Recycling Degree only granular recycling materials are regulated. Monoliths are regulated by national technical approvals of the Center of Competence in Civil Engineering (Deutsches Institut für Bautechnik, DIBt).

Also, with regard to the transport term there are differences in the model approach (geochemical model with advection dispersion in NL versus Kd/Freundlich retarded advection-dispersion model in DE). The geochemical approach followed in NL is based on thermodynamic sorption parameters of substances to individual sorbents in soil according to Dijkstra et al (2004, 2009). In this approach, no prior “fitting” of sorption parameters is necessary. This feature makes the approach suitable to make reactive transport predictions for individual soils with different characteristics. In the Netherlands, three representative soil profiles were selected (sand, peat and clay) from which detailed profile characteristics were available. The transport model predictions of the impact assessments were done for all three soils, and a single source term was fitted such that all substances in all soils were compliant to limit values at the POC. The resulting “fitted” source term (using the kappa approach in figure 3.4) was expressed in mg/kg at L/S 10 and form the limit values in the NL Soil Quality Decree.

The German approach for sorption modeling is based on a different concept (Kd/freundlich). For substances that are either non-sorbing (soluble salts) or strongly adsorbing (some metals), differences between the Kd/Freundlich and the geochemical approach are small. For “intermediate” sorbing metals, differences between both approaches can be considerable. However, the authors are of the opinion that the most important differences are

not caused by the sorption/attenuation modeling approach, but by political boundary conditions, which are set more conservative by the German precautionary soil- and groundwater policy:

- 1 meter (DE) versus 2 meter (NL) “effective soil” for attenuation and retardation processes;
- 200 years (DE) versus 100 years (NL) appraisal period;

The upper two factors are of major importance for differences with respect to resulting release limit values as will be illustrated further below. Other factors that play a less important role, but that cannot be neglected are:

- averaging of concentrations from 1 to 2 meter in the saturated zone (NL) ;
- A distinction of an critical case in DE (groundwater distance < 1 meter below construction bottom line), where the materials have to meet $GFS \times 1,5$ (factor 1,5 is the result of examination of German Federal ministry for environment) in the LS 2 - eluate directly.

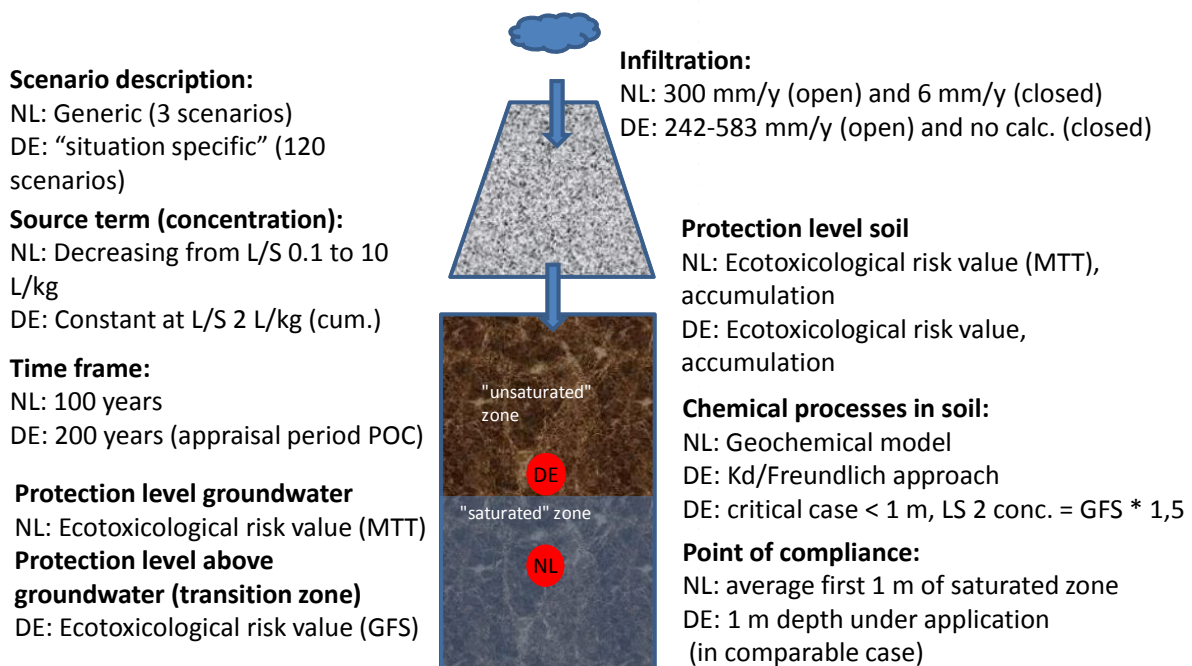


Fig. 3.5: General differences in scenario definition in Germany and the Netherlands. A situation specific differentiation of source term assessment in Germany leads to more than 120 different construction limits. The most important differences are not caused by the different modeling approaches (geochemical model versus retarded advection-dispersion model) but by political boundary conditions, which are set more conservative by the German precautionary soil- and groundwater policy.

3.2.3 Influence of differences between impact assessment methods in DE and NL

The influences of differences in model approach were evaluated for this project and subsequently discussed in the presentation given on the 25th of April at TC 351 WG1 in Berlin. Of course, differences between the protection criteria in soil and groundwater (Figure 3.1 and 3.2) will affect the established release criteria directly, but below the differences of the impact assessment methods.

To hold this report readable, in the following section only an exemplary overview of the influence of the factors that were considered most important are given: the position of the point of compliance, the source term assumption (time dependent versus constant), the appraisal time and the averaging of concentration in the first upper meter of the saturated zone is given (Figure 3.6).

