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# Keu Contamination in Tuscany: The Life Cycle Assessment of Remediation Project as a Decision Support Tool for Local Administration

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**Abstract:** In this study, a Life Cycle Assessment (LCA) was conducted on a project to clean up a heavy metals-contaminated site located in central Italy (Tuscany) in order to define the less impactful solution. The study evaluated the contamination in the soil, derived from the leaching of backfill materials composed of quarry aggregates and sintered granules referred to as "Keu", a waste derived from the pyrolization of sewage sludge from the Tuscan tannery district, rich in chromium. Three action scenarios were compared, namely the no-action scenario, an excavation and landfill disposal scenario, and a permanent material capping and sealing scenario. The LCA results show the lowest impact for soil capping operations due to the high impacts of heavy metal emissions and landfilling of materials for the first and third scenarios. The third scenario has the lowest impact for ten of the eleven categories analyzed by the CML-IA baseline method. The only exception is the ozone layer depletion category due to the binder synthesis process used for the pavement to protect the membrane. Future studies could be devoted to the study of Keu, through extensive characterization, as well as a study of the fate of this material in landfills to define an appropriate process for future LCA analysis.

**Keywords:** LCA; Keu; contamination; remediation; heavy metals; tannery wastes; chromium contamination

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## 1. Introduction

Contamination of soils and groundwater due to uncontrolled waste disposal and the depauperation of resources are some of the major problems of our time, as identified both nationally and internationally [1–3].

More than 2.5 million sites are thought to be potentially polluted in Europe alone, 5379 of then located in Italy [4], and 14% of those are anticipated to need treatment. The majority of contaminants (60%) discovered on contaminated sites in Europe are mineral oil and heavy metals, with an estimated 6 billion euros needed each year to manage these contaminations [5,6].

In recent years, the problem of "Keu" has emerged in Tuscany, a waste derived from the processes of pyrolysis and sintering of sewage sludges produced from the wastewater treatment plant of the tannery district of Santa Croce dell'Arno, as note rich in chromium. Generally, the chromium concentration for sewage sludges is typically less than 50 mg/kg [7]. Part of the waste produced every year has been destined for the construction sector and used as backfill material. The use of these materials has led to the emergence of numerous potentially contaminated sites, including the one in question, defined as contaminated following characterization by the competent authority. Due to high toxicity, the diffusion of chromium in the environment is a serious threat, with consequences such as reduced germination of seeds and carcinogenic effect in humans [8,9]. The US Environmental Protection Agency listed it as one of 129 priority pollutants, and defines Cr (III) as moderately toxic from oral exposure and specifies Cr (VI)'s serious chronic effects, classifying it as a human carcinogen and fixing the maximum contamination limit for drinking water at 0.1 mg/L [10–12].

Innovative management strategies based on increasingly refined analytical methodologies have been developed to reduce the health risk to people who are exposed, and the toxicity to ecosystems [13,14]. To devise decontamination strategies that are as sustainable as possible, numerous approaches have been studied, including the Life Cycle Assessment (LCA) for defining environmental impacts [15–19].

Other than the primary effects, which are related to the site's condition prior to remediation, secondary effects are related to the site's remediation itself [20], such as greenhouse gas emissions from excavation activities. When choosing between site remediation choices (which include one or more remediation technologies), sustainability principles were taken into account out of concern for these secondary impacts. The idea first surfaced in Europe, where it was supported by decision-makers and business organizations to establish several frameworks [5].

LCA allows the quantification of these secondary impacts arising from the remedial activities, contributing to defining an operational strategy that minimizes the total impacts of remediation operations and the amount of waste produced [18]. It enables us to identify a way to improve the sustainability and circularity of processes, limiting landfilling operations and the resulting land consumption as much as possible [21–23].

According to this view, this study aims to compare three operational choices, assess which alternative is the best and provide decision support to local government. Estimating the potential impacts generated by reclamation activities will allow a comparison of the effects of all the processes involved, granting the best intervention strategy to be defined, with a sustainable approach [24,25].

## 2. Materials and Methods

The Life Cycle Assessment (LCA) method has been used to evaluate the optimum remediation option. According to ISO 14040:2006 and ISO 14044:2006, the study's goal and scope were defined first, then the life cycle inventory and life cycle impact assessment were conducted, and finally, the results were interpreted [25,26].

### *2.1. Goal and Scope Definition*

The project aims to define the best strategy for the remediation of a site contaminated by backfill materials, located in Peccioli (Tuscany), and evaluate the environmental performance of three different scenarios in the entire life cycle. Specifically, the option analyzed are; (i) DN-Do Nothing scenario; (ii) DD-Dig and Dump, and (iii) SC-Soil Capping.

There is no certainty about the exact composition of the backfill material, but it is known that an important part of this is composed of the specific waste denominated "Keu". It derives from the tannery industry of Santa Croce dell'Arno, which is known for its high chromium content.

For a better understanding of the material involved and the definition of the conceptual models of the area, the complete characterization of the soil characterized by the presence of the backfill material has been performed. The results shown in Table 1 report the contamination of the superficial part of the soil (C1 to C7), consisting of the material itself and the average concentration weighted by the mass of soil to remove for each zone. This has been compared with the Italian limit defining site contamination [27] (CSC) and a natural soil sample.

