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Research Paper

LCA of recycling aluminium incineration bottom ash, dross and shavings in a rotary furnace and environmental benefits of salt-slag valorisation

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ABSTRACT

Recycling aluminium in a rotary furnace with salt-fluxes allows recovering valuable alloys from hard-to-recycle waste/side-streams such as packaging, dross and incinerator bottom ash. However, this recycling route generates large amounts of salt-slag/salt-cake hazardous wastes which can pose critical environmental risks if landfilled. To tackle this issue, the metallurgical industry has developed processes to valorise the salt-slag residues into recyclable salts and aluminium concentrates, while producing by-products such as ammonium sulphate and non-metallic compounds (NMCs), with applications in the construction or chemical industries. This study aims to assess through LCA the environmental impacts of recycling aluminium in rotary furnaces for both salt-slag management routes: valorisation or landfill. It was found that this recycling process brings forth considerable net environmental profits, which increase for all the considered impact categories if the salt-slag is valorised. The main benefits arise from the production of secondary cast aluminium alloys, which is not unexpected due to the high energy intensity of aluminium primary production. However, the LCA results also identify other hotspots which play a significant role, and which should be considered for the optimisation of the process based on its environmental performance, such as the production of by-products, the consumption of energy/fuels and the avoidance of landfilling waste. Additionally, the assessment shows that the indicators for mineral resource scarcity, human carcinogenic toxicity and terrestrial ecotoxicity are particularly benefited by the salt-slag valorisation. Finally, a sensitivity analysis illustrates the criticality of the metal yield assumptions when calculating the global warming potential of aluminium recycling routes.

1. Introduction

Recycling aluminium is considerably more sustainable than its primary production, both environmentally and economically (Damgaard et al., 2009; Olivieri et al., 2006). The main reason is that primary production consumes vast quantities of energy and resources during the initial stages of mining the raw materials and extracting from them the aluminium metal (first aluminium oxide is extracted from bauxite mineral through the Bayer process, and subsequently, the oxide is reduced into aluminium metal through the Hall-Héroult process). Another critical benefit of recycling is avoiding the generation of bauxite residue commonly known as “red mud”, which can pose significant environmental risks (Mayes et al., 2016). According to a recent study, producing 1 tonne of primary Al releases between 14 and 17 metric tonnes of CO_2 equivalent from the bauxite mine to casting the metal. From these, the dominating part of the emissions are indirect, released in electricity production for electrolysis, and will therefore highly depend on the source of electricity (Saevarsdottir et al., 2023). The Hall-Héroult electrolytic process is the most energy-intensive step of primary production, with approximately 14 MWh of energy consumed per tonne of Al produced (IAA, 2022). The secondary production route consumes much less energy in comparison, and it consists of re-melting aluminium-containing scrap at temperatures around 700–750 °C, adjusting the melt composition, and solidifying the metal as slabs or ingots which can then be shaped into new products. According to the current Best Available Techniques in Europe (Delgado Sancho, 2017), the process of re-melting aluminium scrap in a rotary furnace uses 0.55–0.70 MWh (2–2.5 GJ) of energy per tonne of produced Al, and the total energy required for producing 1 tonne of secondary aluminium ranges between 0.55–2.50 MWh (2–9 GJ), depending on the quality of

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the scrap and the processes involved. The standard re-melting processes are carried out in reverberatory furnaces or rotary furnaces fired with natural gas, and in the second case mixed with substantial amounts of salts (NaCl/KCl/ fluorides) (Schlesinger, 2013).

This study assesses the environmental impacts of re-melting via a rotary furnace. This process allows recovering the aluminium present in contaminated or oxidised scrap, such as some internal wastes from production (dross, skimmings, shavings), or post-consumer scrap (incineration bottom ash, packaging). It involves mixing the scrap with substantial amounts of salt-fluxes, which separate the contaminants (e.g., oxides, carbides) from the molten metal, promote the coalescence of the metallic droplets and protect it from oxidation (Milani and Timelli, 2023). These interactions are promoted by the rotational movement of the furnace. However, a downside is the generation of salt slag – a mixture of non-metallic compounds (NMCs) such as oxides, carbides and nitrides, salt, and residual droplets of aluminium metal. These residues are classified as hazardous waste (European-Comission, 2015) and they can lead to significant environmental risks if disposed into landfills (EPA., 2015). An alternative to tackle this issue is to valorise the salt slag residues by crushing and dissolving the salts in water (Delgado Sancho, 2017). Examples of salt-slag valorisation processes have been previously investigated in (Padilla et al., 2022) (Li et al., 2013). The present study considers the industrial valorisation of the salt-slag into, on one side, salts and Al concentrates which can be fed back into the rotary furnace, and on the other side ammonium sulphate with application in fertilizers (Rodrigues et al., 2022) and NMCs which the chemical or construction industries can use. The main constituent of the recovered NMCs is alumina (Lucheva and Petkov, 2005), that can be used for example to replace fine aggregates in self-compacting concrete (Sua-iam and Makul, 2017), which is more sustainable than traditional concrete (Joseph and Tretsiakova-McNally, 2010), showing acceptable performances even at 1:1 substitution (Sua-iam and Makul, 2013). Alumina waste can also be used in the production of refractories, mineral wool, or in road construction (López-Alonso et al., 2019), where it improves the long-term mechanical performance of roads made of recycled aggregates. The implementation of industrial wastes/by-products in construction is gaining popularity, since it reduces the environmental burdens associated with the manufacturing of materials and landfill of waste while also tackling the challenge of the shortage and increased prices of traditional construction raw materials, as discussed in (Joseph and Tretsiakova-McNally, 2010).

In the aluminium industry, there is an ongoing discussion regarding whether aluminium re-melting in rotary furnaces is the most sustainable recycling process or whether it merely displaces the environmental burden to the production of salts and generation of salt-slag (Xiao Y, 2005). A recent environmental profile report (European-aluminium, 2018) contains environmental indicators of re-melting scrap in an integrated cast house. Still, it does not include detailed data regarding the process of recycling the dross generated during re-melting, which is done by re-melting in rotary furnaces with salt-flux (mixed with other aluminium-containing waste/side-streams), stating that a dedicated task force should be dedicated to this work. Thus, this study delves into this topic and provides an assessment of the potential environmental impacts of an industrial process to recycle a mix of hard-to-recycle aluminium streams (dross, IBA, shavings) in a rotary furnace with salts based on production data from 2021. The study also compares scenarios where salt-slag residues are either treated for recovery or landfilled.

