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LCA OF WASTE MANAGEMENT SYSTEMS

Preparing the ground for an operational handling of long-term emissions in LCA

Ioannis Bakas[1] · **Michael Z. Hauschild**[1] · **Thomas F. Astrup**[2] · **Ralph K. Rosenbaum**[3]

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Abstract
Purpose Currently, there is no meaningful methodology for the estimation of environmental impacts from long-term heavy metal emissions in a life cycle assessment (LCA) context, when an assessment of landfill and mining technologies is performed. In this paper, the aims are to investigate the main issues hindering the standardisation of a methodology to account for potential impacts from long-term metal emissions, and to describe the characteristics of a robust framework for an operational impact assessment methodology.
Methods In order to demonstrate the issues around potential impacts from long-term emissions in LCA and derive a scientific basis for developing an adequate LCA methodology to address these impacts, a two-part review on long-term metal emissions is performed that (a) identifies a suitable time-dependent life cycle inventory (LCI) while underlining the problems in existing emission prediction attempts and (b) describes the existing LCA approaches for accounting of toxic potential impacts from these emissions while explaining the reason that the identified proposals have not been adopted from the LCA community. These approaches are then compared upon the basis of a common LCI and their differences are highlighted.
Results and discussion A suitable dynamic LCI is identified for landfill emissions, which calculates Ni, Zn, Cd and Pb emissions as a function of time, based on assumed developments of the leachate pH. The results of the application of the different impact assessment methods on that LCI differ by up to 8 orders of magnitude. Therefore, the decision-making process supported by an LCA becomes very confusing. None of the approaches consider future changes in the receiving environment and are accompanied with any uncertainty considerations.
Conclusions In order to move towards a robust environmental assessment of long-term emissions, it is necessary to (i) represent future potential impacts more accurately by estimating time-dependent characterisation factors (CFs) corresponding to changing environmental conditions, (ii) develop more robust estimations by addressing uncertainty and (iii) refer to actual potential impacts, by taking into account the current and future background concentrations.

Keywords Heavy metals · Landfill · LCA · Long-term · Toxicity

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✉ Ioannis Bakas
ioba@dtu.dk

[1] Division for Quantitative Sustainability Assessment, Department of Management Engineering, Technical University of Denmark, Kgs. Lyngby 2800, Denmark

[2] Department of Environmental Engineering, Technical University of Denmark, Kgs. Lyngby 2800, Denmark

[3] ELSA-PACT—Industrial Chair for Environmental and Social Sustainability Assessment, UMR ITAP, Irstea, 361 rue Jean-François Breton, BP 5095, Montpellier Cedex 5 34196, France

1 Introduction

Life cycle assessment (LCA) is a decision support tool aiming at assessing the environmental impacts associated with products or systems (ISO 2006a; ISO 2006b). An important element of LCA is the quantification of emissions, representing an environmental exchange between the product or system and the ecosphere, and the subsequent characterisation of the emissions into potential environmental impacts by applying characterisation factors (CFs) to the emissions. LCA is widely

used in decision-making processes in waste management by assessing integrated waste management systems or comparing different alternatives for treatment of waste (Laurent et al. 2014a; Laurent et al. 2014b). Contrary to product LCA, waste LCA focuses only on the end-of-life stage of products' life cycles. In waste LCA, the technologies involved are relatively limited (corresponding to a chain of waste treatment facilities). The main direct emissions are thus typically caused by a few installations, the location of which is easier to identify as well as the conditions of the receiving environment. This implies that a more site-specific approach is possible, taking into account the local conditions in contrast to the typical situation for product LCAs. A site-specific assessment is also highly relevant in the case of landfills, where the most important leachate impacts are expected to be on a local scale (i.e. within a few km^2).

Landfilling of waste (from e.g. households, industrial sites, institutions and construction and demolition sites) constitutes a predominant waste treatment technology globally. However, landfill leachate poses an important environmental concern. The leachate composition has been well investigated for many types of landfills, and its heavy metal content comprises one of the major environmental pollutants in waste management (Christensen et al. 2001). Besides toxicity, a problematic property of metals is their environmental persistence, which is practically infinite and thus far superior to any organic chemical. In an LCA context, the toxicity potentials of heavy metal emissions have hence been estimated as relatively high compared to those of other compounds (Huijbregts et al. 2000). Given the high toxic potential, an accurate representation of potential impacts from landfill metal leaching in LCA is essential to ensure the credibility of the tool in assisting informed decision-making, neither ignoring nor overestimating their importance.

When trying to estimate the impacts from landfilling in an LCA context, an important characteristic of landfill metal emissions is the very long time horizon of thousands or even hundred thousands of years over which their release takes place in contrast to the other processes in a system which typically emit within minutes to days. The widely used ecoinvent inventory database defines long-term metal emissions as occurring after 100 years until 60,000 years from now ("until the next plateau-covering glacial period [in Switzerland] is estimated") (Hischier et al. 2010). During this period, small amounts of leachate or run-off containing pollutants (especially heavy metals) are emitted from the landfill to the surrounding soil, aquifer and eventually the surface water.