**Table 1.** On-site contaminant concentration [28].

| Samples | Unit | C1 | C2 | C3 | C4 | C5 | C6 | C7 | WAC [1] | CSC [2] | | Natural Soil [3] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Mass** | kg | 928.7 | 764.7 | 4362.2 | - | 581.5 | 719.9 | 419.6 | | Col. A | Col. B | |
| **Cd** | mg/kg | 0.5 | 1.1 | 0.6 | 0.2 | 0.6 | 0.9 | 0.5 | 0.7 | 2.0 | 15.0 | 0.2 |
| **Cr** | mg/kg | 1232.0 | 2371.0 | 1853.0 | 38.0 | 3309.0 | 2775.0 | 1812.0 | 2021.8 | 150.0 | 800.0 | 107.0 |
| **Cr(VI)** | mg/kg | 9.0 | 15.0 | 9.4 | 0.6 | 8.9 | 5.0 | 6.0 | 9.3 | 2.0 | 15.0 | 2.0 |
| **Ni** | mg/kg | 55.0 | 53.0 | 58.0 | 22.0 | 48.0 | 66.0 | 34.0 | 55.8 | 120.0 | 500.0 | 102.0 |
| **Pb** | mg/kg | 26.0 | 63.0 | 26.0 | 17.0 | 30.0 | 31.0 | 24.0 | 30.3 | 100.0 | 1000.0 | 15.0 |
| **Cu** | mg/kg | 53.0 | 124.0 | 135.0 | 29.0 | 101.0 | 192.0 | 107.0 | 125.3 | 120.0 | 600.0 | 41.0 |
| **Zn** | mg/kg | 135.0 | 285.0 | 135.0 | 36.0 | 234.0 | 214.0 | 167.0 | 166.2 | 150.0 | 1500.0 | 90.0 |
| **As** | mg/kg | 5.8 | 7.8 | 7.6 | 3.1 | 6.5 | 6.9 | 6.6 | 7.2 | 20.0 | 75.0 | 8.7 |
| **Sb** | mg/kg | 7.3 | 18.0 | 9.0 | 1.0 | 13.0 | 15.0 | 12.0 | 10.7 | 10.0 | 30.0 | <1 |
| **Be** | mg/kg | 0.5 | 0.6 | 0.6 | 0.5 | 0.5 | 0.5 | 0.4 | 0.6 | 2.0 | 10.0 | 1.5 |
| **Co** | mg/kg | 8.4 | 9.3 | 9.0 | 5.0 | 9.3 | 13.0 | 6.7 | 9.2 | 20.0 | 250.0 | 13.0 |
| **Fe** | mg/kg | 25.488 | 25.928 | 20.905 | 10.052 | 32.567 | 36.184 | 22.204 | 24.302 | - | - | 31.775 |
| **Mn** | mg/kg | 442.0 | 350.0 | 473.0 | 321.0 | 387.0 | 423.0 | 301.0 | 436.9 | - | - | 582.0 |
| **Mo** | mg/kg | - | 3.1 | 1.9 | 0.3 | 3.6 | 1.9 | 2.7 | 2.0 | - | - | 6.5 |
| **Hg** | mg/kg | 0.6 | 0.7 | 0.1 | 0.1 | 0.2 | 0.3 | 1.5 | 0.3 | 1.0 | 5.0 | <0.1 |
| **Se** | mg/kg | 0.6 | 2.5 | 1.4 | 0.3 | 2.1 | 42.0 | 0.9 | 5.2 | 3.0 | 15.0 | 0.8 |
| **Sn** | mg/kg | - | 22.0 | 9.5 | 1.3 | 10.0 | 21.0 | 12.0 | 10.8 | 1.0 | 350.0 | 3.1 |
| **Tl** | mg/kg | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 1.0 | 10.0 | 0.3 |
| **V** | mg/kg | 25.0 | 22.0 | 29.0 | 23.0 | 25.0 | 23.0 | 18.0 | 26.4 | 90.0 | 250.0 | 78.0 |

[1] Weighted Average Concentration of site's contamination. [2] Italian Legislative Decree 152/2006 established two types of limits: Sites for park, private and residential use (Col. A) and Sites for Commercial and Industrial use (Col. B). In yellow values over Col. A limits, in red values over Col. B limits. [3] Soil sample taken outside the site perimeter.

The analysis was conducted by the regional agency for environmental protection (Arpat) for all the non-organic compounds. Column A (Col. A) and Column B (Col. B) report the minimal values for Italian regulation that define a site as potentially contaminated. Highlighted in red are the values greater than column B limits, while in yellow are the values greater than Column A.

The comparison between the analysis of the backfill material and the natural soil contamination shows that the contamination derives from the presence of chromium (III), chromium (VI), copper, and zinc. In particular, the concentration of Cr (III) for the contaminated samples ranges from 1232 mg/kg to 3309 mg/kg, while the maximum concentration in the natural soil is 107 mg/kg, and the limit for industrial sites is 800 mg/kg. An eluate test confirmed the high presence of Keu in the materials, characterized by the presence of antimony and sulphate (typical Keu components), and highlighted a high concentration of Cr (VI) [28]. The absence of contamination in the natural soil (except for a single sample, view Table S1 in the supplementary material for the complete characterization results), placed under the backfill material, indicates a low leaching effect, probably due to the limited time range between the positioning and the characterization. The leaching tests of the backfill material demonstrate a high leachability potential; the results are contained in Table S2 in the supplementary materials.

According to the majority of site remediation, LCA studies [16], for this study, the identified functional unit is the entire amount of material to manage. It has been identified through supplementary analysis in 7777 t of material, composed of recycled backfill material and other raw materials derived from quarries, not separable from each other.

