Use of life cycle assessments to evaluate the environmental footprint of contaminated sediment remediation

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ABSTRACT

Ecological and human risks often drive the selection of remedial alternatives for contaminated sediments. Traditional human and ecological risk assessment (HERA) includes assessing risk for benthic organisms and aquatic fauna associated with exposure to contaminated sediments before and after remediation as well as risk for human exposure, but does not consider the environmental footprint associated with implementing remedial alternatives. Assessment of environmental effects over the whole life cycle (i.e., Life Cycle Assessment, LCA) could complement HERA and help in selecting the most appropriate sediment management alternative. Even though LCA has been developed and applied in multiple environmental management cases, applications to contaminated sediments and marine ecosystems are in general less frequent. This paper implements LCA methodology for the case of the polychlorinated dibenzo-*p*-dioxins and -furans (PCDD/F)-contaminated Grenland fjord in Norway. LCA was applied to investigate the environmental footprint of different active and passive thin-layer capping alternatives as compared to natural recovery. The results showed that capping was preferable to natural recovery when analysis is limited to effects related to the site contamination. Incorporation of impacts related to the use of resources and energy during the implementation of a thin layer cap increase the environmental footprint by over one order of magnitude, making capping inferior to the natural recovery alternative. Use of biomass-derived activated carbon, where carbon dioxide is sequestered during the production process, reduces the overall environmental impact to that of natural recovery. The results from this study show that LCA may be a valuable tool for assessing the environmental footprint of sediment remediation projects and for sustainable sediment management.

Introduction

Selection of sediment management alternatives for contaminated sediments is often based on human and ecological risk assessment (HERA) frameworks (*1*). The Grenland fjord in Norway, which is contaminated by polychlorinated dibenzo-*p*-dioxins and -furans (PCDD/Fs), exemplifies this risk based approach for selection of remedial solutions. In this case, capping of the contaminated sediments has been proposed to mitigate risk above the HERA-derived threshold values in fish and shellfish (*2*). The risk-reducing effectiveness of different capping alternatives in current studies is based on the ability to reduce the flux of PCDD/F from the sediments below threshold levels, thus neglecting the environmental footprint of these materials originating from production, use and disposal. As result, energy and resource intensive advanced capping alternatives may be recommended solely based on HERA.

Whereas HERA is suitable for assessing whether the contaminated sediments constitute an unacceptable human and environmental risk, it does not address environmental consequences aggregated over the whole life cycle of the remediation project and from intended future site use. Even though high-end capping alternatives may reduce the risk associated with sediment contamination, the material production and placement necessary for implementing these alternatives, as well as the energy and equipment use they necessitate, may result in environmental hazards that have not been quantified by traditional HERAs. One common way to determine the relative environmental impact between product systems occurring over the whole life cycle is by use of life cycle assessments (LCA). In this method the inputs, outputs and the potential environmental impacts of a product system are compiled and evaluated throughout the product's life span (*3*). In LCA of contaminated sites, impacts have normally been referred to as primary, secondary and tertiary effects (*4*). Primary effects originate from the contamination source, in this case intended effects of reducing PCDD/F uptake in sea food, local ecotoxicological effects on the benthic fauna and physical local impacts of the capping operation. Secondary impacts are the effects related to the use of resources and energy during the implementation of a thin layer cap. Tertiary aspects of the remediation may include increased recreational use of the area or increasing commercial fishing after lifting the dietary notice. However, these tertiary effects were considered to be too uncertain and speculative to be included in the study.

Use of LCA in soil remediation projects has shown that the risks originating from the remediation process often exceed the environmental impacts associated with the site contamination (*5*,*6*). Even though life cycle impacts of environmental management in aquatic ecosystems are gaining interest in both academia and industry (*7*), LCA has rarely been used in sediment management. One explanation may be that LCA was originally developed primarily for land applications and the current impact models are therefore only partially applicable to aquatic conditions.

In this paper we use the Grenland fjord remediation case to investigate the feasibility of using LCA to assess the environmental footprint of contaminated sediment remedial alternatives. Based on the results, we generalize and discuss the possibilities for the future use of LCA in contaminated sediment management.

