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Review

A perspective on LCA application in site remediation services: Critical review of challenges

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ABSTRACT

The remediation of contaminated sites supports the goal of sustainable development but may also have environmental impacts at a local, regional and global scale. Life cycle assessment (LCA) has increasingly been used in order to support site remediation decision-making. This review article discusses existing LCA methods and proposed models focusing on critical decisions and assumptions of the LCA application to site remediation activities. It is concluded that LCA has limitations as an adequate holistic decision-making tool since spatial and temporal differentiation of non-global impacts assessment is a major hurdle in site remediation LCA. Moreover, a consequential LCA perspective should be adopted when the different remediation services to be compared generate different site's physical states, displacing alternative post-remediation scenarios. The environmental effects of the post-remediation stage of the site is generally disregarded in the past site remediation LCA studies and such exclusion may produce misleading conclusions and misdirected decision-making. In addition, clear guidance accepted by all stakeholders on remediation capital equipment exclusion and on dealing with multifunctional processes should be developed for site remediation LCA applications.

Contents

1. Introduction

In urban and industrial areas there are many sites used for industrial purposes that are now underutilized or derelict. These sites, generally labeled as brownfields, are often contaminated or contain infrastructures. The environmental impacts of brownfields can be attributed to the site's degraded physical state, which results in potential risk to human health and ecosystem quality, and to the fact that the sites are economically inactive, which results in the loss of available land for development and hence increases development pressure on peripheral land [1]. Site remediation activities support the goal of sustainable development, however, such activities have their own economic, social, and environmental impacts. For a remediation service to be truly considered "sustainable"

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its impacts should not surpass the benefits of decontamination [1,2].

Life cycle assessment (LCA) is gaining widespread acceptance in order to support environmental decision-making. LCA enables the inventory and quantification of environmental interventions and related impacts over the whole life cycle of a product, process or activity. In the site remediation decision-making, LCA can help in choosing the best available technology to reduce the environmental burden of the remediation service or to improve the environmental performance of a given technology. However, LCA remains a fairly new tool as the methodology is still under development and therefore LCA, though scientifically valid, still needs to depend on several technical assumptions.

In this paper, a critical review of challenges concerning the LCA application to site remediation services is presented. This review article discusses existing LCA methods and proposed models focusing on critical decisions and assumptions of the LCA framework for site remediation activities.

2. LCA framework

The structural and procedural components of LCA are determined by the international standard series ISO 14040–43 [3–6]. The mandatory initial steps include the definition of goal and scope, functional unit and the system boundaries, which are crucial to establish the context in which the evaluation is to be made [7]. The ensuing steps are inventory analysis, impact assessment and interpretation. The life cycle inventory analysis (LCI) examines and compiles all relevant environmental interventions, i.e. land transformation and occupation and energy and material inputs and outputs, of processes during the life cycle of a good or a service, hereafter called product. The life cycle impact assessment (LCIA) is carried out to translate the collected emissions and consumptions into environmental and/or health effects and is commonly expressed by representative impact category indicators. Finally, within the interpretation phase, the results of inventory analysis and impact assessment are discussed to extract the environmental hot spots and to derive recommendations.

Some of these steps relate to certain points within the LCA process where either the LCA practitioner must make a decision about how to proceed or a choice must be made regarding methodological unknowns [8]. The former case relates to decisions where there is no single correct way to proceed in a given LCA study. For example, to set the functional unit of a product system is essentially a decision point and clearly a matter of choice, although the following stages of the LCA should be conducted in a consistent way with this choice. The second type of issue occurs where variations in current practice exist and the LCA community has not yet established a clear guidance, such as the choice of impact category assessment models. Often these variations in methodology are described as "assumptions" and are recognized to have a great impact on the study results [8,9].

In the present study a review of past LCA studies in site remediation context is undertaken, focusing on critical decisions and assumptions of the LCA framework. The discussion is divided among the following four topics:

(a) Goal definition.
(b) Functional unit.
(c) Scope and boundaries (cut-off rules and exclusions).
(d) Taxonomy of impact categories and models.

In the next sections, it is discussed how these crucial LCA elements have been adopted in site remediation LCA literature.

3. Goal definition

The first step in an LCA is to clearly establish the goal of the study. Once the goal is defined, the scope and boundaries for the study can be drawn in a consistent manner with the goal. For example, if the goal is to assess the regional impact of a site remediation system service, then data should consider the regional average, rather than the national average. Presently there is no detailed guidance on how to match the goal with the study in LCA applications [8].

At this point two very distinct categories of LCA goals exist: the LCA practitioner can conduct an "attributional" or a "consequential" study. Rebitzer et al. [10] proposes the term "attributional LCA" to denote a description of a product life cycle and the term "consequential LCA" to denote a description of the expected consequences of a change in the product life cycle. The distinction between these two types of LCA has important consequences for the way the product system should be further modeled.

