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Beyond the material grave: Life Cycle Impact Assessment of leaching from secondary materials in road and earth constructions

Oliver Schwab [a,b], Peter Bayer [c,*], Ronnie Juraske [a], Francesca Verones [a,d], Stefanie Hellweg [a]

[a] Swiss Federal Institute of Technology Zurich, Institute of Environmental Engineering, John-von-Neumann-Weg 9, 8093 Zurich, Switzerland
[b] Karlsruhe Institute of Technology, Institute for Geography and Geoecology, Adenauerring 20, 76131 Karlsruhe, Germany
[c] Swiss Federal Institute of Technology Zurich, Geological Institute, Sonneggstrasse 5, 8092 Zurich, Switzerland
[d] Department of Environmental Science, Institute for Water and Wetland Research, Radboud University Nijmegen, P.O. Box 9010, 6500 GL Nijmegen, The Netherlands

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ABSTRACT

In industrialized countries, large amounts of mineral wastes are produced. They are re-used in various ways, particularly in road and earth constructions, substituting primary resources such as gravel. However, they may also contain pollutants, such as heavy metals, which may be leached to the groundwater. The toxic impacts of these emissions are so far often neglected within Life Cycle Assessments (LCA) of products or waste treatment services and thus, potentially large environmental impacts are currently missed. This study aims at closing this gap by assessing the ecotoxic impacts of heavy metal leaching from industrial mineral wastes in road and earth constructions. The flows of metals such as Sb, As, Pb, Cd, Cr, Cu, Mo, Ni, V and Zn originating from three typical constructions to the environment are quantified, their fate in the environment is assessed and potential ecotoxic effects evaluated. For our reference country, Germany, the industrial wastes that are applied as Granular Secondary Construction Material (GSCM) carry more than 45,000 t of diverse heavy metals per year. Depending on the material quality and construction type applied, up to 150 t of heavy metals may leach to the environment within the first 100 years after construction. Heavy metal retardation in subsoil can potentially reduce the fate to groundwater by up to 100%. One major challenge of integrating leaching from constructions into macro-scale LCA frameworks is the high variability in micro-scale technical and geographical factors, such as material qualities, construction types and soil types. In our work, we consider a broad range of parameter values in the modeling of leaching and fate. This allows distinguishing between the impacts of various road constructions, as well as sites with different soil properties. The findings of this study promote the quantitative consideration of environmental impacts of long-term leaching in Life Cycle Assessment, complementing site-specific risk assessment, for the design of waste management strategies, particularly in the construction sector.

1. Introduction

Mineral wastes of industrial processes significantly contribute to anthropogenic material flows. In Germany, for example, the metal industry, the energy sector and waste incineration processes produce about 40 million tons of industrial mineral wastes annually (Förstner, 2012). The largest volumes are slags (steel slag, blast furnace slag, and others), ashes (such as municipal solid waste incineration ash and brown coal fly ash) and sands (e.g., foundry sand). As displayed in Fig. 1, only a relatively small fraction, around 6%, of these enormous amounts ends up in landfills. To save natural resources and for economic reasons, the remaining amounts are mainly re-used in the cement and ceramic industry, the construction industry, or in fertilizers (Dehoust et al., 2008). The German construction sector, with about 36%, is the biggest sink for industrial mineral wastes. Most often, industrial mineral wastes are applied as Granular Secondary Construction Materials (GSCM). GSCM replace or complement natural gravel as filling material, for example in road dams or other road and earth constructions.

Application of waste and recycling materials in construction has been a long discussed topic in the research community (see for example the Elsevier WASCON conference series), and in some nations legal regulations have been put into practice. The Netherlands was the first country to regulate the use of waste material in construction (Eikelboom et al., 2001). Also in other European

* Corresponding author. Address: Department of Earth Sciences, Engineering Geology, Swiss Federal Institute of Technology Zurich, Switzerland. Tel.: +41 44 633 68 29.
E-mail address: bayer@erdw.ethz.ch (P. Bayer).

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countries, such as Finland (Mroueh and Wahlström, 2002), strict regulations, based on material quality conditions, led to a restricted processing of critical material in constructions. An upcoming recycling directive in Germany (BMU, 2012) introduces even more conservative limits. However, in the USA, which has less strict regulations, as much as 58% of mineral industrial wastes are reused in road and earth construction (Carpenter et al., 2007). There is also rising interest in the reuse of industrial mineral wastes in emerging economies (Chin-Ming et al., 2006; Pappua et al., 2007).

The environmental benefit from recycling can come along with new environmental impacts. The assimilated recycled materials carry varying loads of pollutants, such as heavy metals, metalloids, polycyclic aromatic hydrocarbons (PAHs), and salts. Road and earth constructions are in many cases at least partially environmentally open systems (Beyer et al., 2009). Thus, materials, such as GSCM, are percolated by rainwater and surface runoff water. Due to potential leaching and release of pollutants from the waste material, this may impose risks to the environment. Over time, leaching is likely to shift pollutants from waste materials to the underlying soil and groundwater. There are ongoing research efforts to survey and evaluate the behavior of these pollutants and to derive new concepts which aim at an advanced scientific assessment of the use of mineral recycling materials in construction, such as the German joint research project "Sickerwasserprognose" (Percolation water prognosis, "Siwap") (Susset and Grathwohl, 2011). Objectives of Siwap are the characterization of potential sources of pollutants, the description of release processes, and understanding the transport processes of pollutants through soil to groundwater.

Even though comprehensive studies and models of different sophistication exist for estimating leaching and transport, so far no approach has been integrated into Life Cycle Assessment (LCA) to estimate the environmental impacts of GSCM, emphasizing fate assessment and specifically the role of soil. However, the inclusion of a pragmatic approach, modeling these as a function of construction type, material quality and spatial conditions, can be crucial for policy makers and industrial actors to quantify the environmental impacts related to GSCM. Also, there is still a lack of studies that address the overall environmental impacts of the reuse of mineral industrial wastes. One reason can be that wastes often occur in a societal sector different from the residual disposition (Roth and Eklund, 2003), which can obscure substance pathways and thus complicate choices on system boundaries in environmental assessments. Current environmental assessments often do not go beyond the fact that these wastes are recycled, stockpiled, or landfilled (Bayer and Finkel, 2006; Norgate et al., 2007; Fthenakis et al., 2009; Tongpool et al., 2010), rarely include the further fate of pollutants in the natural environment, and do not include comprehensive modeling of pollutant fate but focus on solute and water movement through materials (Kosson et al., 2002; Apul et al., 2005). However, this seems relevant as considerable emissions and toxic impacts may occur in the long term (Mroueh et al., 2001), and the fate in the natural environment is decisive for the evaluation of environmental impacts.

One standard methodology for comprehensive environmental assessment of products, processes or services is Life Cycle Assessment (LCA). LCA is a standardized systems-thinking method (ISO 14040 and 14044) to assess potential environmental impacts from "cradle to grave". It accounts for environmentally relevant input and output quantities, from extraction of raw materials, production, and use phase/consumption to end-of-life disposal. It is used to translate these quantities into environmental impacts by applying Life Cycle Impact Assessment (LCIA) methods. Different approaches and models have been suggested for quantifying the impacts of primary and secondary construction materials, such as the approach by Chowdhury et al. (2010) or EASEWASTE (Bhander et al., 2010). Further, the impacts of leaching were assessed with the LCIA methods CML (Huijbregts et al., 2000) and EDIP97 (Wenzel et al., 1997) in the model ROAD-RES (Birgisdóttir, 2005; Birgisdóttir et al., 2007).

Although end-of-life leaching is regarded as a life cycle stage of importance for comprehensive environmental assessment, previous findings are not always satisfying. First, hydrological conditions and leaching behavior of waste materials in field conditions were often barely known and addressed (Olsson et al., 2006), and fate assessment was based on default transfer coefficients rather than on site- and substance-specific coefficients. Also, LCA was found to be limited when it comes to long-term pollutant release and impact assessment (Hauschild et al., 2008a). Hence, it is not surprising that leaching from waste disposal options other than landfilling (Hellweg et al., 2005a; Hauschild et al., 2008b) is often considered to be beyond the system boundaries. However, if LCA