Materials and Methods

Case description

The contamination in the Grenland fjord area is primarily due to historical industrial activities occurring from 1951 to 2002. The fjords system consists of an inner system (Figure 1, area 0-2) and an outer fjord (area 3-4), separated by the Brevik sill, which significantly reduces the flux of contaminants from the inner to the outer part of the fjord system. The present paper investigates the effect of capping the sediments in the most contaminated inner area of the fjord (areas 1 and 2).

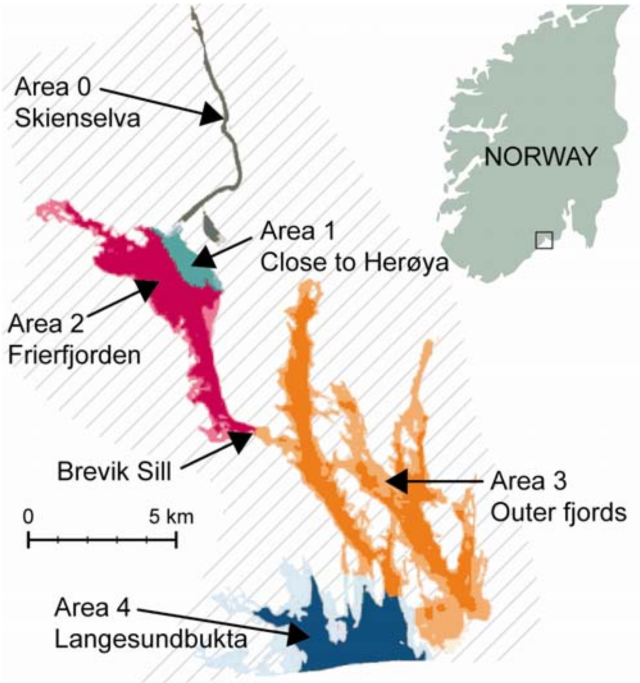

Figure 1 Bathymetric map of the horizontal compartment division in the model application to the Grenland fjords (12). Different colors indicate the horizontal division of five compartments, while the shading within a color indicates the different bottom depth intervals used in the vertical compartment division.

The fate of contaminants has been modeled by using a multi-compartment fate model, linking the abiotic processes describing the fate of chemicals from the sediments into the ecosystem, with the biotic process describing the fate of chemicals in selected marine species (*2*). The performed HERA uses toxic-equivalent-based (TEQ) factors to calculate the risk originating from exposure to PCDD/Fs by expressing concentrations in 2,3,7,8-tetrachloro dibenzo-p-dioxin (TCDD) units (*2*).

Due to elevated levels of PCDD/Fs (app. 200-300 ngTE/kg ww) (*8*) in fish and crayfish above the threshold established by the Norwegian Climate and Pollution Agency, the Norwegian Food Safety Authority has issued a dietary advisory for consumption of fish and shellfish from the area. In the management plan (*9*), sediment capping has been proposed to further reduce the risks associated with sediment contamination. The long-term objective is to remediate the sediment and transition the site to unrestricted use for public recreation and commercial fishing. The model results indicate that capping has to cover a substantial part of the fjord in order to be effective (*2*).

<u>Remediation alternatives</u>

Due to the size of the remediation area, only thin layer capping of the contaminated sediments has been considered as a feasible remediation method (*9*). The use of either passive material to reduce the PCDD/F flux or active carbon containing materials adsorbing PCDD/F (*10*) have been suggested as viable options. An ongoing large-scale pilot project in the Grenland fjord is currently evaluating the feasibility of using this method as a remediation method for the site. In this pilot project three materials are used: locally dredged clay, crushed limestone from a regional source and activated carbon (AC).

The capping materials used in the pilot study are also used in this LCA study with one exception; in the field trials, AC is mixed with clay; however, here AC alone is assumed as a plausible future scenario. Two different sources for the production of AC are also included in this LCA study: a fossil anthracite coal-based product from China and a biomass-derived AC from India utilizing coconut waste as starting material. In the field trial only anthracite AC is used. From a holistic environmental perspective, the biomass derived AC differs from anthracite-produced AC, since it is based on a renewable material. In addition, a net carbon sequestration effect may result from the amendment of the biomass-derived AC to the seafloor instead of its combustion as a fuel (*11*,*12*).