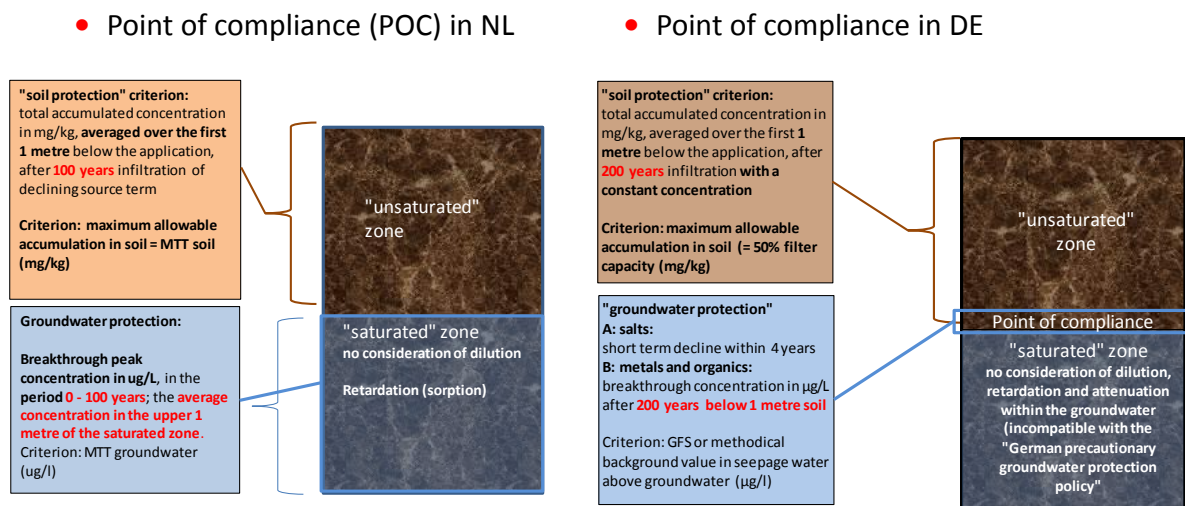


Fig. 3.6: Differences in the positions of the point of compliances, appraisal periods and concentration averaging in Germany and the Netherlands.

Influence of differences in source term definition on release limits

Figure 3.7 shows the effect of the source term definition (left part of the graph) on the shape of the breakthrough curve at the POC (right part of the graph). The declining source term used in the Netherlands for many substances results in a “bell shaped curve” at the POC. The concentrations increase to a maximum value and then they will decrease again. When an averaged constant concentration is used as the source term (Germany), the concentrations increase to a maximum value and they will remain at the maximum value over time.

In the Dutch approach, the maximum concentration of the source term (C_0) is highly determining the magnitude of the concentration at the POC (i.e. the concentration at $L/S=0.1$ L/kg in the Netherlands). In the German approach for salts it is more or less similar, for metals and organics, the average concentration at $L/S=2$ L/kg in Germany will be highly determining. Of course, the degree of sorption (retardation), the distance to the POC will determine whether the maximum concentration in the groundwater is reached or not within the appraisal time.

As explained in the previous chapter, in the Netherlands, some substances have an almost constant source term (solubility controlled leaching over a wide range of L/S) and other substances (salts and mobile substances) have a steep declining source term; the behavior depends on the generic kappa- value. In Germany, a constant source term is used for metals and organics. Only for chloride and sulphate, a time dependent declining source term is used, which is described by a substance-specific and product-specific function.

The moment of breakthrough at the POC will not be strongly affected in both approaches. And, as long as the maximum concentrations of a declining source term and a constant source term are equal, also the concentration at the POC will not be greatly affected. However, when the same actual measured column test result (open squares in left figure of Figure 3.7) is treated in the NL way or in the DE way, the NL way will result in initially higher concentrations at the POC that will decrease afterwards, while treating the same column test data in the DE way will lead to a lower concentration at the POC, which does not decrease over time. In the end, the source term must be fitted such that concentrations will not exceed the groundwater criterion, in the NL case this is a factor that will in principle work towards more stringent release criteria but this, again, only for “intermediate” sorbing metals. However, because the final release criteria in NL are more tolerant than in DE, there are

other more important factors that work on the contrary, as will be demonstrated for the appraisal time below.

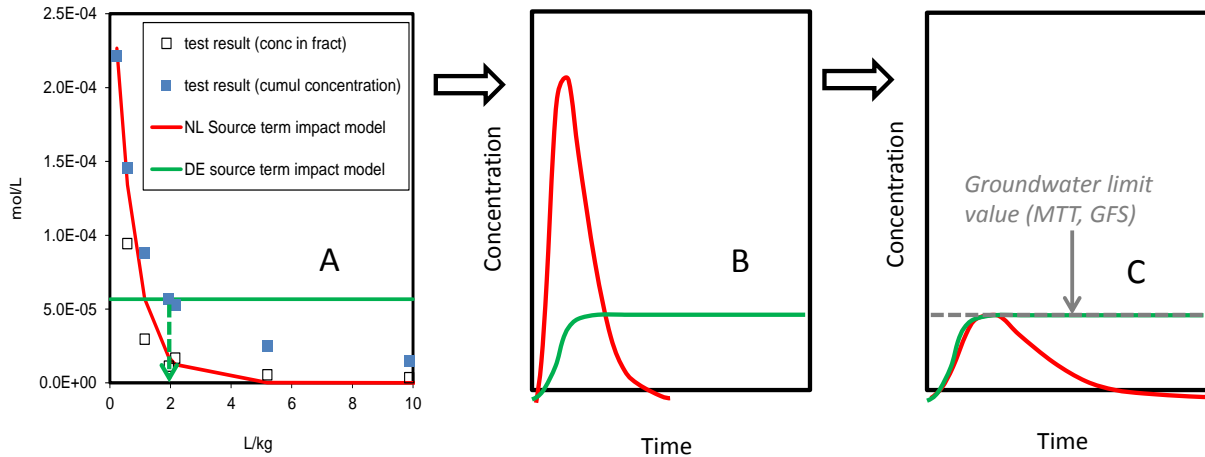


Fig. 3.7: Effect of different source term assessment (NL: declining or nearly constant depending on substance, DE: constant for all metals and organics). For illustration purposes, the situation is shown for mobile substances that completely break through at the POC. **Figure A** shows how theoretically the source term is derived from column test data (open squares) for the same data (concentrations in fractions, open squares). In NL this is done with a first order function that describes the complete curve (red line, averaged kappa- values are used for each substance so the fit is not always perfect). In Germany, the cumulative L/S 2 concentration is derived (blue dots) and assumed constant (green line). **Figure B** shows the concentrations that arrive after some time at the POC. In the NL case, for some “intermediate” sorbing substances concentration peaks are higher than in the DE case due to the high initial concentration in the source, in the DE case the initial concentrations are lower due to the averaging of source term concentration between L/S 0-2, but remain constant. Note that both cases refer to the same column test data. **Figure C:** in setting limit values for release, the source term must be such that all concentrations at the POC are below the groundwater criterion. For the NL case, for some “intermediate” sorbing substances, in principle larger corrections (i.e. more stringent release criteria) are necessary due to the higher initial concentrations. For either the very mobile substances (e.g., chloride and sulfate) or immobile substances (e.g. arsenic), there will be no strong effect. However, there are also (more) important factors that work on the contrary such as the appraisal time. Together these additional factors lead to more tolerant release criteria in NL.

