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RESEARCH ARTICLE

Alternative Method for Treating Electric Arc Furnace Dust: Simulation and Life Cycle Assessment

Marja Rinne[1] · **Petteri Halli**[1] · **Jari Aromaa**[1] · **Mari Lundström**[1]

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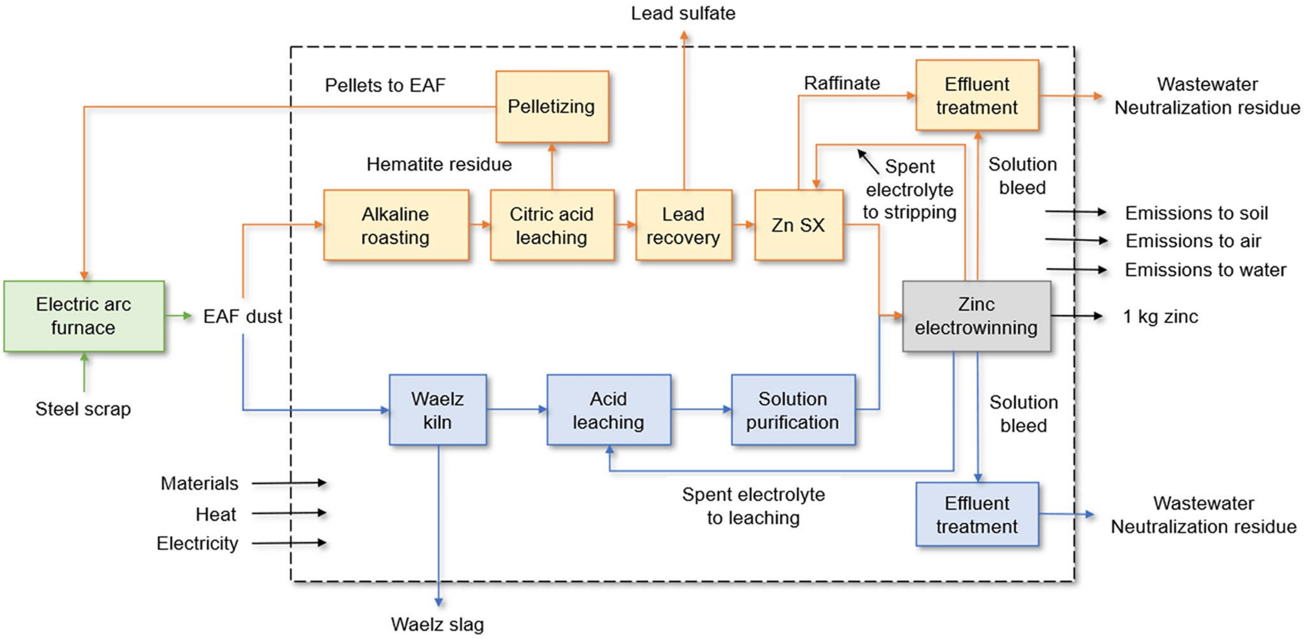
Abstract

The technical and environmental aspects of treating electric arc furnace dust (EAFD) using a novel process including alkaline roasting and organic acid leaching were evaluated and compared to the globally applied conventional Waelz process. The global warming potentials of the roasting–organic acid leaching process (with co-product credits) and the Waelz processes were 7.48 and 4.71 kg CO_2-eq with 33% Zn dust as the feed material. However, with decreasing Zn content, the novel process become environmentally more competitive, and with 12% Zn, the corresponding GWP values were 6.31 and 9.26 kg CO_2-eq. Co-product recovery decreased the net environmental impacts from the processing of the dust, which gave some benefits to the novel process. The Waelz process was observed to be energy intensive; however, the high consumption of leaching and roasting chemicals was the issue in the novel process. The excess caustic soda in the roasting step was observed to be a significant factor in the overconsumption of the organic acid in the leaching stage, and therefore, optimization of the roasting step is crucial. Other ways to decrease leaching chemical consumption include investigating the possibility to reuse the raffinate in leaching, or scrubbing the leach calcine from acid-consuming alkali. Evaluating the impacts of novel processes early enables iterative and agile process development and the directing the focus on the problem areas.