LCA Approach

The LCA investigates the environmental footprint of the active and passive capping materials considered as plausible remediation alternatives and compares them with the footprint of a natural recovery scenario from natural resedimentation. The assessed system can be divided into production, use and disposal phases (Figure 2). The production phase is relevant for passive and active capping materials and relates to impacts from material production, transportation and the capping operation. The use phase includes contaminant release during the phase when the cap will be active in reducing the contaminated flux from the sediments. Impacts in this phase are relevant also for the natural recovery scenario. Public recreational activities and fishing are assumed for all alternatives in the use phase. Impacts related to monitoring the performance of the cap are considered to be outside the scope of this analysis, since it is governed through national monitoring programs independent of remedial strategies. Since the capping materials will eventually be a part of the natural seabed, no environmental impact connected with disposal is foreseen.

The inflow consists of the use of raw materials and energy consumption to produce, transport and apply materials. The outflow consists of emissions to the various relevant compartments: air, water, soil and sediment. Resource use and effects due to the physical impacts of land and sediment use are also addressed in the analysis.

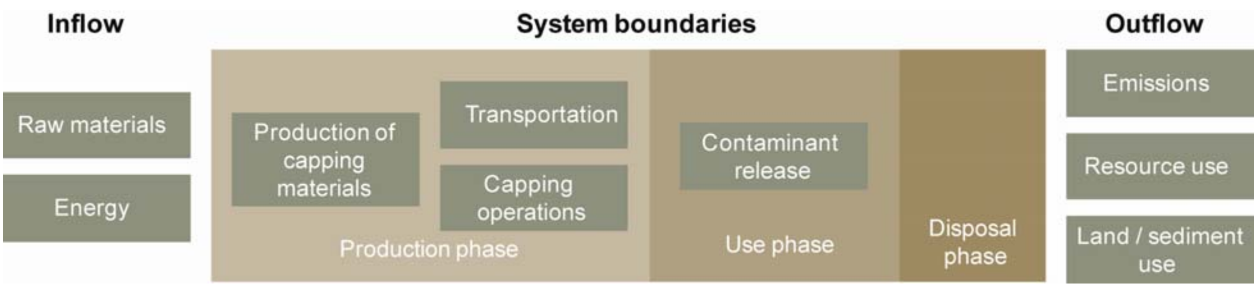

Figure 2 System boundaries for the different capping scenarios assessed in the study. The natural recovery scenario will only have impacts related to contaminant release in the use phase.

Functional unit

Based on recommendations for a life-cycle framework for the assessment of site remediation (*13*), the functional unit is set equal to the remediation of an area of sediments the same size as to the whole inner fjord (23.4 km^2), conservatively assessed for a 90 year time period. This is assumed to be longer than necessary for a successful natural recovery scenario estimated to be approximately 35 years (*2*).

<u>Inventory analysis</u>

The life cycle inventories, i.e. the aggregated environmental data collected for the modeled system, are derived from three main sources. The main source used for the majority of processes is the Ecoinvent 2.2 database. This includes production data for limestone, transport data and energy data. Contaminant fluxes have been calculated with the local fate model using the same settings as in earlier studies (*2*). All production and emission data for AC production, as well as estimates for diesel consumption during dredging and capping, have been obtained from the vendor (Jacobi Carbon. Ragan S and Agder Marine Høyvold P; personal communication 2010). An overview of the inventory data used in the analysis, with reference to their source is given in Supplementary Information (SI), (figure S1-S2 and table S1- S8).