The system boundaries include the materials consumption, transport, and energy necessary for the exploitation of whole processes, among which there is the fuel consumption of trucks and work vehicles, different from each scenario analyzed. Excluded from them there are the impacts derived from the presence of a piezometer, the presence of only three and the same configuration for each scenario would result in three identical contributions that can therefore be excluded in a comparison LCA study [29].

Scenario Analysis and System Boundaries

As suggested by Laurent et al. [30], the characteristics of each scenario and of the system boundaries are carefully described and are depicted in Figure 1.

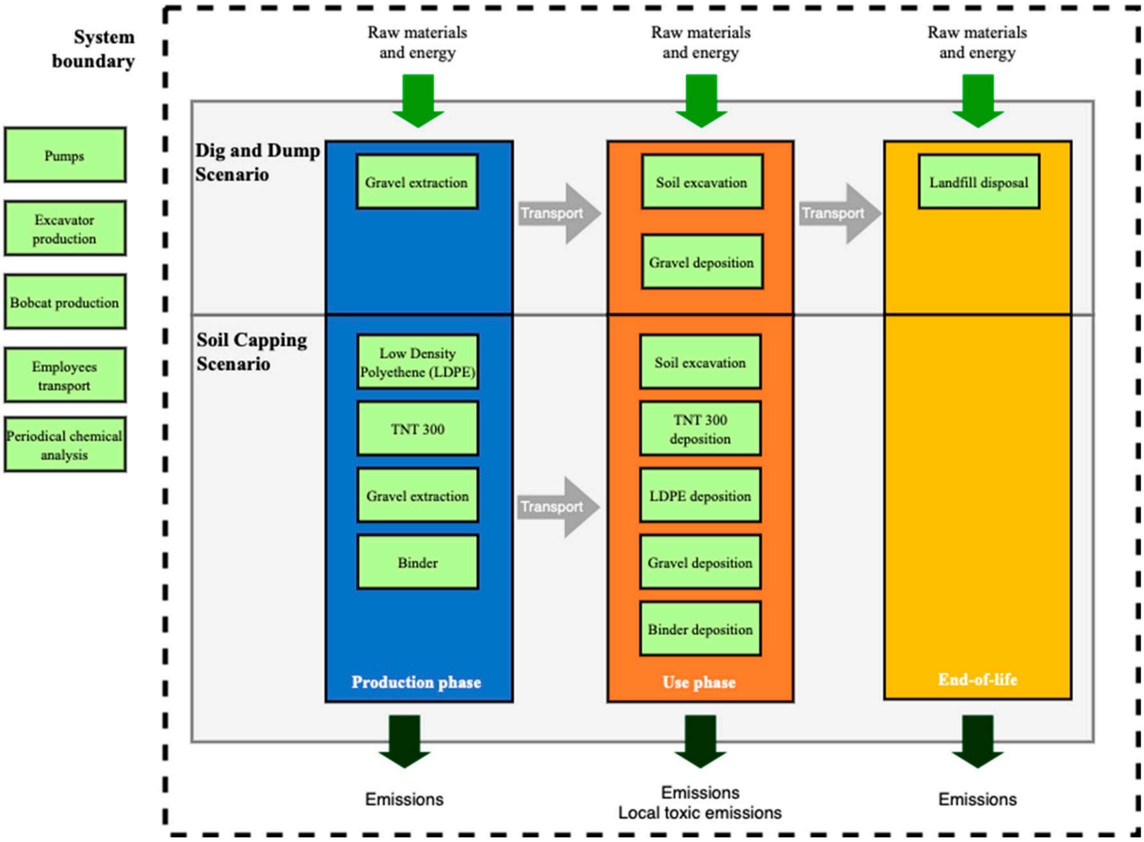

**Figure 1.** System boundaries of the life cycle assessment for Dig and Dump Scenario and Soil Capping Scenario.

In DN, no operations of any kind were planned to remove materials. In this case, the model includes the direct emission in the soil of the chemical compounds included in the materials, esteemed by the chemical characterization previously made. As mentioned above, the investigations did not reveal any obvious signs of contamination in the soils underlying the excavated carryovers, nor in the groundwater. However, from a life-cycle perspective, the input of pollutants into the soil can also generate potential environmental impacts in aquatic ecosystems (both marine and fluvial). This is because atmospheric precipitation and seasonal groundwater fluctuation phenomena can lead to the migration of some of the contaminants to the water matrix. To take this aspect into account as well, the mathematical models and characterization factors already implemented in the calculation method were used.

DD includes the excavation of the materials and the landfilling. For the excavation, the use of the “New Holland 245 q.li” excavator was planned, alimented with diesel fuel and located 2.4 km from the site. The relevant transportation was carried out by vehicles owned by the same company. The excavation operations included the removal of the backfill material quantity and about 1220 t of additional soil for preventive purposes, for a total of 8997 t. For this purpose, 25 working days and an average excavator consumption of 33 L/h are estimated. The volumes removed would be destined for a suitable landfill, identified as the Cassero Landfill, located in the municipality of Serravalle Pistoiese (PT), 59.5 km away and transported by an authorized company located at the distance of 42.9 km in the province of Pisa. Next, a road embankment is planned through the laying of about 9200 t of material, taken from the PCM Quarry, located in Ponte di Camporena, Iano, Montaione (FI), 23.2 km near the site. The transport of this additional material would be carried out through the previously mentioned vehicles, while the laying is planned to be done using a Bobcat 40-quart loader type mod; 463 High Flow, which is already present on the site. The consumption of the vehicle is estimated at 8 L/h of diesel fuel, for an estimated work time of 10 working days.