Graphical Abstract



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Keywords Process modeling · Environmental impacts · Zinc · Waelz process · HSC Sim

Introduction

Approximately 30% of steel is produced globally by recycling steel scrap in the electric arc furnace (EAF) process [1]. This process is known to generate significant amounts of electric arc furnace dust (EAFD). EAFD is a heterogeneous industrial waste with wide variation in composition; the literature shows that the dust has a high zinc content, which typically varies between 10 and 35%, averaging 20% [2]. Due to the presence of harmful components in the material, such as Cr^{6+}, Cd, Pb, and halogens, EAFD is considered a hazardous waste that cannot be disposed of as landfill without further treatment [3]. In addition, EAFD contains significant amounts of valuable elements, such as zinc, which makes the dust a significant secondary resource. However, currently only zinc is commonly recovered from the dusts, whereas the other major components including iron are scarcely recycled.

The methods used or investigated in EAFD recycling can be divided into hydrometallurgical and pyrometallurgical processes. The pyrometallurgical Waelz process is by far the dominant process, with over 80% of recycled steel dust treated by the method [4–6]. It is also mentioned in the best available techniques (BAT) for non-ferrous metal industries [7]. Many possible alternatives to the Waelz process have emerged without comparable commercial success, the rotary hearth furnace (RHF) process being the most mature [1, 8]. One of the advantages of the process is that iron may be recovered as direct reduced iron instead of low-value slag. RHF plants have, however, in practice suffered from performance issues, and Piret [9] has argued against the perceived successes of the technology. Hydrometallurgical processes, where aqueous solutions are used for metal recovery, have lately received some interest. They could provide less capital-intensive alternatives for the treatment of smaller feed volumes or low-zinc dusts that cannot economically be treated pyrometallurgically [5, 10]. Some hydrometallurgical processes, such as the EZINEX and Zincex processes, have also been commercially developed and piloted [11, 12], but only the ammonium chloride-based EZINEX process has operated industrially [4].

Hydrometallurgical processes are more sensitive to the mineral species in the EAFD than pyrometallurgical processes, which complicate process development. Most of the zinc present in the dust is typically either in the form zincite (ZnO) or zinc ferrite ($ZnO\bullet Fe_2O_3$), of which the former is easily soluble, but the latter requires aggressive conditions to dissolve. Furthermore, the leaching of zinc ferrite also dissolves iron, which is a harmful impurity in hydrometallurgical circuits [2, 13]. The generation of secondary waste and the presence of halides in the dust create further challenges for recycling [6, 14]. Alkaline ammoniacal or sodium hydroxide solutions have been shown to have good selectivity for zinc over iron, but zinc ferrite does not dissolve in alkaline conditions [2]. Mineral acid solutions, such as sulfuric, nitric, and/or hydrochloric acid, can break down zinc ferrite but with rather poor selectivity, which results in excess iron in the solutions and generation of secondary waste [2, 15]. Organic lixiviants, such as citric acid [2, 16], oxalic acid [17], and monosodium glutamate [18], have been suggested in the literature as environmentally benign alternatives. However, avoiding the use of harmful lixiviants or high temperatures in processing does not guarantee sustainability. Furthermore, problems may also arise from excessive chemical consumption or wastewater management, which may create significant indirect environmental burdens.

Life cycle assessment (LCA), a standardized framework for the assessment of environmental impacts, can be used to determine the environmental footprint of a product or a process from cradle-to-grave [19, 20]. In the case of novel processes, process modeling or other data estimation methods have been possible tools in the generation of life cycle inventory (LCI) data [21]. Some previous studies exist on the treatment of galvanized steels and EAFD [22–24]. Viklund-White [22], for instance, compared several recycling routes including Waelz and EZINEX processes for zinc from galvanized steels to landfilling and found that the impacts from recycling are higher than landfilling in the studied categories, unless direct current (DC) furnace route is applied with a clean Norwegian electricity mix. The study by Viklund-White [22] only considered primary energy consumption, global warming, and acidification categories, where landfilling has no meaningful impacts. Exergy analysis has also been used to study the resource efficiency of pyrometallurgical EAFD treatment routes: Waelz, RHF, and novel in-process separation [23]. The Waelz process was the least exergy efficient of the studied processes due to the production of waste slag, whereas RHF and the novel process were able to recover iron.