There are several published environmental life cycle assessments (LCA) of aluminium production and recycling processes. A recurrent takeaway is that recycling brings forth significant environmental benefits (Damgaard et al., 2009; Olivieri et al., 2006). This is mainly due to the assumption that by producing secondary aluminium, the need for primary production and its associated energy use and emissions (e.g., CO_2, PAHs, and PFCs) is mitigated. This approach is defined as system expansion (substitution), and is recommended by the ISO guidelines when dealing with multifunctional processes, or those that provide multiple functions such as recycling systems (which treat waste while producing materials), as long as further subdivision of the system is not possible (ISO, 2006a). By expanding the system, the impacts of the alternative production of the secondary function in the most likely way of producing it (primary production of aluminium in this study) are discounted from the total impact. As discussed in (Vadenbo et al., 2017) the choice of such underlying assumptions is critical for the interpretation of the results, since the avoided burdens credited based on the expected displacement of other product systems can dominate the overall results, as it occurs for aluminium in the assessment of waste management options by Manfredi (Manfredi et al., 2011). The assumptions related to the energy source can also play an important role in the interpretation of the LCA, as shown by the aluminium primary production emissions presented by McMillan (McMillan and Keoleian, 2009), with regional variances between 7.07 and 21.9 kg CO_2 eq. kg $^{-1}$ metal in 2005. A critical review by Liu (Liu and Müller, 2012) also highlights the energy use as a source of uncertainties in LCAs in the aluminium industry. In this review, the emissions associated with producing a tonne of primary aluminium across regions varied as much as from 5.92 to 41.10 tonnes CO_2 equivalent. Other mentioned challenges include the use of industry-wide inventory data, different system boundaries and diverse assumptions for the allocation of recycling (e.g., recyclability, product lifetime). Damgaard (Damgaard et al., 2009) concluded that recycling aluminium brings forth large overall reductions of Global Warming Factors, but since these highly depend on the type and amount of energy used and its sourcing, the reductions can vary between 5.0 and 19.3 tonne CO_2 tonne $^{-1}$ aluminium scrap processed. The current LCA study attempts to minimise these uncertainties by using data from a recycling plant instead of generic data and by carrying out a sensitivity analysis of one of the processes determining assumptions: the recycling metal yield (mass of metal recovered per initial mass of the scrap treated), which varies for each type of scrap and waste management route.

2. Material and methods

The framework to develop this LCA is the International Standard Organization (ISO) framework 14040–14044 (ISO, 2006a, b). It consists of four phases: the definition of the goal and scope, in which the context and modelling aspects are defined; the inventory analysis, where inputs and outputs to the process are accounted for; the life cycle impact assessment, with the calculations of associated potential environmental impact; and finally, the life cycle interpretation phase, where results are analysed, and recommendations are drawn.

2.1. Goal and scope definition

This LCA has three main goals. First, to quantify the environmental impacts of treating a tonne of hard-to-recycle aluminium containing side/waste streams (dross, IBA, shavings) in a rotary furnace. Second, to compare between the impacts of two waste management scenarios for the salt slag residues: landfill or valorisation treatment. And third, to identify hotspots or potential areas where the environmental sustainability of the processes could be improved.

The functional unit (F.U.) selected for comparison is 1 tonne of aluminium containing material ready to be recycled, consisting of a mix of 1/3 wt dross, 1/3 wt IBA and 1/3 wt industrial shavings. The industrial recycling route in which this study is based includes two processes. The first one, re-melting in the rotary furnace, produces secondary aluminium and salt-slag residue, as well as furnace off-gas. The second process, the salt-slag valorisation treatment, allows recovering some of the salts (NaCl/KCl) and the entrapped metallic particles (aluminium concentrates), which are reused internally by feeding them again into the next rotary furnace cycle. In addition, other by-products recovered from the salt-slag treatment are non-metallic-compounds (NMCs) and ammonium sulphate ($(NH_4)_2SO_4$). To allocate these co-products, a system expansion approach is applied, the procedure recommended by the ISO (ISO, 2006b). It was assumed that these byproducts substitute the Simapro flows specified in the LCI tables (primary cast Al alloy, aluminium oxide non-metallurgical, and ammonium sulphate) displayed in the Appendix with a substitution ratio 1:1. However, the allocation of the input of aluminium containing material (dross, IBA, and shavings) is considered burden-free, because as it is defined hard-to-recycle, this flow would not currently have an alternative use. To facilitate the comparison with other published studies which assess the production of 1 tonne of secondary aluminium, the results of Scenario 2 were re-calculated changing the F.U. to 1.384 t Al-containing materials treated, and are presented in the Appendix.

The LCA study compares between two scenarios, a hypothetical scenario where the salt-slag residue is disposed at the landfill (Scenario 1: Al recycling + salt-slag landfill), and the current practice of the European refiner (Scenario 2: Al recycling + salt-slag treatment), where salt-slag residues are treated as described above.

The system boundaries of this study are cradle-to-gate, considering the stages from the raw material extraction to the production of the semi-finished product (secondary cast aluminium ingots). This is a common choice for LCA studies focused on metallurgical or mining processes, due to the uncertainty around the subsequent life-stages (Santero and Hendry, 2016). For instance, an aluminium ingot can be used for multiple applications, as household appliances or car components. On the contrary, when assessing finished products, e.g. cookware, a holistic cradle-to-grave approach, including the transport, usage, and disposal stages, is recommended.

Fig. 1 shows the system considered, illustrating an example input material mix with 70 % metallic content and an output of secondary aluminium. It is here assumed that all the metal present in the scrap is recovered after re-melting (metal yield equal to metal content in the scrap), and that the aluminium alloys entering the system are the same as the secondary cast alloys produced, omitting the need for adjusting the melt composition by refining, dilution, or addition of alloying elements.

The scheme shows the raw materials, the products and wastes produced, and the gas emissions monitored during the re-melting processes. For better visualisation, it excludes the energy sources and process input gases (diesel, natural gas, electricity, N_2, O_2), detailed in the LCI tables provided in Appendix Tables A-1 and A-2. The mass balance of the aluminium alloys (metal), oxides and fluorides entering and exiting the system were calculated to discuss uncertainties linked to potential flows that may not have been considered, for example, due to process inefficiencies or non-monitored emissions. The flows of aluminium metal and NMCs were selected because they are the main contributors to environmental impacts.

The amount of aluminium metal and NMCs entering the furnace was calculated based on the metal yield values provided by the recycling plant. The guidelines to sample and analyse the metal content of scrap/side-streams are described in EN 13920–1 (CEN, 2003), and the Metal Yield is calculated according to Equation (1).

$$Metal\ Yield_{scrap}(\%) = \frac{Weight_{metal\ recovered}}{Weight_{scrap}}*100 \tag{1}$$

Assuming that the metal yield values (70 wt%) are equal to the material's metal content and that there are no re-melting metal losses, the output flow of secondary Al when charging 1 tonne of our material mix into the furnace would be 700 kg. However, the standards mention that the laboratory tests employed to calculate the metal yield usually render slightly higher values than those yields obtained during industrial production. Thus, it is likely that the process metal losses have been underestimated to some extent in this system. For instance, at least 24 kg of aluminium in concentrates (3.4 wt% with respect to the initial 700 kg of metal present assumed) are lost during re-melting by entrapment in the salt-slag, since such is the amount recovered from the salt-slag treatment. In addition, some metals may be transformed into NMCs due to oxidation or reaction with other compounds such as C or N. This would agree with the mass imbalance of the NMCs displayed in Fig. 1, where 300 kg of non-metallics enter the system, but 350 exit it (338 in the NMC fraction and 12 in the concentrates fraction). The input flow of NMCs was calculated by Equation (2), and the output flow was based on average production data, detailed in the following section (inventory