Apart from leachate from landfills of municipal, inert or hazardous waste, long-term heavy metal emissions can be found in the tailings from mining operations leading to significant environmental burdens (Stüben et al. 2001) that are not properly addressed in LCA mainly due to the lack of reliable data (Althaus and Classen 2005) and an appropriate impact assessment methodology. This problem has been underlined and investigated through a sensitivity study that showed the strong dependence of LCA results from mine tailing management on the choice of time frame (Reid et al. 2009). Also, the management of radioactive waste can cause long-term leaching of metals (Singh and Hendry 2012), but due to the special properties of radioactive waste, this source of heavy metal emissions is not investigated in this paper, although some of the considerations described below could be relevant.

Out of the leachate pollutants from landfills, long-term metal emissions constitute a particularity in the LCA context. Inventory modelling in LCA applies a time integration principle, according to which all emissions should be treated as a pulse emission occurring at one point in time, whether they in reality occur in different parts of a life cycle or extended over long time periods (Guinée and Heijungs 1993). The latter case poses problems when emissions occur in relatively low concentrations over very long time periods leading to large cumulated pulse flows. Their impacts are potentially overestimated when the resulting impact scores from emissions in low concentrations integrated over tens of thousands of years are interpreted in the same way as impact scores for emissions occurring in significantly shorter periods and thus integrated over shorter periods of time, which is the case for transportation processes and many industrial processes. There is a "dilution in time" which is important for the observable impacts from the long-term emissions but not represented in the calculated potential impacts in life cycle impact assessment (LCIA).

In LCIA, toxic impacts are modelled using steady-state conditions, applying integration over a defined time horizon. Integrating the potential impacts of long-term emissions over a relatively short time horizon like 100 or 500 years (as done for e.g. global warming potentials), and thus neglecting impacts from emissions occurring later, potentially leads to an underestimation of their impacts. On the other hand, including them fully through integration over very long or even infinite time horizons would lead to a strong overestimation, as the (perhaps negligible if from very low emission concentrations) potential impacts occurring over a long period of time would be fully attributed to the product as if they were occurring right now (as one large emission) in the same way as an emission from e.g. a waste incinerator. While the latter approach does not account for the dilution in time of the impact, the first approach completely neglects the long-term potential impacts. These two extremes present a dilemma for which an operational and accurate solution is needed in LCIA. Some LCIA approaches specifically deal with long-term emissions (Pettersen and Hertwich 2008; Hauschild et al. 2008). However, currently, there is no consensus among researchers on a common methodology, which is illustrated by the differences in this perspective of the various LCIA methodologies (Hischier et al. 2010). Moreover, there are no characterisation

factors available for long-term emissions of metals, which thus have to be represented by the generic characterisation factor for the metal. Further research is called for so that a more accurate representation of long-term emissions is achieved (Hischier et al. 2010).

In summary, when attempting to integrate this kind of emissions and their potential impacts in an LCA, two main hurdles need to be overcome: (1) estimating/predicting the future emissions in the inventory phase and (2) characterising the emissions in a meaningful way that puts them into perspective relative to the impacts caused by emissions that occur over a much shorter time. In order to better understand the perspectives for an adequate representation of long-term emissions, a consistent overview of past experience and current practice within the LCA field is needed. This is done through (i) reviewing the status, reliability and common ground of emission prediction methodologies from an LCA perspective; (ii) reviewing the current proposals for characterisation of long-term emissions in LCIA, comparing them in terms of the results they provide; and, based on that, (iii) deriving main principles for a scientifically robust framework for an operational impact assessment for potential impacts of long-term emissions in the LCA context.

2 Methodology

For addressing the aforementioned objectives, a literature review is conducted. Central studies which have been published in peer-reviewed journals and those that address the issue of long-term emissions from landfills are presented.

The first part of the review aims at identifying studies mainly within the waste management field, which can provide a time-dependent life cycle inventory (LCI) for long-term metal emissions from landfills. A scientifically sound study that provides metal emission profiles for Ni, Cd, Zn and Pb, suitable as an input to an LCI, is selected. The emission profiles correspond to the leaching of these metals from an air pollution control residue (APCR) landfill with a total capacity of 1,000,000 tonnes.

The second part maps research within the LCA field to identify the various approaches that have been developed to address the challenges in assessing potential impacts from heavy metals emitted in low quantities but over long periods of time. The review illustrates the current state of the art and analyses the existing approaches in terms of the perspectives they adopt on future emissions.

The results from the two parts of the literature search are combined to form a case study where the different LCA approaches are applied on a selected emission profile of a metal leaching out of an APCR landfill over a long time period. The same CF (or comparative toxicity potential) is used for both short- and long-term emissions, adopted from recent literature which estimates CFs for Ni and Cu with respect to terrestrial ecotoxicity (Owsianiak et al. 2013). The CF for Ni is chosen as an emission profile for Ni can be found in literature (Astrup et al. 2006). The available CF is relevant for terrestrial ecotoxicity impacts, but from a nickel emission to air. Thus, this CF is corrected for emissions directly to soil using USEtox (Rosenbaum et al. 2008), and the resulting CF is 4.86×10^{3} $m^3/kg_{emitted\ to\ soil}\times day$.

Based on a common emission profile and an identical CF, the existing LCIA approaches can be compared on the same basis. The time-dependent emission profile for Ni provides emission quantities per 200-year time step. This quantity is integrated over 100 years (the first time step is divided into two) and over an infinite time horizon, and the potential impacts in each case are then estimated by multiplying with the CF. The difference between the impacts from the 100-year integration and the infinite time horizon is the impact allocated to a separate impact category, named stored toxicity.