<u>Impact assessment methods</u>

The marine application of LCA has implications on the choice of methodology used to convert the inventory data into information about environmental effects. Marine aquatic toxicity, which is important for this study, is scarcely addressed in available impact models for toxicity (*14*). Sediments, if included in the models, are normally seen as a sink and not as a source for marine contamination. The ReCipe impact model (*15*) which utilizes USES-LCA (*16*) is at present the only readily available impact assessment method that includes a marine release compartment and was therefore selected for this study. The UNEP-SETAC UseTox initiative (*17*) targeted to develop a multimedia chemical fate, exposure, and effect model does not address marine ecotoxicity presently and has therefore not been used here.

An endpoint method was used for the impact assessment in order to achieve maximal agreement with the comparative and management-oriented objectives of the study (Figure 3). Endpoint indicators describe the integrated damage of the components from the inventory, in contrast to midpoint indicators which address effects only. For global warming, a typical midpoint indicator would be the effect of radiative forcing (global warming potential), whereas the endpoint approach would assess the human and environmental damage based on radiative effects. Use of endpoint indicators facilitates the interpretation of results for management purposes and allows integration of results to a single score indicator. However, endpoint indicators are expected to have a higher degree of uncertainty compared to midpoint indicators (*18*).

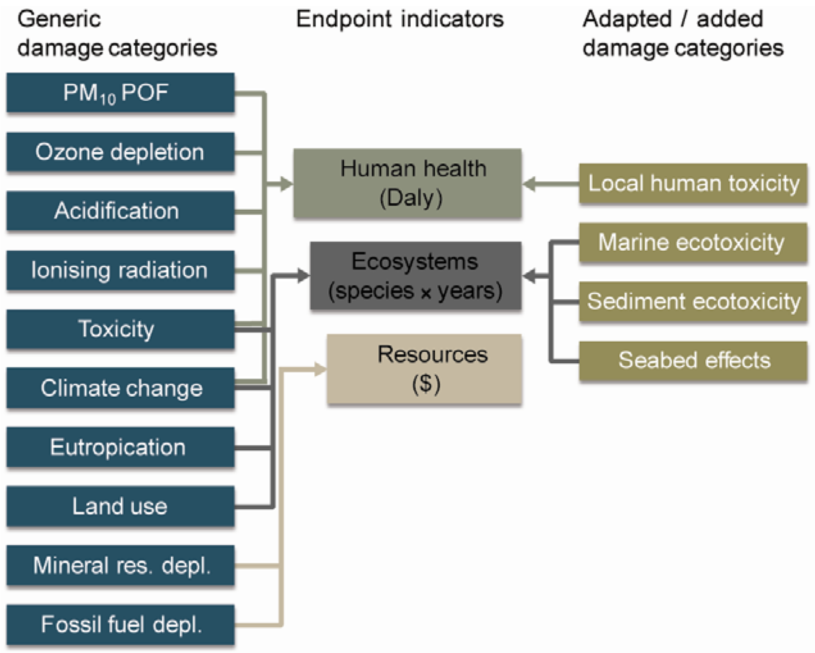

Figure 3 Combination of the generic and adapted/added damage categories into endpoint indicators for the ReCipe impact model used in the study

<u>Local model adaptations with regard to marine and human toxicity effects</u>

The USES-LCA is a multimedia effect model combining a contaminant fate model and an effect model for the estimation of toxicological effects by use of characterization factors (CFs) for human toxicity and ecotoxicity. The CF is an integrated value based on factors describing the contaminant fate (FF) and toxicological effect (EF) and is calculated for each substance (j) and emission compartment (i); soil, water and air:

$$CF_{i,j} = FF_{i,j} \times EF_{i,j}$$

The strategy in the present study was to use the best available information to adapt CFs to assess toxicity to the local fjord system and to add these locally derived CFs to the generic CFs from the USES-LCA model, which assesses consequences on a continental scale as the minimal resolution (*19*). The contaminant flux between the inner and outer fjord was assumed to be the interface between the local adapted model and the default USES-LCA model. Fluxes in the inner fjord were assessed as a part of the local system, whereas the fluxes to the outer fjord were assessed to be a part of the continental scale and incorporated in the default model (Figure 4).

