CALCULATING SITE-SPECIFIC EMISSION CRITERIA FOR SUSTAINABLE LANDFILLS: THE DUTCH APPROACH

J.J. DIJKSTRA*, A. VAN ZOMEREN*, R.N.J. COMANS**, E. BRAND° AND J.W. CLAESSENS°

Energy Research Centre of the Netherlands (ECN), P.O. Box 1, 1755 ZG PETTEN, The Netherlands
*** Wageningen University, Dept. Environmental Science, P.O. Box 47, 6700AA, WAGENINGEN, The Netherlands*
° National Institute for Public Health and the Environment (RIVM), PO. Box 1, 3720 BA BILTHOVEN, The Netherlands

SUMMARY: Recently, a project has been started in the Netherlands to investigate whether source-oriented treatment techniques over a period of one or more decades will lead to a significant reduction in emission potential of a number of pilot landfills. In order to judge whether the leaching of contaminants from a treated landfill has reached an environmentally acceptable level to release the landfill from aftercare, emission criteria for the landfill leachate need to be established. A novel generic methodology is developed to calculate site-specific emission criteria, expressed as maximum concentrations (µg/l) of target contaminants in the landfill leachate, while considering free infiltration of rainwater. Presently, site-specific emission criteria are being calculated for three different pilot landfills in the Netherlands. The methodology is based on a "source-path-point of compliance" approach, and takes into account site-specific factors that influence the transport rate of leached contaminants on the "path" between the "source" (the landfill) and the downstream point of compliance, where site-specific groundwater limit values have to be met. The project is considered to be in line with the Water Framework Directive (WFD, 2000/60/EC) and Groundwater Directive (GWD, 2006/118/EC). The choices made concerning the environmental objective, background concentrations and location of POC are related as much as possible to the Dutch interpretation of the WFD and GWD. The focus of this paper is on the technical approach by which the emission criteria are calculated, as well as the sensitivity of calculated emission criteria for different choices in chemical-, physical and policy-oriented parameters and boundary conditions. Final emission criteria for three pilot landfills will be published in the second half of 2013.

1. INTRODUCTION

Although more waste materials are recycled or re-used nowadays, landfilling of waste materials is still necessary in the future. The necessary everlasting aftercare of landfills raises concern. In

the Netherlands, the Sustainable Landfill Foundation was set up in order to stimulate the development of new landfill technologies to minimize impact and thus reduce or eliminate the need for long-term aftercare. A five-year research program on development of newly designed landfill sites (predominantly inorganic waste, organic waste, bioreactor and stabilised waste) was performed in the Netherlands (Luning et al., 2006;Mathlener et al., 2006;Van Zomeren et al., 2006;van Zomeren and van der Sloot, 2006) The concept of sustainable landfills aims at creating a situation where a biogeochemical equilibrium between a landfill and the environment is reached within a period of 30 years, in order to reduce the long-term risk and the aftercare needs. This can be accomplished by constructing a landfill that has chemical properties that will minimize leaching of contaminants (e.g. neutral pH, slightly reduced conditions, low organic matter content and relatively low salt loads).

However, there are many landfills that are currently in operation or that have already been closed. Hence, there is also a need for the development and application of treatment techniques to reduce the emission potential of existing landfills. Recently, the Dutch Sustainable Landfill Foundation and the Dutch Ministry of Infrastructure and Environment have started up a research project called 'Sustainable emission reduction at existing landfills', in order to test if source-oriented treatment techniques will lead to a significant reduction in emission potential of a number of pilot landfills (Kattenberg and Heimovaara, 2011). The aim of these pilot projects is to investigate whether long-term treatment of the waste body by irrigation and recirculation, and/or subsequent aeration over a period of one or more decades, are sufficiently effective to reduce the remaining emission potential in the landfill to a level that does not pose an undesired risk to the environment. In that case, the landfill might be released from or the aftercare (Kattenberg and Heimovaara, 2011), or the aftercare measures can be adjusted to the actual remaining risk potential (e.g. less monitoring, less stringent requirements on top liner).

In order to judge whether the leaching of contaminants from a (treated) landfill has reached an environmentally "acceptable" level to release the landfill from aftercare, emission criteria need to be established. To this end, a generic methodology is developed to calculate site-specific emission criteria that are expressed as maximum concentrations (µg/l) of target contaminants in the landfill leachate, while considering free infiltration of rainwater. Presently, site-specific emission criteria are being calculated for three different pilot landfills in the Netherlands. The methodology is based on a "source-path-point of compliance" approach, and takes into account site-specific factors that influence the transport rate of leached contaminants on the "path" between the " source" (the landfill) and the downstream point of compliance ("POC", see Figure 1). The choices made concerning the environmental objective, background concentrations and location of POC are related as much as possible to the Dutch interpretation of the Water Framework Directive (WFD, 2000/60/EC) and Groundwater Directive (GWD, 2006/118/EC).

The emission criteria should be interpreted as the maximum concentrations (µg/L) of contaminants in the landfill leachate, that do not exceed the local groundwater quality criteria at the POC (in the current scenario a vertical planar surface located at 20 metres downstream from the landfills, POC2). Site-specific factors that are taken into account for the unsaturated zone include the soil-specific pH and content of mineral and organic reactive surfaces to which released contaminants can bind, the natural background concentration of contaminants in groundwater, as well as hydrological dilution of the contaminants on the "path" towards the POC.

The emission criteria are calculated for a group of 12 inorganic contaminants (soluble salts, metals, metalloids and oxyanions) and over 40 organic contaminants (BTEX, PAH, VOX and mineral oil fractions). An important part of the calculation of emission criteria is that of the retardation in the soil between the landfill and the POC. This part of the approach was modelled using the reactive transport code Orchestra (Meeussen, 2003). For inorganic contaminants, the retardation in soil was modelled in Orchestra based on the generic multi-surface approach as

described by Dijkstra et al. (2009). This approach is based on generic thermodynamic reactions and parameters, and has been independently validated on contaminated soils (Dijkstra et al., 2004;Dijkstra et al., 2009). The approach was previously used in the Netherlands to calculate emission criteria for construction products in the Soil Quality Decree (Verschoor et al., 2006). For organic contaminants, linear distribution coefficients (Kd) were used for reactive transport modelling based on the site-specific organic matter content in the soils and the log Koc of the contaminants.