The impact is then estimated per time step, and discount rates of ±0.01 % are applied to future potential impacts. The magnitude of the rates is set to 0.01 % as this is shown to correspond to time horizons of up to 100,000 years (Hellweg et al. 2003). One positive and one negative discount rate are modelled, in order to account for uncertainties regarding future economic development which determines the level of the compensation required for future damage.

Lastly, the emission concentrations in the landfill leachate, provided also by the Ni profile per time step, are compared to the European median background concentration of Ni in soils. The median situation is chosen for demonstrative reasons, but it is not recommended as the background concentrations for metals vary greatly geographically and according to soil types (Zhao et al. 2007). The median Ni extractable concentration in European soils is estimated at 16 mg/kg (Lado et al. 2008). This is used to estimate the background Ni dissolved concentration by using the equations below and the K_{dNi} (280 l/kg) default values found in the USEtox™ multimedia toxicity model (Rosenbaum et al. 2008):

$$K_d = \frac{C_{sorbed}}{C_{dissolved}}$$

It is also assumed that

$$C_{total} = C_{sorbed} + C_{dissolved}$$

where C_{sorbed} (mg/kg_{soil}) is the sorbed metal concentration, $C_{dissolved}$ is the dissolved metal concentration (mg/kg_{soil}) and C_{total} is the extractable metal concentration (mg/kg_{soil}). *$C_{dissolved}$ is converted into milligrams per liter by using the soil bulk density ρ_{bulk} (mg/l) and the soil water content w (% v/v):*

$$C_{\text{dissolved}}\left[\frac{\text{mg}}{\text{l}}\right] = \frac{\rho_{\text{bulk}} C_{\text{dissolved [mg/kg]}}}{1000\ w}$$

By combining the above equations, $C_{\text{dissolved}}$ can be calculated as

$$C_{\text{dissolved}}\left[\frac{\text{mg}}{\text{l}}\right] = \frac{C_{\text{total}}}{K_{\text{d}} + \frac{1000w}{\rho_{\text{bulk}}}}$$

The median global bulk density for soils, based on more than 15,000 samples, is estimated at 1.4 t/m^3 (Batjes 2008), and it is assumed to apply for Europe. The water content value is adopted from the USEtox default value, which is 0.2. Therefore, the dissolved median Ni concentration in European soils is estimated at 0.038 mg/l.

Only the Ni concentration that is above the background level is transformed into mass and integrated over time, then multiplied with the CF for Ni in order to provide the total actual potential impact.

A comparative analysis of the results of the application of the various LCA approaches helps identify the effect that the specific assumptions and perspectives of each approach have on LCA results, thus directly affecting decisions supported by waste LCA. Based on the analysis, a listing is made of the requirements for developing a robust, common framework for addressing long-term emissions in LCA, by combining different characteristics of the existing approaches.

3 Modelling long-term emissions of heavy metals

Different types of landfills have been investigated in terms of pollutant release from the leachate. Inert landfills such as an APCR landfill tend to be investigated more regarding metal emissions (e.g. Hellweg et al. 2005), while landfills richer in organic matter are normally investigated in terms of organic pollutants (e.g. Kjeldsen et al. 2002). Long-term metal emissions are present in all types of landfills, but the primary focus for toxicity might change with landfill type. Also, the waste composition in the landfill influences the pattern of metal emissions (Bolton and Evans 1991). The literature review conducted here is not confined to a specific landfill type in order to be comprehensive and with the overall aim at identifying a time-dependent leachate inventory.

Heavy metal concentrations in landfill leachate have been extensively monitored and simulated in batch tests for a surveyable time period, usually capped at 100 years (Finnveden et al. 1995; Flyhammar et al. 1998; Kjeldsen and Christophersen 2001). A review of published monitoring and testing results concludes that the leachate metal concentrations within such a short time frame do not pose a great environmental risk (Kjeldsen et al. 2002). This observation fits well with the generally accepted assumption that typically the vast majority of the landfilled waste content of heavy metals remains in the landfill 100 years after the waste deposition (Hauschild et al. 2008). The heavy metal retention in landfills can be explained by mechanisms such as sorption and precipitation (Kjeldsen et al. 2002).

On the other hand, the long-term heavy metal concentration in leachate is difficult to predict in a solid way as laboratory simulation is not possible and mathematical models hence cannot be validated for time frames as long as 100,000 to 250,000 years and should, therefore, not be overinterpreted (Finnveden et al. 1995). Theoretically, after the stable methanogenic phase of a landfill rich in organic matter, the oxygen intrusion facilitates metal mobility due to decreasing pH and oxidation to more soluble metal species (Christensen et al. 2000; Bozkurt et al. 2000). However, existing experimental work leads to contradictory results even for the medium time frame (Kjeldsen et al. 2002).

Given the importance of oxygen intrusion on the landfill chemistry, a modelling approach has been developed based on the oxygen effect (Bozkurt et al. 2000). In order to predict future heavy metal concentrations in the leachate, this model is based on the fact that, after the methanogenic phase of an organic landfill is completed, the subsequent humic phase is characterised by much slower organic carbon degradation, which means that oxygen diffuses into the landfilled waste (Bozkurt et al. 2000). The conceptual model is based on an analysis of the main processes governing each landfill phase after the methanogenic phase, with the presence of oxygen considered as the determining factor for metal mobility (Bozkurt et al. 1999).