In site remediation LCA literature the impacts related to the site's physical state are labeled "primary impacts", and the impacts associated to the remediation service are labeled "secondary impacts". Recently, Lesage et al. [1] proposed the label "tertiary impacts" to define the environmental impacts associated with the effects of the post-rehabilitation fate of the site. The purpose of an attributional LCA is to provide information concerning the environmental properties of an investigated life cycle and of its subsystems. For example, the LCA of a remediation service system may consider only the primary and secondary impacts, not including the effects of the remediation on other stages of the site's life cycle. On the other hand, the joint quantitative assessment of primary, secondary and tertiary impacts are interrelated in a consequential manner. Lesage et al. [1] identifies three interrelated decisions in the scope of site remediation:

(a) The choice of objectives regarding the physical state of the site (e.g., the clean-up target).
(b) The choice of objectives regarding the fate of the site (e.g., if and how it will be redeveloped).
(c) The choice of means to meet these objectives (e.g., the choice of remediation technology).

In LCA goal definition careful attention must be paid to these interrelated choices. For example, the clean-up target determines the fate of the site, which displace alternative site management options (e.g., housing). That is, the residual contamination may lead to the indefinite mothballing of the site with minimized exposure, or may allow the site's residential development. In this case, the LCA should provide information on the environmental consequences, i.e. tertiary impacts, of the remediation service system. The processes where the most important consequences occur should therefore be included in the LCA system's boundary. Moreover, changes in the physical state of the site may affect indirectly the use of a wide variety of sites since rehabilitated brownfields compete directly with suburban greenfields. The LCA practitioner must decide whether to include or not on the scope of the study the environmental assessment of such consequences.

The great majority of past LCA studies in the field can be described as attributional LCA's. The scope of those studies is usually based on the environmental comparison between alternative remediation technologies towards a clean-up target or an exposure threshold. Tertiary impacts have been systematically excluded even when the compared remediation technologies generate different physical states of the site (e.g., soil sealing *versus* decontamination) that subsequently may lead to different uses of the site [7,11–20]. The inclusion of tertiary impacts would introduce a greater complexity on the LCA models, but the potential significance of those impacts was not assessed in the referred studies.

The work of Lesage et al. [1] is a first attempt to develop a consequential LCA methodology. The methodology is limited to the case where a contaminated urban site resulting from industrial occupation is rehabilitated for residential redevelopment. This consequential LCA approach was further compared with an attributional approach in terms of conclusions directed to decision-making [21]. The site rehabilitation for residential redevelopment scenario involved excavation and landfill, infrastructure material recycling and site backfilling. This scenario was compared with an "exposure minimization" consisting in covering the site with clean soil and subsequently idling the site. The attributional LCA results showed no clear preference for either option because of trade-offs between the benefits of decontamination and the impacts of the rehabilitation system service. On the other hand, the consequential LCA supported rehabilitation if it is followed by residential reuse, as long as development of suburban sites is avoided, because tertiary environmental benefits dominated other types of impacts. The authors conclude that when site intervention decisions can affect the fate of the site, the scope of the study should be expanded to include tertiary impacts.

The methodology is limited to residential redevelopment of brownfields; other contexts of redevelopment still need to be set up. In addition, the tertiary impacts of brownfield rehabilitation depend on the type, context and location of other sites that are affected by the rehabilitation [1]. The methodology was simplified in considering only vacant urban sites and suburban greenfields. Moreover, the evaluation of tertiary impacts is subjected to a high data uncertainty. The authors note that (i) the identification of which type of sites can be marginally affected is inherently region-specific; (ii) it is difficult to determine the types of houses and public infrastructures that are situated at these sites; (iii) the data on price elasticity of housing services is quite uncertain.

In terms of the decision-making system, the joint assessment of these three types of impacts translates in a decision support where a wide range of stakeholders are involved, each with their own and potentially conflicting perspectives and objectives [22]. The different stakeholders and decision-making contexts will place a different importance on each of the aforementioned types of impacts. A framework for comparing brownfield management options was proposed by Lesage et al. [22] in regard to these types of environmental impacts and for interpreting the results from different perspectives.

4. Functional unit

The definition of the functional unit is a critical step in LCA because it determines the reference flows and dictates the upstream and downstream process alternatives to be included in the study [8]. Presently there is not clear guidance on how to specify a system's functional unit. The ISO standards only require that the functional unit is dependent on the goal and scope of the study and that it is clearly defined and measurable.

When an LCA is used to compare alternative products the basis of comparison must be equivalent use. That is, each system should be defined so that an equal amount of product or equivalent service is delivered. For example, beverage containers may be delivered in a variety of sizes having different life cycle characteristics. Subsequently, a comparative assertion LCA study might consider 1000 l of beverage as an equivalent use basis. Thus, in order to measure the environmental interventions of a product, the inventory data for a system must be mathematically normalized to a functional unit, which has to be set *a priori* and is not a decision variable.