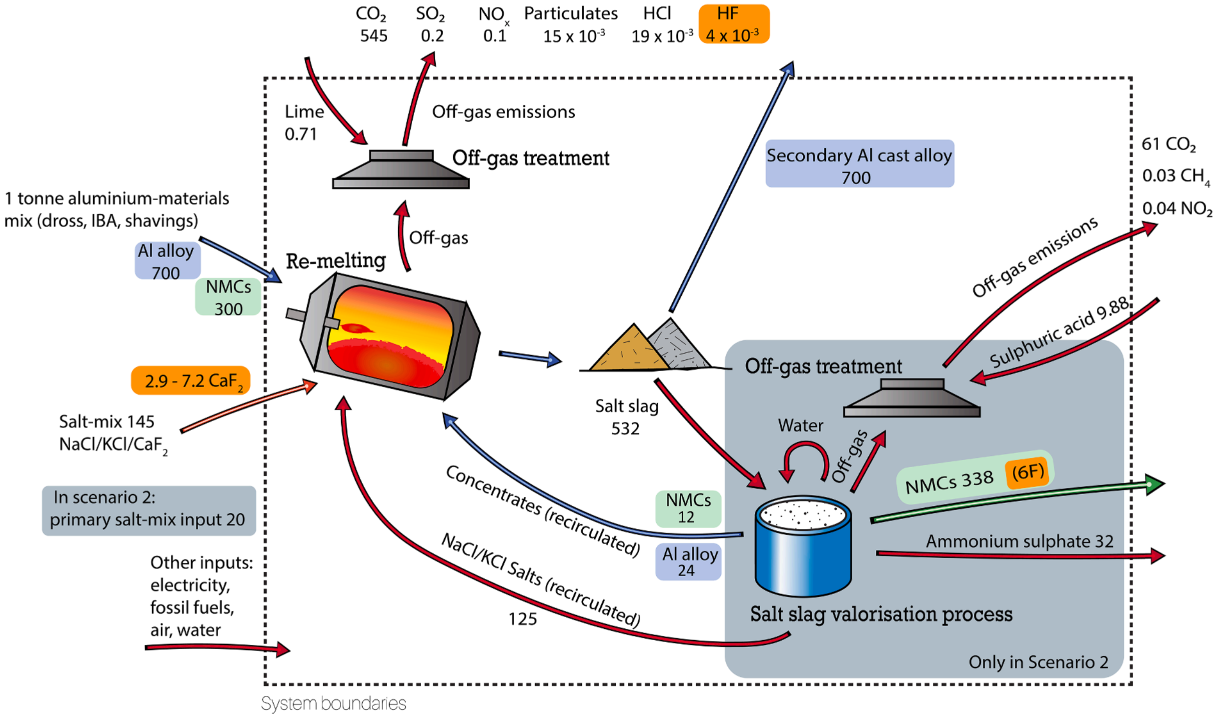


Fig. 1. System considered for Scenario 2 with inputs and outputs for the two processes: recycling 1 tonne of aluminium-containing material mix in a rotary furnace and salt-slag residue valorisation treatment. The units are kg.

analysis).

$$NMC_{scrap} = Weight_{scrap}\left(\frac{100 - Metal\,Yield\,(\%)_{scrap}}{100}\right) \tag{2}$$

The mass flow of the fluorides is also interesting to discuss. It is common practice to add small amounts of fluorides (2–5 wt%) into the salt-flux to promote the coalescence of the metal and hence reduce the risk of small aluminium droplets being entrained by the salt-slag residues and lost (Capuzzi et al., 2018; Peterson, 1990; T. Utigard, 1998; Thoraval and Friedrich, 2015). Still, some aluminium recyclers avoid using fluorides, even at the expense of increasing the amount of aluminium lost, due to their higher environmental impact and costs (Delgado Sancho, 2017). For instance, fluorides could form perfluorinated compounds (PFCs, such as CF_4 and C_2F_6) which have large global warming potentials (Damgaard et al., 2009; McMillan and Keoleian, 2009) and controlling these emissions is a primary concern for the aluminium industry. Since the fluorides added into the fluxes cannot be recovered by the current salt-slag valorisation process (because CaF_2 is not soluble in water), as opposed to KCl/NaCl salts, the fluorine-containing compounds should largely end up in the NMCs in the system considered. In this study, the average composition of the NMCs (wt %), based on producer data, are: Al_2O_3 (60–72 %), SiO_2 (7–12 %), MgO (5–10 %), CaO (2–3 %), C (1–3 %), F (0–4 %) and moisture/volatiles (9–11 %). Considering the above, a mass balance of the F-compounds entering and exiting the system was conducted to assess whether part of the fluorides could evaporate and/or escape the furnace as PFCs without being monitored during re-melting. According to the producer environmental report, only trace amounts of fluorine exit the rotary furnace through the off-gas treatment as HF (<4 g per cycle), and the rest would end up in the NMCs fraction. The analysis of the smokestacks from the re-melting plant did not measure any CH_4, HFC, PFC, NF_3 or SF_6 emitted. Chemical analyses of three types of produced NMCs gave an average F concentration of 1.8 wt%. Considering that the amounts of CaF_2 charged into the furnace range between 2–5 wt% of the weight of the salts added, the theoretical F content on the NMCs, following the mass balance, should range between 0.42–1.05 wt%. Since the theoretical concentration of F expected in the NMCs is lower than the measured one, it seems reasonable to accept that no hidden flows of F-containing compounds are exiting the system.

2.2. Inventory analysis

The data was provided by European aluminium recycling and salt-slag treatment plants from their 2021 operations and environmental reports, where most data was expressed relative to the annual secondary aluminium produced or the salt-slag residue treated, e. g. tonne CO_2 emissions/tonne Al produced. The study considered the treatment of 1 tonne of material mix consisting of 1/3 incineration bottom ash (IBA), 1/3 dross, and 1/3 industrial shavings, which is a representative example from the industrial recycling processes, where it is common to mix different types of scrap together, depending on the scrap available and the alloy specifications for the secondary aluminium. Since the expected metal yield (Al alloy) of each of these material types individually was 74.6 wt% for IBA, 64.7 wt% for dross and 70.3 wt% for shavings, the average metal yield of the scrap mix was 70 wt%. However, for the scenario where the salt-slag residues are treated, and additional aluminium concentrates are recovered, the metal yield increases to 72.3 wt%. This is because, based in the industrial data explained below, when 700 kg of Al is recovered, 532 kg of salt-slag is also generated, from which it is possible to recover ca. 36 kg of aluminium concentrates. Since the expected metal content of the aluminium concentrates is 67.3 wt%, approximately 24 more kg of aluminium would be produced after recirculating them into a second re-melting cycle, as represented in Fig. 1.