Influence of difference in appraisal time on release limits

Figure 3.8 shows an example for the calculated concentration of copper (a metal with a moderate retardation) at the POC as a function of time. The horizontal red line shows the ground water limit value and the vertical dashed lines show the appraisal times used in NL (100 years) and DE (200 years). In this example, the ground water limit value is just met after 100 years and the release limits in the laboratory test can be calculated on the basis of the assumed source term for this example (Dutch situation). However, Figure 3.8 does also show that moderately mobile metals like copper may exceed the ground water limit value after the appraisal period of 100 years (examples available in extra sheets). This is a natural consequence of using a finite appraisal time. Copper acts as an intermediate mobile

substance, of which the calculated emission limit is very sensitive to appraisal time as will be shown below. Note that either the mobile substances such as chloride and sulphate, or the very immobile substances such as lead, do not show the sensitivity to appraisal time. When a decreasing source term is used for the impact assessment, the exceedance of the ground water limit value will, consequently, be temporary and it can be decided by the regulator to accept this temporary exceedance in view of other policy decisions (e.g. creating a market for recycling, adjustment of limit values during revision of regulatory criteria etc.).

As the appraisal period in Germany is 200 years, there is a strong effect on the allowable source term concentration, i.e. the assumed source term concentration in Germany should be substantially lower in order to meet the groundwater limit values after a period of 200 years. In summary: the longer the appraisal time, the more stringent source terms (and release limits) are necessary.

Although more factors influence the differences in release limits, it can be assumed that mainly the difference in the appraisal period results (for moderately mobile substances) results in more tolerant release limit values in the Netherlands in comparison with Germany (difference is depending on scenario a factor 2-8 for Cu, see also Figure 3.8).

The appraisal period has such a large effect on the calculated release (or concentration) limits in the laboratory test that even the shown differences in the groundwater limit concentrations and source term definition are of relatively small importance (e.g. the German GFS of copper is 14 µg/L and the Dutch NL MTT-gw is 1,1 µg/L).

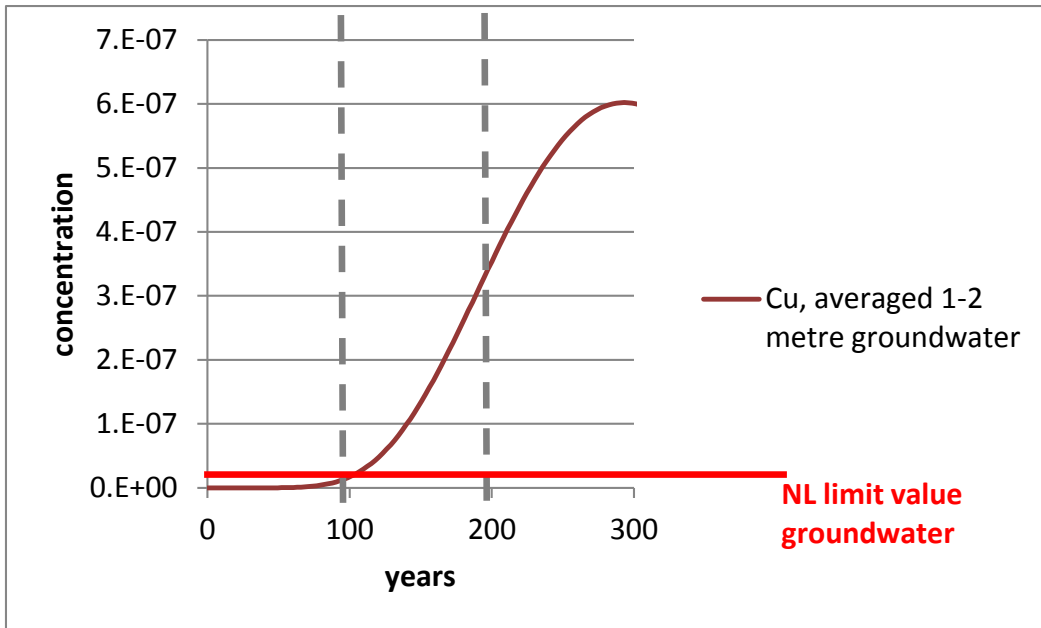


Fig. 3.8: Calculated concentration of copper (mol/l) at the POC as a function of time. The horizontal red line shows the ground water limit value and the vertical dashed lines show the appraisal times used in NL (100 y) and DE (200 y). In the example, copper acts as an intermediate mobile substance, of which the calculated emission limit is very sensitive to appraisal time (see text). Note that either the mobile substances such as chloride and sulfate, or the very immobile substances such as lead, do not show the indicated sensitivity to appraisal time.

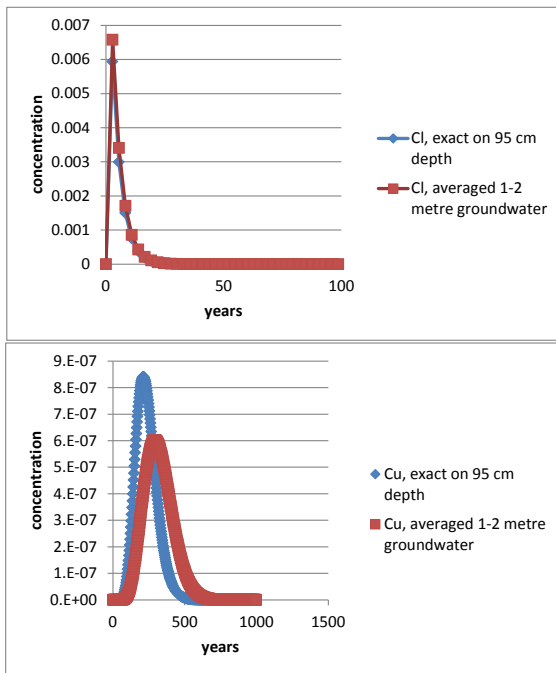
Effect of averaging concentrations in the saturated zone

Figure 3.9 shows the effect of averaging the concentration at the POC in the upper meter of the saturated zone between 1 and 2 meters depth (the Netherlands) versus the concentration at the POC exactly on 1 meter depth at the interface between the unsaturated and saturated zone (representative for scenarios in Germany).

For a soluble salt such as chloride (upper figure), this conceptual difference is not of any importance. Also for substances that are extremely strongly retarded, this is not of importance. For the group of “moderately” sorbing substances such as most metals, there is a relatively small effect visible on the peak concentration in groundwater (lower figure, example for copper, of which the peak concentration is about 25% lower). It can be concluded that the effect of “averaging” in the upper meter of groundwater between 1 and 2 meter depth (as is done in the NL case) is not of great importance. Although this result may be somewhat counterintuitive, it can be fully explained (see figure 3.9), however it will not be detailed in this report.