In site remediation LCA, Diamond et al. [23] recommend that "the functional unit should relate to the production of an amount of treated soil". The majority of the site remediation LCA studies consider, as the functional unit, the treatment of an amount of soil or groundwater towards a site-related impact metrics, usually a given regulatory criterion [7,11,13,14,18,20–22]. Accordingly, a typical function unit in site remediation LCA studies would be "the legal legacy contamination of 1 ha of treated soil". The estimation of final mass or volume of remediated soil may be a point subjected to uncertainties, because accurate estimates would be obtained for options involving soil excavation, but these values are likely to be uncertain for *in situ* treatment or containment options, or for the no-intervention scenario [12].

Moreover, the functional unit results in a relative comparison, so that LCA provides no estimate of the absolute mass of releases. The absolute magnitude of the service delivered is generally considered irrelevant because the system is assumed to exhibit linear behavior [24–26]. That is, the relative differences in LCA are assumed not to change per product unit when calculated for 1000, 10,000, or 100,000 product units. However, primary impacts (e.g., ecotoxicity and human toxicity) are non-linearly dependent from the pollutants mass load and simply relate them to a functional unit of the remediation system service may produce flawed results for different scale factors. In this trend, some authors, who included the assessment of primary impacts in their studies, defined as the functional unit the total amount of treated site toward a regulatory criterion or some other contamination level [7,12,19], although the non-linear dependency from pollutants mass load was only considered when risk assessment (RA) models were applied to evaluate primary impacts [12,19].

The choice of the time horizon is very important when comparing remediation system services. For example, thermal treatment has a short but intense impact while alternative options may have milder but longer impacts (e.g., landfilled soil) [27]. Some authors included the time horizon in the functional unit, disregarding eventual potential environmental impacts beyond the defined time period [7,17]. This issue is further discussed in Section 5.2.

5. Scope and boundaries (cut-off rules and exclusions)

The scope definition establishes the main characteristics of an intended LCA study, covering issues such as the temporal, geographical and technology coverage, the mode of the analysis, and the overall level of sophistication of the study [28]. The scope of a product system can be limited to include only processes that make relevant contributions to the environmental flows. The further inclusion of processes that are inputs to other processes (i.e., background processes) may not be considered if their contribution is found to be insignificant. However, determining significance can be an arbitrary procedure. To ensure that such exclusions do not affect the accuracy or the application of the results, one must quantify the input or output in question in order to determine its relevance [8].

The level of detail of the inventory analysis depends on the purpose of the study and on the size of the systems. Large systems that include several industries may exclude specific details found not to be significant, e.g., computer use in consultancy services. Moreover, the lack of inventory data may be a consideration in defining the product system, although such constraints should not compromise the scientific basis of the study [8].

5.1. Spatial coverage

The geographic boundary is central to the original intent of using the LCA methodology for the environmental assessment of site remediation. This is because activities associated with the remediation do not take place only on the site but also may take place in the region (e.g., extraction of backfill from a quarry) or in the world (e.g., crude oil extraction for the production of diesel to fuel the heavy machinery used during remediation) [22]. Moreover, the environmental impacts that are relevant can be of a local scope (e.g., ecotoxicity impacts due to the site's residual contamination), of a regional scope (e.g., acidification associated with NO_x emissions from heavy machinery) or global (e.g., climate change due to greenhouse gas emissions). In short, LCA allows for consideration of activities at and beyond the contaminated site itself and addresses environmental impacts that can occur at a global, regional, and local level.

Generally, in life cycle inventories, emissions are summed up per pollutant regardless of their geographical place of occurrence, and therefore the inventory outcome lacks any retrievable relation with a particular region [29]. Consequently, the local and regional scales are not used in the calculation of pollutant characterization factors. It is recognized that the lack of spatial dimensions in inventory data is a point of uncertainties relatively to impact results [3,30]. This concern is especially relevant in primary impacts assessment since they are inherently site-specific. Impact assessment models of non-global cumulative impact categories may therefore take account of geographic differences in fate factors, background levels and sensitivity of the pollutant-supplying or pollutant-receiving environment. This issue is further discussed in Section 6.

5.2. Temporal coverage

The goal of LCA is to aggregate and assess environmental interventions independent of when and where they occur. A theoretically complete life cycle system would start with all raw materials and energy sources in the earth and end with all materials back in the earth or at least somewhere in the environment but not part of the system [31]. Defining a different system boundary represents a decision to limit the product system in some way. Constraints on cost, time, or other factors may argue in favor of a more limited boundary, but a too limited boundary may exclude consequential activities or elements (see Section 4). Main guidelines allow limitations in the time frame that is applied for emissions [3], but the LCA community maintains that life cycle assessment is a tool for sustainability assessment and therefore must incorporate the principle of temporal justice.

In site remediation consequential LCAs the environmental interventions of site's fate are a function of time. According to Lesage et al. [1] time should ideally reflect the total life-expectancy of the new houses (in case of the site's residential development). Lesage et al. [21,22] assumed 40 years of residential occupation of the rehabilitated site. However, the choice of this type of value is quite arbitrary, and hardly has a real relation to the life-expectancy of house services [1,32].