The inputs and outputs to the LCI inventory are collected in tables A-1 and A-2 in the Appendix for the scenarios with aluminium recycling with salt-flux landfill and salt-flux treatment. The process assumptions, based on discussions with the recycling plant, are described below:

The salt-flux was a mix of 70 wt% NaCl and 30 wt% KCl, and 2 wt% additions of CaF_2. The valorisation treatment of 1 tonne of salt-slag produces 236 kg of salts, 636 kg of NMCs, 67 kg of aluminium concentrates and 60 kg of ammonium sulphate. The NaCl and KCl recovered from salt-slag treatment are recirculated into the re-melting process. Consequently, 90 % of the weight of salts needed is considered without burden, and only the remaining 10 % of the required salt additions are included in the impact calculations. This is based on the following data for 1 cycle of the rotary furnace: 140 kg of salts are added per tonne of scrap treated and 760 kg of salt-slag are generated per tonne of secondary aluminium produced. Lastly, the concentrates recovered from the salt-flux treatment are recirculated, which increases the effective material treated in Scenario 2 to 1.036 tonne instead of 1 tonne.

The systems evaluated consider European market conditions. The impact of some assumptions such as treating different types of scrap with varied metal yield is tested through sensitivity analysis, where hypothetical batches of the individual material streams are compared (UBCs, mixed packaging, dross of varied metal content).

2.3. Impact assessment and interpretation

The impact method used for this study is ReCiPe 2016 (Huijbregts et al., 2020). Calculations were developed in SimaPro v. 9.5.0.0 and background data was considered through ecoinvent 3.6 with allocation at the point of substitution (APOS).

3. Results and discussion

3.1. Net environmental impacts

Table 1 displays the environmental impacts, calculated by the ReCiPe (Huijbregts et al., 2020) method for 18 impact midpoint indicators, when treating 1 tonne of Al-materials through both scenarios. The results could also be expressed as end-point indicators, aggregated into three categories: human health, ecosystems, and resources. The impact of each mid-point indicator into the end-point impact indicators is included in the Appendix Table A-4. The third column of Table 1 shows the relative improvement when implementing a salt-slag treatment with respect to landfilling the residue.

The results reveal that implementing a salt-slag recovery treatment improves the environmental performance of the recycling process; reducing the net impacts by between 5 and 25 %. The most benefited indicators were mineral resource scarcity, human carcinogenic toxicity and terrestrial, marine, and freshwater ecotoxicity. However, these are just relative improvements may not be necessarily linked to the largest actual impacts on the environment.

The “Global warming potential” (GWP) is a well-established indicator usually discussed in metallurgical LCA studies (Santero and Hendry, 2016). In this study, treating 1 tonne of the considered Al-containing streams reduces the GPW by approximately 12 t CO_2 eq. when the salt-slag residue is disposed at the landfill, and by 13 tonnes of CO_2 eq. if the salt-slag residues are valorised. This falls within the ranges reported by Damgaard (Damgaard et al., 2009): GWP reductions between 5.0 and 19.3 tonne CO2 eq. per tonne of Al scrap processed. All the contributions per process input are provided for all midpoint impact indicators in the supplementary material, and the main contributions to the GWP are listed below.

When treating 1 tonne of Al-containing materials through this recycling process, the impacts to the GWP arising from process emissions were 170 kg CO_2 eq. for the salt-slag landfill route, and 234 kg CO_2 eq. for the salt-slag treatment route. Using natural gas to fire the furnaces led to 183 kg CO_2 eq. for salt-slag landfill and 250 kg CO_2 eq. for salt-slag treatment. The impacts of electricity consumption were also lower for salt-slag landfill than for salt-slag treatment; 27 kg CO_2 eq. vs. 48. This is

Table 1
LCA results for 18 midpoint indicators for recycling 1 tonne of aluminium-containing material mix in a rotary furnace for salt-slag residue landfill or valorisation treatment and relative improvement when salt-slag is treated.

Midpoint indicator	Salt-slag landfill	Salt-slag treatment	Improvement (%)	Unit
Global warming	−12,267	−13,221	7.8	kg CO_2 eq
Stratospheric ozone depletion	0	0	9.8	kg CFC11 eq
Ionising radiation	−212	−229	8.0	kBq Co-60 eq
Ozone formation, Human health	−32	−34	7.2	kg NO_x eq
Fine particulate matter formation	−26	−27	5.3	kg PM2.5 eq
Ozone formation, Terrestrial ecosystems	−32	−34	7.0	kg NO_x eq
Terrestrial acidification	−56	−59	5.5	kg SO_2 eq
Freshwater eutrophication	−5	−5	8.6	kg P eq
Marine eutrophication	0	0	6.1	kg N eq
Terrestrial ecotoxicity	−9,254	−11,078	19.7	kg 1.4-DCB
Freshwater ecotoxicity	−350	−411	17.5	kg 1.4-DCB
Marine ecotoxicity	−483	−567	17.6	kg 1.4-DCB
Human carcinogenic toxicity	−2,616	−3,141	20.1	kg 1.4-DCB
Human non-carcinogenic toxicity	−11,340	−13,058	15.2	kg 1.4-DCB
Land use	−1,001	−1,131	13.0	m_2a crop eq
Mineral resource scarcity	−163	−203	24.6	kg Cu eq
Fossil resource scarcity	−2,596	−2,855	10.0	kg oil eq
Water consumption	−83	−89	6.8	m^3

logical since the salt-slag treatment includes the additional step of treating the salt-slag for recovery, which uses fuels and energy and generates emissions. However, the salt-slag treatment prevents significant impacts of the waste landfill (176 kg CO_2 eq.). In addition, recovering NMCs saves −394 kg CO_2 eq., ammonium sulphate −63 kg CO_2 eq., and recovering and recirculating NaCl/KCl salts into the rotary furnace reduces the emissions associated with salt production by 31 kg CO_2 eq. Finally, producing secondary aluminium reduced the GWP by −13,355 for the salt-slag treatment and by −12,909 for the salt-slag landfill, being the difference of −445 kg CO_2 eq. due to the Al recovered from concentrates.

However, the rest of indicators may also represent significant environmental impacts and those factors affecting them must be identified. Therefore, for the contribution analysis presented in the next section, five midpoint indicators were selected based on 2 criteria: those that display a higher relevance for the endpoint categories (global warming, fine particulate matter and human carcinogenic toxicity, based on the analysis displayed in Table A-4 in the Appendix) and those that are more greatly affected by a change in scenario (mineral resource scarcity and terrestrial ecotoxicity, as seen in Table 1).

3.2. Contribution analysis

Fig. 2 displays the normalised contributions of each input to the selected impact indicators for Scenario 1 (salt slag landfill) and 2 (salt-slag treatment).

The results show that the impacts of producing secondary aluminium dominate. The contribution of secondary aluminium is higher for the salt-slag treatment route because, as explained in the inventory analysis, this involves recovering additional aluminium from the concentrates in the salt-slag, which then can be recycled instead of landfilled. Recovering non-metallic compounds (NMCs) also show significant benefits for the human carcinogenic toxicity and mineral resource scarcity. To discuss the contributions of the rest of the inputs to the process, Fig. 3 omits the recovered aluminium. The results are also normalised with respect to the salt treatment route.