Bozkurt et al. modelled several scenarios in order to determine the total depletion time for organics and metals in the waste mass and the influence of factors such as the type and characteristics of the top cover (Bozkurt et al. 2001). However, no results are presented regarding the quantity of heavy metals leaching out of the landfill as a function of time. Therefore, this work is not suitable for compiling an LCI.

Finnveden (1996) attempted to produce emission rates for metals contained in a municipal waste landfill based on a collection of mass balance approaches from literature. The author produced best estimates of emission rates for a surveyable time period (about one century from waste deposition), which may constitute a dynamic LCI. However, the mass balance approach for an infinite time frame, which is in line with the scope of this paper, claims that all metals will eventually leave the landfill. Therefore, this study does not provide a suitable time-dependent LCI.

Other attempts to predict the evolution of metal concentrations out of a landfill involve geochemical models. One study attempted a prediction of the evolution of Cu and Cd concentrations in a municipal incineration residue landfill, based on

soil infiltration rate, share of macropore flow and sorption capacity of the waste (Hellweg et al. 2005). With the help of scenarios that determine the presence of these two heavy metals in waste and the use of the geochemical model PHREEQC (Parkhurst and Appelo 1999), the authors tried to predict the development of emission concentrations over the next 20,000 years for Cd and over 100,000 years for Cu. The results of this work could be used in an LCA context, as illustrated in the same study which also presents results of an LCIA. On the other hand, only information for two metals is given and the time frames for the emission development are different for both metals. A different geochemical landfill model, LandSim, has also been employed to predict the depletion of heavy metals (among others) from a typical municipal solid waste landfill over a 20,000-year period, based on a variety of landfill parameters (Slack et al. 2007). Results of the simulation show that heavy metals leach at very low concentrations over a long period of time and that only chromium and mercury at any point in time exceed current drinking water standards proposed by health organisations. Moreover, the results indicated that the heavy metal content remaining inside the waste mass is negligible after 4000 years. This conclusion contradicts other evidence of literature suggesting long attenuation of heavy metals in landfills and also findings of laboratory tests (Flyhammar et al. 1998; Hyks et al. 2009). Therefore, the reliability of these results for compiling an LCI is questionable.

A laboratory test aiming at simulating the landfill leaching process can also be found in literature (Hyks et al. 2009). The test used a column percolation set-up, which applied forced infiltration of eluent in the column, filled with samples from an inert landfill, in order to simulate leaching until high liquid to solid (L/S) ratios were reached, corresponding to a leaching duration of thousands of years (Hyks et al. 2009). The test was performed on samples from a landfill for municipal solid waste incineration air pollution control residues, up until the L/S ratio reached 200–250 l/kg, in order to determine the leaching behaviour of a large variety of heavy metals. The L/S ratio could then be translated into time based on site-specific landfill characteristics (Hjelmar 1990), and in this case, the total simulated duration corresponds to more than 10,000 years. Results of the test showed that less than 3 % of the initial heavy metal mass had leached during the simulated time period of 10,000 years. This test cannot, however, be used in the context of LCI as it does not cover the entire leaching time period of a landfill.

If a relationship between pH and the L/S ratio is established for fly ash landfills, it is possible to predict Al, Ca, Cd, Ba, Mg, Ni, Pb, S, Pb, V and Zn concentrations in the leachate, using pH as a determining factor, as shown in another study (Astrup et al. 2006). Two types of incineration residues (semi-dry and fly ash) were treated in batches while they were either in a carbonated or in an uncarbonated form. The pH was measured in the batches, and for each measurement, the leaching of metals and the L/S ratio were calculated. In this way, leaching concentrations were able to be connected with the pH level. The L/S ratio was translated into time based on average Danish environmental conditions. Based on these assumptions, the authors produced emission profiles for the selected heavy metals until an L/S ratio of 5000 which corresponds to 100,000 to 250,000 years. The results show different behaviours for different metals, which is also due to the type of residue investigated and the presence of carbonation. Therefore, it is difficult to group metals according to their leaching patterns.

The graphs in Fig. 1 present the leaching concentration profile over time modelled using the results of Astrup and co-workers for four selected metals and averaged over the two waste streams modelled (non-carbonated fly ash and semi-dry residue). The model has a temporal resolution of 200 years, and therefore, this study could easily be used to provide a time-dependent LCI for this type of landfills. The total time frame is chosen at the lower end of the range, set at 100,000 years.

The extrapolation of information obtained through leaching tests into considerations regarding metal leaching behaviours in real landfills is hindered by restriction posed in the leaching simulations in the laboratory (van der Sloot et al. 1996). However, for the scope of long-term emissions where actual measurements are impossible to obtain, laboratory tests are the only source of information available regarding metal leaching development patterns in the distant future.

It is important to notice that for a very long initial stage, the concentrations of leaching metals remain at a low level but then start to rise, with Ni and Zn reaching their maximum predicted concentrations at the end of the modelling period. This observation has important consequences when assessing potential impacts and monitoring of pollution around landfill sites.