Finally, SC envisaged the construction of trenches in the soil in place, to the side of the intervention areas, without affecting the excavation of the backfill materials, having a width of about 60 cm and extending in depth to about 30–40 cm below the thickness of the backfill material. The operation would involve the movement of about 290 $m^3$ of soil in place through the use of the same type of excavator, for a working time of about 3 consecutive days. The excavation equipment, as well as the machinery transport service, would always be provided by a company located nearby. Next, the following materials would be laid-in the following order:

- Non-woven fabric-TNT 300 $g/m^2$. The membranes would be laid manually over an area of about 11,000 $m^2$ (double layer) and sent to the site from the supply warehouses, by courier; for the supply, it was assumed that they would be purchased from a company located in the Municipality of Santa Maria a Monte (PI).
- Low-density polyethene sheet-LDPE (black silage film with a thickness of 200 microns and a specific weight of 197 $g/m^2$). The membranes would be spread manually over an area of about 5500 $m^2$ and sent to the site from the supply warehouses, by courier; purchase from a company located in the Municipality of Camaiore (LU) was assumed for the supply.
- Cementation of the trench excavation with the soil originated from the excavation performed (clayey silt).

Above the intervention thus made will be laid, depending on the specific areas of intervention:

- Approximately 670 $m^3$ (1350 t) of stabilizer are planned to be taken from the PCM Quarry for the formation of a road embankment not more than 30 cm thick. The transport of the stabilizer from the quarry would always be carried out by vehicles owned by an excavation and demolition company based in the municipality of Peccioli (PI), while for the laying, it is planned to use the previously mentioned shovel located on site. The consumption of the vehicle in question is estimated at 8 L/h of diesel fuel, while the stabilized paving operation will take approximately 3 working days.
- Approximately 325 $m^3$ (552.5 t) of closed binder, with a thickness of about 5 cm. The intervention involves the use of a paver machine and a static roller, owned by a company located in the municipality of Ponsacco (PI), 14.4 km away, whose diesel fuel consumption was estimated at 24 L/h (at 2/3 load) and 11 L/h, respectively. Transportation of the work vehicles (total weight of about 10 t) and binder at the intervention site will always be done by vehicles owned by the aforementioned company. The intervention will take approximately 2 working days.

The system boundaries establish which life cycle processes are included or excluded from the evaluated system [25,31].

For this study, are excluded from the boundaries of each scenario the following elements:

- The pump for the monitoring of groundwater;
- The transport and the process involved in the chemical analysis for the “certification of complete remediation” and for the following site monitoring;
- The process involved in the production of the machinery (excavator and bobcat) used for the operations;
- The periodic transport of the site manager;
- The transport of the employee.

The first two are excluded because the process and the operation would be quite similar for each scenario. The third has been excluded due to its longer life cycles compared to the time of the works. Finally, personnel transport was excluded because fuel consumption was estimated to be much lower than that of trucks during the intervention period.

Figure 1 reports a schematic diagram of the system boundaries for each scenario.

### 2.2. Life Cycle Inventory (LCI)

The analysis incorporated all the information about the energy and material streams involved in the system boundaries into the inventory. The inventory was created using background information from the project requirements, and was obtained from the characterization of the relevant matrices. Project and characterization data from the matrices involved were combined. Simapro 9.3 software conducted the LCA, using the Ecoinvent database as a guide to simulate the underlying processes.

Given the complexity of the contaminated material and the absence of comprehensive data, the landfill disposal process of sludge from paper production (Pulp & Paper) was used in the DD scenario, as it has comparable contaminant concentrations to the material under study. This approximation, however, allows the modeling of the process and a proper assessment of the magnitude of the impacts generated by the treatment of this waste.

Table 2 shows the Ecoinvent processes used for the model, while the complete environmental data used are listed in Tables S3–S5 of the Supplementary Materials.

**Table 2.** List of Ecoinvent 3.8 processes used for the LCA.

| Processes | Ecoinvent 3.8 Records |
|---|---|
| Transportation of heavy equipment and materials | Transport, freight, lorry >32 metric ton, EURO5 {RER [4]} \| transport, freight, lorry >32 metric ton, EURO5 \| APOS [5], U |
| Transport of TNT and LDPE membranes | Transport, freight, light commercial vehicle {CH} \| processing \| APOS, U |
| Contaminants ground emission | Emission to soil |
| TNT 300 membrane production | Textile, nonwoven polyester {RoW3 [6]} \| textile production, nonwoven polyester, needle-punched \| APOS, U |
| LDPE membrane production | Polyethylene, low density, granulate {RER} \| production \| APOS, U |
| Quarry stabilizer production | Gravel, crushed {CH} \| production \| APOS, U |
| Binder production | Mastic asphalt {CH} \| production \| APOS, U |
| Diesel fuel production and consumption | Diesel, burned in building machine {GLO4 [7]} \| market for \| APOS, U |
| Landfill disposal | Sludge from pulp and paper production {Europe without Switzerland} \| treatment of sludge from pulp and paper production, sanitary landfill \| APOS, U |

[4] Europe. [5] At Point Of Substitution. [6] Rest of the World. [7] Global.

### 2.3. Environmental Sensitivity Analysis

According to the International Life Cycle Data (ILCD) Handbook guidelines [31], a sensitivity analysis was performed for each scenario, aimed to evaluate the influence of model inputs on Life Cycle Impact Assessment (LCIA) results. This type of analysis was used to assess the sensibility due to the variation of input data and modeling choice. Scenario analyses are one-factor-at-a-time (OFAT) approaches used to investigate the robustness of the results and identify the most sensitive aspects that could influence the LCA results and, as a result, the recommendations presented to decision-makers.