The calculation of the emission criteria is an iterative process (see Figure 1), and involves many assumptions and policy-related choices. Examples of assumptions that have an influence on the calculated emission criteria include the time-dependency of the source term (e.g., constant or declining concentrations in the leachate as a function of time) and local soil chemistry (e.g., pH and organic matter content). Examples of policy-related choices that influence the emission criteria include the time period during which concentrations in groundwater should remain below the groundwater quality criterion (e.g., 100 or 1000 years) and the distance of the landfill to the POC.

The approach of calculating emission criteria, an overview of the most important assumptions and policy-related choices and their possible influence on the emission criteria will be discussed in more detail below. The focus of this paper is on the technical aspects of the approach. Presently, calculations are on-going, and final emission criteria for three pilot landfills will be published in the second half of 2013.

2. APPROACH

2.1 General approach to establish site-specific emission criteria

Figure 1 schematically shows the general approach that is followed to establish site-specific emission criteria for landfills. In the approach, the source term concentration (step 1) is adjusted in such a way that the concentration of a contaminant, after transport through soil and groundwater (step 2), is below the groundwater limit value within a certain time frame (step 3 is the comparison with the groundwater limit value, and step 4 indicates the next iteration if necessary). When the concentration at the POC equals the groundwater limit value, no further iterations are necessary, and the source term concentration in step 1 equals the site-specific emission criterion (step 5).

It is important to note that similar methodologies have been followed to develop emission criteria for construction products and large-scale applications of (excavated) soil and sludge in the Netherlands (Soil Quality Decree) as outlined in Verschoor et al. (Verschoor et al., 2008;Verschoor et al., 2006). Also, emission criteria for recycled construction products in Germany have been calculated in conceptually a very similar way (Dijkstra et al., 2013;Susset and Grathwohl, 2010), as well as the Annex II Landfill Directive limit values. These approaches are, however, generic with respect to choices and parameters, while the present approach was designed to allow for more site-specific features.

Conceptually, the approach to establish site-specific emission criteria consists of three main model compartments:

(1) The "source term". The source term is the landfill that potentially emits contaminants to the soil and groundwater environment through leaching of (rain)water through the landfilled waste. It is the "source term" for which emission criteria need to be established. The source term may be constant over time (a constant concentration as a function of time), or may have a different temporal pattern (decreasing, increasing etc.).

(2) The "pathway". After the contaminants have entered the environment, they will be

transported through soil and groundwater. Except for soluble salts, the velocity by which contaminants travel through soil and groundwater is for most contaminants slower than that of the water in which they are dissolved because of sorption processes. This process is referred to as "retardation". The soil and groundwater environment between the landfill and the "point of compliance" (see below) is referred to as the "pathway".

(3) The "point of compliance (POC)". The POC is the location at a certain distance from the source, somewhere on the pathway, where the local groundwater quality criterion is supposed to be met. The "breakthrough" time of a contaminant is the time needed for a contaminant to arrive at the POC.

The estimate of the retardation in the soil along the "pathway" is based on a geochemical speciation model (Dijkstra et al., 2009), in which different inorganic contaminants compete with each other for sorption "sites" to soil particles. Because of this so-called competition, as well as the fact that binding of contaminants to soil particles is in itself a non-linear process (i.e., doubling the dissolved concentration of a contaminant does not lead to doubling the amount of sorbed contaminant), the process of calculating an emission criterion requires a number of iterations. When the maximum concentrations at the POC with the desired time frame are equal to the groundwater criterion, the concentrations emitted by the source term are equal to the site-specific emission criteria.
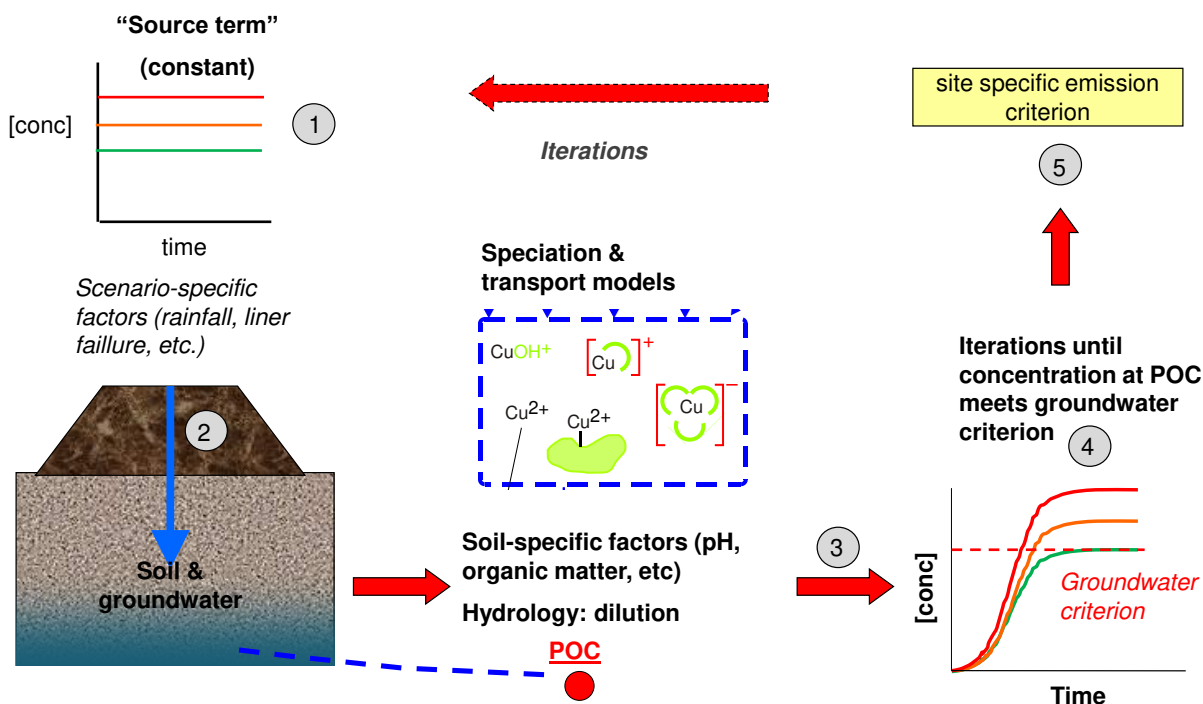

Figure 1. Methodology to calculate site-specific emission criteria for the three pilot-landfills. An iterative calculation process (step 1 to 5) results in limit values for the source (landfill leachate concentration) that do not exceed groundwater quality criteria at the point of compliance (POC) downstream the landfills. Step 1 is the starting point, with an estimated concentration in the source term. In step 2, reactive transport in soil and groundwater is calculated. In step 3, resulting concentrations at the POC are compared to the groundwater limit value. If the concentration is not equal to the groundwater limit value (e.g., the red and orange line in the figure), a next calculation is needed (iteration) until the concentration in the source term in step 1 is such that the concentration at the POC in step 4 meets the groundwater limit value (green line). That concentration is then defined as the site-specific emission criterion in step 5.