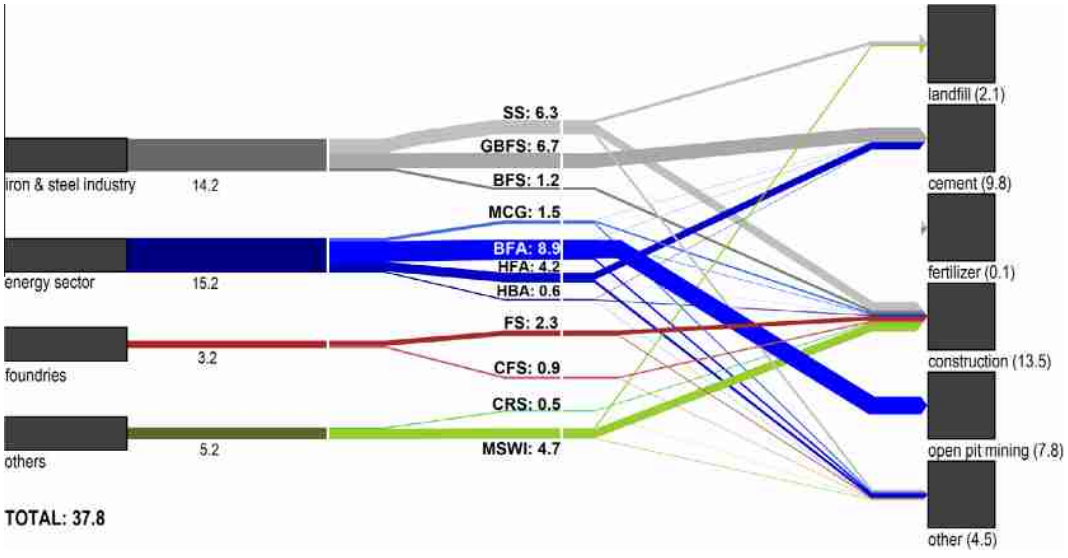

Fig. 1. Annual mass flows of industrial mineral wastes in Germany (in Mio. tons). SS (steel slag), GBFS (granular blast furnace slag), BFS (blast furnace slag), CFS (cupola furnace slag), MCG (melting chamber granulate), BFA (brown coal fly ash), HFA (hard coal fly ash), HBA (hard coal bottom ash), FS (foundry sand), CRS (copper residual slag), MSWI (municipal solid waste incineration ash) according to flow rates for 2004 by Dehoust et al. (2008) and annual quantities reported by Förstner (2012).

intends to be complete and capture all environmental impacts "from cradle to grave", there is a need for quantifying and assessing leaching from mineral waste materials, including the economically attractive reuse options in road and earth construction.

For protection of groundwater, a special focus on the role of subsoils is essential in the context of leaching (Hellweg et al., 2005b). Since the release of heavy metals is desorption-limited and hence not a function of total solid pollutant content in the waste material, recent developments have focused more on approaches quantifying the released concentrations of heavy metals in the seepage water (Dijkstra et al., 2008; Susset and Grathwohl, 2011). The transport of pollutants through soil and the evolution of pollutant concentrations at the groundwater surface are of rising interest and better understood (Susset and Leuchs, 2008). Current integrated approaches for environmental assessment are based on experience from leaching tests, which combine mathematical release and fate modeling, and include technical and geographical factors (Beyer et al., 2009; Susset and Grathwohl, 2011). Long term heavy metal release is regarded as critical in material life cycles (Pettersen and Hertwich, 2008), and different approaches for assessing the impact of metals on the environment have been developed (Pizzol et al., 2011). So far, there is no model that integrates the recent findings on leaching (Beyer et al., 2009; Susset and Grathwohl, 2011) and fate assessment (Utermann et al., 2005) in a LCA approach.

With this background, we present a LCA approach for quantifying leaching from secondary construction materials and assessing the fate and effects based on the scenarios (soil properties, seepage water rates, constructions) of the German site-specific risk assessment concept for the upcoming Recycling Degree (Susset et al., 2011). We then apply this model to the total of German GSCM. To do so, we examine material flows of major industrial mineral wastes to the construction sector, and derive substance flows of selected heavy metals and metalloids into the environment. Special focus is set on the influence of soil properties on leaching to the groundwater and the environmental impact. Regarding the case of leaching from constructions, the application of waste materials is not already the actual "grave" of critical substances as those are likely to be released to the environment and thus potentially beyond system boundaries. We regard this end-of life system – "leaching from graveyard to grave" (Fig. 2) – and thus lay the foundation for subsequent end-of life system expansions in LCA of construction materials.

2. Materials and methods

2.1. System description and framework

Figs. 2 and 3 show the system and the procedure suggested for the assessment of leaching from GSCM in a LCA framework (A list of abbreviations and parameters with units is provided in supplementary data S1). The four steps of LCA are (1) goal and scope definition, (2) Life Cycle Inventory Analysis (LCI), in which inputs (resource uses) and outputs (emissions) are quantified, (3) Life Cycle Impact Assessment, in which the resource and emissions from the LCI are assigned to environmental impact categories, like "resource use" or "ecotoxicity", and assessed, typically by applying Life Cycle Impact Assessment (LCIA) characterization factors (*CF*s), and (4) interpretation. A CF is a pollutant and environmental medium specific cause-effect factor (Pizzol et al., 2011). As pollutants can be distributed over several environmental compartments like air, soil or freshwater, it can be necessary to evaluate the environmental fate of emissions prior to effect assessment. Fate depends on the pre-defined system boundary of a study. In the context of LCA, a system boundary defines the scope of the studied system and includes all processes and material or substance sources and sinks of interest.

Our proposed procedure (Fig. 3) integrates technical construction-specific factors, geographical factors, as well as material and substance specific factors (Fig. 2). As reference, a functional unit (FU) is defined, which is 1 ton of GSCM. GSCM covers a specific area per construction type (m^2/t). This area is percolated by rainwater, and pollutants are released over time (mass per functional unit).

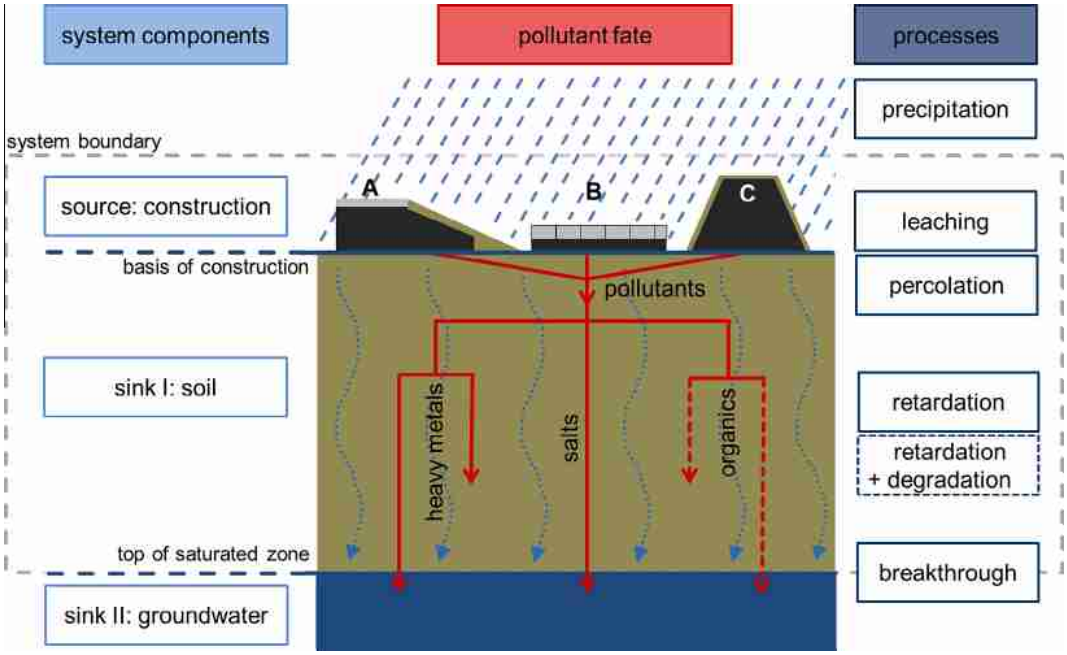

Fig. 2. "Graveyard to grave" system components, pollutant fate paths and processes. Three construction scenarios: noise protection dam (A), parking area (B), road dam (C). Construction compartments that can contain GSCM are indicated in black, and modeled by the horizontal and vertical size layer thickness and covered area per mass unit GSCM.

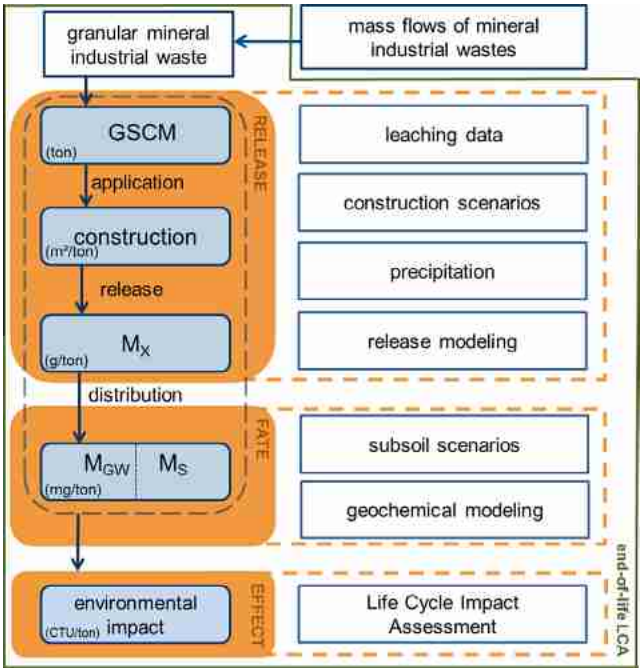

Fig. 3. Framework of environmental assessment of leaching from Granular Secondary Construction Materials (GSCM) with release, fate and effect modeling. Total leached mass (M_X) disperses to compartments soil (M_S) and groundwater (M_{GW}).