In addition to EAFD treatment, the impacts from secondary zinc recovery from other types of zinc-rich wastes have been investigated. Muica et al. [25] conducted a life cycle assessment on the recycling of zinc smelting slags in the Waelz process and compared the option to the smelting of zinc concentrates in imperial smelting process (ISP). While the recycled zinc from ISP slags had lower impacts than primary zinc from concentrates, the study did not consider the fact that primary zinc is predominantly produced hydrometallurgically. Briffaerts et al. [26] investigated recycling routes for zinc batteries, concluding that the impacts and benefits are different between the options and that there are no typical pyrometallurgical or hydrometallurgical processes.

In light of the fact that each process results in a different environmental profile, it is vital to critically evaluate also novel processes for their impacts. Halli et al. [16] developed an entire flowsheet for the electrolytic recovery of zinc from thermally pre-treated EAFD using citric acid solutions by conducting lab-scale experiments. The advantage of the process is that the hematite residue may be recycled back to the EAF for iron recovery. In the current work, the development stage roasting–citric acid process presented by Halli et al. [16, 27] was compared against the conventional pyrometallurgical Waelz process to recognize possible weaknesses and merits in the novel process. Both processes were simulated with Metso Outotec's HSC Sim 9 software [28], and the mass and energy balances were converted into internally consistent life cycle inventories. The environmental impacts were then calculated using Sphera's GaBi software [29].

Table 1 Quantitative mineral composition (wt%) of the dust used in process models: *Base* [2]; *Low* [34], and *Average* [15]

Mineral species	Base [%]	Low [%]	Average [%]
$ZnFe_2O_4$	23.1	7.5	11.0
ZnO	31.2	10.1	14.9
ZnS	3.1	3.1	3.1
Fe_3O_4	14.6	46.5	30.6
PbO	1.6	1.9	1.4
MnO	2.6	2.6	1.3
Cr_2O_3	0.6	0.6	0.6
NaCl	6.4	6.4	6.4
KCl	3.7	3.7	3.7
CaO	3.9	3.1	6.2
SiO_2	6.7	12.8	18.3
MgO	1.2	1.2	1.2
C	1.4	1.4	1.4
Zn / Fe / Pb grade	33.4 Zn, 21.3 Fe, 1.5 Pb	12.2 Zn, 37.1, Fe, 1.7 Pb	17.1 Zn, 27.2 Fe, 1.3 Pb

Materials and Methods

The impacts and process performance of the conventional pyrometallurgical EAFD treatment process were compared against roasting–citric acid processing for the recovery of zinc and possible by-products. The LCA methodology involved four phases: 1. goal and scope definition, 2. life cycle inventory (LCI) analysis, 3. life cycle impact assessment (LCIA), and 4. interpretation. The analysis was conducted according to ISO 14040:2006 [19].

Process simulation, using Metso Outotec's HSC Sim 9 software, was used to calculate the mass and energy balances of the processes and to compile the LCI, while the impact assessment was conducted using Sphera's GaBi (v. 10.5.0.78) software. A similar methodology has been used before to investigate the impacts of rare earths production [30], copper production [31], and the treatment of secondary raw materials [32, 33].

Raw Materials Used

The nominal composition of the EAFD (Ovako Imatra Oy Ab, Finland) was obtained from Halli et al. [2] and modified into a mineral composition to model the reactions. Experimental work conducted on the roasting, leaching, and solution purification on the original (*Base*) composition is presented in other publications [2, 16, 27].

The recycling models were tested with three different EAFD compositions, shown in Table 1. The zinc contents were 12.2% (*Low*) [32], 17.1% (*Average*) [15], and 33.4% (*Base*) [2]. The elements that were not analyzed in the characterizations of Dutra et al. [34] and Kukurugya et al. [15] were assumed to be equivalent to those analyzed by Halli et al. [2]. In addition to the included major elements, smaller amounts of other elements, such as copper, aluminum, vanadium, nickel, and phosphorus were present as well as sodium and potassium as chlorides. Fluoride may be present in small quantities, but it was not analyzed, and it was assumed to behave the same as chloride in the process. The minerology analysis was balanced with inert quartz (SiO_2). A similar method has also been used to estimate the mineral composition of gold ores and secondary raw materials [33, 35].