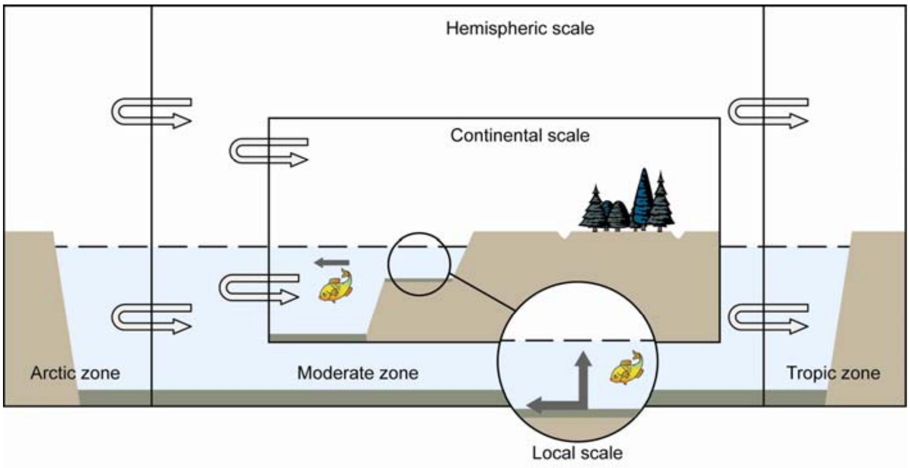

Figure 4 Incorporation of environmental effects into the USES-LCA model by introducing a local scale. The dark arrows show direction of contaminant fluxes to water and sediment-pore water. Fluxes through the Brevik sill are considered to be the connection between the local scale and continental scale models. Adapted from (*19*).

FFs for the local-scale-impact-model adaptations have been based on TCDD flux, water and sediment concentrations using the local abiotic transport model (*2*), see SI (figure S5). For sediments, ecotoxicological effects are assumed to be related to the pore water only (*14*), converting sediment concentrations into pore water concentrations using the sediment pore water partition coefficient (K_d), see SI (table S9). For all effect calculations, the standard EFs from USES-LCA 2.0 were utilized.

For the characterization of human toxicity, the USES-LCA model assumes the consumption of fish as the single exposure pathway. In this case, an intake fraction of fish (IF) was calculated using locally derived values for contaminant fate and exposure. Of note is the fact that the intake rate (IR) of fish, which depends on the ratio between areal population and the volume of the water compartment, is significantly higher for the local fjord compared to generic values (SI table S11). As for ecotoxicity, the fate calculations are combined with the generic USES-LCA 2.0 effects factor (EF) values describing the toxicological effects via oral ingestion of PCDD/F exposed fish. The locally calculated CFs are given in SI table S10.

Local model adaptations with regard to sediment use

One topic not previously introduced in LCA is changes in the benthic fauna caused by the physical impact of a capping operation. Effects may be caused by e.g. depletion of oxygen due to degradation of capping material, sediment burial or variations in grain size between the cap and the natural seabed (*20*). For capping with clean materials, oxygen depletion due to degradation is not relevant. However, sediment burial, referred to as *sediment occupational effects*, and variations in grain size, referred to as *sediment transformational effects*, are necessary to consider. In both cases a five-year time horizon may be anticipated for these post-capping effects (*21*). By using the relationship between the cause of hazard and the ecological effect, expressed as potential affected fraction of species (PAF), the CF for seabed effects was calculated as follows:

$$CF_{seabed\_eff} = 5 \times \frac{0.5PAF}{HS50} \quad (22)$$

The cause of hazard for *occupation* (HS_o) is given by thickness of the cap and for *transformation* (HS_t) is given by the difference in grain size between the capping material and the natural seabed. HS_o and HS_t were determined based on work performed by Smit et. al (*23*) (SI table S12).

Normalization and weighting

Using a normalization process allows damage effects to be transformed into unitless indexes (ecopoints) and thus allows a comparison between impact categories. Both *external normalization* relating effects against an external reference situation and *internal normalization* where results are related internally are relevant methods to apply in LCA. In this case external normalization was selected to facilitate the relative significance of results across categories, even though this also assumes a delineation of effects within a spatial and temporal resolution (*24*). The estimated effects from the study were normalized against the effects from the annual contaminant releases of 28 European countries during the year 2000 scenario (*25*), using endpoint characterization factors from ReCipe (www.lcia-recipe.net) for effect calculations (SI table S15).