4 State of the art in LCA

The life cycle impact assessment of long-term heavy metal emissions is currently problematic. The uncertainty of including long-term heavy metal emissions in toxicity assessments within LCA has been identified as a problem for quite some time, from both a methodological and case study perspective (Finnveden and Nielsen 1999; Hauschild et al. 2008; Hischier et al. 2010). Many LCA studies of solid waste management systems underline the uncertainty in their results stemming from the inability to accurately account for these potential impacts, while others choose to ignore toxicity impacts altogether (Laurent et al. 2014a; Laurent et al. 2014b). The uncertainty discussed relates mainly to modelling uncertainty (both inventory and impact modelling). However, there is also a

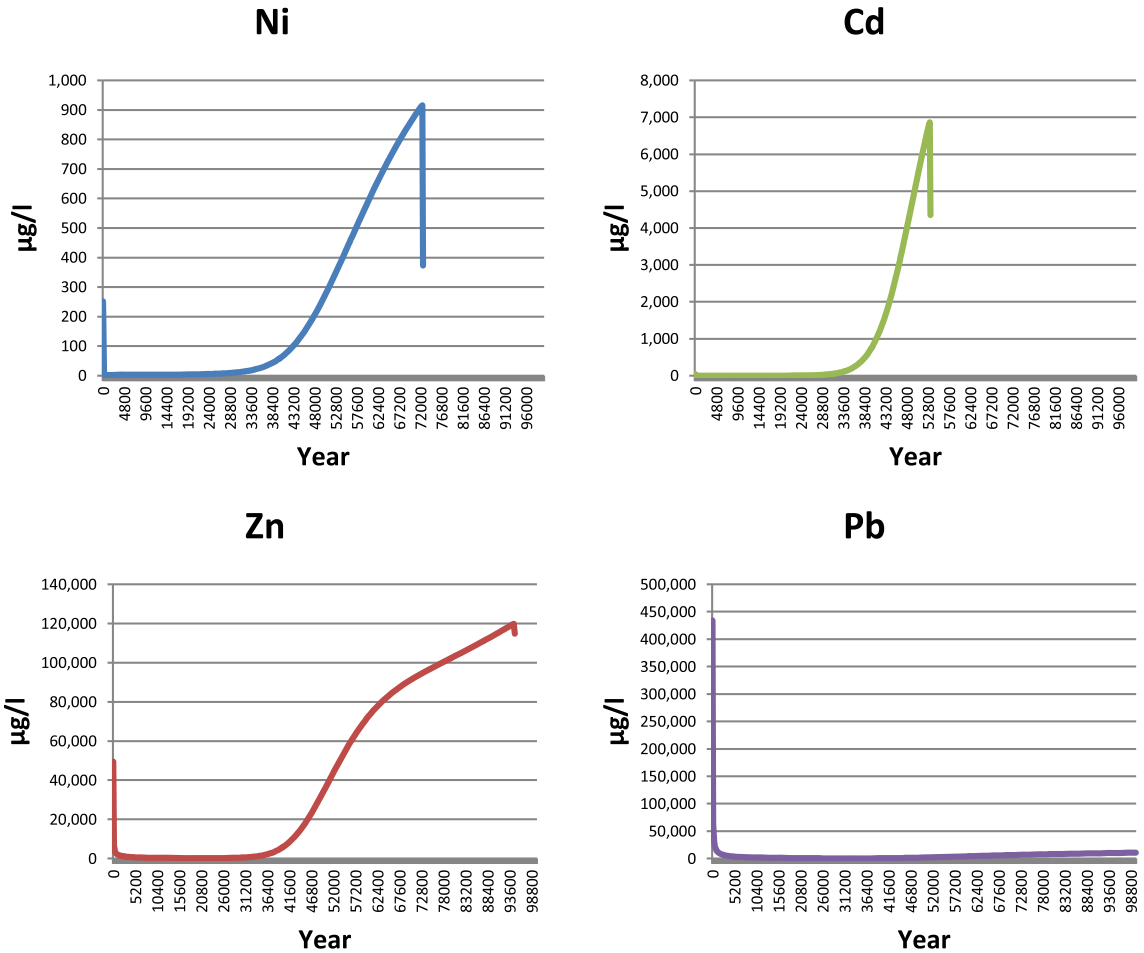

Fig. 1 Development in average leaching concentrations for four heavy metals over 100,000 years (based on Astrup et al. 2006). Note: The proposed patterns do not fully cover the time frame of 100,000 years, as the metal content in the waste is depleted earlier, except for Pb

methodological uncertainty present which is based on the choice of time aggregation principle for the emissions (Finnveden and Nielsen 1999; Udo de Haes et al. 1999).

Long-term emissions are currently treated in LCA either by modifications in the inventory modelling or by hybrid methods adopted from other fields such as risk assessment (Hischier et al. 2010). The state of the art in the inventory registration of long-term emissions is represented by ecoinvent, which is one of the most widely used LCA databases, providing inventories for many processes, including waste management (Frischknecht and Rebitzer 2005).

The ecoinvent background report for landfills provides a comprehensive set of arguments for and against inclusion of long-term emissions in the inventories and consequently in impact assessment (Doka 2009), but no consensus is reached by the authors (Hischier et al. 2010).

Therefore, ecoinvent inventories report both short- and long-term emissions from landfills, but their inclusion or not in the impact assessment depends on whether or not the chosen impact assessment methodology allows for it. Impact assessment methodologies such as CML2001, EDIP 97/2003 as well as IMPACT 2002+ (Hauschild and Wenzel 1997; Guinée et al. 2001a; Guinée et al. 2001b; Jolliet et al. 2003; Hauschild and Potting 2005) can produce differentiated impact assessment results by including or excluding long-term (beyond 100 years) emissions. Other methodologies, such as Eco-indicator 99 and ReCiPe, apply social perspectives reflecting the social and environmental values of current generations towards future generations (Goedkoop and Spriensma 2001a; Goedkoop and Spriensma 2001b; Goedkoop et al. 2009). Some of these perspectives include both short- and long-term emissions and apply the same characterisation factor for the two emission durations in the impact assessment.