As construction materials leach different heavy metals at a time, release modeling is used to feed an inventory. The pollutants distribute in soil and groundwater, where they can cause environmental impacts. Ecotoxicological impacts are calculated for the quantification of GSCM impacts on the environment. They are obtained by multiplication of pollutant mass and Characterization Factor (*CF*) according to the USEtox method (Rosenbaum et al., 2008) and are expressed in comparative toxicity units per mass unit (CTU/t). The comparative toxicity unit of USEtox is "potentially affected fraction of species" (PAF, in m^3 day/kg).

The here presented framework is based on approaches of the project Siwap for release and fate modeling (Susset et al., 2011), and complemented by Life Cycle Impact Assessment. Please note that Siwap and the planned German Recycling Directive (BMU, 2012) propose concentration-based, site- and case-specific micro-scale risk assessments. The here proposed macro-scale LCA approach is mass-based and cannot be as specific as the risk assessment of Siwap. Though relying on similar assumptions, the motivations of these two concepts are inherently different. BMU (2012) aims to exclude specific materials with severe potential environmental loads from sensitive construction types by assessing the risk for a particular situation. In contrast, the presented LCA framework is intended to include site-dependent leaching assessment with a particular focus on environmental fate in future construction LCAs. While the intention of BMU (2012) is to exclude potentially critical cases, this study offers a generalized view as common in LCA. The results of the LCA based approach thus do not anticipate the conditions when the planned directive would become operative. Average material flows and contaminant fate are calculated independent of risk. Major links between the two concepts are fundamental transport model assumptions and input parameter specifications. However, it has to be emphasized that these are more specific in BMU (2012), allowing for making more case-specific assessments. Thus, total environmental loads are expected to be reduced once BMU (2012) is legally binding.

Although conceptually the proposed framework accounts for the physico-chemical processes relevant for fate assessment, many model parameters can only be approximated. Accounting for highly case and site specific environmental impacts in a macro-scale LCA thus is hardly feasible, and instead a simplified approach is suggested. Consistent with previous work and as proposed in Siwap, we favor averaged parameter values for subsequent calculations. Still these values may be variable and induce uncertainties in release, fate and impact modeling. The factors most sensitive to parameter uncertainties will be highlighted. For example, the sensitivity of transfer coefficients to changes in soil layer and leachate concentrations will be discussed. Underlying concentration ranges from laboratory-scale percolation tests are provided as supplementary data (Table S2).

2.2. Materials and construction types

Mineral industrial waste materials include blast furnace slag (BFS), granulated blast furnace slag (GBFS), steel slag (SS), municipal solid waste incineration bottom ash (MSWI), copper refinery slag (CRS), cupola furnace slag (CFS), foundry sand (FS), melt chamber granulate (MCG), hard coal fly ash (HFA), hard coal bottom ash (HBA), and brown coal fly ash (BFA) (Susset et al., 2011). Before applied in constructions, these materials may be processed. This involves granulation, classification, washing and pH adjustment (Dehoust et al., 2008). Material quality can be described by pollutant solid concentration (mg/kg) and leachate concentration (μg/l).

GSCM are applied in several road and earth constructions that can be sealed to a certain degree. Open and partly-open (and thus percolated) constructions can be described by three standard scenarios (Susset and Leuchs, 2008): road dam (partly open), parking area (open) and noise protection dam (partly open). These construction scenarios (Fig. 2) differ in geometry, ground cover and degree of sealing, thickness of GSCM layer, porosity, and density. Considered percolation rates are 486 mm/a for road dams, for parking areas 313 mm/a (open cover) or 583 mm/a (pavement cover), and for noise protection dams 313 mm/a (Susset et al., 2011). A number of varieties in construction of road dams, parking areas and noise protection dams have been standardized for Germany (Susset et al., 2011, see S3). A crucial parameter in this context is the area *A* (m^2/ton) that is covered by one mass unit of GSCM, which is a function of GSCM layer thickness and material density. The larger the covered and open area, the more rainwater percolates through one mass unit of material. This is in general larger for parking areas (open) than for road dams (where water percolates through the unsealed, open embankments), and least for noise protection dams (partly-open). Construction varieties are listed in more detail in supplementary data S3, as defined in Susset and Leuchs (2008) according to German construction practice. In this study, these varieties are grouped into three representative construction scenarios A, B and C (Fig. 2), and the results of the varieties are averaged within each of the three construction scenarios.

2.3. Release modeling

Precipitation and construction type are site-dependent and determine the percolation rate *PR*, which governs the liquid–solid ratio, *LS*, in the GSCM layer. Material-specific eluate concentrations (laboratory scale) and leachate concentrations (in situ) are described by *LS*. It is suggested that cumulative concentrations measured at column percolation of *LS* = 2 l/kg correspond to time scales of 8–40 years and are most appropriate for conservative medium- to long-term risk assessment (Grathwohl and Susset, 2009; Delay et al., 2007). Accordingly, these are also applied in the presented study. It has to be considered that these lab-based

model specifications may be of limited transferability to the field and have to be updated based on future experience after long-term monitoring over decades.

The contact time between percolation water and GSCM matrix is limited, and thus leaching shows substance-specific characteristics. Substances can be grouped by two basic release schemes, that are (1) availability-controlled, which applies for salts, and (2) sorption-controlled, which applies for heavy metals and organics (Hyks et al., 2008; Susset and Leuchs, 2008). In the following, we focus on heavy metals only, and include further selected metalloids that are assumed to follow the same leaching scheme (Sb, As, Pb, Cd, Cr, Cu, Mo, Ni, V and Zn, in the following "heavy metals") (Susset and Leuchs, 2008). There may be further relevant substances such as Se and Ba, but currently there is not sufficient information available for including those equivalently in the analysis. For availability controlled substances, source concentration $C_S(t)$ is assumed to decline over time. $C_S(t)$ is assumed to be constant for sorption controlled substances within a time frame of 200 years (Susset and Grathwohl, 2011). The simplification of constant source concentration is part of a regulatory concept (Susset and Grathwohl, 2011), and can be questioned for example regarding potential pH changes in the construction material under field conditions, and subsequent changes in pollutant release behavior. However, with reference to the regulatory concept, $C_S(t)$ is regarded to be constant for the here considered heavy metals within a time frame of 100 years. The mean values and, if too low to be measured, limits of quantification, as applied in previous studies (Juraske et al., 2009), are considered in Eq. (1) for $C_S(t)$ (see Table 1). Information on concentration ranges is provided in supplementary data S2.

The inventory lists the total mass M_X of emissions per functional unit (FU). Leached total mass is obtained by summing up the results for t_{max} time steps (for a parameter list and units, see S1):

$$M_x = \sum_{t=t_1}^{tmax} (C_S(t)) \cdot PR \cdot A \cdot D \quad (1)$$

M_X (g) is a product of source concentration ($C_S(t)$) (μg/l), percolation rate (PR) (l/(m^2 a)) according to Susset et al. (2011), and horizontal area (A) (m^2) related to the functional unit (Susset and Leuchs, 2008). The construction specific geometry and the sealed surface share are considered by a further factor, the dilution coefficient (D), as modeled by Beyer et al. (2007) and applied by Susset et al., 2011 (see S3). The assumption of constant cumulative concentration medians (LS = 2 l/kg) is very conservative for two reasons. That is, (1) source concentrations for the assessed heavy metals are considered constant within a time period of 100 years, also for construction types with thin GSCM layers, i.e. parking areas. And (2), materials are not divided in subcategories with lower leaching (as this is done in BMU, 2012).

2.4. Fate assessment

The fate is first assessed separately to the effect in this study. This allows for considering site-specific soil geography and substance-specific fate characteristics in the model, and thus goes beyond existing LCIA practice. M_X is divided into the share that is absorbed in the subsoil (M_S) and the share that reaches the groundwater (M_{GW}). During construction, the topsoil layer is usually removed. Thus, water percolates only through the remaining soil or subsoil. In order to account for different conditions, two subsoil scenarios (Fig. 3) with specific adsorption behavior and distinct parameter specifications are considered: (1) sand and (2) clay (Table 2). Crucial parameters determining the fate of metals in soils are pH value, clay content (f_{clay}), organic matter content (f_{oc}), bulk soil density (ρ_s) and porosity (θ_s) (Blume et al., 2010). Subsoil parameters differ from topsoil layers, especially in f_{oc}, which is commonly lower in the subsoil.