Given the absence of a specific waste disposal process in question within the databases available and the absence of data to reconstruct it, a comparison of the originally chosen process was made with other processes contained within the Ecoinvent database. The choice was made by analyzing the description of the process within the database, specifically by comparing the chemical characterization provided by Ecoinvent with the backfill material characterization shown in Table 1.

In addition, downstream of the LCA analysis, a comparison was made between the impacts generated by the binder production process and those from the clay and pavement cement production processes. This was done to define whether binder, a material chosen at the design stage, represents the best alternative from an environmental point of view.

## 3. Results

Table 3 shows the results of the LCA analysis according to the CML-IA baseline method. Data are expressed in percentage differences between the potential impact generated by each analyzed scenario to the corresponding reference scenario. Lower potential impacts are defined by a negative value; on the contrary, a positive value indicates a higher potential impact.

The comparison between the DN and the other two scenarios (DN-DD and DN-SC) shows the lack of results for some impact categories, this is due to a potential zero impact for the DN scenario for the categories in which "nd" falls. The first scenario has a potential impact higher for all four categories, this is due to the total amount of contaminants in the matrix that could be leached and transported to the water table, which would adversely affect the soil, groundwater and, at the end of the life cycle, the sea.

The DD and SC scenario have no direct emission to the water or to the soil as in the DN scenario. For the DD scenario these impacts derives from the process involved in the end-of-life treatment, such as the treatment of the leachate. For the SC scenario, the main contributor is the binder production.

For the reason previously explained, there is a second comparison between the DD and the SC scenarios, showing a greater impact for all categories in the first one, except for "Ozone Layer Depletion". Based on the assumptions and data supplied for this study, this means that applying a membrane that isolates the contaminated material is a better choice than doing nothing, or than excavation and landfill disposal. Figure 2 shows the contribution of the process to the total impact for each category; for the DD and SC scenarios. It is evident how the total potential impact contribution is very different between the first and the second:

- In the first case, the greater contributor is the End-of-Life process (landfill disposal), with small contributions of material production and transport for some categories. This is due to the few materials used for this scenario (quarry materials) and the use of vehicles already on the site for operations and a landfill located near the site.
- In the second case, the impact contributions are more variegated, but the greater contributors are the material production, represented mainly by TNT 300 production process (8–40%) and the binder production process (50–87%).

Remediation operation was finalized to reduce the potential local environmental threat [21], so the local potential impacts of the two scenarios were compared through the difference with the potential impact of the first scenario, generated by the direct emission of contaminants in the soil.

**Table 3.** Life cycle impact assessment of the scenarios.

| **Scenario** | **Abiotic Depletion [kg Sb eq.]** | **Abiotic Depletion (Fuel) [MJ]** | **Global Warming [kg $CO_2$ eq.]** | **Ozone Layer Depletion [kg CFC-11 eq.] *** | **Human Toxicity [kg 1.4-DB eq.] **** | **Fresh Water Aquatic Ecotox. [kg 1.4-DB eq.] **** | **Marine Aquatic Ecotox. [kg 1.4-DB eq.] **** | **Terrestrial Ecotox. [kg 1.4-DB eq.] **** | **Photochemical Oxidation [kg $C_2H_4$ eq.]** | **Acidification [kg $SO_2$ eq.]** |
|---|---|---|---|---|---|---|---|---|---|---|
| DN-DD | nd | nd | nd | nd | −69% | −25% | −45% | −99% | nd | nd |
| DN-SC | nd | nd | nd | nd | −98% | −98% | −97% | −100% | nd | nd |
| DD-SC | −88.8% | −70.5% | −99.4% | 10.8% | −92.6% | −97.9% | −94.7% | −94.0% | −98.9% | −94.8% |

* CFC-11 = Trichlorofluoromethane. ** 1.4-DB = 1.4-dichlorobenzene.