A similar approach of differentiated time periods has been developed previously, by distinguishing between two time periods, namely the surveyable time period (set to 100 years corresponding to the end of the methanogenic phase of a typical municipal waste landfill) and the infinite time period (set to the time necessary for all emissions to occur) (Finnveden

1999). The author recommends assessing potential impacts over both time periods in a full LCA, since it should consider impacts from all relevant emissions, instead of the common practice of cutting off emissions occurring after 100 years.

Several approaches have been developed beyond the LCI phase in order to tackle the issue of long-term emissions' potential impacts. These approaches aim at developing specific LCIA methodologies that estimate impacts from emissions beyond a point in time (stored toxicity) (Hauschild et al. 2008), or at weighing differently impacts from future emissions compared to current impacts (environmental discounting) (Hellweg et al. 2003). The last methodology presented here functions as an intercept between LCI and LCIA by eliminating emissions beyond a certain threshold. Dynamic LCIA methods that have been developed but do not address specifically emissions during very long time frames are not examined here as the long-term perspective requires a special framework compared to simple time-differentiated approaches (Levasseur et al. 2010).

4.1 Stored toxicity

In order to distinguish between potential toxic impacts from short- and long-term emissions, a new impact category has been proposed, which builds upon current ecoinvent practice: the emissions occurring during the first 100 years of a landfill are treated together with the emissions released from all other processes in the life cycle (Hauschild et al. 2008). The emissions beyond that point in time are classified as belonging to a new impact category, named stored toxicity (or stored human toxicity and stored ecotoxicity), following the EDIP 2003 method (Hauschild and Potting 2005). In order to estimate the potential toxic impacts, emissions inventoried in the stored toxicity categories are characterised in the same way (and based on the same factors) as in the traditional toxicity categories. The characterised results are then normalised based on an area's total annual stored toxicity, which is determined from the mass flow of heavy metals within all waste streams in the year 1994, divided by the area's population. The normalisation is performed in order to assess the relative magnitude of the examined system's impact.

The proposed stored toxicity approach was applied in an LCA study on landfilled bottom ash, and the results showed that stored toxicity impacts are 325 times greater than the highest result in the traditional impact category spectrum (namely $Ecotoxicity_{water}$) (Birgisdóttir 2005).

This approach, although in line with current practice, is still problematic since the introduced impact category lacks interpretation and weighting factors needed in the comparison with potential impacts in the other categories. However, it provides a first step in separating between (i) emissions occurring within a time period where landfill chemistry, and therefore heavy metal emissions, is well known and monitoring or testing is possible and (ii) emissions occurring in a distant future where uncertainty in their development is high and no monitoring or testing is possible.

4.2 Environmental discounting

Hellweg and co-workers attempted an application of economic discounting in environmental assessment under the context of LCA (Hellweg et al. 2003). The authors based their analysis on four main arguments for environmental discounting (changes in magnitude of damage, pure time preference, productivity of capital, uncertainties). These arguments, based on the economic theory, can be quantified in order to produce discount rates, applied on potential impacts from future emissions, so that the weight of future impacts is differentiated compared with impacts from emissions occurring instantly.

The choice, however, of a discount rate is subjective and very decisive, and in order for it to be meaningful, it needs to be close to 0 ($-1\ \% \leq r \leq +1\ \%$). Under a higher positive discount rate, even potential impacts from relatively short-term emissions (below 100 years) would practically disappear. Even in the case of low discount rates, the value of the discount rate is the sole most determining factor for the resulting potential impacts, because of the exponential nature of discounting. The choice of a discount rate presupposes the monetisation of environmental impacts and depends on assumptions on the development of the future global economy, as well as value choices of the LCA practitioner.

The adoption of moral archetypes from Cultural Theory for LCA modelling purposes (Hofstetter et al. 2000) could be a guide for selecting an appropriate discount rate according to the selected cultural perspective (Hellweg et al. 2003). However, even within each cultural perspective, the subjectivity of the choice is still not eradicated which endangers the objectiveness of decisions supported by an LCA, while it has been shown that the influence of an archetype choice is significant for the overall LCA results (Schryver et al. 2012).

4.3 Background concentrations

Another approach for tackling the issue of long-term emissions has been inspired by the field of risk assessment. According to this, emissions occurring at leachate concentrations that are below the background concentration levels for each heavy metal can be discarded as they cause no effect (Finnveden 1999). Therefore, it has been proposed to cut off emissions of heavy metals that occur after the point in time when their concentrations fall below the background concentrations (Finnveden and Huppes 1995). This approach has inspired studies related to landfill aftercare period and technologies related to an effort for a fast reduction of metal leaching concentrations (Brand et al. 2014).

This approach neglects that heavy metal emissions do not necessarily follow a decreasing trend, as illustrated in Fig. 1, and that background concentrations might not stay at the same level during the long time frame considered. For some heavy metals, emissions have been shown to decrease steadily and then to increase again in the future (Astrup et al. 2006). On the other hand, heavy metal background concentrations could increase substantially in the future because of accumulation in specific environmental compartments (van der Voet et al. 2000).