2.2 Pilot landfills

Presently, three pilot landfills are selected for which site-specific emission criteria are calculated. With respect to important parameters in calculating site-specific emission criteria, the landfills differ with respect to factors such as depth of the unsaturated zone, groundwater flow and resulting dilution of landfill leachate. An important feature is also the physical and chemical characteristics of the underlying and surrounding soil, which are different for each of the landfills.

2.3 Model approach and main assumptions

The scenario approach of calculating site-specific emission criteria has a generic basis (i.e. equal for all participating landfills), but is parameterized as much as possible in a site-specific manner. Figure 2 provides an overview of a number of important boundary conditions of the model approach that are highly determining for the value of the emission criteria. Figure 2 also identifies the main parameters that are generic and site-specific. The boundary conditions will be discussed in the paragraphs below.

Choices and decisions with respect to the model scenario are the result of cooperation between the National Institute for Public Health and the Environment (RIVM) and the Energy research Centre of the Netherlands (ECN), in consultation with the Landfill sector, coordinated by the Dutch Ministry of Infrastructure and the Environment.
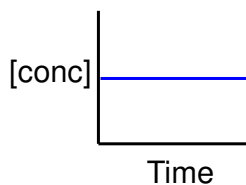
- Generic source term: constant and 300 mm/y
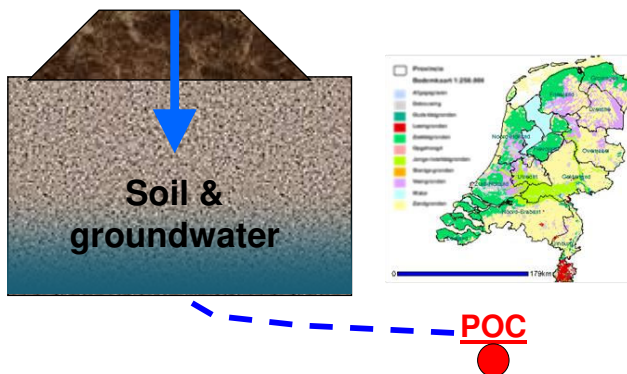
- Site specific soil properties: database of Dutch soil profiles (STONE database);
- Generic profile thickness: 1 m unsaturated + 1 m saturated;
- Generic: no retardation between 1st metre groundwater to POC;
- Site specific dilution factor between 1st metre groundwater and POC;
- Generic POC: planar surface at horizontal distance 20 m from groundwater table
- Site-specific criterion at POC: MPC (ug/l) in groundwater (inorganic), negligible risk value (organic)
- Generic time frame: 500 years

Figure 2. Important choices and boundary conditions of the model approach of calculating site-specific emission criteria.

2.2.1 Source term

A choice was made to calculate site-specific emission criteria that are protective for the period of 500 years. The emission criteria are calculated for a (worst) case where the liner systems of the landfill show complete failure from t=0 onwards, in which t=0 is the moment at which the landfill treatment period is ended and the aftercare or completion phase starts. For the model scenario this implies that all precipitation that falls on the landfill will be conducted through the landfill body into the soil below the landfill. A net downward flux of 300 mm/year is assumed through the bottom liner of the landfill, which is equal to the net infiltration of natural soils in the Netherlands.

Furthermore, it was decided that the concentration in the landfill leachate is constant over the duration of 500 years from the moment the treatment period is ended. This choice was made on the assumption that the generally large waste body in a landfill will lead to small temporal changes in leachate concentrations on the perspective of the time period. Given the average height of the landfills of about 15 metres, an assumed dry bulk density of 1550 kg/m3 and a net infiltration of 300 mm/year, an L/S ratio of about 6.5 l/kg is reached after 500 years, which is roughly two-third of the L/S value (L/S 10) of a regular percolation test such as CEN/TC 292 TS14405.

The concentrations of target contaminants in the leachate (source term) are subject to the iterative calculation (see Figure 1 and 2).

2.2.2 Pathway

The generic modelling scenario includes, below the landfill, a vertical soil profile with a thickness of 2 metres. The upper 1 metre is defined as the unsaturated zone, the lower 1 metre is defined as the saturated zone. Sorption of contaminants can take place over the full thickness of the soil profile. The transport of water and contaminants is based on a relatively simple 1-D saturated downward transport scheme consisting of 2 model soil layers. The calculated groundwater concentrations in cell between 1 and 2 metres depth are representative for the "upper 1st metre" of groundwater below the landfill. The choice for 2 cells instead of a fine grid of e.g. 20 soil layers results in somewhat earlier breakthrough but more dispersed concentration profiles, compared to late breakthrough and sharp concentration gradients in case of a finer grid. This choice was made based on a preliminary model evaluation that takes into account, in a generic manner, the potential effects of preferential flow, which are known to be extremely important but heterogeneous in their nature and spatial occurrence (unpublished results).

Because no sufficient detailed information has become available on the binding properties of the soil between the upper metre of the saturated zone and the POC, it is assumed that no sorption takes place between the upper metre of the saturated zone and the point of compliance (a planar surface at 20 metre distance). However, site-specific hydrodynamic dilution factors are taken into account for dilution with local groundwater, which range in the present study from a factor of 1 (no dilution) to 4.7 (considerable dilution).

In summary, the total "attenuation" of emitted contaminants consists of two parts: (1) the retardation due to sorption in the upper 2 metres of soil, followed by (2) only dilution towards the point of compliance.

In particular the chemical properties of the soil below the landfill are important for the degree of "retardation" of contaminants before these enter the saturated zone. The mobility of contaminants in soil predominantly depends on the pH and the amounts and type of reactive surfaces to which contaminants may bind: reactive dissolved organic matter (i.e. fractions of Dissolved Organic Carbon, DOC) and particulate organic matter, iron- and aluminium hydroxides and clay minerals (Dijkstra et al., 2009).