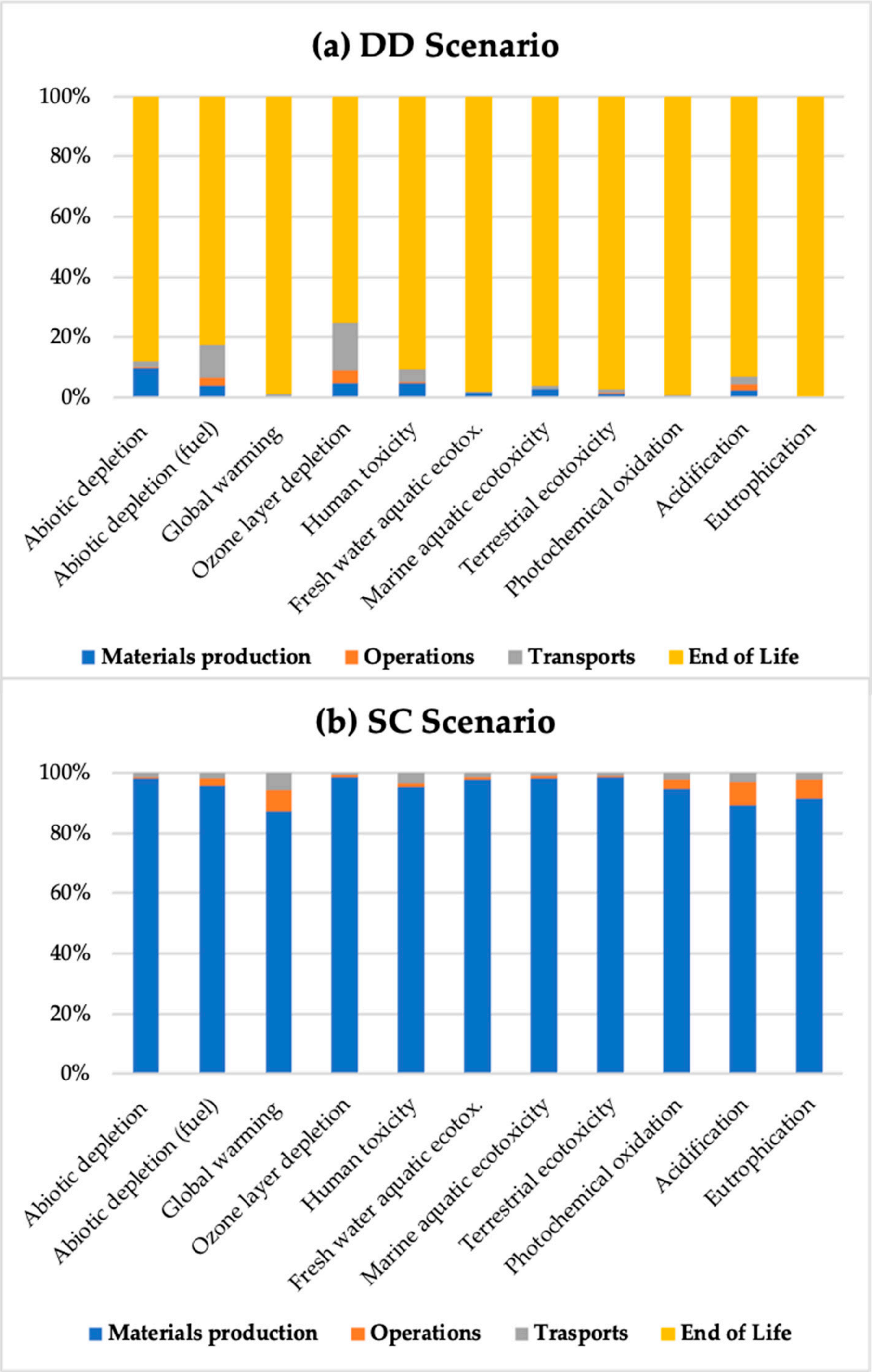

**Figure 2.** Contribution analysis for the two scenarios, expressed in percentage for each impact category (**a**) Dig and Dump scenario; (**b**) Soil Capping Scenario.

The results shown in Figure 3 express a much greater potential impact for the zero scenario, confirming that the choice of remediation is better than a no-action scenario. They also show how landfilling may pose a problem for neighboring areas, considering the non-negligible impact for three out of four categories, especially for surface water. For further details, Tables S3–S5 of the Supplementary Materials reported the results of the LCIA of each scenario, for all the processes involved.