In the past attributional LCA studies the time horizon is usually equal to the rehabilitation time, which may vary widely. For example, rehabilitation by "excavation and landfill" is usually much shorter than one involving *in situ* bioremediation [1]. In this trend all the environmental interventions identified in the inventory analysis for the rehabilitation time are normalized to the functional unit. For example, Toffoletto et al. [17] defined a 2-year period of biopile treatment of a diesel-contaminated site towards a regulatory criterion. Cadotte et al. [19] assessed four rehabilitation scenarios for a diesel-contaminated site using an evolutive LCA, in which the environmental assessment was performed on a yearly basis ending in the longest rehabilitation time scenario (300 years for natural attenuation).

The leaching of contaminants of landfilled soil or of the rehabilitated site itself, due to residual contamination, can endure for thousands of years if toxic metals or highly persistent organic compounds are present. In site remediation LCA literature, the long-term emissions of contaminants that were not degraded or contained are disregarded, which may be unacceptable to many stakeholders considering the quantities of toxic substances that can be present. Lesage et al. [21] assumed a time horizon of 44 years in assessing an excavation and landfill treatment scenario of a contaminated site with metals, petroleum hydrocarbons (PHCs), and polycyclic aromatic hydrocarbons (PAHs). The authors state that this time horizon is too short for long-term emissions from landfills to be significant, enhancing the environmental benefits of the remediation system service. Diamond et al. [23] developed a life cycle framework to address burdens associated with contaminated sites and issuing from remediation activities, suggesting 25 years of time horizon. Page et al. [12] examined a lead-contaminated site remediated by excavation and landfill and also used 25 years although the authors state that was a too short time horizon. Bender et al. [11] and Godin et al. [7] used 50 years.

The basis in which the time horizon is defined in the above-mentioned studies seems quite subjective. It is recognized in the literature that the exclusion of long-term emissions may have a relevant influence on the outcome of an LCA study [33–36]. Furthermore, Finnveden and Nielsen [33] go so far to state that the time horizon should be possibly extended to the hypothetical infinite.

The assessment of long-term emissions in LCA is problematic since such processes are unsteady-state (i.e., vary with time). Thus, non-linear methods to project inputs, outputs and impacts should be used at the expense of increasing the impact assessment complexity. Moreover, the uncertainties in predicting those emissions in the distant future are very large [37]. In order to make LCA sizeable and manageable various approaches of temporal cut-off rules to be applied in life cycle inventory of long-term emissions are reported in the literature:

(a) One approach is to introduce a separation between near-term emissions and long-term to infinite emissions. The short-term period is often referred to as the surveyable period, set at 100 years or similar, usually interpreted as the follow-up or operating time of a deposit or landfill site [36]. The LCA practitioner may choose to neglect emissions beyond this stage [38,39], although this approach will often be unacceptable to many stakeholders since only a very small fraction of the toxic substances in, for example, a landfill may be emitted in the foreseeable time horizon, and the largest potential for toxic releases escapes the assessment [33]. Alternatively, the LCA practitioner may have the emissions aggregated and interpreted separately [40–42]. In this trend, Hauschild et al. [37] state that the uncertainties in modeling long-term impacts are so large that it is meaningless to apply some average situation and try to model long-term emissions. Instead, the authors propose a new impact category called 'stored toxicity' by using the same characterization factors as for the conventional ecotoxicity and human toxicity impact categories.
(b) Another approach is to assess long-term release potentials which may be based on mass or time. In the former case, the total mass of the residual contaminant or the total mass initially placed in the landfill is presented as a midpoint impact category [43]. If they are based on time, all releases may be neglected if they occur after, for example, the expected time of the next ice-age [44], or, alternatively, a critical time period may be introduced at which all significant effects are considered to be completed [45]. Another time-based approach is to base the cut-off for long-term releases on the resulting exposure concentrations. That is, by assuming the continuously decreasing leachate concentrations, releases occurring after a set threshold leachate concentration, which may be based on inert waste cri-

teria, background concentrations or effect-based thresholds, are then excluded from the emissions inventory [36]. This approach requires knowledge of the leaching kinetics which is presently not available for many of the toxic metals and persistent organic compounds, particularly under the changing conditions of a distant future [37].

(c) A third approach is to use discount rates to reduce the significance of long-term impacts by introducing a weighting between future and present impacts. This approach contradicts fundamental ethical values by imposing environmental damages to future generations based on pure time preference [35].

The relevance of long-term emissions on an LCA output depends on the scope of the study, but simply to exclude them *per se*, as hitherto observed in site remediation LCA literature, is quite an arbitrary procedure. However, no common approach has emerged yet on how to address long-term emissions and even such schemes are inherently based on subjective valuations of future potential environmental impacts. In short, the definition of the temporal coverage is problematic since the exclusion of long-term emissions may represent relevant uncertainties in the outcome of a LCA study for scenarios entailing actions with long-term emissions. In addition, the choice of a scheme to address such emissions is still a subjective assumption.

5.3. Capital equipment and infrastructure exclusions

The inclusion, or exclusion, of capital equipment has been a subject of debate among the LCA community. Capital equipment includes the buildings and machinery that are needed to produce the product under analysis. The common practice is to exclude capital equipment in an LCA [8]. Generally it is assumed that an item, such as heavy equipment or a building, has such a long lifespan that its contribution to the LCA would be insignificant after being apportioned by its years of use or by the number of products it produced.