For each of the landfill sites, properties from a nearby soil profile were selected from a large

Dutch soil database (STONE database), for which these data are known for layers of 10 centimetres to a depth of 2 to 3 metres. The pH-dependent sorption of inorganic contaminants to these surfaces is modelled using the reactive transport code Orchestra (Meeussen, 2003) and is based on the generic multi-surface approach as described by Dijkstra et al. (2009). This approach is based on generic thermodynamic reactions and parameters, and has been independently validated on contaminated soils (Dijkstra et al., 2004; Dijkstra et al., 2009). An advantage of this approach is that the adsorption can be computed for soils with different binding properties, and no fitting of sorption parameters to specific soils is necessary. Also, important interactions that influence contaminant mobility are taken into account, such as precipitation of minerals and competition between contaminants for sorption sites. The approach was previously used in the Netherlands to calculate emission criteria for construction products and large-scale applications of (excavated) soil and sludge in the Soil Quality Decree (Verschoor et al., 2006). For more detail on the chemical part of the model is referred to Dijkstra et al (2009).

For organic contaminants, linear distribution coefficients (K_d) were used for reactive transport modelling based on the site-specific organic matter content in the soils (STONE database) and the log K_{oc} of the contaminants (Brand et al., 2007).

2.2.1 Requirements at point of compliance

The POC consists of a vertical planar surface located at 20 metres downstream from the upper metre of the saturated zone below the landfill. It was decided that the model does not take explicit transport into account between the upper metre of the saturated zone and the POC, only a dilution factor that is site specific, and which is based on separate hydrological studies. Site-specific background concentrations (estimated from long-term upstream monitoring data at each landfill site) is taken into account in the dilution.

The concentration at the POC must fulfil site-specific groundwater criteria: (1) For inorganic substances the groundwater criteria are determined by the Maximum Permissible Concentrations (MPC). These MPC values consist of a site-specific part, i.e. the local background concentrations, and a generic part, which are the Maximum Permissible Addition (MPA) values. (2) For organic substances, the groundwater criteria are based on the level of negligible risk for ecosystems. The level of negligible risk is in principle defined as the MPA value divided by a factor of 100. However, in the case that the drinking water standard is lower than the level of negligible risk, the drinking water standard determines the groundwater criteria for both organic and inorganic substances. For chloride, sulphate and ammonium, the groundwater criteria are determined by the lowest of the MPC or drinking water standard. In the cases that the background concentration in groundwater is higher, the groundwater criteria are determined by the background concentration in groundwater.

Depending on the degree of retardation in the soil profile, contaminants arrive at different times at the POC. The highest concentration that arrives at the POC (i.e. after retardation and dilution) within a period of 500 years must fulfil the MPC value.

3. RESULTS AND DISCUSSION

Presently, calculations are on-going and final emission criteria for three pilot landfills will be published in the second half of 2013. Below, a number of diverse and representative examples are shown of modelled behaviour of contaminants as a result of certain site-specific properties, but site specific emission criteria cannot be inferred from this data.

3.1 General observations

Figure 3 shows a typical model result and is used to demonstrate a few important principles. First of all, it is important to note that regardless of the degree of binding of a contaminant, a "constant source term" implies that sooner or later all contaminants will reach the same concentration at the POC as in the source term (in case also hydrological dilution is taken into account, the concentration at the POC will be proportionally lower; see black vertical arrow in Figure 3).

Next, it is important to understand that calculation of an emission criterion requires a time frame in which the (modelled) concentration at the POC must be below a certain groundwater criterion. If no time frame would be used (implying an infinite or eternal time frame), the emission criteria from the landfill would become equal to the groundwater protection criteria, corrected for the hydrological dilution factor. The effect of time frame on the calculated emission criteria also depends on factors such as the assumed thickness of the (un)saturated zone in which sorption of contaminants is assumed to occur, the groundwater flow rate, and the distance between the source and the POC. In this study, no sorption is taken into account between the upper metre of the saturated zone and the POC (see 2.2.2). Given the boundary conditions of this study, the site-specific emission criterion will mainly be different from the groundwater criterion due to:

- Retardation (binding) of the contaminants in the unsaturated zone and the first metre of the saturated zone, in such a way that the maximum concentration at the POC is not reached within the time frame. For contaminants that are not subject to binding to soil (e.g., salts), the maximum will be easily reached within time frames longer than a few years. Contaminants that show strong binding, will only partially arrive at the POC within the time frame, or in some cases will not arrive at the POC at all.
- Hydrological dilution of the source term (dilution factor).

The principles with respect to time frame dependency of emission criteria are not unique to this study, but are identified previously in the calculation of emission limit values for construction products for the Dutch Soil Quality decree (Dijkstra et al., 2013; Verschoor et al., 2006) and the emission limit values for the German recycling decree (Dijkstra et al., 2013; Susset and Grathwohl, 2010).

The relative moment at which a contaminant arrives at the POC, depends on its binding affinity with the soil. The binding affinity is determined by intrinsic chemical properties of the contaminant, and specific soil chemical factors.
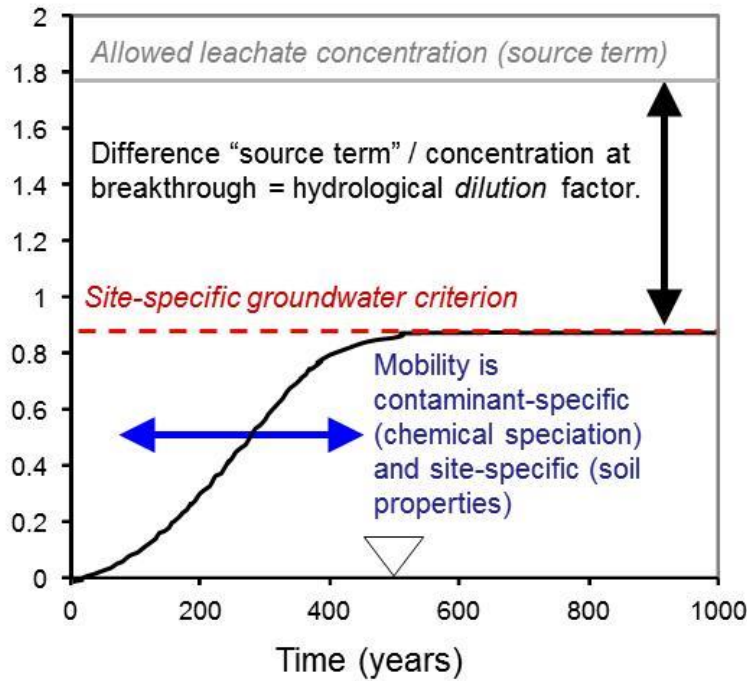
Figure 3. Typical result of a modelled concentration of a contaminant at the point of compliance (POC). In this case, the contaminant reaches its maximum concentration after a period of 500 years (indicated with a triangle on the time axis) and fulfils exactly the groundwater limit value. Depending on the chemical reactivity of the contaminant and the local chemical soil properties, the contaminant may arrive earlier or later (blue arrow). Because the source term is constant, the difference between the maximum concentration at the POC and the source term concentration equals the site-specific dilution factor.