Equally important is the effective transport zone for retardation processes with 2 meters in the Netherlands versus 1 meter in Germany. This is important especially for moderately sorbing metals like copper, chromium and vanadium. The effect on release limits is not a factor 2, but less. Due to the “averaging” the effective depth at which concentrations are judged in the Netherlands is 1,5 meters; for moderately mobile metals the effect of the extra retardation is a factor of 1,5 that peaks arrive later at the POC and concentrations are slightly lower due to some more dispersion.

In all of the above cases, the effect of appraisal time is estimated to be much more important than “averaging” or extra retardation. This is in particular the case for moderately mobile metals that arrive at the POC just around the appraisal times of 100 or 200 years. Under those conditions, small differences in substance mobility or choices with respect to “averaging” concentrations are “amplified” and lead to large differences in release limit values.



Difference between:

- POC exactly on 1 meter depth, or
- POC = “averaged” concentration in upper meter saturated zone
- Conclusion: not very different

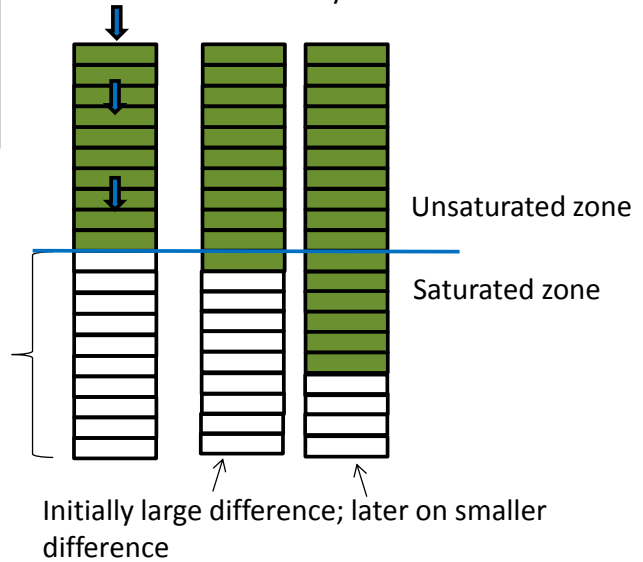


Fig. 3.9: Effect of averaging method for concentrations at the POC. Blue diamonds represent the results of the method used in Germany, the brown squares represent the results of the method used in the Netherlands.

3.3. Comparison of application limit values for construction products in Germany and the Netherlands

3.3.1 Pre-processing for comparison

The construction limits in the Netherlands and in Germany are not directly comparable because there are:

- **limit values in the Netherlands and Germany are strictly spoken incomparable** as they are derived at LS 10 (Netherlands) and LS 2 (Germany) , respectively.

- **many different scenarios**

Netherlands: 3 scenarios (generic for all products including recycled), no pre-classification of material qualities with material values:

Granular, all open applications, unit: cumulative mg/kg at L/S 10 (percolation test, NEN7373/NEN7383; < 4 mm, 3 weeks)

Granular, all closed applications, unit: cumulative mg/kg at L/S 10 (percolation test, NEN7373/NEN7383; < 4 mm, 3 weeks).

Monolithic products, all applications, NEN7375, unit: cumulative mg/m².64 days

Germany: About 120 scenarios (different constructions and underground constellations – groundwater distance from 0.1 to < 1 meter: critical case, groundwater distance > 1 meter: favorable case, distinction of sand or silt/loam/clay) with media related application values which are generic for all recycled products. Pre-classification of material values for each recycling product: Identification of relevant substances and expected concentrations from statistics of a huge number of measurement data. Material values depend on allowable applications and vary for different materials / classes):

Granular, open applications, unit: cumulative µg/L at L/S 2 (percolation test, DIN 19528; 0-32 mm, extensive test up to LS 4 (4 fractions): 1 week; for factory control: LS 2 - short test: 2-3 days, acc. to DIN 19528 or batch acc. to DIN 19529)

Granular, partly open applications, unit: cumulative µg/L at L/S 2; (testing like granular open)

Granular, closed applications, unit: cumulative µg/L at L/S 2; (testing like granular open)

Monolithic: no regulation within upcoming recycling Degree

An important difference between the final judgment in the Soil Quality Decree (NL) and the upcoming Recycling Degree (DE) is that in the Netherlands all different materials (new

construction products and recycled products), constructions (application height), soil types and groundwater distances are covered by only three scenarios (open granular, closed granular and monolithic). This means that deviations from the standard case (application height, different groundwater distance, etc.) in daily construction and recycling practice is accepted, with the big advantage of simplification of a regulation.

In Germany each construction is evaluated separately based on the calculated limits from the impact assessment, which are also the same for every material in a specific construction but very differentiated among different applications. The media related application values depend on the construction itself, the soil zone (sand or silt/loam/clay), groundwater distance, critical case from 0.1 to < 1 m groundwater distance), the material values of each material and/or material class depend on the allowable or achievable construction and are pre-classified based on measurement data. This means, that in many cases different materials or material classes have different material values, which are linked to the media related application values (which are situation-specific).

Therefore following comparison approach was developed to create a basis for comparison of limit values:

- Selection of the most relevant and comparable scenarios for comparison
- Dutch values (mg/kg L/S 10) were “backwards” translated to ug/L at L/S 2 using a “reversed” kappa approach (see formula in 3.2.1 and Verschoor et al., 2006).

Comparable scenarios:

The most relevant application in Germany is the open application with open cover for the favorable case of a groundwater distance of > 1 meter and a soil zone of sand and the critical case with a groundwater distance between 0.1 and 1 meter. Isolated constructions in Germany can be directly compared with the Dutch isolated constructions.

For salts, where the media related application values depend additional of the thickness of the construction, relevant cases of ballast layers < 0,5 m and fillings of > 1 meter thickness under open covers were chosen for comparison.

Backwards calculation of LS 2 values from Dutch LS 10 values:

The back-calculation of the release limit values that are expressed in mg/kg at L/S 10 towards ug/l at L/S 2 is based on a simple conversion of the integral of the formula given in 3.2.1 given the default application height (0.5 m), annual precipitation (300 mm/year) and

bulk density (1550 kg/m³). The kappa-values are substance-specific and mentioned in Verschoor et al. (2006). For calculation the original SQD model files of the Dutch impact assessment were used. Note that this is a generic function that applies to all materials in the Soil Quality decree, so it ignores material-specific differences.