Moreover, this approach could be considered as conflicting with the general LCA principles according to which all emissions should be taken into account whether above or below thresholds (Udo de Haes et al. 1999), although attempts to include background levels in LCIA can be found in literature (Hellweg et al. 2005). This holds in particular in the extreme case of landfill leaching in contaminated soil where landfill emissions could potentially dilute the soil pore water, thus reducing its toxicity to some extent, depending on the affected organisms and the dose-response curves for the leached metal. It could, however, be argued that a cut-off determined by the natural background pore water concentration gives a more relevant representation of actual potential toxic impacts resulting from long-term heavy metal emissions than a cut-off at an arbitrarily set time limit or an infinite time horizon, as concentrations of metals below the background level have no toxic effect on the local ecosystems. Due to the natural variation in soil water concentrations of metals, a high degree of site specificity would be relevant in the modelling (Lado et al. 2008).

5 Comparison of different methods dealing with long-term emissions

The LCA methods described above differ in many aspects: (i) the approaches to handling the short- and long-term emissions; (ii) the respect of the time equity principle; and (iii) the inclusion of subjective (value) choices in the accounting of potential impacts. To investigate the importance of these differences, the different LCA approaches are applied on the same emission profile of Ni leaching out of an air pollution control (APC) residue landfill. Based on the assumptions on the initial content of the landfill, Ni will have leached out in about 73,000 years. The cut-off of emissions after 100 years, the adoption of an infinite time horizon, the handling of emissions in a separate stored toxicity impact category, the concept of environmental discounting and the estimation of potential impacts only from emissions above the background concentrations are all applied on the Ni profile.

The CF used for the analysis is estimated at 4.86×10^3 $m^3/kg_{emitted\ to\ soil} \times day$, and it represents the impact on terrestrial ecotoxicity from an emission of Ni to soil.

The results of the application are shown in Fig. 2. The lowest impact given by the cut-off of emissions at 100 years and the highest impact from the negative discount rate differ by approximately 8 orders of magnitude. The stored ecotoxicity approach (which is practically the difference between the cut-off and the infinite time horizon approaches) gives an impact around 5 orders of magnitude higher than the results of the cut-off approach, demonstrating the significance of long-term against short-term emissions. The use of a negative discount rate, which gives a higher importance to impacts occurring in the future, provides the highest results. The difference between impacts with a small positive and a small negative discount rate is about 5 orders of magnitude, highlighting the importance of the choice of discount rate, which relies on subjective criteria. The second lowest overall impact is produced by the inclusion of emissions only above the background concentrations. This is because the pattern of the emission profile for Ni shows a sharp increase in the leached metal only after 35,000 years and exceeding the background Ni concentration after 55,000 years approximately.

A similar pattern would result from the comparison of the methods for emissions of other metals, as only the magnitude would change (based on different emission levels, CFs and background concentrations). Therefore, it is safe to say that the cumulative effect of the choice of LCIA approach towards long-term emissions from all metals leaching out of the landfill would be much higher.

If the results from this comparison are put in the context of a full LCIA, the selection of method becomes critical as it would potentially lead to very different decisions supported by the corresponding LCA study. For example, the infinite time horizon method would lead to a dominance of toxicity impact categories over the full LCIA profile (Hauschild et al. 2008).

6 Discussion

The prediction of the development of emissions over time for heavy metals in landfills is shown to be difficult due to hindering factors such as (i) the complexity and dynamics of landfill chemistry where many processes and the waste characteristics (waste composition) influence heavy metal mobility, (ii) many different parameters (e.g. pH, organic matter in the deposit) that govern metal kinetics and (iii) the inherent uncertainty when attempting to predict future conditions in landfills determining the chemical behaviour for very long time periods. Due to this complexity, there is little common ground among the different methods reviewed. No study has been identified that has made comprehensive use of all information available regarding the governing mechanisms of metal mobility—instead, the studies tend to focus on a selected

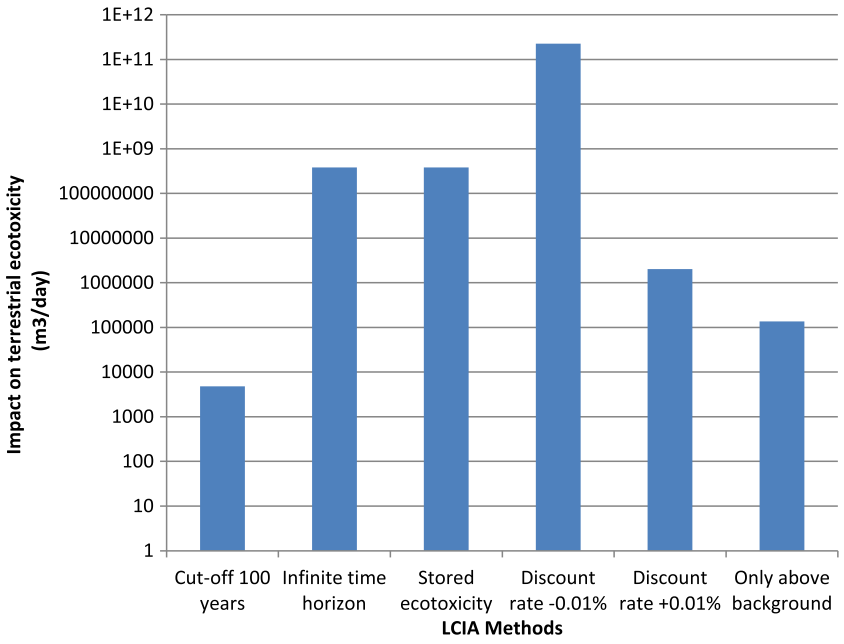

Fig. 2 Assessment of the terrestrial ecotoxicity impact for a given emission pattern of Ni leaching out of an APC residue landfill, applying five different approaches towards handling of long-term emissions

few of these mechanisms and adopt them in the construction of a prediction model.