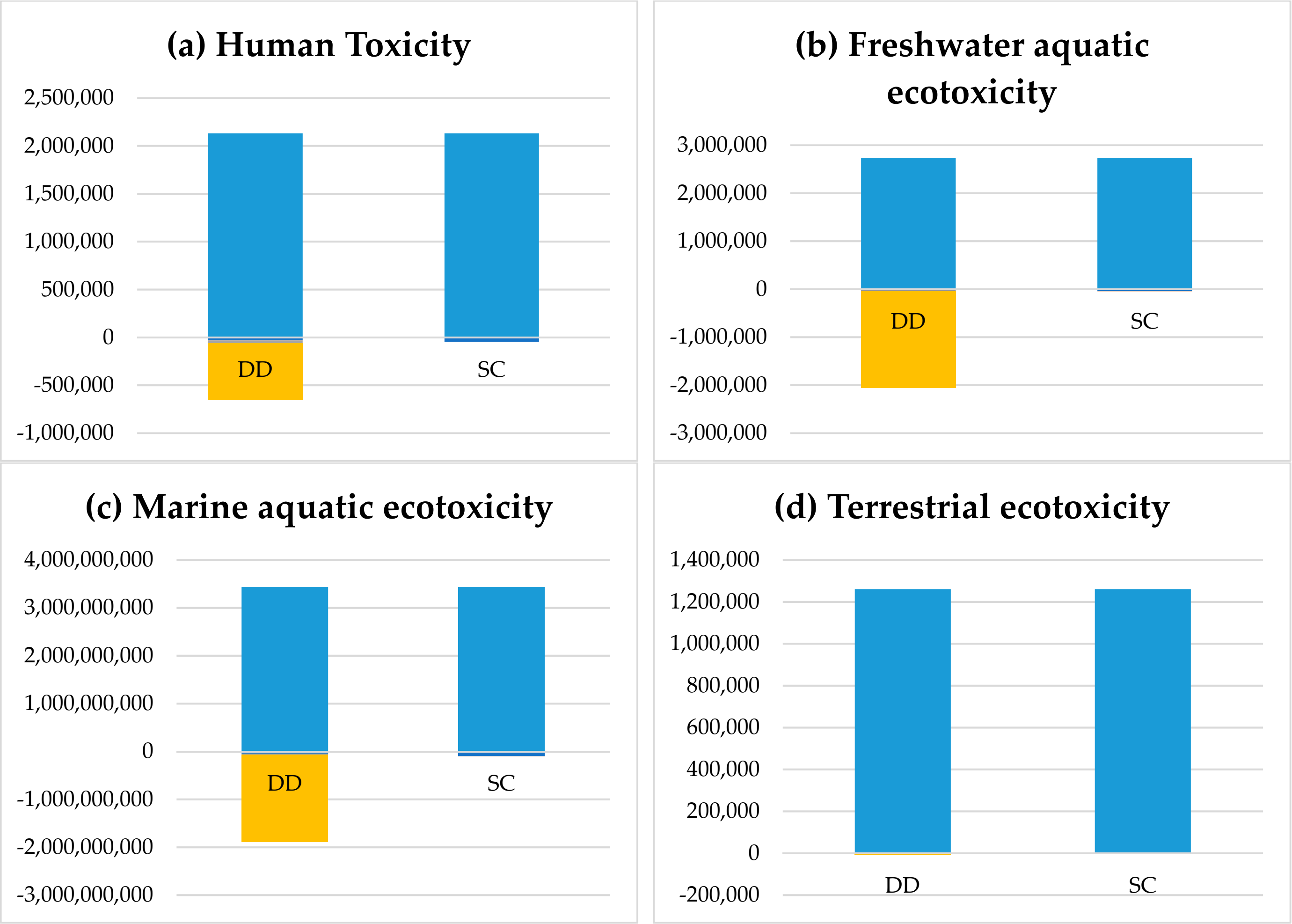

**Figure 3.** Focus on the contribution analysis for the four local impacts (**a**) Abiotic depletion; (**b**) Ozone layer depletion; (**c**) Acidification and (**d**) Terrestrial ecotoxicity.

The results shown so far define the third scenario as the best, but there are some considerationse:

- The disposal process is not waste-specific, but has used the "Sludge from pulp and paper production disposal", because the concentration described in SimaPro is similar to its composition;
- In the third scenario, the higher contributor to the potential impacts is the binder production process, which could be potentially replaced with other materials.

For these reasons, a sensitivity analysis has been conducted, to compare another disposal process with similar composition and compare the potential impacts generated by the use of other construction materials for the floor.

## *3.1. Sensitivity Analysis*

### 3.1.1. Dig and Dump Scenario

Four additional waste landfilling processes have been compared: "Hard coal ash disposal", "Lignite ash disposal", "Wood ash disposal" and "Municipal Solid Waste disposal". The processes have been selected by comparing the chemical concentration of the backfill material and the concentration defined by the Ecoinvent description. Specifically, the benchmarks used are Cr, Cu, Zn and Pb, while trying to have the other chemicals not too high.

The results presented in Figure 4 expose a similar contribution to the impact for the local categories and less impact for the others, especially for the abiotic depletion, global warming, and acidification.