3.3.2 Results

Figure 3.10 compares the release limit values for *open applications* in the Netherlands (backwards calculated to LS 2 in ug/l) and in Germany (ug/l at L/S 2). The demonstrated comparison is exemplary for the relevant and comparable German scenarios: sand, groundwater distance > 1 meter and critical case: groundwater distance 0.1 to 1 meter.

Figure 3.11 compares limit values for *isolated* constructions. It is important to note that there is no modelling of impact assessment for isolated constructions in Germany (no media related modeling)¹. Per convention of the federal ministry of environment in Germany generally all regulated materials are accepted in isolated, closed constructions, assuming that there is negligible seepage water flowing through the recycling material (e.g. core constructions under asphalt pavement or below cobblestone with bituminous watertight joints). The way the release limits for closed applications are established in DE, is the following. If there are materials with high concentrations in LS 2 eluates which don't allow an application in open constructions, the material values are set to the 90th percentiles of concentration contributions in LS 2 eluates. These material are only accepted in isolated applications. For the comparison in figure 3.9, the maximum material values for each substance of all regulated materials are given.

Note that in NL, impact assessment modeling is done for "closed applications" with an annual precipitation of 6 mm/year (compared to 300 mm/year for open applications).

Figure 3.12 compares the Dutch and German release limits for the above mentioned scenarios expressed in factors difference.

¹ Note: E.g. street dams with asphalt pavement, noise protection walls with technical measures of protection are not closed or isolated constructions. These partly open constructions were evaluated by impact assessment taking into account hydraulic processes with a numerical hydraulic model. By-passing may lead to reduced concentrations in the seepage water averaged along the cross-sectional area of the construction (see Grathwohl & Susset, 2011).

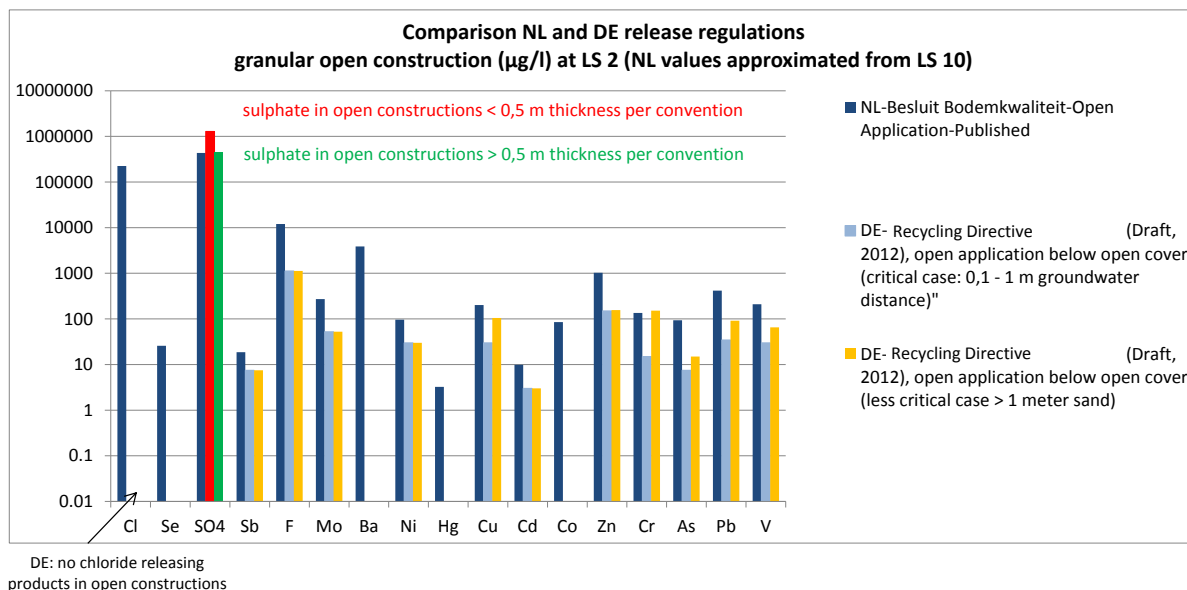


Fig. 3.10: Comparison of Dutch and German limit values (µg/L) for open applications.

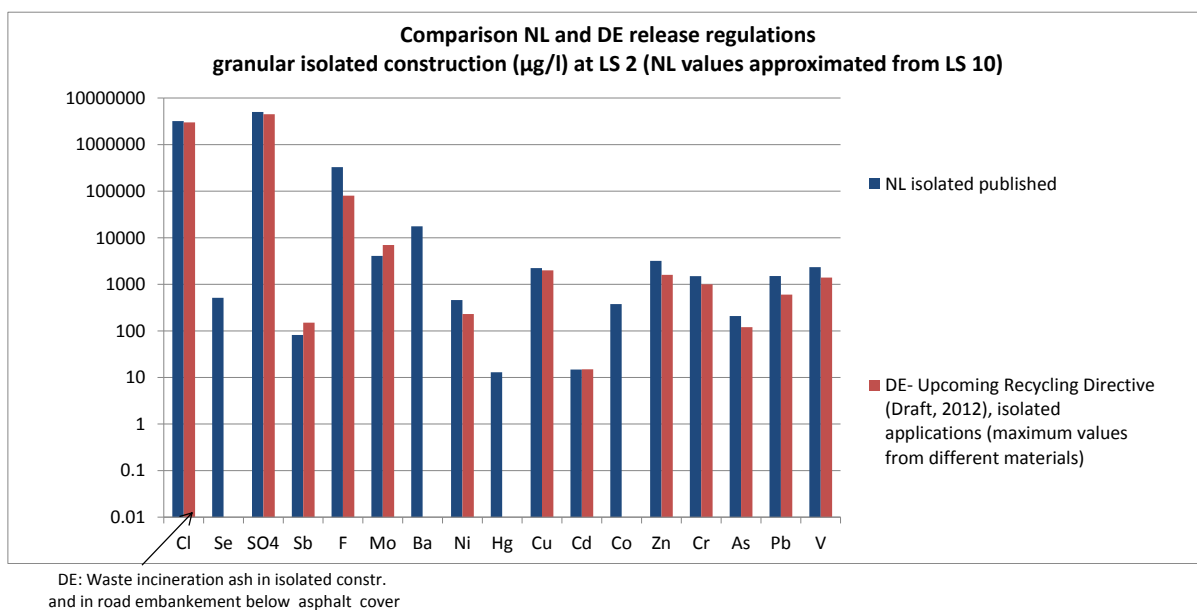


Fig. 3.11: Comparison of Dutch and German limit values (µg/L) for isolated (closed) applications.