In site remediation LCA literature the capital equipment and infrastructure was systematically excluded from the system boundaries. The main pointed reason is that the remediation equipment (e.g., a bulldozer) can be reused on other product systems so its significance is assumed to be negligible (e.g., [19]). However, Frischknecht et al. [46] studied the environmental relevance of capital equipment in LCA of goods and services. The findings of their study shows, for example, that major or significant contributions of capital equipment and infrastructure to impact categories, such as mineral resources, land use or terrestrial ecotoxicity, were observed in processes such as residual material landfill, construction materials, and waste incineration. The construction of materials can be relevant, for example, in stabilization/solidification remediation techniques, and waste incineration can be a significant process in thermal treatment techniques. Of course, this potential relevance depends on the relative importance of those foreground processes to the overall product system. Nevertheless, the work of Frischknecht et al. [46] suggests that capital equipment and infrastructure cannot be excluded *per se* without solid proof. Heijungs et al. [47] proposes an initial indicator of the relative importance of capital equipment and infrastructure based on the cost of its maintenance and depreciation. If these costs are a substantial part of the good or service price, the environmental impacts of capital equipment and infrastructure should not be excluded *a priori*.

5.4. System expansion and allocation in open-loop recycling

As discussed in Section 4, the product system should be expanded if the remediation system service imposes a consequential environmental pressure outside the initial system boundary. To limit the system boundary in order to not include the consequences of remediation system service may produce misleading conclusions and misdirected decision-making.

The system expansion can also be applied when a process within the product system is multifunctional, that is, it delivers more than one good or service as exported functions. The boundaries are expanded to include the alternative production of a co-product. Therefore, a necessary requirement of system expansion is the existence of an alternative way to produce that co-product. For example, if a given material is removed from the site and recycled back into the economy, such as cement, metals, or even treated soil as a material for downstream products (e.g., construction materials), the product system may include an alternative way of producing that material as an avoided burden. In short, the main-product 'A' generates a credit equal to the credit saved by not producing the material that the co-product 'B' is most likely to displace.

The recycling of materials into other product systems is usually out of the scope of the most of LCAs in site remediation, but at least one exception exists. Lesage et al. [21,22] assumed that the recovery and recycling of cement and bituminous concrete of a site remediation service displaces an equivalent amount of crushed gravel production, and consequently the authors further expanded the system to include such process as an avoided burden. The major difficulties is that not only does system expansion require more data to be collected but also it can be difficult to find exact substitutes to co-products. Moreover, the LCA practitioner can face multiple choices for co-products replacement, which can provide an arbitrary range of credits to be assigned to the primary product. It is clear that this choice may significantly influence the final results.

The LCA practitioner may face the dilemma of avoiding system expansion by applying an allocation method when dealing with multifunctional processes. Allocation can be defined as the partitioning or assignment of material inputs and environmental releases of a system to the functions of that system in proportionate shares. The choice between an allocation method and system expansion may be arbitrary since the LCA community has not reached a consensus on how to deal with multifunctional processes [48]. Such an arbitrary choice in an LCA study may significantly influence or determine the final results [49].

In attributional LCA applications another allocation problem arises, whereas the burdens of remediation must be allocated to the "legacy contamination" function and "land production" function. The exported land production function has been excluded in the past LCA attributional studies. Lesage et al. [1] observed that this allocation could be based on the economic value of the two functions of the rehabilitation.

6. Taxonomy of impact categories and models

The selection of impact categories and models is one of the major sensitive issues in LCA studies. A number of impact assessment methodologies are available to the LCA practitioner which includes a model for each impact category. LCIA methods include traditional impact categories at global level, such as global warming, and at regional and local levels, such as acidification and ecotoxicity. It is recognized that different goals and scopes require different impact categories, data sets, and impact assessment models [3,8,50]. The choice of impact categories is a subjective issue and therefore there might not be a consensus on impact categories to assess. Guidelines on how to match the goal and scope with the choice of impact categories are available in the literature [28,51].

In site remediation service systems, primary impacts occur mainly on a local level while secondary and tertiary impacts can occur at different levels. There is a general consensus on how to model the global level impacts (climate change and ozone depletion), but impact models that occur at a regional level (acidification, photochemical formation, aquatic and terrestrial eutrophication) and at a local level (human toxicity and ecotoxicity) vary greatly [8,52]. Table 1 shows the impact categories and models applied in site remediation LCA literature.

6.1. Regional impacts assessment

The variation on assessment models of regional impact categories relates mainly to their capability to deal with spatial dimensions. Emissions can be simply aggregated per pollutant regardless of their geographical place of occurrence, or site-dependent pollutant characterization factors obtained by more detailed models may be used. The former case results in an inventory outcome that lacks any retrievable relation with a particular region and consequently, the local and regional scales are not used in the calculation of fate factors [29]. As referred in Section 5.1, the lack of spatial dimensions in the inventory data used for impact assessment introduces uncertainty in impact results. In site remediation LCA studies the assessment of regional impacts with only a quantification of emissions was applied by Blanc et al. [15] and Harbottle et al. [18]. Generic characterization factors were applied by Volkwein et al. [13] and Bayer and Finkel [16].