3.2 Examples of different concentration-time profiles of contaminants at the POC

In general, four types of contaminant behaviour at the POC can be distinguished, that have a strong influence on the calculated emission criterion. They are indicated by case A, B, C and D (Figure 4). In Figure 4, the (constant) source term concentration is in all cases calculated such that the concentration in the groundwater at the POC meets the groundwater limit value within the time period of 500 years, i.e. the source term concentration represents the site-specific emission criterion and no further iterations are necessary. In these examples, the model calculations were performed up to 1000 years to indicate effects of breakthrough after the chosen time frame of 500 years.
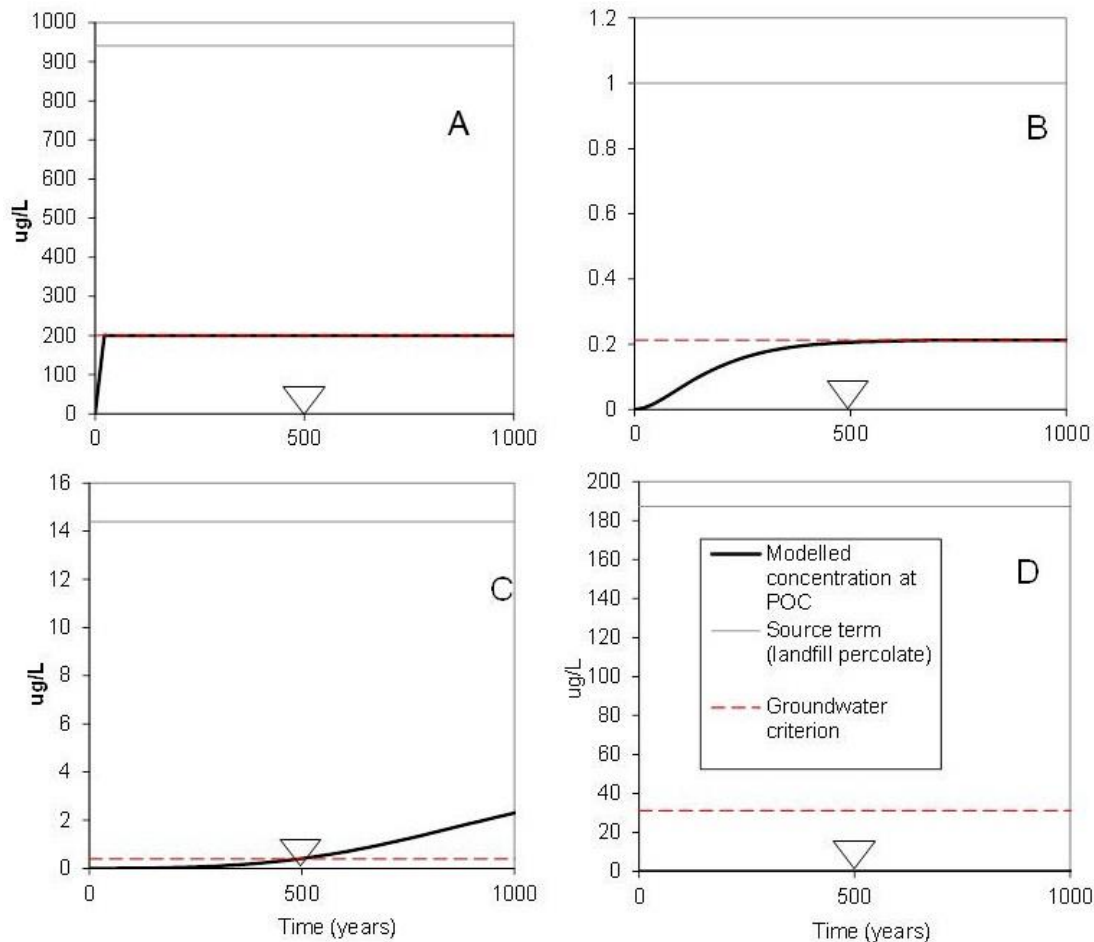
Figure 4. Four different concentration-time profiles of contaminants at the POC. The triangles indicate the end of the time frame (500 years) during which the concentration at the POC must be below the groundwater criterion. Behaviour A: rapid breakthrough, representative for mobile contaminants such as soluble salts; behaviour B: contaminants that show mild retardation, but still complete breakthrough within the time frame of 500 years; behaviour C: strong retardation, only partial breakthrough within 500 years; Behaviour D: extremely strong retardation, no breakthrough at the POC.

Case A is representative for mobile contaminants such as soluble salts (e.g., chloride). These contaminants travel with the same velocity as water and reach their maximum concentration in groundwater already after a few years (Figure 4A). Also contaminants that show considerable retardation according to behaviour B reach their maximum within 500 years.

Both in case A and B, the difference between the (constant) source term concentration and the maximum concentration in the groundwater equals the dilution factor at this location, which is 4.7 (as can also be estimated from Figure 4A and 4B). In other words; the maximum tolerable source term concentration (= emission criterion) is a factor of 4.7 higher than the groundwater criterion; which is solely due to the assumed hydrological dilution at this location. For case A and B, retardation is not strong enough to play a role in calculating the site-specific emission criterion.

On the contrary, contaminants that show behaviour C are strongly retarded, resulting in only partial breakthrough after 500 years. Also in this case, the source term is calculated such that the concentration after 500 years meets the groundwater criterion. In this case, the factor between the

concentration at the POC after 500 years and the maximum tolerable source term concentration is a factor of about 36, which is build up from a dilution factor (4.7) and a remaining factor due to retardation and partial breakthrough (~8).

From Figure 4C it is clear that the emission criterion is very sensitive to the chosen time frame. A further illustration of the sensitivity of the fitted emission criteria for the chosen time frame (100, 500 or 1000 years) is given in the next paragraph. It should be stressed that in case C the concentrations of contaminants at the POC will continue to increase after 500 years to values well above the groundwater criterion. Case C is commonly found for "medium mobile" contaminants, among which heavy metals such as copper and chromium.