Weighting may be applied in order to summarize damage effects into single score indicators. This study has weighted the different effect categories using the following weights: ecosystem 40%, human health 40% and resource use 20%, thus reflecting the time horizon and the objectives of common policy principles emphasizing ecosystem damage and human health to resource use (*15*).

The use of indicators, normalization and weighting has been heavily debated (*26*-*28*), since all approaches have advantages and disadvantages. For this exploratory and comparative study, a pragmatic view utilizing recommended values has been used. The results are however discussed with respect to model sensitivity and it's applicability to contaminated sediment remediation.

Results and Discussion

Primary effects affecting the fjord system

The normalized impacts values of the different remediation alternatives affecting the fjord system are given in Table 1. Based on primary effects, all active remediation scenarios were favorable compared to a natural recovery scenario. Impacts of human toxicity dominated over impacts of marine and sediment ecotoxicity. Local toxicity impacts were also higher than regional impacts. These findings are as expected due to the chronic nature of PCDD/Fs toxicological effects and the higher exposure in the local fjord system model as compared to the background level. The physical impact of the capping operation on the benthic community is also relatively high and outweighs the ecotoxicological effects. These findings are supported by experimental data indicating that the physical effects of a capping operation may have a significant short-term impact on the benthic fauna compared to the chronic toxicological effects (*29*,*30*).

Table 1 Normalized impact values (ecopoints) for primary effects of contaminated sediments. This includes local and regional effects for human toxicity and marine ecotoxicity as well as local sediment ecotoxicity of PCDD/F. It also includes local sediment transformational (difference in grain size) and occupational (cap thickness) effects of the capping operation.

Impact effect	Compartment [b]	NR	Clay	Limestone	Anthracite AC	Biomass AC
Human toxicity[a]	Local	122	24	24	6	61
	Regional	4	$7 \cdot 10^{-2}$	$7 \cdot 10^{-2}$	$2 \cdot 10^{-2}$	0.2
Marine ecotoxicity[a]	Local	$3 \cdot 10^{-4}$	$5 \cdot 10^{-5}$	$5 \cdot 10^{-5}$	$1 \cdot 10^{-5}$	$1 \cdot 10^{-4}$
	Regional	$1 \cdot 10^{-5}$	$2 \cdot 10^{-6}$	$2 \cdot 10^{-6}$	$6 \cdot 10^{-7}$	$6 \cdot 10^{-6}$
Sediment ecotoxicity	Local	$2 \cdot 10^{-5}$	$5 \cdot 10^{-6}$	$5 \cdot 10^{-6}$	$1 \cdot 10^{-6}$	$1 \cdot 10^{-5}$
Sediment transformation	Local	-	-	86	-	-
Sediment occupation	Local	-	12	12	0.9	0.9

[a] The reduction of accumulated contaminant flux due to active capping in comparison to the natural recovery scenario is: Clay and lime 80%, Anthracite AC 95%, Biomass AC 50%

[b] Local compartment refers to the fjord specific characterization factor, whereas regional refers to use of generic impact factors from USES-LCA 2.0

Overall impacts including secondary effects

Figure 4 presents the overall normalized and weighted results; detailed results, including unweighted data, are presented in SI (tables S13 and S14). Each stack in the figure contains the integrated weighted value of the potential effects on human health, ecosystem damage and use of non-renewable resources. In contrast to the primary impact results, the overall impact was higher for the active capping alternatives than for natural recovery, thus the resources used for active remediation (see SI table S18) were not compensated for by the gains from toxicity source reduction. This is consistent with LCA studies for contaminated soil (*31*) and indicate that the amount of energy and resources necessary to remediate contaminated sediments result in a large environmental footprint, especially for use of anthracite based activated carbon. Evidently the carbon sequestration effects of using biomass-based AC (*11*,*12*) is important with respect to overall life cycle impact and if this effect is incorporated in the

LCA this alternative exhibits a reduced environmental footprint that allows it to be compared with a natural recovery scenario. The degree of allocation of carbon sequestration for use of biomass-derived AC is a subject of discussion (*12,32*) and Figure 4 therefore shows a case with and without this allocation.