Sorption intensity is reflected by the substance-specific retardation capacity of a soil, which depends on physical and chemical soil parameters, thickness of soil, leachate and background concentration, and percolation rate. The effect of pH is considered in the calculated K_d (Eqs. (2) and (3)) and by comparing two different soil scenarios. Sorption and desorption processes in soil can be described by Freundlich isotherms. These are approximated by pedo-transfer functions with pollutant and soil specific coefficients K^*, a, b and c determined by linear regression of laboratory soil analyses (Utermann et al., 2005; Gäbler et al., 2009, see supplementary data S4). The Freundlich coefficient K_{fr} is dependent on soil properties:

$$\log K_{fr} = \log K^* + a \cdot \mathrm{pH} + b \cdot \log f_{clay} + c \cdot \log f_{oc} \quad (2)$$

With respect to the background concentration in the soil (C_b) (μg/l), a concentration dependent linear distribution coefficient K_d (ml/g) is derived, where C_{CB} (μg/l) ($C_S(t)\cdot D$, see Eq. (1)) is the percolate concentration at the basis of a GSCM construction and n the Freundlich exponent (Utermann et al., 2005; Gäbler et al., 2009):

$$K_d = \frac{1 \cdot K_{fr}(C_{CB}^{n+1} - C_b^{n+1})}{(C_{CB}^2 - C_b^2)(n+1)} \quad (3)$$

Table 2
Two default soil scenarios that represent selected German subsoil properties. Parameters pH, clay content (f_{clay}), organic matter content (f_{oc}), bulk soil density (ρ_s) and porosity (θ_s) (according to Beyer et al., 2009).

Soil scenario	Soil type	pH –	f_{clay} %	f_{oc} %	ρ_s g/cm^3	θ_s –
I	Sand	4.8	2.46	0.1	1.42	0.17
II	Clay/silt	6.6	12	0.1	1.45	0.26

Table 1
Medians of measured cumulative eluate concentration (μg/l) at liquid-to-solid ratio LS = 2 from column percolation tests (Susset et al., 2011). For values below limit of quantification (LoQ), the LoQs according to Lange and Knödel (2003) are taken as reference values (in italics). For min–max concentration ranges, see *supplementary* data S2.

Material/pollutant	Sb	As	Pb	Cd	Cr (tot)	Cu	Mo	Ni	V	Zn
Steel slag (SS)	*0.01*	*0.1*	*0.01*	*0.01*	18.0	*0.1*	74.0	*0.1*	166.0	*0.1*
Gran. blast furnace slag (GBFS)	*0.01*	*0.1*	*0.01*	*0.01*	0.05	*0.1*	0.01	*0.1*	6.0	*0.1*
Blast furnace slag (BFS)	*0.01*	*0.1*	*0.01*	*0.01*	2.0	*0.1*	4.0	*0.1*	5.0	*0.1*
Melting chamber gran. (MCG)	*0.01*	*0.1*	*0.01*	*0.01*	*0.05*	*0.1*	*0.01*	*0.1*	*0.05*	*0.1*
Brown coal fly ash (BFA)	*0.01*	*0.1*	*0.01*	*0.01*	*0.05*	*0.1*	188.0	*0.1*	*0.05*	*0.1*
Hard coal fly ash (HFA)	7.0	5.0	3.0	*0.01*	537.0	0.1	2000.0	*0.1*	14.0	*0.1*
Hard coal bottom ash (HBA)	3.0	5.0	*0.01*	*0.01*	22.0	5.0	79.0	*0.1*	76.0	*0.1*
Foundry sand (FS)	*0.01*	12.0	2.0	*0.01*	8.0	6.0	16.0	5.0	17.0	9.0
Cupola furnace slag (CFS)	*0.01*	*0.1*	11.0	*0.01*	13.0	9.0	*0.1*	*0.1*	18.0	8.0
Copper residual slag (CRS)	*0.01*	20.0	*0.01*	*0.01*	*0.05*	6.0	69.0	*0.01*	*0.05*	*0.1*
Mun. solid waste in. ash (MSWI)	37.0	*0.1*	*0.01*	*0.01*	53.0	191.0	132.0	*0.1*	19.0	*0.1*

The retardation coefficient R is a function of this distribution coefficient, bulk soil density ρ_s (g/cm^3), and soil porosity θ_s:

$$R = 1 + K_d \frac{p_s}{\theta_s} \quad (4)$$

After passage of a subsoil with the given thickness L(m), percolates reach the next compartment, groundwater. This is evaluated at the top of the saturated zone (Fig. 2), neglecting the potential existence of a capillary fringe. The total mass leached to groundwater, M_{GW} (g), is calculated for $t = t_1,\ldots,t_{max}$ (Δt = 1 year), based on the analytical solution of the Advection–Dispersion-Equation as applied by Grathwohl and Susset (2009):

$$M_{GW} = A \cdot PR \cdot \Delta t \cdot \frac{C_S}{2} \sum_{t=t_1}^{tmax} \left(erfc \frac{L - \frac{vt}{R}}{\sqrt{\frac{4\alpha vt}{R}}} + e^{\frac{L}{\alpha}} erfc \frac{L + \frac{vt}{R}}{\sqrt{\frac{4\alpha vt}{R}}} \right) \quad (5)$$

where $C_S(t)$ (µg/l) is the source concentration, PR (l/(m^2 a)) is percolation rate, v (m/s or m/a) is percolation velocity, L (m) is the soil depth to groundwater surface, and α (m) is longitudinal dispersivity. Subsequently, one can calculate transfer coefficients (TC), which represent the share of M_X (g) that reaches soil (TC_S) and groundwater (TC_{GW}):

$$M_S = M_X \cdot TC_S \quad (6)$$

$$M_{GW} = M_X \cdot TC_{GW} \quad (7)$$

$$M_X = M_{GW} + M_S \quad (8)$$

$$TC_{GW} + TC_S = 1 \quad (9)$$

2.5. Effect assessment

Effects of heavy metals are modeled as ecotoxicological effects on freshwater organisms. This is done using the LCIA method USEtox (Rosenbaum et al., 2008). Impacts per emitted masses of heavy metals are quantified in the category "Freshwater Ecotoxicity" with factors from the USEtox inorganic database 1.01 (Rosenbaum et al., 2008). USEtox is recognized as a consensus model for the actual state of the art of impact quantification of organic and inorganic chemicals. Since USEtox does not provide specific characterization factors for the groundwater compartment, the ecotoxicological effect on groundwater (ET_{GW}) was approximated with characterization factors CF_{FW} of the USEtox path "emission to freshwater" (Table 3) ($ET_{GW} = M_{GW} \cdot CF_{FW}$, where M_{GW} is the mass leached to the groundwater in the first 100 years after the construction). Freshwater factors are applied as little is known about ecotoxicological effects on groundwater organisms.

Table 3
Characterization factor Freshwater Ecotoxicity – emission to freshwater (CF_{FW}) for different heavy metals according to USEtox (USEtox inorganic database 1.01, Rosenbaum et al., 2008). CF_{FW} is applied to model ET_{GW}, $ET_{SOIL,p}$ and $ET_{GSCM,p}$.

Category Path Pollutant/CF	Freshwater Ecotoxicity Emission to freshwater CF_{FW} (PAF per m^3 day/kg)
Sb(V)	$1.9 \cdot 10^5$
As (III)	$1.5 \cdot 10^4$
Pb(II)	$3.7 \cdot 10^2$
Cd(II)	$9.7 \cdot 10^3$
Cr(VI)	$1.0 \cdot 10^5$
Cu(II)	$5.5 \cdot 10^4$
Mo(VI)	$2.9 \cdot 10^2$
Ni(II)	$1.5 \cdot 10^4$
V(V)	$1.1 \cdot 10^5$
Zn (II)	$3.9 \cdot 10^4$

Emissions to soil are regarded as "stored ecotoxicity" (Hansen, 2004; Hauschild et al., 2008b) in the sense that these emissions are temporarily stored in soil but can have an impact on groundwater in the future. Note that the USEtox characterization factors for emission to soil are not applicable here. This is because USEtox does not apply specific soil effect factors for emissions to soil, but fate assessment from soil to freshwater and freshwater effect factors. Application of USEtox CFs for emission to soil in this study would result in a double fate assessment with regard to leaching and it would also include unlikely pathways like surface runoff. Thus, the more specific fate model applied in this study is combined with freshwater characterization factors.

Since plant uptake can be neglected in this paper as emissions reach soil layers underneath constructions, we assess the emissions to soil as potential ecotoxicological impact on freshwater ($ET_{SOIL,p}$). As both the emissions to groundwater and emissions to soil are evaluated in terms of their potential impacts on the aquatic environment, they may be added up according to subsequent Eq. (10). ET_{ENV} hereby quantifies the ecotoxicity of heavy metals that are released from the industrial waste material to the environment. Note that fate assessment (Section 2.4) is not needed for the calculation of ET_{ENV} (PAF m^3 day), but for the distinction between ET_{GW} and $ET_{SOIL,p}$.

$$ET_{ENV} = M_S \cdot CF_{FW} + M_{GW} \cdot CF_{FW} \quad (10)$$

where M_S and M_{GW} represent the masses stored in soil and transported to groundwater and CF_{FW} (PAF m^3 day/kg) the freshwater Characterization Factors (Table 3).