Fig. 3 allows identifying the hotspots or most relevant potential areas for process improvement apart from minimising the aluminium metal losses. To reduce the process impacts into the GWP, efforts should focus into minimising process emissions and reducing the use of natural gas for thermal energy (e.g. substituting it by green hydrogen), since these appear as the two greatest negative contributors. Avoiding the landfill of the salt-slag and recovering the NMCs also provides substantial benefits. Recovering ammonium sulphate is another positive aspect of the salt-slag treatment, to a much lower extent but enough to compensate the impacts related to the use of oxygen and electricity, which are also visible. The rest of the contributions seem negligible in comparison, although their values can be found in the supplementary material. For the midpoint indicator “fine particulate matter formation”, the relative contributions of the process emissions, the usage of oxygen, sodium chloride, electricity, natural gas, and the recovery of ammonium sulphate are of similar magnitudes. The impact of the salt-slag disposal is roughly three times as much as those mentioned, and the savings from recovering NMCs are the largest. The recovery of NMCs has the most substantial benefits for the indicators “human carcinogenic toxicity”, and “mineral resource scarcity”. The impact of preventing salt-slag landfill is visible for all indicators but especially beneficial for the “terrestrial ecotoxicity”. For this category, the reuse of NaCl and KCl are also significant benefits of the salt-slag treatment. Finally, the recovery of aluminium sulphate also saves substantial impacts on the “terrestrial ecotoxicity”, and the impacts related to using oxygen to fire the furnaces are also visible across all the selected indicators, although they are not affected by the choice of recycling route since the rotary furnace remelting is present in both scenarios. The results in absolute values are plotted in Figure A-1 in the Appendix and included in the supplementary material.

3.3. Discussion

This LCA study describes a realistic scenario for a European recycler and allows comparing the salt-slag management routes and discussing potential areas where the efforts to improve the environmental performance of recycling via a rotary furnace should focus.

The largest contributor to all environmental impact savings was the recovered aluminium. Through the system expansion approach, by producing a secondary aluminium co-product, the market could avoid the production of primary aluminium, which has a high contribution to all impact categories, so discounting it from the total leads to great net environmental impact savings. Still, using GWP as an example, if the allocation of the recovered aluminium is omitted, the recycling process contributions would be 0.64 tonne CO_2 eq. for the scenario where salt-slag is landfilled and 0.14 tonne CO_2 eq. for the route with salt-slag recovery. Damgaard attributed global warming impacts of the same magnitude (ranging between 360–1,260 kg CO_2 eq. t^{-1} aluminium scrap treated) to the recycling processes for aluminium post-consumer scrap in refiners (Damgaard et al., 2009). In (European-aluminium, 2018), there is some data from an environmental assessment for secondary cast alloy production in 2010 in Europe (refining model, using the software GaBi), where the GWP of the recycling processes was 510 kg CO_2 eq. tonne $^{-1}$ secondary aluminium, without considering the benefits from substitution. These numbers confirm that the GWP impacts of secondary aluminium production process are much lower than those from the

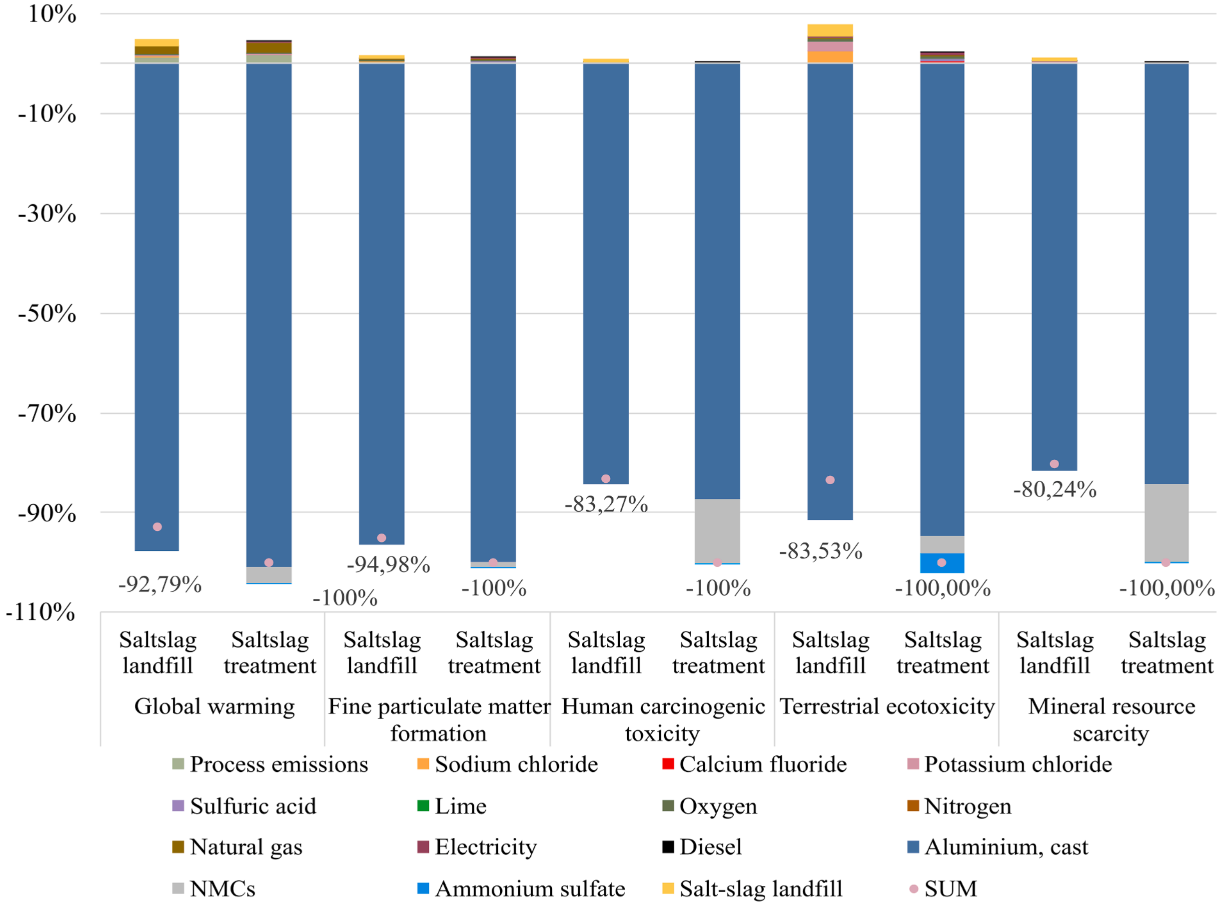


Fig. 2. Contribution analysis to selected indicators during aluminium recycling in a rotary furnace for salt-slag landfill or salt-slag valorisation. Results normalised to 100% of the maximum relative impact per impact category.

primary production route, which was reported by (Liu and Müller, 2012) to generate between 5.92 and 41.10 tonnes of CO_2 eq., depending on the location, energy source and technologies used.

This study has shown that even small increases of 2.3 wt% in the metal yield bring forth significant environmental benefits. Thus, minimising the aluminium losses should be a priority to improve the environmental performance of the recycling and waste management processes. In addition, to improve the sustainability performance of re-melting scrap in a rotary furnace and its consequent salt-slag treatment processes, the results suggest focusing the efforts on maximising energy efficiency, cutting down process emissions and optimising the recovery and usage of NMCs.