Case D is an example of a contaminant that that does not reach the POC at all within the calculation period. Case D is sometimes found for substances that are predicted to form a precipitate in the soil below the landfill, such as the formation of $Pb(OH)_2(s)$ at high pH; also arsenic is in some cases sorbed so strongly to the soil (as arsenate), that it does not reach the POC. Under those conditions, the contaminants are predicted to accumulate below the landfill and will not be transported towards the POC. This behaviour implies that it is not possible to determine a site-specific emission criterion based on a groundwater criterion at the POC, because the concentration in the leachate would become infinitely high. In Case D, other environmental criteria must be sought as a basis for determining a site-specific emission criterion. In those cases, it was decided that the emission criterion is established based on limit values of total content in the soil (mg/kg) as was done in a similar way previously in the Dutch Soil Quality decree for construction products and large-scale applications of (excavated) soil and sludge (Verschoor et al., 2008; Verschoor et al., 2006).

Whether a specific contaminant shows behaviour according to case A, B, C or D depends on the intrinsic chemical properties of the contaminant (chemical reactivity) and to site-specific properties, such as the pH and the amounts of reactive surfaces present (organic matter, Fe/Al hydr) oxides and clay content). Therefore a general subdivision of contaminants in each of the above categories is not possible, as they are site-specific. An exception are soluble salts, which are chemically non-reactive and will show in all scenarios a mobility according to case A.

3.2 Sensitivity of site-specific emission criteria to choices and parameters

Model predictions are sensitive to uncertainty in the value of important input parameters such as the representativity of the assumed soil properties from the selected STONE soil profiles compared to the real soil properties below the landfill on-site. Also, the predictions are also to a large extent sensitive to choices and boundary conditions with respect to the model scenario (time frame, groundwater criterion, downward water flow, etc.).

Although the sensitivity of emission criteria for specific parameters is highly landfill site-specific, there are a number of important general features. Therefore, we will show for one of the landfill sites, for which (draft) emission criteria are calculated, the estimated sensitivity of the emission criteria for a "realistic" variation of a few of the most important input parameters (chemical and physical), and the sensitivity of the emission criteria for policy-related choices and parameters. The results shown in Figure 5 and discussed below are limited to inorganic substances.

Chemical properties source term and soil

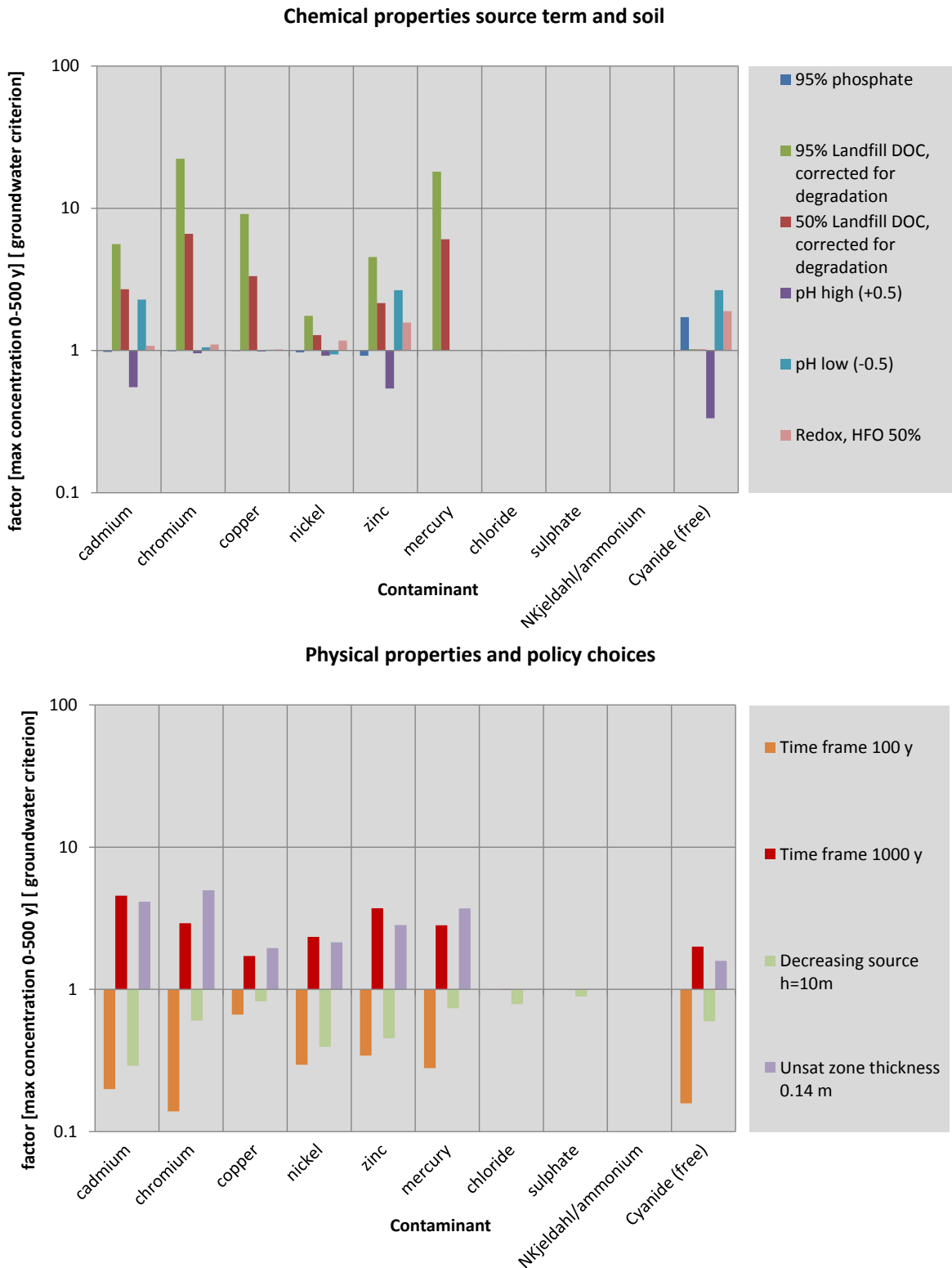**Physical properties and policy choices**

Figure 5. Example of the calculated sensitivity of site-specific emission criteria for different choices and values of important input parameters. Sensitivity is expressed as a factor between the maximum calculated concentration at the POC within 0-500 years and the groundwater criterion at the POC. A factor of 1 = no change (emission criterion remains unchanged); factor > 1: concentration above groundwater criterion, i.e. would lead to more stringent emission criterion; factor < 1: concentration below groundwater criterion, i.e. choice would lead to more tolerant emission criterion.