An additional "stored toxicity" is the fraction of substances in the construction materials that are not leached during the first 100 years. To quantify a maximum impact scenario, $ET_{GSCM,p}$ was quantified as the potential ecotoxicological impact when the total heavy metals in GSCM ($t = 0$) is released to freshwater.

For the estimation of total annual environmental loads in Germany, a hypothetical application scenario is developed. It defines application rates AR (Table 4) of GSCM per construction, as approximated from mass flow data (Dehoust et al., 2008) and suggestions of material-specific maximum allowed flows of GSCM to construction types according to the recent German regulation draft (BMU, 2012). The value of AR represents which share of a material is applied in which construction scenario. This influences the magnitude of emitted heavy metals as constructions are percolated differently, depending on the scenario. For a conservative assessment, it is assumed that GSCM that are legally allowed for different construction scenarios (BMU, 2012) are applied entirely in the most percolated one. For this reason, the value of AR of the least percolated scenario, noise protection dam, is assumed zero in this scenario. As high heavy metal concentrations of leachate from HFA were found to break through to groundwater, HFA is recommended to be applied in closed constructions only (Susset et al., 2011) and is thus not considered relevant in this assessment of open and partly-open constructions (see Table 4). Thus, our hypothetical application scenario considers recommendations to reduce the leaching environmental pressure. Note that this hypothetical application scenario does not, as recommended by BMU (2012), exclude critical cases (i.e. materials with source concentrations that impose a particularly high risk to the environment) from the assessment. As the directive proposed by BMU (2012) has not come into force yet, such cases are considered possible. In combination with the conservative assumptions for source concentration $C_S(t)$, this will yield higher mass flows than it is to be expected once the regulatory concept of BMU (2012) is put into practice.

Table 4
Assumed GSCM application rates (–) in road dam (A), parking area (B), noise protection dam (C) and closed constructions in the hypothetical application scenario (based on Dehoust et al., 2008 and BMU, 2012).

Construction scenario	Material										
	BFS	GBFS	SS	CRS	CFS	FS	MCG	HBA	HFA	BFA	MSWI
Road dam	0.4	0.3	0.3	0.3	0.4	0.3	0.3	0.5	0.0	0.4	0.4
Parking area	0.1	0.3	0.3	0.3	0.0	0.3	0.3	0.0	0.0	0.0	0.0
Noise protection dam	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Closed	0.5	0.3	0.3	0.3	0.6	0.3	0.3	0.5	1.0	0.6	0.6

3. Results and discussion

3.1. Substance flow analysis

A primary question to judge the relevance of heavy metals in GSCM is the amount recycled in constructions. Table 5 lists the total annual masses of heavy metals in German mineral industrial wastes. Out of the nearly 70,000 t of heavy metals in industrial mineral wastes per year, around 68% end up in constructions. Highest fractions (>75%) and masses are computed for the metals Cu and Zn, whereas, for example, relatively small fractions are given for As, V and Mo. By representing around two thirds of the total mass, Cu and Zn dominate the heavy metals in the construction material. This is due to their high contents especially in copper residual slag (CRS) (Table S6).

In the three scenarios, the share of heavy metals that leaches within 100 years is highest for parking areas (scenario B), followed by road dams (A) and noise protection dams (C) (Table 6). The lifetime of road and earth constructions is assumed to be 100 years, as it was in a previous life cycle approach (Birgisdóttir et al., 2007). One mass unit of GSCM covers a larger area in scenario B than in A and C. Thus, B has higher percolation rates and consequently higher leaching rates. For substances, the highest leached shares over time are found for Mo, Cr, As and Sb (Table 6). This reflects that Mo and Sb are anionic, mobile metals, As is a highly mobile metalloid, and Cr is highly mobile as chromate (see Matschullat et al., 1997). In the worst case, nearly all (93%) mass of Mo is leached from the parking area already within the given period of 100 years. Total leached masses after 100 years were calculated for the conservative hypothetical application scenario and are listed in Table 7. Quantities range between small mass values below 50 kg (for Cd), and 79 t (for V). In total, more than 150 t of heavy metals leach from GSCM over the first 100 years after construction in the hypothetical application scenario. Please note that we regard millions of tons of GSCM, which are distributed over considerably large areas.

Table 6
Average leached pollutant share per construction scenario after 100 years. Incorporating GSCM is accounted for in three scenarios: noise protection dams (A), parking areas (B) and road dams (C). More detailed data is provided in supplementary data S5.

	Sb	As	Pb	Cd	Cr	Cu	Mo	Ni	V	Zn
Leached share (%)										
A	0.12	0.16	0.02	0.02	0.34	0.03	4.77	0.02	0.12	0.02
B	5.65	7.28	1.14	0.76	17.73	1.53	92.63	0.86	5.43	0.80
C	0.07	0.09	0.01	0.01	0.22	0.02	1.13	0.01	0.07	0.01

The largest amounts of heavy metals leach from foundry sand (FS), steel slag (SS), municipal solid waste incineration ash (MSWI), and cupola furnace slag (CFS). Due to high mobility, Mo and V often dominate the leached masses even though their relative share in the source (the construction) is small or average (see Table 5). Raw iron or steel slag shows high deposits of V. This is reflected in Table 7, where V from steel slag is the most quantity-rich substance flow from construction materials to the environment in this scenario. The upcoming German recycling directive (BMU, 2012) suggests limiting the application of these critical materials in specific applications, which is supported by this study. Foundry sand (FS) is difficult to assess as these sands are either resin-coated or clay-coated. This significantly influences the leaching behavior, and is thus still subject to German legislative process (BMU, 2012). Note that the hypothetical application scenario comprises the assumption that HFA application in open and partly-open constructions is not allowed. This is why HFA shows zero leaching, due to limitations in HFA application rates (*AR*) in percolated constructions for groundwater protection reasons (see Table 4).

3.2. Fate assessment

As introduced in Table 2, we model two subsoil scenarios with different parameters and thus different retardation potential (Eqs. (2)–(4)): sand (scenario I) and clay/silt (scenario II). In a sensitivity

Table 5
Annual masses of heavy metals in industrial mineral waste material in Germany, and share in construction sector (calculated based on Dehoust et al., 2008).

Material/n pollutant	Sb	As	Pb	Cd	Cr	Cu	Mo	Ni	V	Zn	Total
Mass of mineral wastes (t/year)											
Steel slag (SS)	95	63	227	1	964	488	126	246	1323	309	3841
Gran. blast furnace slag (GBFS)	5	7	27	<1	208	34	13	20	1407	121	1842
Blast furnace slag (BFS)	<1	1	5	<1	37	6	2	4	252	22	330
Melting chamber gran. (MCG)	6	16	111	<1	189	174	14	149	85	315	1059
Brown coal fly ash (BFA)	130	123	217	18	363	105	267	89	2341	356	4008
Hard coal fly ash (HFA)	61	218	378	5	315	395	126	336	1105	819	3758
Hard coal bottom ash (HBA)	2	4	13	<1	19	30	6	28	34	34	169
Foundry sand (FS)	1	18	35	2	493	48	30	24	173	175	998
Cupola furnace slag (CFS)	<1	4	7	<1	97	10	6	5	34	34	195
Copper residual slag (CRS)	6	101	2743	3	205	4568	18	65	34	13,810	21,551
Mun. solid waste in. ash (MSWI)	113	30	3652	28	2045	12,051	38	663	150	10,810	29,587
Total	420	584	7414	59	4934	17,908	646	1626	6937	26,803	67,330
In construction sector (%)											
	40	31	73	48	62	77	27	53	23	76	68

Table 7
Leached mass of heavy metals per secondary construction material and total (kg), 100 years, hypothetical application scenario.

	Sb	As	Pb	Cd	Cr	Cu	Mo	Ni	V	Zn
Material	Leached mass (kg)									
SS	4.3	43.4	4.3	4.3	7812.3	43.4	32117.1	43.4	72046.4	43.4
GBFS	0.9	8.8	0.9	0.9	4.4	8.8	0.9	8.8	529.5	8.8
BFS	0.1	0.5	0.1	0.1	10.1	0.5	20.2	0.5	25.2	0.5
MCG	0.5	5.4	0.5	0.5	2.7	5.4	0.5	5.4	2.7	5.4
BFA	<0.05	0.1	<0.05	<0.05	<0.05	0.1	158.7	0.1	<0.05	0.1
HFA	0	0	0	0	0	0	0	0	0	0
HBA	5.0	8.4	<0.05	<0.05	36.9	8.4	127.6	0.2	127.6	0.2
FS	2.9	3438.1	2.9	2.9	2292.1	1719.1	4584.2	1432.6	4870.7	2578.6
CFS	0.7	7.1	782.4	0.7	924.6	640.1	7.1	7.1	1280.3	569.0
CRS	0.5	916.7	0.5	0.5	2.3	275.0	3162.6	4.6	2.3	4.6
MSWI	354.3	1.0	0.1	0.1	507.5	1828.8	1263.9	1.0	181.9	1.0
Total (kg)	400	4400	800	<50	11,600	4500	41,400	1500	79,100	3200

analysis, we found that in both scenarios soil parameters show the same tendencies in how importantly they affect transfer coefficients, i.e. pH > L > ρ_s > f_{clay} > C_b > f_{oc} > θ_s. In a sensitivity analysis (parameter variation ±10%), the effect of pH was found to be two orders of magnitude higher than the effect of θ_s, and double of the effect of transport distance L and soil density ρ_s. For the two soil scenarios, Fig. 4 displays the relationship between transfer coefficients and leachate concentration. The values of TC_S range between 0 (0% of leached mass absorbed in soil, but leached to groundwater) and 1 (100% of leached mass absorbed in soil). The sum of TC_S and TC_{GW} per substance is 1 (Eq. (9)) and both are displayed on the vertical axes in Fig. 4.