The comparison between the salt-slag management scenarios showed that although adding the step of valorising the salt-slag also implies increasing the use of energy, water and raw materials of the process, the recovery of by-products from the salt-slag and its commercialisation or internal recirculation make this route overall more sustainable. The environmental benefits from valorising the salt-slag from aluminium recycling were also reported by Olivieri (Olivieri et al., 2006). Furthermore, salt-flux recycling makes it possible to recover aluminium from scrap which is complicated to recycle through other re-melting methods due to being partly oxidised and/or contaminated (e.g. dross, IBA, post-consumer food packaging). However, if certain scrap types could be recycled efficiently without salts, e.g., by applying a thermal pre-treatment to clean the scrap from organics and then re-melting in a reverberatory furnace, a specific assessment should be conducted to determine the most sustainable and profitable route. This could apply to shavings and packaging waste, but not to partly oxidised scrap as dross or IBA where salt additions are needed to liberate the metal from the oxides it is surrounded by.

The addition of alloying elements or the need to refine the melt was not included in the study, as this will depend on the composition of the scrap and the requirements of the alloy under demand. The typical secondary cast alloys produced at the plant are $AlSi_9Cu_3$ / $AlSi_7Mg_{0.3}$, where Si is the main alloying element, with main applications in the transportation sector. In addition, it should be mentioned that the pre-treatments of scrap, such as shredding and sorting in material recycling facilities (MRF), were not considered. Damgaard (Damgaard et al., 2009) found that these contributions are negligible (6.8 kg CO_2-eq. $tonne^{-1}$ scrap treated) compared to the re-melting process. The emissions associated with the collection and transport of scrap, calculated in (Eisted et al., 2009), were also not considered, as they are not part of the scope of the study. Future work could expand this study by varying the energy/fuel sources or the applications of the recovered oxide by-products (NMCs) in different industries and locations.

4. Uncertainty and sensitivity evaluation

4.1. Sensitivity to the material type and recycling yield

The following sensitivity analysis assesses the influence of varying the metal yield on the GWP of recycling 1 tonne of materials/scrap via the salt-slag treatment route (Scenario 2). The metal yield of the material/scrap depends on one side on its intrinsic content of non-metallic contaminants (moisture, organics, oxides), and on the other side on the metal losses during the high-temperatures processes, which are affected by factors such as its Mg content, specific surface area or the furnace operation (Rossel, 1990; Xiao and Reuter, 2002; Xiao Y, 2005).

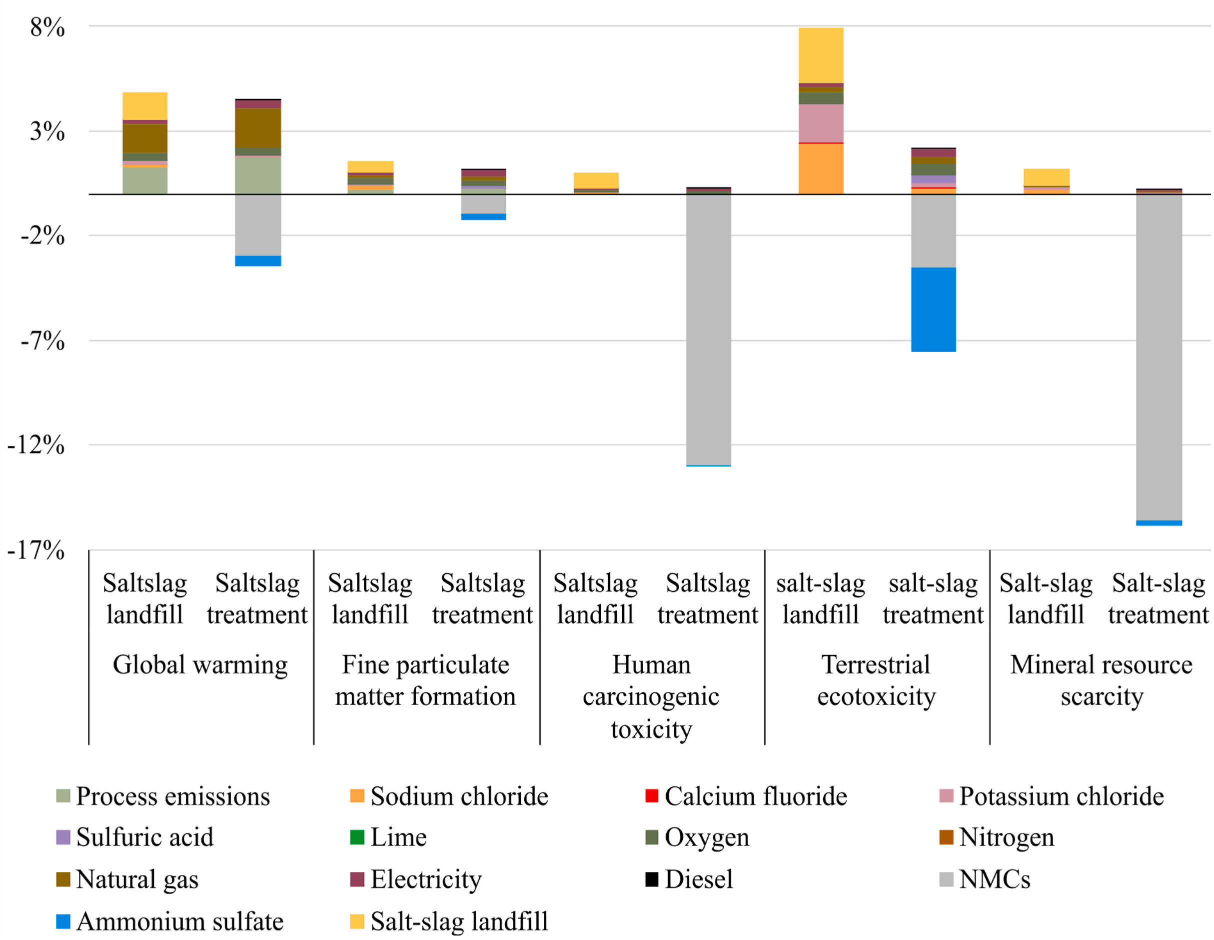


Fig. 3. Contribution analysis after omitting the allocation of recovered aluminium selected midpoint indicators during aluminium recycling in a rotary furnace for salt-slag landfill or salt-slag treatment.