Most of the site-dependent LCIA models are related to specific geographical contexts, such as Western Europe (e.g., EDIP97, IMPACT2002+), United States (e.g., US EPA TRACI), or Japan (e.g., LIME). This limitation led to the application of site-specific factors that are not completely appropriate to the geographical context of some site remediation LCA studies [7,17,19,21,22].

There is currently a trend towards making regional impact assessment models on acidification, eutrophication (aquatic and terrestrial), and photochemical oxidation, more site-dependent, providing pollutant characterization factors for other regions than Western European countries or the United States, or even for regions within these countries (e.g., [53–59]).

6.2. Local impacts assessment

The assessment of ecotoxicity and human toxicity impacts is particularly problematic in site remediation service systems because they are inherently site-specific. The site-specific conditions have a dominant influence on the behavior of contaminants in the subsurface, and the understanding of this behavior is essential for the assessment of ecotoxicity and human toxicity impacts. Depending on the application, the lack of site-specific data could result in misleading conclusions and misdirected decision-making [7].

The scope of primary impacts is essentially local and their inclusion within the LCA depends on the goal of study. For a comparison of different remediation system services towards a regulatory criterion, the usual practice in site remediation LCA literature is the exclusion of primary impacts assessment (Table 1). The assessment of a no-intervention scenario is out of the scope of the referred studies and, therefore, the potential environmental benefits of remediation services are not evaluated. Moreover, the impacts associated with the residual contamination have been generally excluded leading to an incomplete assessment.

The traditional approach in LCIA of ecotoxicity and human toxicity is to sum up the emissions per pollutant regardless of the environmental receptors media and the exposure pathways. This aggregation carries a fundamental assumption that these emissions can be combined into a single effect and one overall, simultaneous exposure, or based on simplified assumptions regarding the contaminants transport and fate among environmental receptors and exposure pathways [14,17,19–22]. Owens [24] reported that the combination of no threshold, linear dose response, simultaneous exposure, and additivity assumptions extend the assessment beyond a worst case scenario to an impossible scenario. This point has been a long controversial issue in LCA (e.g., [60]).

A more simplistic approach is the case when the choice of target values is part of the decision process regarding the remediation service. The residual contamination is simply estimated and the target values presented as a midpoint impact category. In this perspective, some studies compared the residual concentration with the national target or limit values [11,13].

It was early recommended that the complete assessment of ecotoxicity and human toxicity impacts should be based on risk assessment or even to integrate RA in the LCA process [24,26,61,62]. The speciation of pollutants among environmental receptors is difficult to model in LCA since detailed information is required about spatial parameters, such as infiltration rate, macropore flow, pH value, content of organic material, and distance to the groundwater. This information is not available without an elaborate analysis of the soil, which is generally beyond the scope of a LCA but it is a component of RA. The standardized RA procedure identifies threshold contaminant concentrations for adverse effects on ecosystems and human health, and examines the fate and transport of those contaminants along source-to-receptor pathways. A review of RA models applied to contaminated sites was undertaken by Bardos et al. [2]. Even though LCIA can use the models and the methodologies developed for RA, LCA is designed to compare different products and systems and not to predict the maximal risks associated with single substances [63]. However, some authors applied RA in order to assess primary impacts in site remediation LCA studies. Page et al. [12] used the Mackay Level III multimedia RA model [64–66] in order to assess the toxicity burden of a contaminated site with lead after an excavation and landfill treatment. Godin et al. [7] applied LCA to evaluate alternative remediation options of a spot lining landfill. The EDIP LCIA model was used to assess local impacts and, in order to technically improve the assessment, the procedure included the conceptual modeling of groundwater flow and contaminant transport (cyanide, fluoride, iron, and aluminium), typically a component of RA. Both studies relied on intense site-specific investigations and data. In addition, since RA is oriented toward the absolute mass releases while LCA is a function-oriented tool that assesses impacts for a selected process unit, both studies defined the total volume of the remediated soil as the basis to which the inventory data was collected and the further LCIA of secondary impacts was performed. Although these efforts, an integrated RA/LCA framework is yet to be developed for site remediation system services. The SETEMIP-Environment [67] has highlighted discrepancies between RA and LCA at the conceptual level and has draw general recommendations for the development of a decision support tool combining the complementary analyses of RA and LCA for contaminated site management.

LCIA models are still in development and do not yet take all important contaminant-specific properties and processes into account. They can be improved to provide a more meaningful result in human toxicity and ecotoxicity assessment by critically adopting and adapting advanced knowledge and models from RA [63]. Recently, the increasing complexity of LCIA models, such as EDIP2003 [68] and USES-LCA 2.0 [29,69], led to the inclusion of characterization factors considering the fate of pollutants by applying multimedia and multiple pathway exposure models. These impact assessment models address environmental receptors such as freshwater, marine, and terrestrial but they fail to address emissions to deep soil layers and the groundwater. These environmental media are therefore still neglected in LCA applications [34].