The soil types selected for the two scenarios differ mainly with respect to the pH, f_{clay} and θ_s. The higher retardation capacity of the clay/silt soil (scenario II), which is mainly due to the greater f_{clay} and smaller pH, yields a generally higher share of metals remaining in the soil. Pb, Cr, Cu, Cd, Ni, V and Zn show close to zero leaching to the groundwater (Fig. 4). For the sandy soil and in particular for high leachate concentrations, Cd, Ni, V and Zn partially reach, and at high concentration of 1000 µg/l they all reach the groundwater compartment by more than 50%. The most mobile compound is As, with only minor differences for both soil types. Only Mo shows more substantial leaching in the clay/silt soil, while this is only marginally visible for Sb. This is attributed to the important role of the higher pH of 6.6 in comparison to the pH of 4.8 in the sandy soil scenario I.

Aside from the source concentration (Fig. 4), the transport distance significantly affects the transfer coefficients. Fig. 5 shows the computed values of TC_S as a function of transport distance for fixed source concentration of 10 µg/l. Most heavy metals are absorbed by soil layers of 1 m thickness in both soil scenarios. As indicated in Fig. 4, especially As, Sb, Ni and Cd are critical for groundwater in the less sorptive sandy soil. In the clayey soil, the mobile compounds As, Sb and Mo pass through even 2 m soil

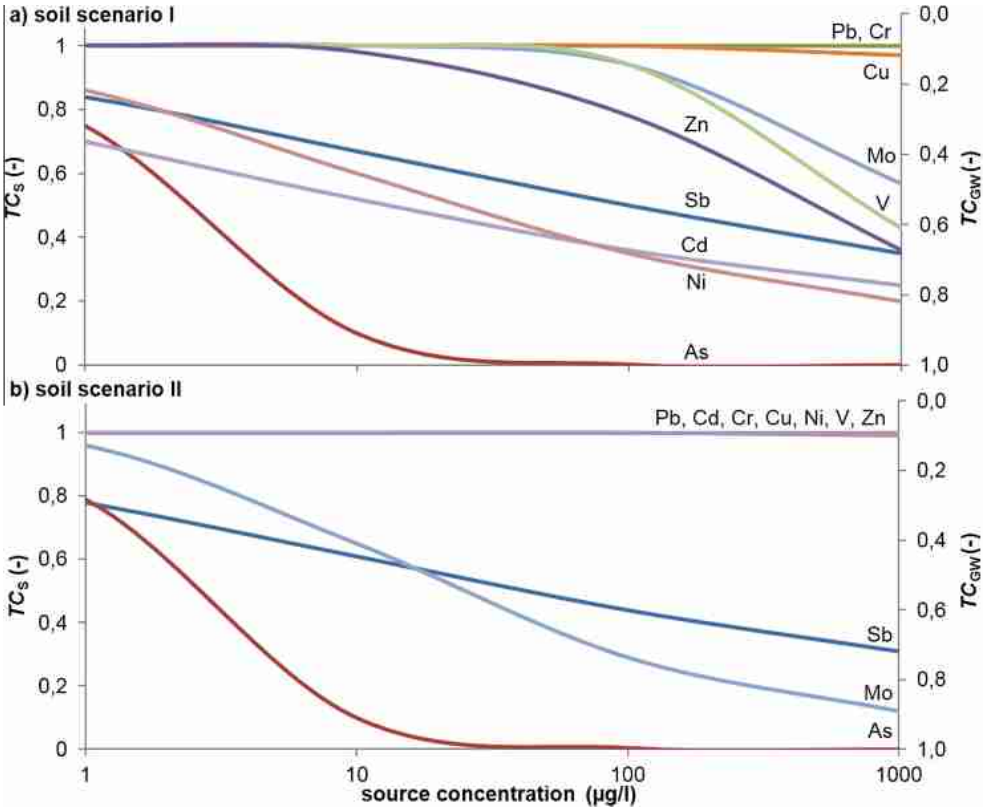

Fig. 4. Transfer coefficients, cumulated over 100 years, as a function of increasing source concentration for soil scenario I and II, L = 1 m.

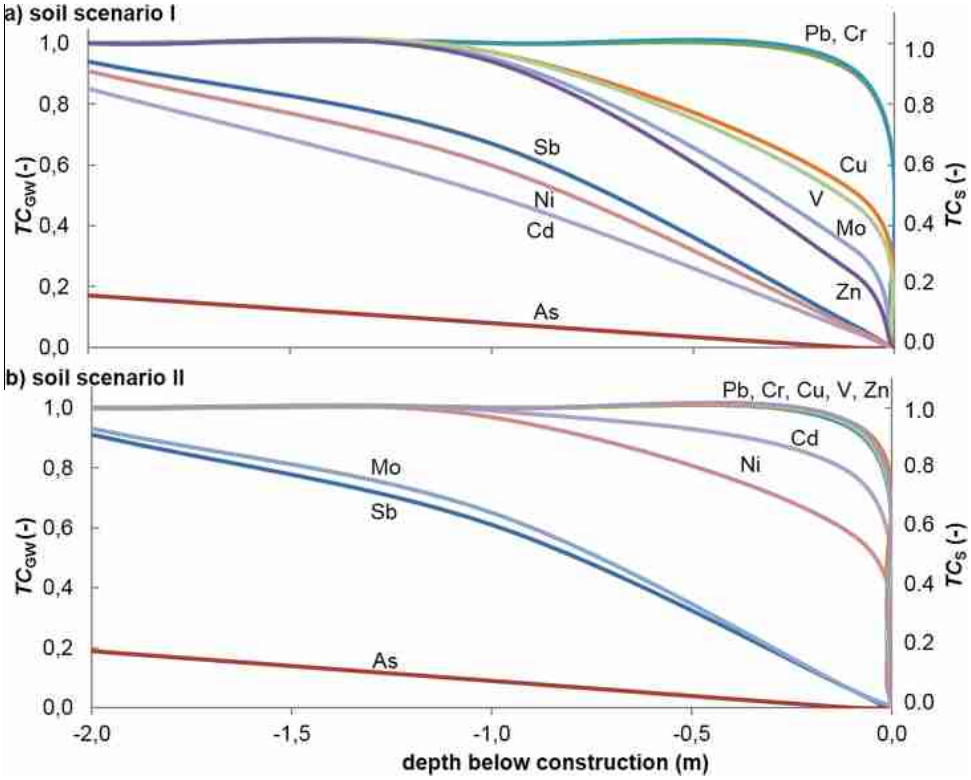

Fig. 5. Transfer Coefficients, cumulated over 100 years, as a function of increasing soil depth for soil scenario I and II, $C_S(t)$= 10 μg/l.

layers and break through to the groundwater. The metals Cd, Ni, V and Zn show the highest sensitivity to the soil type, while Pb and Cr are in both scenarios least likely to break through to the groundwater. These scenarios deliver insight into the fate of the heavy metals, however, they are only exemplary. The computed transfer coefficients are dependent on (1) substance, (2) soil parameters, (3) source concentration and (4) transport distance, and thus highly case-specific.

3.3. Ecotoxic impact

Freshwater Ecotoxicity characterization factors of the LCIA method USEtox (Table 3) are applied to heavy metal leaching (Table 1) from the three construction scenarios (Fig. 2). If not indicated otherwise, this is for subsoil scenario I (Table 2), a transport distance of 1 m and transfer coefficients as illustrated in Fig. 4a. Fig. 6 displays the ecotoxic impacts of waste materials per FU versus the total annual material quantity of mineral industrial wastes. It compares the "ecotoxicity potential of groundwater emissions" (ET_{GW}) and "total released ecotoxicology" (ET_{ENV}) to "potential ecotoxicology" ($ET_{GSCM,p}$). For $ET_{GSCM,p}$, all containing heavy metals are assumed to leach to freshwater. High values on the vertical graph axis indicate high ecotoxicity, and high values on the horizontal axis reflect high quantities of GSCM. Thus, dots in the lower left corner identify materials that have a low ecotoxicological impact and are applied in low quantities, while dots in the upper right corner identify materials with high ecotoxicological impacts that are applied in large quantities.