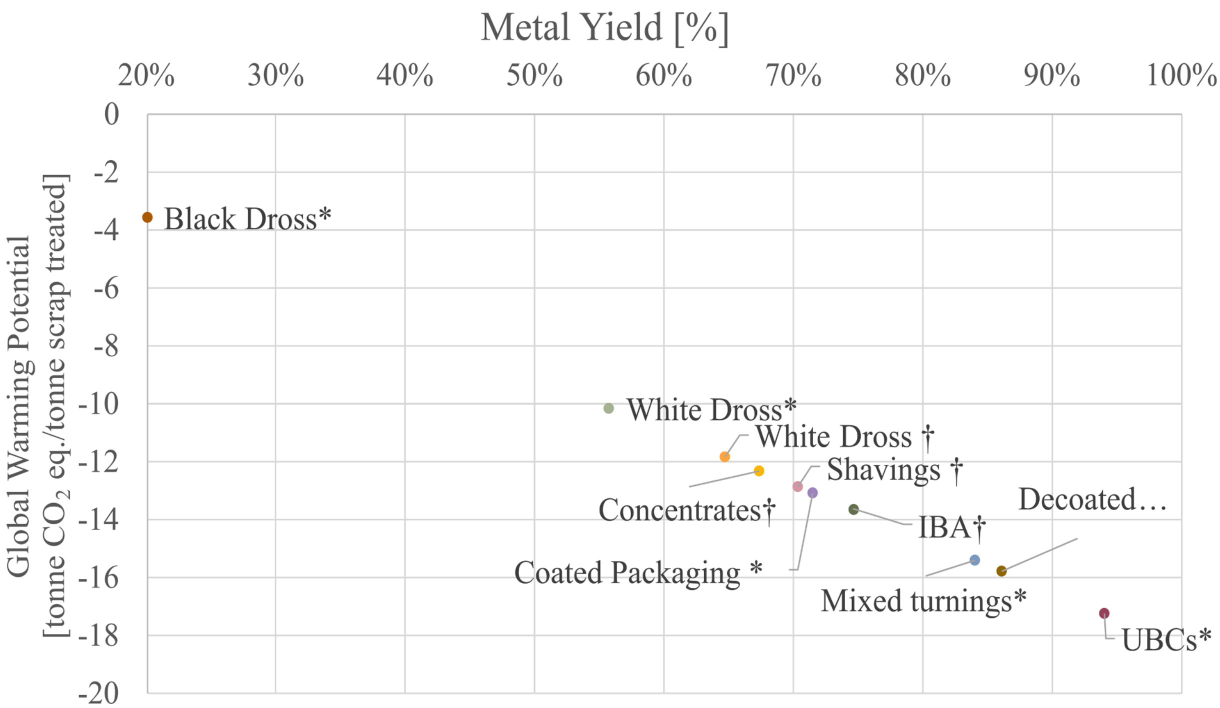


Fig. 4. Calculated global warming potentials for varying scrap types and metal yields. *Average metal yield values from Standard EN13920. †Metal yield values reported by recycling plant.

The average values for metal yields for different scrap categories are collected in Standard EN13920 (CEN, 2003). Some of those values were used for the sensitivity analysis, where the GWP of recycling different scrap types is compared to the materials considered in the present study in the hypothetical case where they would be re-melted in individual batches. The results are presented in the Appendix Table A-5 and Fig. 4 below.

The results show the importance of the metal yield assumptions; recycling 1 tonne of scrap with metal yields between ca. 20 and 95 % varies between −3.5 and −17 tonnes CO_2 equivalent. Therefore, it is advisable that LCA studies which aim to compare recycling routes use fixed metal yield values or include a similar sensitivity analysis. It can be challenging to assess the environmental performance of recycling specific scraps because the metal yields can vary even within the same scrap type. Especially for dross, the metallic content varies drastically depending on the production route. White dross, also known as wet dross, is generated during primary production and has typical metal contents between 15–80 % (Peterson, 2011). However, black dross, also known as dry dross, which originates from secondary production routes, has typical metal contents as low as 10–20 % (Tsakiridis et al., 2013). Still, even for black dross with metal contents as low as 20 % of its weight, recycling saves 3.5 t CO_2 eq./tonne treated. The metal yield of incineration bottom ash (IBA) can also vary depending on its origin and size fraction. For example, in a laboratory study (Gökelma et al., 2021), recycling three different size fractions of IBA from the UK and USA gave average metal contents between 76–79 % for the 2–6 mm size fraction, 83–85 % for sizes between 6–12 mm and 88–89 % for sizes between 12–30 mm. It is also interesting to observe that, according to this sensitivity analysis, recycling 1 tonne of de-coated packaging saves approximately 3 more tonnes of CO_2 eq. compared to recycling coated packaging. According to (Capuzzi et al., 2017; McAvoy B; McNeish, 1990), applying a de-coating treatment reduces the amount of metal lost during re-melting. However, the emissions associated with the additional thermal de-coating pre-treatment process should be controlled by similar off-gas systems as those utilised in the rotary furnace. The emissions from thermally de-coating aluminium products were studied in (Bateman et al., 1999). All considered, since the sensitivity analysis shows that slightly increasing the metal yield brings forth significant environmental benefits, the authors propose that successfully de-coating the scrap before re-melting it or preparing the scrap in other ways that decrease the re-melting losses, even just to a small extent, would improve the net environmental performance of the recycling process.

4.2. Pedigree, uncertainty, and one-at-a-time sensitivity analysis

A pedigree analysis (Ciroth et al., 2016), displayed in Table 2, was carried out for the evaluation of uncertainty. After the pedigree evaluation, the squared standard deviation is incorporated into SimaPro to test how it affects the results of the assessment. A one-at-a-time (OAT) sensitivity analysis is carried out as shown in Fig. 5.

As it was expected and in accordance with the contribution analysis, products and co-products hold the greatest influence in the OAT and can account for more than 3 % of variation. When the product ratio is increased, the environmental performance increases (is more negative) and vice versa. All other parameters effect stays below the 1 % of variation. Regarding the different impact categories, they are similarly affected by the change in parameters, excluding the salt-flux that affects mostly terrestrial ecotoxicity. A significant variation is not found when comparing the different scenarios.

5. Conclusions

This LCA study provides a detailed analysis of the environmental impacts of recycling Al-containing waste/side-streams in a salt-based rotary furnace process with subsequent salt-slag treatment, producing secondary cast Al alloys and byproducts. The main results were:

Treating 1 tonne of hard-to-recycle aluminium-containing streams (mix of dross, IBA and shavings with metal yield∼ 72 wt%) through this recycling route saves 13.2 t CO_2 eq. of contributions to GWP. The main contributors to reducing the global warming potential are the recovery of aluminium concentrates, NMCs, the avoidance of landfill, and to a lower extent the recovery of ammonium sulphate.

The implementation of a salt-slag valorisation process reduces the GWP by 1 tonne of CO_2 eq. and brings forth significant environmental benefits (between 5–25 %) for all other midpoint indicators considered compared to the hypothetical scenario where salt-slag would be disposed at landfill. The midpoint indicators most benefitted by the salt-slag treatment were, in descending order: mineral resource scarcity and human carcinogenic toxicity (due to NMCs recovery), and terrestrial ecotoxicity (due to prevention of landfill, ammonium sulphate and salts recovery). Further efforts to improve the environmental performance the process should focus on minimising metal losses and optimising the recovery and usage of byproducts (NMCs).

Finally, the sensitivity analysis showed how metal yield assumptions have a substantial effect on the environmental performance of the recycling process, e.g., variations between 20 and 95 wt% lead to global warming potentials ranging from −3.5 to −17 t CO_2 eq.