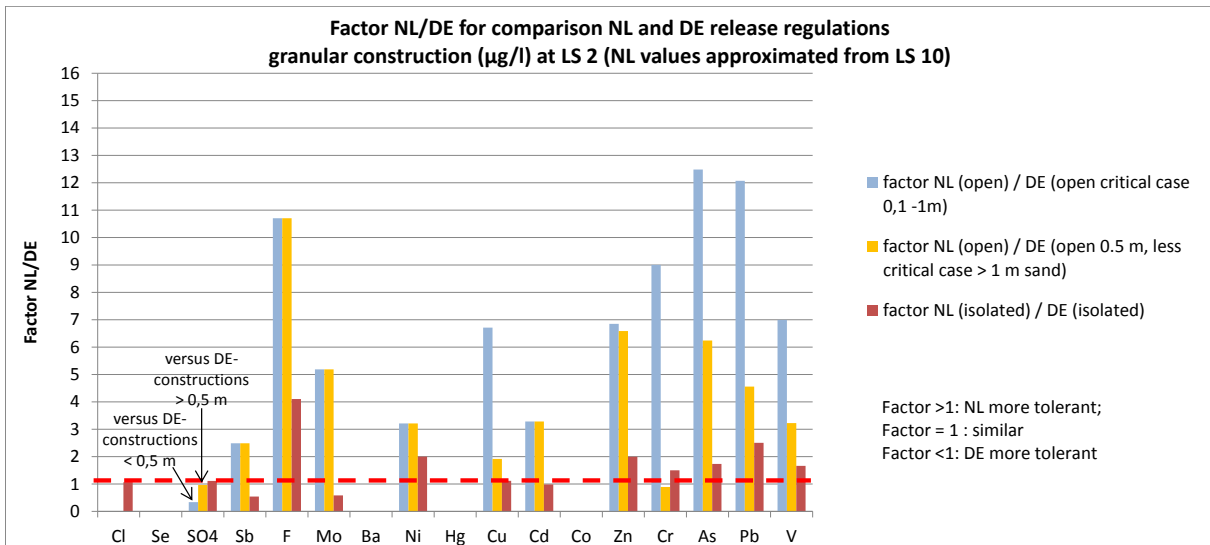


Fig. 3.12: Factors of difference between the Dutch and the German limit values for all scenarios.

Conclusion of comparison of DE and NL release limits

Granular, open construction:

The Dutch release limits for open constructions are generally factor 3 to 12 more tolerant than the German limits.

There is one exception for sulfate in constructions of moderate thickness < 0.5 meter, with a factor 3 more tolerant limits in Germany. For constructions > 1 meter, the sulfate-limits are similar.

Granular, isolated construction:

The Dutch release limits for closed constructions are factor 0.5 to 5 more tolerant than the German limits.

There are two exceptions for molybdenum in the singular case of coal fly ash with factor 1.7 more tolerant application limits in Germany and for the singular case of waste incineration ash (quality class 2) with factor 2 more tolerant limits in Germany. Note that DE limits are not derived with impact assessment modeling, the Mo- and Sb-values are 90th percentiles of concentration distribution of measured LS 2-eluates.

3.4. Important factors that cause differences between release limits in Germany and the Netherlands

The most important factors influencing the differences between release limits as evaluated within this project are as follows:

- Only 3 “Generic” scenarios in the Netherlands versus over 120 “situation specific” evaluations in Germany; many of the DE scenarios are more strict than the generic scenarios in NL. In the NL Soil Quality Decree deviations from the generic scenarios with respect to application height, distance to groundwater etcetera are accepted.
- Source term functions using a constant metal (source with cumulative LS 2) concentration versus Dutch L/S dependent concentration behavior. Although the NL source term in the impact assessment modeling is in principle a more stringent way of evaluating the source term because of the initially high concentrations for substances ($C_{LS 0,1}$ versus $C_{LS 2}$), the overall effect on differences in release limits between DE and NL compared to other factors is considered to be relatively small. The effects of source term definition on the observed differences between NL and DE release limits are limited mainly to “intermediate” sorbing metals. For salts and strong sorbing substances there is no strong effect from the source term definition. Remaining relevant effects from the source term definition are presumably cancelled out / compensated against other more important factors such as appraisal times (see below) and the larger diversity of scenarios in DE among which is a “critical case” (see below).
- The time frame/appraisal time (100 years in NL, 200 years in DE). This is considered by the authors to be a very determining factor for differences between release limits of DE and NL. The large effect of appraisal times was previously identified in Verschoor et al. (2006).
- The thickness of soils effective for retardation/attenuation processes (2 meters NL, 1 meter DE). This factor is of a relatively minor importance, as it causes a minor shift of breakthrough curves in time (about a factor of 1,5) and it hardly influences the concentrations at the POC. However, when this factor is seen in conjunction with appraisal time, a factor of 1,5 shift in breakthrough time (e.g., breakthrough at 80 or 120 years, with an appraisal time of 100 years) this factor could amplify differences in release limits between NL and DE.
- The distinction of a critical case in Germany. If the groundwater distance is between 0.1 and 1 m, there is no calculation (no retardation taken into account) of impact

assessment, and the LS 2 cumulative concentration equals GFS or background value times factor 1.5. And there is no recycling allowed at less than 0.1 m or within the groundwater.

3.5. Comparison of judgment and quality control in Germany and the Netherlands

A comparison of the judgment and quality control systems gives insight, how the measurement results from leaching tests are applied in the different member states. Notwithstanding, that standardization does not cover “impact assessment”, it is important to understand the relation between test method and impact assessment and what member states ask from standardization bodies in relation to boundary conditions to the test methods. Figure 3.13 gives a comparative overview.