In the LCA domain, the use of predictions of long-term emissions is limited, as the current state of the art either uses emissions from robust models or laboratory findings that extend to the first 100 years after deposition or uses mass balance calculations to estimate the aggregated emissions over infinite time horizons. The proposed approaches to account for potential impacts from long-term emissions present some interesting insights but carry individual, inherent problems. An overall conclusion is that a distinction in the way potential impacts are calculated for short- and long-term emissions is required.

None of the approaches considers the changes in environmental conditions or ecosystem equilibrium when estimating future potential impacts. The CF used is the same for short- and long-term emissions, although the environmental compartments receiving long-term emissions are bound to change if such long time perspectives are investigated. Currently, the predominant approaches for integrating emissions over time do not allow for a time-differentiated CF, although that would increase accuracy. For example, fluctuations in the soil pH would change drastically the metal mobility (Richards et al. 2000), increasing or decreasing their toxic potential. The presented dynamic inventory allows for investigating impacts in every future point in time and paves the way for investigations regarding changing over time soil properties that influence the calculations of metal toxicity.

The comparison of existing LCIA approaches on the same metal emission profile shows that the resulting predicted potential toxic impacts differ quite substantially, thus leading to very different assessments of the toxicity potential of landfill leachate. The common practice in LCA, of cutting off emissions after 100 years, is problematic as it ignores the vast majority of potential impacts, regardless of the selected approach towards long-term emissions as shown in Fig. 2 and supported by other studies (Huijbregts et al. 2001). However, given the importance of heavy metal emissions in the total toxicity-related impacts, it is essential that a robust, accurate and realistic approach is developed so that properly informed decisions can be made based on the LCA results. A robust approach needs to include an accompanying module on uncertainty in both the prediction of emissions (LCI) and the subsequent toxicity modelling (LCIA), which, although very complex, is essential for putting the impact assessment results into context and avoiding their over-interpretation.

In order to produce an accurate representation of potential impacts from long-term emissions, the changes in the receiving environment need to be investigated and possibly reflected in a time-dependent toxicity CF for each metal being emitted. This should be based on a modelling of the temporal changes in landscape properties over long-term time frames of the environmental compartments receiving the emissions (mainly soil) and the response of the exposed ecosystems, which would result in time-dependent fate, exposure and effect calculations. Site specificity is also crucial, as soil properties that affect metal toxicity, such as pH or soil organic matter, vary across the globe.

On the other hand, the handling of long-term emissions should be realistic, so that only real potential toxic impacts are taken into account. The inclusion of background concentrations (Birgisdóttir 2005) in a comparison with the emission concentrations would disregard emissions that actually dilute existing background levels in the environment. These considerations should reflect the fact that metal emissions from

landfills occur over long time frames but in relatively low concentrations. These low concentrations are not currently considered, due to the predominant practices of integrating emissions over time. The evolution of background levels with time can be addressed by modelling different scenarios (Hellweg et al. 2005). As the background levels have a significant effect on the amount of emissions contributing to toxicity, a spatially differentiated approach is required in order to capture the natural variations in soil concentrations of metals in different regions (e.g. Shacklette and Boerngen 1984; Herselman et al. 2005; Lado et al. 2008).

Some of these method elements may challenge traditional axioms of LCA specifically and sustainability in general. The time equity principle, essential to the sustainability concept aiming to preserve equal development chances for current and future generations, according to which all emissions from a system should be treated equally regardless of the point in time at which they occur, is one of them. This principle functions well in relatively short time frames (1 day, 1 year or even a decade) since the internal and external conditions that determine the potential impacts from an emission do not change substantially. However, in extreme conditions, when emissions occur over a very long time frame, the time equity principle cannot hold, since the conditions determining the estimation of the impacts are bound to change. Therefore, emissions occurring in different time periods should be treated differently in an LCA context, by modifying accordingly the associated CFs. Some existing approaches in LCIA already challenge the time equity principle by explicitly applying discounting in future impacts or differentiating between impacts within a surveyable time period and future impacts (Hellweg et al. 2003; Hauschild et al. 2008). Likewise, the common practice in LCA to ignore impacts from emissions occurring after a specific time frame, without adequate scientific argumentation, clearly violates the time equity principle.

On the other hand, the involvement of background concentrations is not consistent with the general rule in LCA of treating emissions in the same way whether above or below thresholds (Udo de Haes et al. 1999). However, such an approach is necessary not only for producing more realistic future estimates for potential toxic impacts but also for addressing the fact that long-term emissions typically emit in low concentrations but over very long time periods. Overall, it is time a consensus is reached for the issue of long-term emissions in LCA. The complexity of emission prediction, the diversity of approaches and the inherent uncertainty lead most practitioners to ignore long-term emissions altogether. On the other hand, policymakers tend to undervalue or ignore future potential impacts due to pure time preference (Harvey 1994), which is not supported by environmental ethics (Hellweg et al. 2003). These two factors cause a significant misrepresentation of potential toxic impacts in LCA studies involving long-term emissions of metals.

Compliance with ethical standards Our research does not involve any human or animal participants.

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