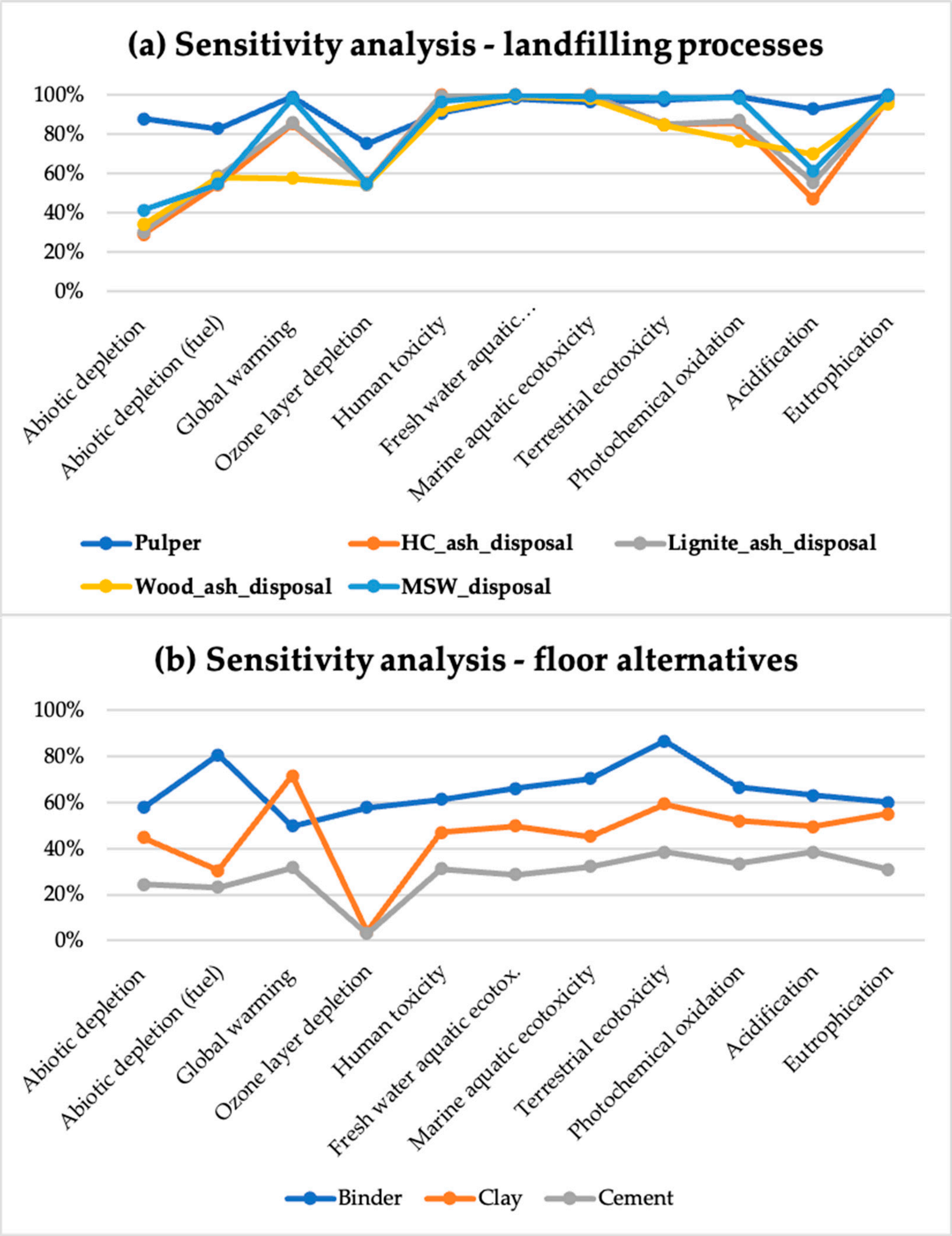

**Figure 4.** Sensitivity analysis results. (**a**) Five landfill disposal processes are compared: Pulper sludge, Hard coal ash; Lignite ash; Wood ash and Municipal Solid Waste (**b**) Three products are compared: Binder (real case); Clay and Cement.

### 3.1.2. Soil Capping Scenario

For this scenario there are different results, as shown in Figure 4. Binder production has been compared with clay and cement production, specifically for the construction of floors. The impacts of the binder are higher for all the categories except for global warming:

the average contribution of binder to the total impact is 65%, while the contributions of clay and cement are 46% and 29%, respectively.

## 4. Discussion

The contribution analysis demonstrates that the SC scenario is the less impactful scenario, mostly if compared with the potential impact of DD. However, attention must be paid to the absence of a specific contaminant dispersion model, useful for better defining the potential impacts derived from the direct emission to soil [32] in the "Do Nothing Scenario". Furthermore, as evidenced by the sensitivity analysis, it would be useful for more studies to define the properties of this waste and its behavior subjected to landfill activities, such as the leaching test [33].

Bardos et al. specify how the LCA is not a complete decision-support tool for remediation activities [17]. For this reason, many specific tools have been developed that analyze the social, economic and sanitary components [34,35]. The sanitary aspects are highly considered by Italian regulation, which indicated the mandatory nature of the risk analysis before the development of a remediation project for a contaminated site. For this aspect, this study evidenced a positive effect for the toxicity categories in the second scenario, but must specify that the LCA is not an affordable tool for evaluating toxicity and ecotoxicity at the local scale. The Risk Analysis results confirm the positive effects of the soil capping, derived from the interruption of direct exposure to the contamination [36].

The transport activities have a poor impact on the global impact of the DD and SC scenarios. This is due to the selection of proximal sites for landfill disposal and for the rental of vehicles. In particular, for the DD scenario, the contribution of transportation is 747,710 tkm, while for the SC scenario it is 39,701 tkm. This relevant difference is due to the transport of the backfilling material to the landfill, which contributes to 534,109.5 tkm. The contribution of whole processes and the relative impacts are carefully described in Tables S3–S5 in the Supplementary Materials.

The sensitivity analysis also highlighted the possible study of an alternative project with the substitution of cement for the binder, allowing the reduction of the potential impact, especially for the category "Ozone layer depletion" with a zero-impact result. This may be due to the use of NaOH in the binder life cycle, the production of which has chlorine gases as its main by-products [37]. Differently than expected, the use of clay has an impact higher than concrete.

LCA does not consider the quality of soil impact or ecological and biodiversity impacts. For the first case, the reference plot of land is intended for the construction of a building, so the soil quality would be impacted equally for each scenario. To assess the ecological impacts of this project, an Ecological Risk Assessment could be performed following the ISO 19204:2017 [38].

The results obtained will help to support the local administration to select the best remediation strategy and every future project for sites contaminated by Keu. Finally, for better results, a leaching test in landfill conditions could be conducted to support the construction of a specific process to assess the landfilling impact.

## 5. Conclusions

This study concerns the definition of the best strategy for the remediation of a site contaminated by backfill materials containing Keu, with an average chromium concentration higher than 2000 mg/kg.

Three different scenarios have been compared: Do-Nothing scenario (DN), Dig and Dump scenario (DD), with the excavation and consequentially disposal of backfill material, and Soil Capping scenario (SC), with the permanent on-site confinement of materials. The environmental impact analysis was conducted by the calculation method CML-IA baseline and Simapro 9.3.

The results showed that no removal operation would lead to negative impacts for only the local impact categories, corresponding to 4 out of 11 of the total categories analyzed. The impacts for these categories are greater than for the other two scenarios.

The SC scenario is the best scenario, with less impact for each category, except for "ozone layer depletion". The substitution of the binder with the cement for the floor above the capping membrane allows a sensible reduction of the impacts of this scenario. The worst performance of the DD scenario is due to the landfilling process, which is the largest contributor as confirmed by the sensitivity analysis.

Keu is a material used on more than one occasion as backfill material in Tuscany, whereby it would be appropriate to conduct a complete characterization. This should include leaching tests, and studying its fate if subjected to a landfill environment so that in future LCAs, a landfill disposal process that more closely reflects the life cycle of this material can be used.

**Supplementary Materials:** The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su142214828/s1, Table S1: Complete characterization of backfilling materials (C1–C7), contaminated deep soil sample (R1), natural soil samples (T1–T7) and white sample (B); Table S2: Leaching test results of backfill material samples (C1–C7) and contaminated deep soil sample (R1); Table S3: Environmental data used for the Do-Nothing scenario and LCIA results; Table S4: Environmental data used for the Dig and Dump scenario and LCIA results; Table S5: Environmental data used for the Soil Capping scenario and LCIA results.

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