Hellweg et al. [70] proposed a procedure for estimating heavy metal transport in soil taking into account the most relevant

Table 1
Impact categories and models applied in site remediation LCA literature.

LCA study	Remediation services	Contaminant	Impact assessment					
			Primary	Model	Secondary	Model	Tertiary	Model
Bender et al. [11]	• Groundwater extraction	Petrol	Area balance (housing and ensuring)	LCI data	Fossil resources	Nordic guidelines on LCA		
	• Vapor extraction • Groundwater extraction, activated carbon and air stripping combined with *in situ* bioremediation				Water Land use	LCI data LCI data		
					Global warming	IPCC 1995		
					Acidification	Nordic guidelines on LCA		
					Toxicity	Prüfwerte Baden-Württemberg		
					Photochemical oxidation	CML 1992		
					Odour	Odour threshold values		
					Noise	LCI data		
Page et al. [12]	• Excavation and landfill	Lead	Land use (balance) Residual toxicity burden	LCI data MacKay Level III	Global warming Solid waste burden	IPCC 1995 LCI data		
					Gross energy requirement	LCI data		
					Toxicity	MacKay Level III		
Volkwein et al. [13]	• Onsite insuring	PAHs	Area balance (housing and ensuring)	LCI data	Fossil resources	Nordic guidelines on LCA		
	• Surface sealing with asphalt • Decontamination (excavation and soil washing, microbiological treatment and thermal treatment)				Water Land use	LCI data LCI data		
					Global warming	IPCC 1995		
					Acidification	Nordic guidelines on LCA		
					Toxicity	Prüfwerte Baden-Württemberg		
					Photochemical oxidation	CML 1992		
					Odour	Odour threshold values		
					Noise	LCI data		
Ribbenhed et al. [14]	• *Ex situ* thermal treatment • *Ex situ* bioslurry	Organic substances, mercury and cadmium	Human toxicity Marine, freshwater, sediment, and terrestrial ecotoxicity	USES-LCA USES-LCA	Global warming Ozone depletion	IPCC 1995 Nordic guidelines on LCA		

	• *Ex situ* soil washing				Acidification	Nordic guidelines on LCA
	• *In situ* electrodialysis				Human toxicity	USES-LCA
					Marine, freshwater, sediment, and terrestrial ecotoxicity	USES-LCA
					Aquatic eutrophication	Nordic guidelines on LCA
Blanc et al. [15]	• Offsite landfill	Sulfur			Air emissions (CO_2, CH_4, NO_x, SO_x, Cd, Pb)	LCI data
	• Onsite containment				Raw materials consumption	LCI data
	• Liming				Energy consumption	LCI data
	• Liming and embankment				Water emissions	LCI data
	• Bio-leaching				Waste production	LCI data
Godin et al. [7]	• No-intervention	Spent pot lining	Chronic and acute water ecotoxicity	EDIP97 with a conceptual model of contaminant transport	Global warming	EDIP97
	• Excavation and treatment		Air, water and soil human toxicity	EDIP97 with a conceptual model of contaminant transport	Ozone depletion	EDIP97
	• Excavation and onsite disposal				Acidification	EDIP97
	• Excavation and incineration				Photochemical oxidation	EDIP97
					Chronic and acute water ecotoxicity	EDIP97
					Air, water and soil human ecotoxicity	EDIP97
					Bulk waste	EDIP97
Bayer and Finkel [16]	• Pump-and-treat (activated carbon)	Acenaphthene			Global warming	UBA Umweltbundesamt
	• Funnel-and-gate (activated carbon)				Photochemical oxidation	UBA Umweltbundesamt
					Acidification	UBA Umweltbundesamt
					Aquatic and terrestrial eutrophication	UBA Umweltbundesamt
					Human toxicity	UBA Umweltbundesamt
					Depletion of Energy Resources	UBA Umweltbundesamt

Table 1 (*Continued*)