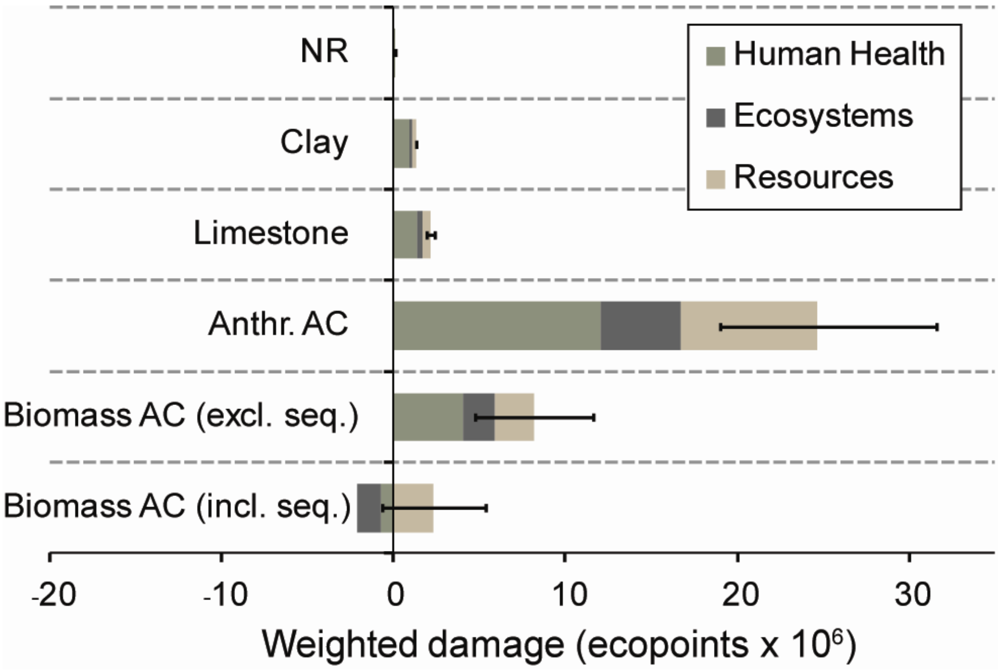

Figure 5 Normalized and weighted results (ecopoints × 10^6) obtained using the ReCipe hierarchist endpoint with the European normalization values and the average weighting set (*25*). The standard deviation (SD) for the alternatives was calculated based on Monte Carlo simulations using the predefined SD for the single unit processes and the SD for the flux calculations (SI figure S4). A distribution of SD between the endpoint indicators is given in SI table S17.

<u>Uncertainty and sensitivity analyses</u>

Uncertainties in LCA may originate from sources related to data, methodological choices and impact assessment model (*26*). In this study, uncertainties connected to inventory data are addressed by the use of standardized inventories and locally derived values. The error bars given in Figure 5 represent the combined uncertainties in qualitatively estimated uncertainty values (*33*) from the unit processes in SI table S5-S8. The error bars for natural recovery are based on standard deviation in the abiotic fjord model, see SI figure S4. Methodological and impact related uncertainties have been addressed through careful choice of the base impact model and through model adaptation to fit the local setting, with the inclusion of site specific effects like sediment use, as described in the methodological section. Different weighing sets will also effect the absolute values of the weighted damage potentials and therefore to a minor degree effect the relative order between the alternatives (SI figure S6).

The results of the LCA are sensitive to variations in the input data, and changes in the inventories may have substantial impacts on the results. In figure S7 and S8 in the SI the sensitivity to changes in the operational dredging efficiency (diesel use) and material efficiency (cap material use) is presented. Even though higher efficiency is beneficial in both cases, operational efficiency is more important for locally derived capping materials, whereas engineered materials with higher life cycle impact in the production phase benefit more from higher material efficiency. In contrast, biomass-derived AC including sequestration is non-sensitive to operational and material efficiency, since the positive carbon sequestering effect outweighs the negative impacts in the production phase.