3.1.2 Sensitivity of emission criteria for chemical choices and parameters

The upper part of Figure 5 shows the sensitivity to chemical parameters and choices for a number of inorganic substances. The following variations to the "default" scenario were made:

- *Phosphate.* A higher concentration of phosphate was used in the source term (95-percentile value of monitoring data of the respective landfill site, 2.6 mg P/L assumed to be present as phosphate) compared to the default scenario (0.05 mg/l PO4 as background value). Phosphate is a strong sorbate to Fe/Al hydroxides in soils, and competes for sorption sites with other anions such as arsenic, sulphate and cyanide which is also taken into account in the geochemical model approach (see 2.2.2 and (Dijkstra et al., 2009)). In the shown example case, a higher phosphate concentration has only a minor influence on other ions (only cyanide: factor 1.7). Arsenic is not shown in Figure 5, as it is in sorbed too strongly to Fe/Al(hydr)oxides in this soil to reach the POC even under the influence of higher phosphate concentrations. However, the mobility of arsenic, and the influence of phosphate on its mobility depends on many site-specific factors; in one of the two other landfill sites included in this project, a strong mobilizing influence of phosphate was found on arsenic. The sensitivity analysis has led to a more precise estimate of the amount of phosphate that should be taken into account in the source term, and an incentive for landfill owners to measure phosphate in their landfill leachate.

- *Dissolved Organic Carbon (DOC).* In particular metal cations such as copper (Cu^{2+}) bind strongly to DOC present in soil pore water. DOC acts as a "carrier" for facilitated transport of sorbed metals towards the POC. Hence, DOC is an important parameter in the model (see 2.2.2). In the standard scenario, the native concentrations of DOC in the soil pore water are taken as invariant in the model, while in reality the DOC from the landfill leachate may lead to much higher DOC concentrations in the pathway between landfill and POC. To illustrate the effects of increased DOC concentrations, the 95 and 50 (median) percentile concentrations of the landfill leachate are used in the model and corrected for degradation over a treatment period of 10 years (based on landfill data evaluation, the degradation is estimated as a factor of 4 reduction of the DOC concentration; the percentage of DOC that is attributed to humic substances important for metal binding is estimated at 50% (Dijkstra et al., 2009)). Both the 95 and 50-percentile of landfill DOC scenarios lead to much higher concentrations of metals in solution and much earlier breakthrough, which inevitably leads to more stringent emission criteria for most metals (factor 2 to 20, Figure 5). Metals that most strongly bind to organic matter are most sensitive to DOC variations (i.e. Copper and Chromium). The analysis has led to the use of median values of DOC from the specific landfill sites corrected for degradation, instead of soil-native DOC. Also, it is important to determine the fraction of DOC that is attributed to humic substances important for metal binding (Van Zomeren et al., 2004).

- *pH.* The pH is of influence to the degree of binding of all reactive contaminants. Hence, variation in pH in the soil (+ or – half a unit, compared to the original site-specific soil pH values in the soil profile) leads to concentrations that are higher or lower than the groundwater criteria at the POC, in particular cadmium, zinc and cyanide (Figure 5). Metals that are relatively less strongly bound to organic matter, such as Cadmium, Nickel and Zinc, are more sensitive for pH variations than metals that are strongly bound to organic matter such as copper and chromium (Figure 5). Copper and chromium are virtually 100% distributed over dissolved and particulate organic matter, and hence, their mobility is primarily determined by the distribution of dissolved and particulate organic matter in the soil (Dijkstra et al., 2009).

- *Redox.* A mildly reducing environment may lead to the dissolution of Fe-(hydr)oxides present in soils, which act as sorbents for metals and anions. A mildly reducing condition was simulated by a 50% lower amount of iron (hydr)oxide in the soils compared to the "native" amount of iron hydroxides in the soil profile at the landfill site. Reducing the amount of

sorbent surface has a mobilizing effect; in this case zinc and cyanide concentrations exceed the groundwater criterion with about a factor of 2 (Figure 5).

3.1.3 Sensitivity of emission criteria for physical and policy-related choices and parameters

The lower part of Figure 5 shows the sensitivity to a few physical properties and policy-related choices. The following variations to the "default" scenario were made:

- *Time frame.* As suggested by Figure 3 and 4, the time frame is of major influence to the site-specific emission criteria. In this project, the time frame during which concentrations at the POC should remain below the groundwater criterion is 500 years. A shorter time frame of 100 years, as was decided for leaching limit values of construction products in the Dutch Soil Quality Decree (Verschoor et al., 2006), leads to much lower calculated concentrations at the POC, as many retarded contaminants have not yet reached the POC. This would lead to more tolerant emission criteria as a higher source term can be allowed than in the default scenario (up to a factor of 10, see Figure 5). Vice versa, a time frame of 1000 years would lead to more stringent emission criteria (Figure 5). It may be noted that the value of 100 years for construction products and large-scale applications of (excavated) soil and sludge in the Dutch SQD was, among other considerations, based on the lifetime and "retrievability" of construction works; this argument would be questionable for landfills. On the other hand, the assumption of a constant source (or any source term behaviour) for a period of 1000 years for landfills would be scientifically and philosophically debatable. It should also be noted that long time frames such as 1000 years will, in combination with relatively thin thickness of the unsaturated zone (such as in the Netherlands) and a POC located relatively close to the source, in many cases lead to complete breakthrough of contaminants at the POC, which implies that retardation of contaminants in the unsaturated and saturated zone is irrelevant for the value of the emission criteria.
- *Decreasing versus constant source.* In the default scenario, the source term concentration is assumed constant. Here, we have allowed the source term to decrease, based on the height of the landfill (e.g., 10 metres in the example case), density (1550 kg/m^3), infiltration (300 mm/y) and a first-order dependence of concentrations on L/S value based on a large database with percolation test measurements (Verschoor et al., 2008; Verschoor et al., 2006). The effect of a decreasing source is strongest for a number of metals (factor 2-5), but is almost absent for soluble salts. The absence of an effect for soluble salts is explained by their high mobility and the fact that a decreasing source has no influence on the initial concentrations. The maximum reached concentration does not change, only the shape of the concentration as a function of time is influenced (i.e. an "S-shaped" curve for a constant source term and a "skewed Gaussian" curve for a declining source term).
- *Thickness unsaturated zone.* In the default scenario, the thickness of the unsaturated zone is 1 metre, being a generic choice (see 2.2.2). In the sensitivity analysis it is tested what the effect is when a location-specific choice is made, in the example case a thickness of 0.14 metres (very shallow groundwater tables are common in parts of the Netherlands). As may be expected because the amount of soil (and sorbing surfaces) decreases substantially, and the distance to groundwater is reduced, this choice leads to concentrations that exceed the groundwater criteria (Figure 5) and as a consequence to more stringent emission criteria. Other landfill sites in this project have an unsaturated zone that is thicker (up to 3.5 metres) leading to more tolerant emission criteria.
- *Choices in background concentrations and groundwater criteria.* From a policy point of view, different (site-specific) choices on background concentrations in the groundwater and different groundwater criteria can be used at the POC. Of course, different criteria in the groundwater have a direct effect on the calculated emission criterion; these possible choices