LCA study	Remediation services	Contaminant	Impact assessment					
			Primary	Model	Secondary	Model	Tertiary	Model
Toffoletto et al. [17]	• Bioremediation: single-use treatment facility *in situ*	Diesel	Chronic and acute water ecotoxicity	EDIP97	Climate change	EDIP97		
	• Bioremediation: permanent treatment center *ex situ*		Air, water and soil human toxicity	EDIP97	Ozone depletion	EDIP97		
					Acidification	EDIP97		
					Photochemical oxidation	EDIP97		
					Bulk waste	LCI data		
					Eutrophication	EDIP97		
					Chronic and acute water ecotoxicity	EDIP97		
					Air, water and soil human ecotoxicity	EDIP97		
Harbottle et al. [18]	• *In situ* stabilization/solidification		Human toxicity	CLEA	Global warming	IPPC 1995		
	• Disposal to landfill		Ecotoxicity	ORNL RAIS	Gaseous emissions	LCI data		
					Raw materials	LCI data		
Cadotte et al. [19]	• Oil removal, natural attenuation and pump-and-treat	Diesel	Ecotoxicity	US EPA TRACI	Global warming	US EPA TRACI		
	• Bioslurping, bioventing and biosparging		Human health cancer effects	US EPA TRACI	Ozone depletion	US EPA TRACI		
	• Bioslurping, bioventing and chemical oxidation		Human health non cancer effects	US EPA TRACI	Acidification	US EPA TRACI		
	• Bioslurping, *ex situ* treatment using biopiles and natural attenuation		Human health criteria	US EPA TRACI	Eutrophication	US EPA TRACI		
					Photochemical oxidation	US EPA TRACI		
					Ecotoxicity	US EPA TRACI		
					Human health cancer and non cancer effects	US EPA TRACI		
					Human health criteria	US EPA TRACI		
Lesage et al. [21,22]	• Excavation and landfill	PHCs, metals and PAHs	Human health	IMPACT2002+	Human health	IMPACT2002+	Human health	IMPACT2002+
	• Exposure minimization with clean soil covering		Ecosystem quality	IMPACT2002+	Ecosystem quality	IMPACT2002+	Ecosystem quality	IMPACT2002+
					Climate change	IMPACT2002+	Climate change	IMPACT2002+
					Resources	IMPACT2002+	Resources	IMPACT2002+
Payet and Gambazzi [20]	• No-intervention	Cadmium, copper, lead and zinc	Human health	IMPACT2002+	Human health	IMPACT2002+		
	• Phytoremediation		Impacts on Water	IMPACT2002+	Impacts on water	IMPACT2002+		
	• Excavation and incineration				Climate change	IMPACT2002+		
					Resources	IMPACT2002+		

processes (sorption, precipitation/dissolution, and surface complexation) in order to derive time and site-dependent ecotoxicity and human toxicity impact characterization factors for heavy metals. The proposed generic procedure serves to classify the mobility of heavy metal cations in a given soil enabling a site-dependent assessment of heavy metal emissions to the groundwater, although a number of simplifications were considered. The mineral composition of the soil and the speciation of metals have only roughly been considered, the influence of temperature has been neglected, and the spatial parameters were assumed to be constant over time. Moreover, the general spatial characteristics of soils were set to Swiss landfill sites and, therefore, default values for other countries, other substances, and other applications than landfills still need to be set up. Nevertheless, the procedure can be adapted to make it compatible to existing LCIA methods.

7. Conclusions

This paper deals with selected issues and challenges regarding site remediation LCA. However, there are other points concerning LCA in general that have not been discussed here, such as the lack of readily available inventory data sources, the lack of data which can be used to assess the potential impacts of the inventory data, the partitioning of inventory data across impact categories in the classification step, and issues regarding interpretation and weighting schemes (normalization, valuation, and uncertainty analysis).

The spatial and temporal differentiation of non-global impacts is a major hurdle in site remediation LCA. It becomes difficult to make a judgment whether or not a given threshold is exceeded, particularly in primary impacts assessment, since LCIA methods make only limited use of spatial and temporal information and therefore the prediction of toxicity effects becomes inherently problematic in LCA. Consequently, the credibility of LCA as an adequate site remediation holistic decision-making tool is affected, considering the relevance that local impacts may have on the outcome of the decision-making process. In order to compare the environmental burdens of different site's physical state scenarios (contaminated *versus* rehabilitated) it is highly recommended to direct the efforts in developing an integrated RA/LCA framework, although the two methodologies are conceptually different since LCA is designed to compare different products and systems, in which impact results are presented as a relative comparison, while RA predicts the maximal risks associated with single substances. The time-scale of assessment may be problematic in scenarios entailing actions with long-term emissions, since LCIA models are based on the underlying assumption of steady state modeling. The integration over time of the concentration suggests an exposure to constant concentration of chemicals, and therefore, depending on the application, the development of dynamic modeling in LCIA may be required if the existing schemes to address long-term emissions produces unsatisfying results in terms of uncertainty. However, LCA is an adequate tool for decision-makers wanting to evaluate the environmental impacts only attributable to remediation activities towards a given legacy contamination.

The assessment of tertiary impacts is also problematic, especially if the further brownfield management option is yet to be specified by the decision-maker, which may depend on the LCA outcome or on other environmental management tools. A number of brownfield management service scenarios (house, commercial, industrial, public infrastructures, or recreation services) should be set, which are associated with important sources of uncertainty. Such scenarios are abstract concepts difficult to measure in reality to which depend the regional specificity of which types of sites are marginally affected and how by the site redevelopment. Moreover, the effort of such a scenario analysis and modeling work would be quite high. A simpler and manageable approach would be the development of generic land use occupation and transformation inventory databases for different brownfield management scenarios. Uncertainty analysis of results may reveal if more regional-specific detail is required and further modeling work is necessary to support the decision at the hand. In addition, clear guidance accepted by all stakeholders on remediation capital equipment exclusion and on multifunctional processes should be developed for site remediation LCA applications.

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