Copper residual slag (CRS) and municipal solid waste incineration ash (MSWI) are found to have the highest $ET_{GSCM,p}$. However, ET_{ENV} of CRS is low compared to other materials, whereas MSWI ranks among the materials with the highest ET_{ENV} (Fig. 6). This indicates that potential $ET_{GSCM,p}$ does not necessarily correlate with ET_{ENV}. This is in accordance with laboratory findings stating that pollutant solid content does not correlate with leachate concentration (Grathwohl and Susset, 2009). Thus, it is essential to clearly distinguish between solid content and leached mass of a material over time, also when it comes to ecotoxic environmental impact assessment.

Absorption capacities of soils substantially influence the fate of heavy metals, as evident from sensitivity of transfer coefficient to soil scenarios (Figs. 4 and 5). In a different interpretation, this means that subsoils can reduce the risk of the ecotoxicological impact on freshwater (ET_{GW}). Fig. 7 indicates that increasing subsoil layer thickness reduces ET_{GW}. In subsoil scenario II, all heavy metals except As, Sb and Mo are adsorbed by 100% over the modeled time frame in a layer of 20 cm. In subsoil scenario I, this is Pb, Cr and V, while Zn, Ni and Sb show the highest mobility of the modeled heavy metals. It can be stated that for homogenous subsoil layers of 2 m all heavy metals accumulate in soil and thus ET_{GW} is reduced by 100% in the modeled scenarios. It was found that beside substance, subsoil properties and percolation rate, the source concentration is also critical here. Subsoil can reduce the ecotoxicological impact on groundwater over 100 years, but heavy metals still remain in the subsoil. Thus, even if overlying polluted construction materials would be removed, there is a risk of heavy metal leaching to groundwater.

Regarding effect assessment in LCA, characterization factors are a large source of uncertainties (Rosenbaum et al., 2008). Also, ecotoxicological effects are regarded to be a linear function of pollutant quantities, while in reality this is a non-linear function of concentrations (see, among others, Olsson et al., 2006; Pettersen and Hertwich, 2008). This has to be considered when interpreting the results of leaching in the context of environmental assessments of GSCMs, since these impose low concentrations over long time frames. This also needs to be kept in mind when interpreting stored ecotoxicity results. There are two orders of magnitude between substance characterization factors (see Table 3). Regard-

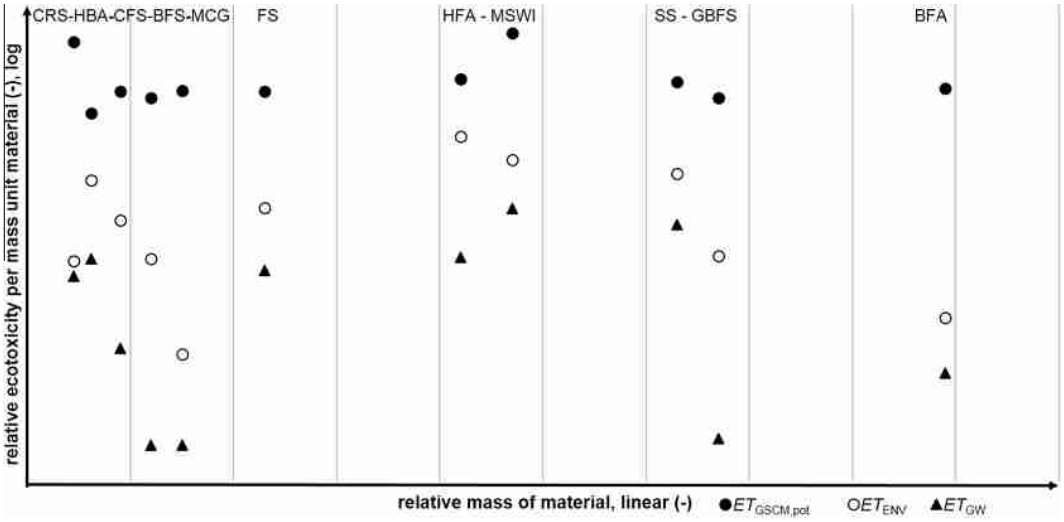

Fig. 6. Material quality versus material quantity. Leaching is assessed for construction scenario B, soil scenario I (1 m), 100 yrs. For $ET_{GSCM,p}$, all heavy metals are assumed to leach to freshwater. Quantity (x) is normalized to BFA = 1 and displayed linear, quality (y) is normalized to MSWI = 1 and log displayed.

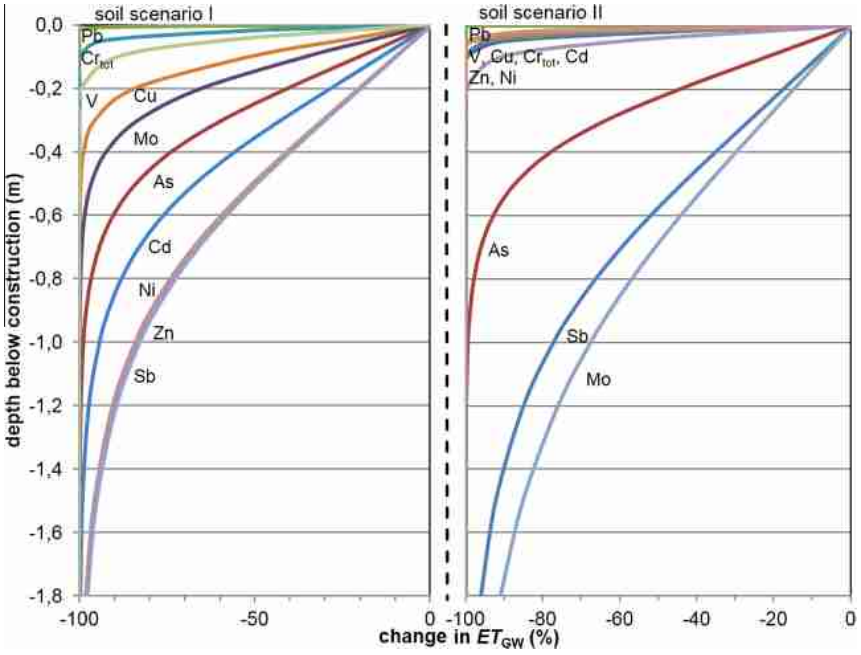

Fig. 7. Effect of transport distance on ET_{GW}, (Construction scenario A, MSWI, soil scenarios I and II).

ing transfer coefficients, we observe three orders of magnitude difference between substances (scenario: L = 1 m, $C_S(t)$= 10 μg/l). This variability in transfer coefficients increases for decreasing soil thickness and for increasing concentrations (Figs. 4 and 5). This distinctively illustrates the importance of site-dependent, rather than generalized, fate assessment in Life Cycle Impact Assessment of leaching from contaminated materials.

Table 8 lists the ecotoxicological effects of heavy metals per functional unit (FU) for the three construction scenarios. Scenario B (parking area) is found to have the highest ecotoxicological impacts. This is due to GSCM covering generally a larger area per mass unit in parking areas (B) than in road dams (A) or noise protection dams (C) (see also Table 6). Table 8 reveals that Mo from 1 FU hard coal fly ash in a parking area bedding (construction scenario B) shows a relatively low ecotoxicological impact per FU, although Mo from scenario B exhibits (1) by far the highest leached share of all regarded substances (93%, see Table 6), (2) the highest source concentration $C_S(t)$ (Table 1) and (3) is also one of the most mobile substances (Figs. 4 and 5). However, it also has the lowest characterization factors (Table 3). By contrast, Arsenic (As) has the highest characterization factor (Table 3), but ET_{ENV} of As is relatively low in most cases (Table 8). This is due to the fact that As shows low leachate concentrations for most materials (Table 1). In comparison, Cr is little mobile in soils (Figs. 4 and 5), but has high source concentrations (Table 1) and high characterization factors (Table 3). Thus, it contributes most to the total ecotoxicity of leaching from several GSCM materials (Table 8). These observations indicate that the ecotoxicological impacts of GSCM are complex functions of source concentration, substance properties, subsoil and effect factor, and thus are highly site- and case-dependent.

Table 8
Ecotoxic Environmental Impact ET_{ENV} in PAF (potentially affected fraction of species, in m^3 day/kg) per functional in unit subsoil scenario I (sand, 1 m) and construction scenarios A (road dam), B (parking area) and C (noise protection dam).