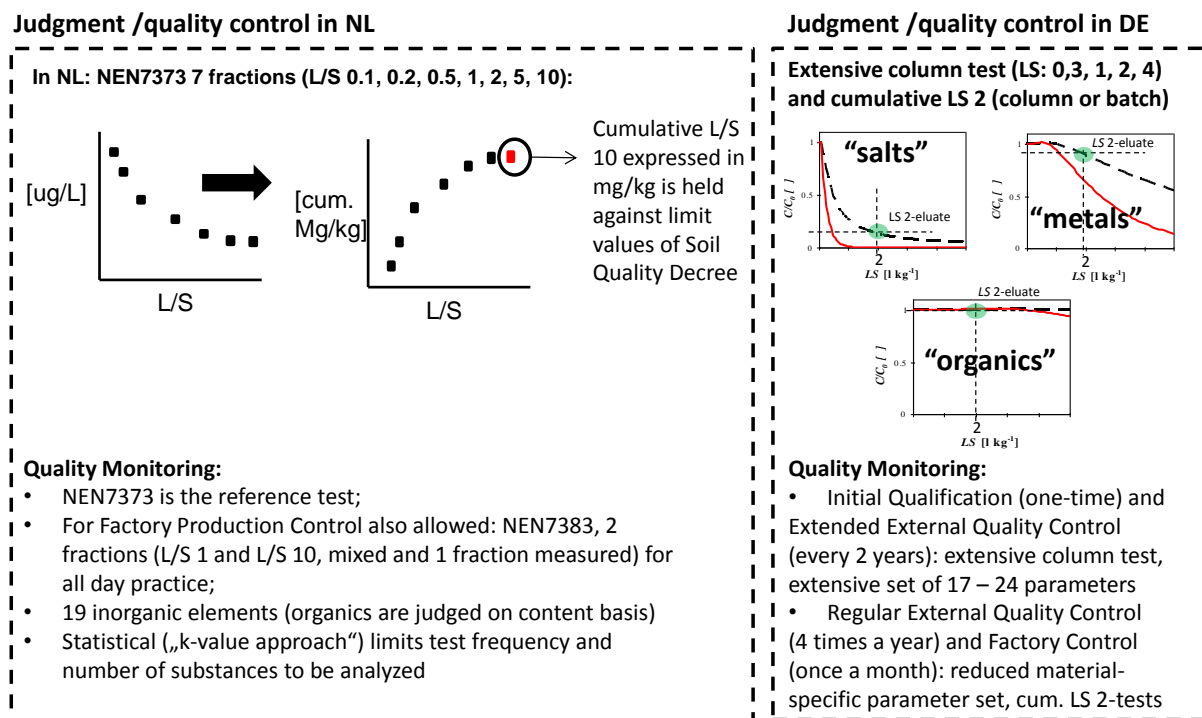


Fig. 3.13: Comparison of Dutch and German judgment and quality control system

In both countries column test are performed in an up-flow mode, as an extensive test for basic characterization (collection of eluate fractions at LS 0.3, 1, 2, and 4 in Germany and at LS 0.1, 0.2, 0.5, 1, 2, 5 and 10 in the Netherlands).

For factory control in Germany, a short-term column test for compliance testing is performed, where the aqueous effluent is collected up to LS 2 in one fraction. The cumulative

concentration in LS 2 eluates of the short-term column percolation test (DIN 19528, 2009) represents an average concentration which averages between initial high concentrations and medium-term lower concentrations or medium-term higher concentrations. Regarding the common applications of mineral recycling materials in technical constructions in the field, LS 2 corresponds to medium time scales of several years to decades (depending on the thickness and seepage water rate, etc.). In most cases concentrations declines with time and therefore the LS 2 concentration is dominated by the initial higher mass release.

A difference between German and Dutch judgment/quality control is, that in Germany the cumulative concentration held against the limits is the LS 2-concentration, which is in most cases dominated by initial high release rates. Long-term release doesn't play a dominant role in the German risk assessment, as LS 2 cum-concentration is judged and taking into account the conservative limits a material "fails" caused by initial concentration contribution not by long-term release (with very conservative limits). A possible concentration increase or decrease that occurs after L/S 2 is therefore outside of the "window" of the standard German test procedure, as a constant (high) concentration is assumed after L/S 2.

With respect to a possible increase, there are certain reactive materials which show increasing concentrations over time (e.g., increasing concentrations of vanadium in the seepage water of waste incineration ash). In German research projects it was demonstrated in various test, that concentrations in LS 2 short-term column percolation tests were still high enough to judge the materials safely (because the initial release was still high enough, concentration rise already after LS 1 and the German limit values are very conservative). For details see Grathwohl & Susset (2011).

The DE approach can be seen as "conservative", because of the assumption that high initial concentrations are assumed to remain constant over time. In NL, a (substance-specific) concentration decrease of the source term over time is part of the approach (in the kappa-approach). This implies that any increase of concentrations to values above the soil and groundwater criteria at the POC after the appraisal time (fig. 3.8), are more easily accepted by policy makers, as such increased concentrations are known to be only temporarily.

In the Dutch judgment/quality control system a cumulative LS 10 concentration (expressed in mg/kg as a sum of 7 fractions) is held against the limit values of Soil Quality Decree. For factory production control it is also allowed, according to NEN7383, to mix fraction L/S 1 and L/S 10 for all day practice.

A widely spread misconception is that in the NL system, apart from the more tolerant release criteria, the judgment of materials is even more tolerant than in DE as the cumulative L/S 10 result “average” between initial high concentrations and long-term usually lower concentrations. This averaging effect would indeed be more tolerant if both the judgment and impact assessment were based on constant concentrations over time (as in DE, see below). However, in NL the generic release criteria are not based on a constant “average” concentration in a certain L/S range between L/S 0 and 10, but on a source term with high initial concentrations C_0 at L/S 0.1 (see Figure 3.4) which decreases between L/S 0.1 and 10 according to the substance-specific kappa-approach (see Figure 3.4). Expressing the value in mg/kg at L/S 10 (or any other L/S value) does therefore not make a difference for the value of C_0 , and hence, does not make a difference for the compliance of products to the limit values due to “dilution” or whatsoever. This also implies that for the “generic” judgment of materials in the SQD, the exact L/S-concentration behavior to L/S 10 is strictly spoken not important as only the L/S 10 cumulative amount in mg/kg “counts”, and the shape of the source term is assumed to behave according to the kappa- formula in paragraph 3.2.1. Of course, for individual products the time dependent concentration behavior may deviate from the kappa-approach (see also below). Initial concentrations measured in the test may be either higher or lower than the assumed C_0 .

The reasons why NL developed NEN 7373 consisting of 7 fractions (and NEN7383 for factory production control) is because NL NEN7373 is a “basic characterization test” which allows the use of the test also for more specific questions. It is important to address that there is also a route parallel of the “generic” risk assessment in NL, which can be addressed as “specific” risk assessments. A new or special material that is brought to the market, or gives rise to a specific concern by regulators, should be tested with all 7 fractions in NEN7373. The concentration pattern derived from the test is informative for the release mechanism, and provides answers whether the observed release is very temporal (e.g., only in the first fractions after which it stops), remains constant over the entire duration of the test, or increases as a function of L/S. The first example may be informative to producers in order to take measures that may eventually lead to acceptance of a product. But in particular the final example may give rise to additional investigations, even when the product complies with SQD regulations on the basis of a cumulative L/S 10 value. An example of products that have recently followed the latter route in NL are certain types of fine steel slag granulates in ponds and parking lots, and artificial turf (rubber infill) in soccer fields.