are highly site-specific and policy- dependent, and will therefore not be shown and discussed in more detail.

5. CONCLUSIONS

- A novel and generic methodology is developed to calculate site-specific emission criteria for sustainably managed and treated pilot landfill sites, expressed as maximum tolerable concentrations (μg/l) of target contaminants in the landfill leachate. The methodology is based on a "source-path-point of compliance" approach, and takes into account site-specific factors that influence the transport rate of leached contaminants along the "path" between the "source" (the landfill) and the downstream point of compliance, where site-specific groundwater limit values have to be met. Presently, calculations are on-going and final emission criteria for three pilot landfills will be published in the second half of 2013.

- In calculating the site-specific emission criteria, a number of generic assumptions are made (e.g., annual precipitation in mm/y, failure of the liner systems, constant source term concentrations) that must be seen as conceptual choices. Many site-specific choices and assumptions are also made (e.g., site-specific chemical soil properties and physical properties such as hydrological dilution) that result in highly site-specific emission criteria.

- With respect to the chemical and physical factors that have the strongest influence on the site-specific emission criteria, the concentration of reactive Dissolved Organic Carbon (DOC) that is assumed in the source term and transported downwards in the underlying soil, is by far the most important factor throughout the different investigated landfills. Therefore, there is a need to gain more insight in the composition of DOC in landfill leachate in order to better parameterise the models used for this type of assessments.

- With respect to the policy- factors that have the strongest influence on the site-specific emission criteria, the time frame during which soil and groundwater are protected is of crucial importance. Generally, short time frames (e.g., 100 years) lead to much more tolerant emission criteria than long time frames (e.g., 1000 years). The effects are caused by the partial breakthrough of contaminants at the point of compliance, and is contaminant- and site-specific. The important effect of time frame on calculated emission criteria requires careful consideration.

REFERENCES

Brand,E, P F Otte, J P A Lijzen. CSOIL 2000: an exposure model for human risk assessment of soil contamination: a model description. RIVM report 711701054/2007. Dutch Institute for Public health and Environment (RIVM).

Dijkstra, J. J., J. C. L. Meeussen, and R. N. J. Comans, 2004, Leaching of heavy metals from contaminated soils: an experimental and modeling study: Environmental Science and Technology, v. 38, no. 16, p. 4390-4395.

Dijkstra, J. J., J. C. L. Meeussen, and R. N. J. Comans, 2009, Evaluation of a generic multi-surface sorption model for inorganic soil contaminants: Environmental Science and Technology, v. 43, no. 16, p. 6196-6201.

Dijkstra,JJ, A Van Zomeren, B Susset. Technical principles underlying limit values for release of substances for the percolation test TS3: comparison DE and NL. 2013. Presentation held for CEN/TC 351 TG1, April 25, 2013.

Kattenberg,W, T Heimovaara. Policy process of allowing research pilots for sustainable

emission reduction at landfills in The Netherlands. Proceedings Sardinia 2011, Thirteenth International Waste Management and Landfill Symposium S. Margherita di Pula, Cagliari, Italy; 3 - 7 October 2011, CISA Publisher, Italy.

Luning,L, E H M van Zundert, O Coops. Bioreactor, stortplaats voor overwegend organisch afval. Den Bosch, The Netherlands, Dutch sustainable landfill foundation, 2006.

Mathlener,RA, T Heimovaara, H Oonk, L Luning, H A Van der Sloot, A Van Zomeren. Opening the black box, process-based design criteria to eliminate aftercare of landfills. ISBN-10: 90-73573-30-0, 1-78. Den Bosch, The Netherlands, Dutch sustainable landfill foundation, 2006.

Meeussen, J. C. L., 2003, ORCHESTRA: An object-oriented framework for implementing chemical equilibrium models: Environmental Science and Technology, v. 37, p. 1175-1182.

Susset, B., and P. Grathwohl, 2010, Leaching standards for mineral recycling materials – A harmonized regulatory concept for the upcoming German Recycling Decree: Waste Management, v. 31, p. 201-214.

Van Zomeren,A, J C L Meeussen, H Oonk, L Luning, H A Van der Sloot. Evaluation of geochemical and biochemical processes and release from landfills. ISBN-10: 90-73573-34-3, 1-100. 2006. Den Bosch, Dutch sustainable landfill foundation.

Van Zomeren,A, E van der Weij-Zuiver, J J Dijkstra, R N J Comans. A new rapid batch procedure for the determination of humic and fulvic acids and its relevance for geochemical modeling of heavy metal leaching, 2004, Sao Pedro, Sao Paulo, Brasil, International Humic Substances Society (IHSS).

Van Zomeren,A, H A van der Sloot. Equifill, landfill site for predominantly inorganic waste. 1-101. 2006. 's-Hertogenbosch, the Netherlands, Dutch sustainable landfill foundation.

Verschoor,AJ, J P A Lijzen, H H van den Broek, R F M J Cleven, R N J Comans, J J Dijkstra. Revision of the Dutch Building Materials Decree: alternative emission limit values for inorganic components in granular building materials. The 9th International Symposium on Environmental Geotechnology and Global Sustainable Development, June 1-4, 2008, Hong Kong.

Verschoor,AJ, J P A Lijzen, H H van den Broek, R F M J Cleven, R N J Comans, J J Dijkstra, P Vermij. Kritische emissiewaarden voor bouwstoffen; Milieuhygiënische onderbouwing en consequenties voor bouwmaterialen. RIVM rapport 711701043 /2006.