In addition, variations in contaminant concentrations may affect the results, especially for the natural recovery scenario. This study averages PCDD/F fluxes over the whole inner fjord system according to the selection of the functional unit. By narrowing the scale further, the effect of natural recovery will vary depending on the local sediment contaminant concentration within the fjord. However, in order for an active remediation scenario to be beneficial from a life cycle perspective, PCDD/F fluxes haves to be two order of magnitude higher than the scenario used (SI figure S9) which is unrealistic (*34*).

Future use of LCA in contaminated sediment management

Sustainable sediment management can only be achieved by a holistic approach towards assessing remedial alternatives. This study shows that LCA may be a valuable tool for assessing the environmental footprint of sediment remediation projects and can be used for prioritization and optimization of remedial alternatives from a life cycle perspective. Even technologies with a relatively low resource-intensity, such as thin layer capping, can have a significant environmental footprint which approaches that of site-specific implementations for some of the more resource intensive solutions (e.g., dredging and disposal), (*35*). The use of LCA in contaminated sediment management would enhance the relative attractiveness of remedial solutions with limited raw material and energy use. LCA may be especially relevant for addressing beneficial sediment and alternative energy uses, such as the use of biomass-derived AC instead of coal based AC as discussed in this paper.

There are many issues that need to be carefully considered in implementing LCA for sediment management. In this paper, the environmental risk factors associated with sediment contamination have been extended to incorporate effects associated with the implementation of sediment management alternatives. The difference between traditional HERA results and results from the LCA are however substantial (*36*), and the LCA can therefore only be attempted for comparative assessment of remedial alternatives found to be acceptable through HERA. The comparative nature of such LCA implementation allows for dealing with the uncertainty that is attracting increasing attention within LCA and ERA communities (*26*). Even though many parameters may be uncertain, they are likely to result in similar over- or underestimation of risks for all considered alternatives and are thus unlikely to affect the final ranking.

The question of relevant scale and focus is important for both LCA and HERA. In general, HERA considers the local scale and focuses on risk of specific stressors, while LCA operates on a global scale, normalizing and weighting impacts for relative comparison. As for other specific LCA applications, (*37*) the results from this study emphasize the necessity of including a local compartment to the impact assessment model for future LCA applications in coastal areas to reach an acceptable resolution in the impact assessment. Even so, based on the standardized normalization and weighting procedures applied in this study, the damage from primary aspects are assessed as relatively minor compared to the secondary aspects. From a life cycle perspective, contaminant levels have to be substantially higher to justify commonly accepted remediation practices, which may contradict public values. Therefore, instead of basing the weighting on standardized damage categories more focus may be given to the perspective of the decision maker, thus giving higher focus to local (primary) effects than global (secondary) effects in the LCA.

In addition, both LCA and HERA do not explicitly consider many factors important in the selection of sediment management alternatives. One way to address this may be to assess the tertiary effects related to the remediation (*38*). Examples of such effects would be increased recreational use of the area or increased commercial fishing after lifting the dietary advisory. This approach would, however, require a more developed system for monetization of social and economical impacts (*39*). Establishing a more complex cause and effect related weighting systems may, on the other hand, reduce the transparency of the study and increase the use of controversial criteria which is undesirable (*40*).

An alternative to avoid controversial weighting procedures is to combine LCA and multi-criteria decision analysis (MCDA). MCDA integration would allow tertiary effects to be added separately to the standardized LCA results and the weighting between impact categories could be assessed using values elicited from stakeholders also incorporating uncertainties in the evaluation (*41*). Further research may be directed towards developing such an integrated framework for sustainable sediment management.

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SUPPORTING INFORMATION

More detailed information about the LCA assumptions, detailed inventory results as well as detailed results from the impact analysis are found in the supporting information for this paper. This information is available free of charge via the Internet at http://pubs.acs.org.

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BRIEF

LCA is a valuable tool for the assessment and prioritization of remediation alternatives for contaminated sediments based on the overall environmental footprint