4. Conclusions

Impact assessment in Germany and the Netherlands

- Common goals in protecting soil and groundwater and the fundamental principles of impact assessment in Germany and the Netherlands are similar.
- There are similarities and differences in absolute concentration of soil protection and groundwater protection values. For groundwater, the protection levels are relatively similar.
- Main influence on release limit values are given by political conventions caused by different soil- and groundwater protection philosophies (time frame/appraisal time with 100 years in NL, 200 years in DE, thickness of soils effective for retardation/attenuation processes with 2 meters NL and 1 meter DE, critical case in Germany) and not by the modeling concepts.
- Even if a soil- or groundwater protection value in different MS is the same, the derived release limit values may differ, caused by different point of compliances and / or appraisal periods and /or model approaches, etc.

Release limit values for construction products in Germany and the Netherlands

- The Dutch release limits are generally more tolerant than the German release limits.
- Granular, open application: Dutch factor 3 to 12 more tolerant than the German limits with exception of sulfate.
- Granular, isolated application: Dutch release limits for closed constructions are factor 0.5 to 5 more tolerant than the German limits with exception of molybdenum. (coal fly ash) and antimony (waste incineration ash).

Judgment and quality control in Germany and the Netherlands

- In Germany, the cumulative concentration ($\mu\text{g/L}$) at L/S 2 is compared to the limit value.
- In the Netherlands, the cumulative release at L/S10 (mg/kg) is compared to the limit value.
- LS 2 averages between initial high concentrations and typically medium-term lower concentrations, LS 10 averages between initial high concentrations and typically long-term lower concentrations. However, the LS 10 values always relate back to by default high initial concentrations due to the kappa-approach, with which the release limits are derived.
- In Germany long-term release doesn't play a dominant role in the judgment of, as in practice, when a measured LS 2 value is compared with the release limits, a material "fails" already caused by initial concentration contribution not by long-term release.

However, the impact assessment with which the limit value is calculated, is based on an appraisal time of 200 years and assuming (for metals) a constant source during the complete appraisal time.

- Main reasons for choosing a test conducted in several fractions to L/S 10 in NL are to build up experience and knowledge on how the leaching of different products would proceed as a function of L/S, and L/S 10 being a value that is reached within a time period of decades for many applications. This information can later be used for both generic and specific impact assessments. For instance, “specific” risk assessments are needed for special materials that are brought to the market, or products/scenarios that give rise to a specific concern. In those cases, products should be tested with all 7 fractions in NEN7373. The concentration pattern derived from the test is informative for the release mechanism, and provides answers whether the observed release is very temporal (e.g., only in the first fractions after which it stops), remains constant over the entire L/S range (and possibly beyond) or increases as a function of L/S. For factory production control the same test (NEN7373) is carried out with less fractions (NEN7383), but both tests run up to L/S 10 and hence, the results of both tests are exactly the same.
- For the above use of the test, local equilibrium is important.
- In Germany for basic characterization 4 fractions to L/S 4 are collected and measured (LS-fractions: 0.3, 1.0, 2.0, 4.0). For special issues the standard is open for measurements at higher LS.

5. Technical requirements of TS-3, options for standardization and regulation issues

Technical requirements

As will be outlined below, the technical requirements to TS3 that NL and DE need for their impact assessment, are very similar:

- A laboratory column up flow percolation test should be suitable for all granular construction products.
- Test result should approach local equilibrium as best as possible (i.e. rather insensitive to contact time), and for NL important: all over the duration of the test;
- Test result should avoid clear artifacts due to size reduction
- The test should quantify the initially high concentrations (i.e., L/S 2 in DE, and L/S 0.1 and further in NL).
- The test should not be based on a single/averaged concentration at L/S 10; this would not be compatible with the DE impact approach; it would also not be compatible with NL approach which takes changing concentrations as function with L/S into account.
- The test should at least allow quantification of L/S 2 eluates (DE) and proceed up to L/S 10 in several fractions (NL) to be compatible both with NL and DE approach.

The above similarities in impact assessment and requirements to TS3 are in contrast with the resulting limit values and scenarios, which are much more stringent in DE. This has the consequence that similar products may be judged much more stringent in DE than would be the case in NL. This discrepancy in judgment may be an important reason why Option A of TS3 will allow application of the product in NL, while the same product based on this assessment would not be acceptable for application in DE. However, option B would more easily allow application of this product under DE legislation. The reason is that option B is assumed to comply more often with DE limit values, as option A may lead to initial higher concentrations (when expressed in average concentration between L/S 0 and L/S 2).

Whether initial concentrations in option A are the result of local equilibrium (NL position) or that artifacts may play a role and/or that true local equilibrium rarely occurs in many scenarios (DE position) has therefore become the focal point of the discussion. It should, however, be stressed that this technical discussion on interpretation of results is the consequence of the limit values being more strict in DE than in NL. A more useful discussion, that would more easily lead to technical agreement, should focus on differences in choices

that have been made during impact assessment, and make judgment of products more similar in NL and DE. This discussion belongs to regulators.

Options

A technical, measurement program in order to evaluate the suitability of a test protocol with respect to the above requirements (option A or B) should take into account:

- Contact time (to evaluate local equilibrium)
- “porous” and “low porous” materials
- Size reduction: Look also for materials that are not too sensitive to crushing + differences in contact time;
- Effects to be quantified for L/S 2 and 10
- address situations in which there is evidently effect of size reduction artifacts (when is size reduction “helping nature” to establish local equilibrium, and when is it not?)

What belongs to regulators

Although the goals of protecting soil and groundwater are similar, and also the way impact assessment modeling is done is quite similar, this project made obvious, that there are very relevant differences in assumptions describing the application scenarios in Germany and the Netherlands (and perhaps in other member states).

These differences in assumptions have led to two assessment systems, from which one leads to very conservative limits, where “some $\mu\text{g`s}$ ” decide between failing and passing on the one hand (DE), and another assessment system leads to much more tolerant limits which are less sensitive to measurement differences in “ $\mu\text{g-range}$ ” (NL).

In general, there is a contradiction between a maximal protection of soil and groundwater and the maximum allowable degree of recycling of products. On the one hand, a strict and worst-case protection of soil and groundwater may limit the (re-)use of construction products. A tolerant policy for the (re-)use of construction products may on the long term limit the quality of soil- and groundwater. The latter processes are very long-term (decades to centuries). It is up to policy makers to find a balance between the different goals, and to take care of an as much as possible unified approach in the EU member states.

6. References

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- Susset, B., Grathwohl, P. Leaching standards for mineral recycling materials – A harmonized regulatory concept for the upcoming German Recycling Decree. Waste Management 31 (2011) 201–214.