Material	Scenario	Sb	As	Pb	Cd	Cr	Cu	Mo	Ni	V	Zn	Total
SS	A	<0.01	<0.01	<0.01	<0.01	3.13	0.01	0.04	<0.01	32.01	0.01	35.19
	B	0.16	0.13	<0.01	0.01	149.52	0.46	1.78	0.14	1670.64	0.34	1823.18
	C	<0.01	<0.01	<0.01	<0.01	1.81	0.01	0.02	<0.01	18.58	<0.01	20.42
GBFS	A	0.01	<0.01	<0.01	<0.01	0.02	0.02	<0.01	0.01	2.01	0.01	2.06
	B	0.26	0.22	<0.01	0.01	0.69	0.77	<0.01	0.24	92.10	0.56	94.86
	C	<0.01	<0.01	<0.01	<0.01	0.01	0.01	<0.01	<0.01	1.13	0.01	1.15
BFS	A	<0.01	<0.01	<0.01	<0.01	0.48	0.01	<0.01	<0.01	1.32	0.01	1.82
	B	0.21	0.17	<0.01	0.01	21.86	0.61	0.12	0.19	60.59	0.45	84.21
	C	<0.01	<0.01	<0.01	<0.01	0.27	0.01	<0.01	<0.01	0.74	0.01	1.02
MCG	A	<0.01	<0.01	<0.01	<0.01	0.01	0.01	<0.01	<0.01	0.01	0.01	0.05
	B	0.21	0.17	<0.01	0.01	0.55	0.61	<0.01	0.19	0.61	0.45	2.79
	C	<0.01	<0.01	<0.01	<0.01	0.01	0.01	<0.01	<0.01	0.01	0.01	0.03
BFA	A	<0.01	<0.01	<0.01	<0.01	0.01	0.01	0.14	<0.01	0.01	0.01	0.20
	B	0.23	0.19	<0.01	0.01	0.61	0.68	7.09	0.21	0.68	0.50	10.20
	C	<0.01	<0.01	<0.01	<0.01	0.01	0.01	0.08	<0.01	0.01	0.01	0.11
HFA	A	3.69	0.24	<0.01	<0.01	142.82	0.01	0.15	<0.01	4.13	0.01	151.05
	B	192.00	16.13	0.15	0.01	6559.91	0.68	7.59	0.21	189.62	0.50	6966.80
	C	1.99	0.17	<0.01	<0.01	80.19	0.01	0.08	<0.01	2.32	0.01	84.76
HBA	A	1.54	0.24	<0.01	<0.01	5.85	0.74	0.06	<0.01	22.41	0.01	30.85
	B	78.81	16.13	<0.01	0.01	268.75	33.89	2.69	0.21	1051.57	0.50	1452.56
	C	0.84	0.17	<0.01	<0.01	3.29	0.41	0.03	<0.01	12.58	0.01	17.33
FS	A	0.01	0.78	<0.01	<0.01	2.41	1.00	0.01	0.25	5.68	1.27	11.43
	B	0.26	46.27	<0.01	0.01	110.76	46.10	0.62	12.77	260.98	53.16	530.93
	C	0.09	15.71	<0.01	<0.01	37.79	15.73	0.21	4.35	89.05	18.18	181.10
CFS	A	<0.01	<0.01	0.01	<0.01	2.26	0.87	<0.01	<0.01	3.47	0.65	7.26
	B	0.16	0.13	0.39	0.01	107.99	41.49	<0.01	0.14	165.80	28.19	344.30
	C	<0.01	<0.01	<0.01	<0.01	1.31	0.50	0.00	<0.01	2.01	0.36	4.18
CRS	A	<0.01	0.82	<0.01	<0.01	0.01	0.58	0.03	<0.01	0.01	0.01	1.46
	B	0.16	47.01	<0.01	0.01	0.42	27.66	1.65	0.14	0.46	0.34	77.84
	C	<0.01	0.55	<0.01	<0.01	0.01	0.34	0.02	<0.01	0.01	<0.01	0.91
MSWI	A	18.78	<0.01	<0.01	<0.01	12.61	25.22	0.09	<0.01	5.01	0.01	61.73
	B	991.62	0.17	<0.01	0.01	579.29	1160.61	4.32	0.19	230.29	0.45	2966.95
	C	9.97	<0.01	<0.01	<0.01	7.08	14.17	0.05	<0.01	2.82	0.01	34.09
Average		39.42	4.41	0.02	<0.01	245.51	41.61	0.81	0.58	119.05	3.21	

4. Conclusions and outlook

In the here chosen hypothetical construction scenario, GSCM construction activities of one year impose up to 150 t of leached heavy metals over the first 100 years, mainly V, Mo and Cr. Leached shares from total solid content are found to be below 1% for road dams and noise protection dams, but 20–50 times higher for parking areas, depending on the substance. Mo leaches by much higher shares to the environment than other here studied metals. In the case-specific upcoming German Recycling Directive (BMU, 2012), Mo is often a limiting factor to material application. This is supported from the macro-scale perspective of this study.

Transfer coefficients show high sensitivities to soil type for the two regarded soil scenarios. It was found that for both scenarios nearly all inspected heavy metals are retarded by more than 90% by a soil layer of 1 m, and by 100% by layer of 2 m. For the modeled concentrations, Sb, As and Mo were found to be most critical to leach to groundwater, even for longer transport distances. However, the transfer coefficients of the studied heavy metals span a three orders of magnitude range (soil scenario I, 1 m). This indicates the fundamental role of subsoil in impact assessment.

The environmental impact expressed as ecotoxicity per mass unit GSCM is found to vary over construction scenarios and deployed waste materials. Among the inspected heavy metals, Cr, Mo and Sb show the highest ET_{ENV}, and Sb, As and Mo the highest ET_{GW} for different construction scenarios and materials. Hard coal fly ash (HFA), municipal solid waste incineration ash (MSWI), hard coal bottom ash (HBA) and steel slag (SS) are identified as the most critical materials per mass unit. For steel slag, this is mainly because of V emissions. Brown coal fly ash, granulated blast furnace slag, steel slag and municipal solid waste incineration ash are found to be the most critical materials when both material quality (ET_{ENV}), and material quantity are considered. Recent developments towards metal extraction from mineral industrial wastes like MSWI bottom ash (see Thomé-Kozmiensky, 2013; Boesch et al., 2013) yield chances not only by recovery of secondary resources, but can potentially also reduce leachate emissions and thus the ecotoxicological impact. As indicated, a subsoil layer of 2 m can reduce ET_{GW} significantly and even result in emissions of some metals to be completely adsorbed in soil. The pH, transport distance and soil density are vital parameters for environmental impact assessment in the context of GSCM. However, the outcomes of the fate assessment is highly variable over soil scenarios and leachate concentrations and thus not to be generalized. It is observed that leached quantity and environmental impact are strongly dependent on time and space, which determine percolation rate, leached mass and soil constellation. Further, material and construction characteristics play a major role. Hence, a case-specific assessment as proposed in BMU (2012) seems adequate for enforcement of regulations in construction practice, while the here proposed macro-scale assessment can help to reveal critical combinations of material, site and application.

While relatively small shares are leached to the groundwater in the first 100 years, most of the pollutants remain in the material.

By application of GSCM, industrial wastes enter the construction material cycle and are likely to impose further environmental threats in subsequent construction settings, if recycled, or after the time horizon of 100 years considered here. Stored ecotoxicity (Hansen, 2004) quantifies the toxicity of the pollutants that remain in the material and could potentially be released over a longer time frame. From the studied mineral wastes, this is highest per functional unit for MSWI ash (Fig. 6). This is because of generally high contents of all metals (see S6), especially of those with high characterization factors, such as Cu (see S6). A significant share of released heavy metals (and thus ecotoxicological impact ET_{ENV}) does not impact on groundwater within the first 100 years, but is absorbed and stored in subsoil ($ET_{SOIL,p}$) and may impact on groundwater later.

Despite its limitations, an LCA approach in the context of environmental impacts from secondary materials can be a relevant guidance for decision makers, as it allows for a comparative assessment of potential ecotoxic impacts. Recent methods in ecotoxicity assessment of metals also consider chemical speciation, metal partitioning and bioavailability (Gandhi et al., 2010). These developments can improve the impact assessment, also in the context of long-term leaching from GSCM. For further research, integrated assessments of other pollutants, especially salts and PAHs, can be of interest. Also, leaching from demolition wastes is of vital relevance, as for most countries the quantitative mass flows of secondary construction materials from demolition wastes exceed those of mineral industrial wastes by far (see Vázquez, 2013). This study can be the basis for further assessments of leaching from traditional filling materials in road and earth constructions, which should be assessed as a benchmark.

Although leaching was regarded to be significant in relation to other life cycle stages of road and earth construction activities in previous studies, this was to a vast degree based on estimations, especially regarding fate assessment (Mroueh and Wahlström, 2002; Birgisdóttir et al., 2007). As proposed in this study, comprehensive leaching impact assessment and the key role of soil in fate assessment should be considered in further road and earth construction LCAs, both for waste and natural materials. However, three critical factors have to be stressed, that is (1) the restrictions of LCA to cope with the high variability in geographical conditions, construction specifics and material quality, (2) the limitations of LCIA, especially regarding groundwater impact assessment, and (3) the fact that ecotoxicological impacts can only be assessed as a function of pollutant masses in LCIA, but are rather a non-linear function of concentration. Future work that refines the spatial resolution for regionalized material- and soil-specific leaching assessment is needed. Additionally, the proposed fate models need to be validated with experience from long-term field monitoring. From a LCA point of view it is desirable to consider, in addition to concentration-based long-term risks (such as BMU, 2012), also mass-based long-term ecotoxicological impacts of GSCM in the construction industry for sustainable design of waste management strategies, and for minimizing degradation of the natural resources soil and groundwater.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.wasman.2014.04.022.

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