CRediT authorship contribution statement

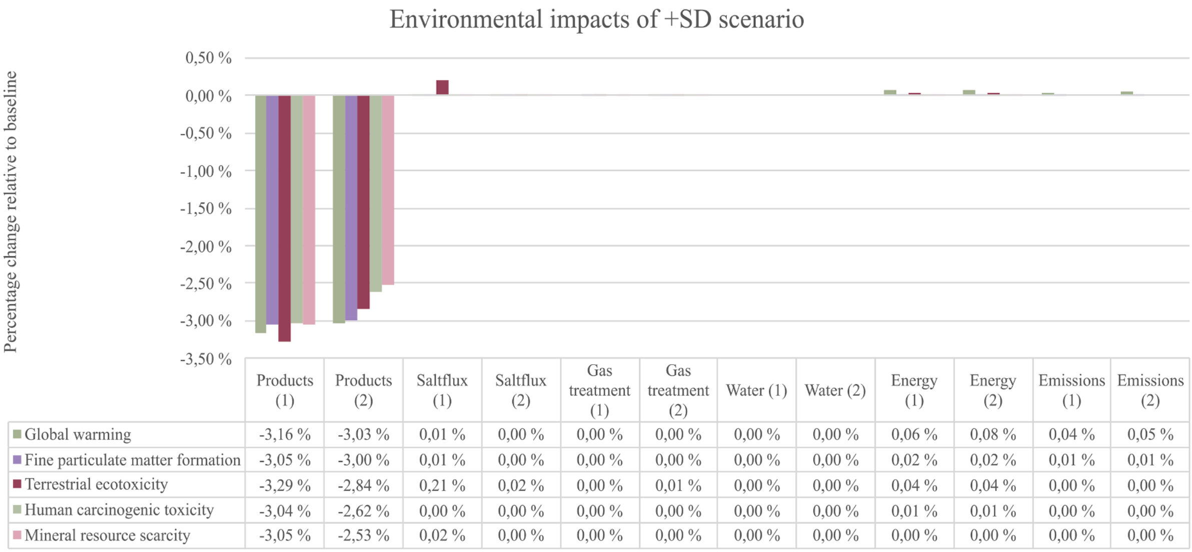
Alicia Vallejo Olivares: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Elisa Pastor-Vallés:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Johan Berg Pettersen:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Gabriella Tranell:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

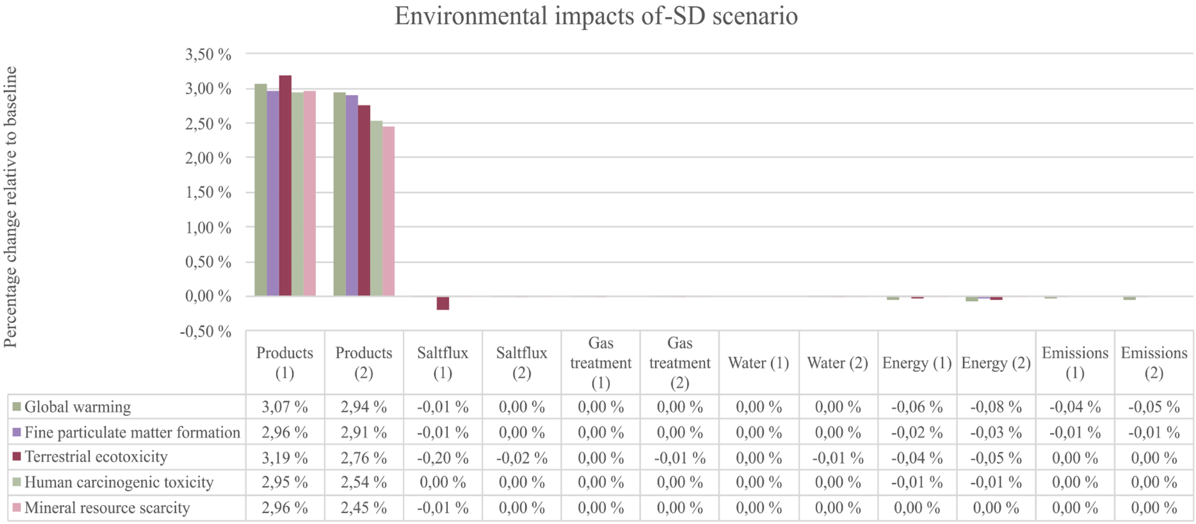
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Table 2
Pedigree analysis (based on Ciroth et al., 2016) of the flow categories used in the LCA study.

Flow	Reliability	Completeness	Temporal correlation	Geographical correlation	Further technological correlation	Squared (geometric) standard deviation (SD2)
Products (Al alloy, NMCs, amm. sulphate)	1	2	1	2	1	1.06
Salt-flux mix	2	2	1	2	1	1.08
Gas treatment (lime and sulfuric acid)	2	2	1	1	1	1.07
Water	2	2	1	1	1	1.07
Energy and fuels	2	2	1	1	1	1.07
Emissions	2	2	1	1	1	1.07
Solid waste (dust and sludge)	3	3	1	3	1	1.13



	Products (1)	Products (2)	Saltflux (1)	Saltflux (2)	Gas treatment (1)	Gas treatment (2)	Water (1)	Water (2)	Energy (1)	Energy (2)	Emissions (1)	Emissions (2)
Global warming	-3,16 %	-3,03 %	0,01 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,06 %	0,08 %	0,04 %	0,05 %
Fine particulate matter formation	-3,05 %	-3,00 %	0,01 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,02 %	0,02 %	0,01 %	0,01 %
Terrestrial ecotoxicity	-3,29 %	-2,84 %	0,21 %	0,02 %	0,00 %	0,01 %	0,00 %	0,00 %	0,04 %	0,04 %	0,00 %	0,00 %
Human carcinogenic toxicity	-3,04 %	-2,62 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,01 %	0,01 %	0,00 %	0,00 %
Mineral resource scarcity	-3,05 %	-2,53 %	0,02 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %



	Products (1)	Products (2)	Saltflux (1)	Saltflux (2)	Gas treatment (1)	Gas treatment (2)	Water (1)	Water (2)	Energy (1)	Energy (2)	Emissions (1)	Emissions (2)
Global warming	3,07 %	2,94 %	-0,01 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	-0,06 %	-0,08 %	-0,04 %	-0,05 %
Fine particulate matter formation	2,96 %	2,91 %	-0,01 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	-0,02 %	-0,03 %	-0,01 %	-0,01 %
Terrestrial ecotoxicity	3,19 %	2,76 %	-0,20 %	-0,02 %	0,00 %	-0,01 %	0,00 %	-0,01 %	-0,04 %	-0,05 %	0,00 %	0,00 %
Human carcinogenic toxicity	2,95 %	2,54 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	-0,01 %	-0,01 %	0,00 %	0,00 %
Mineral resource scarcity	2,96 %	2,45 %	-0,01 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %	0,00 %

Fig. 5. Percentage change compared to baseline by + SD (top) and −SD (bottom). The number in parenthesis represents the scenario considered. (1) = Salt slag landfill; (2) Salt slag treatment.

Data availability

The LCI data is included in the appendix tables (attached as E-component) and the results in the supplementary material

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2024.04.023.

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