Process and Simulation Descriptions

HSC Sim 9 [28] was used to calculate the mass and energy flows in the two processes: the Waelz and roasting–citric acid process (RCAP), to compare the mass and energy balances and to compile an internally consistent LCI for the analysis. More detailed process descriptions, model assumptions, and the design parameters used in constructing the simulation models can be found in the supplementary material.

From here on in this study, the term "Waelz process" is also used to refer to the subsequent hydrometallurgical refining of Waelz oxide, the crude zinc oxide product from the pyrometallurgical process, into cathode zinc by sulfuric acid leaching and electrowinning (EW). Italic is used to refer to the simulation models of the processes.

Waelz Process Description

The Waelz process was first operated industrially by Krupp Grusenwerk and Metallgesellschaft A.G. in 1923 [36], after which the process has continued to see improvement in terms of capacity, costs, and environmental impacts [9]. The process is operated in reducing conditions at elevated temperatures (1150–1200 °C) to separate zinc from the material through reduction and volatilization [37] in a horizontal rotary kiln. Air is blown to the furnace countercurrent to the charge, so the zinc vapor ($Zn_{(g)}$) re-oxidizes above the tumbling furnace bed, and the zinc oxide is carried by the process gas as fine dust that is collected from the gas filters (Waelz oxide). Waelz oxide also contains other volatile impurities, such as sodium, potassium, lead, and halogens, which is why hydrometallurgical refining needs to be applied to obtain pure zinc. In addition to Waelz oxide, the process generates large amounts of solid slag that mainly consists of reduced iron, iron oxides, furnace sealings, and slag formers. The slag is usually disposed of in landfills because the heavy metals remaining in the slag typically prevent its utilization as a construction material [38]. The slag is either acidic (silica-based) or basic (lime-based). In the simulation, silica sand flux was applied to produce an acidic slag with basicity index ($CaO + MgO/SiO_2$ ratio) of 0.4.

Halogens are harmful to the electrowinning of zinc, and therefore, Waelz oxide is not a suitable feed material for electrolytic zinc production without pre-treatment. In this work, an alkaline sodium carbonate solution washing circuit was applied as the pre-treatment method, assumed to decrease the total halogen content below 300 ppm [39], which is the known accepted level in Zn EW. The washed Waelz oxide is leached in sulfuric acid in two leaching stages, resulting in a pH 5 zinc-bearing solution that is fed to solution purification [40]. A standard zinc cementation process was used to remove the metallic impurities and to obtain a pure electrolyte for zinc EW [40].

Roasting–Citric Acid Processing

The citric acid process consists of roasting the dust at 450 °C with NaOH to decompose zinc ferrite ($ZnO{\bullet}Fe_2O_3$) into soluble sodium zincate (Na_2ZnO_4) and citric acid-insoluble sodium ferrite (Na_2FeO_2) [27]. Furthermore, halogens (F, Cl), which are a problem for zinc EW, are also efficiently removed from the raw material during the roasting step. Volatilized halogens may be captured from the gas by adsorption into solution. It should be noted that the roasting step was not optimized, and it was conducted with twice the theoretical stoichiometric amount of NaOH to guarantee full zinc ferrite decomposition. The excess NaOH also resulted in increased acid consumption in the subsequent leaching stage [27]. The leaching is conducted in 0.8 M citric acid solution, which selectively dissolves lead and zinc, leaving an iron-rich residue that is suitable for feeding into iron production after drying and pelletizing. The conditions for leaching were a solid-to-liquid ratio of 100 g/L (not optimized), 0.8 M citric acid (optimized), 40 °C (optimized), and 60 min (optimized) [27].

After leaching, lead is removed by sulfate precipitation. The concentrated sulfuric acid used for lead precipitation also decomposes the citric acid. The separation of manganese and iron from zinc is achieved by SX with D2EHPA, where zinc is selectively stripped by 1 M sulfuric acid solutions. Iron and manganese are also extracted to D2EHPA but not stripped, so the organic needs to be scrubbed with an HCl solution periodically to prevent the accumulation of impurities. The purified zinc-rich solution is then subjected to a conventional zinc EW process [41].

Life Cycle Assessment

The LCA study was performed based on the guidelines of ISO 14040:2006 [19] and ISO 14044:2006 [20]. Contribution analysis and sensitivity analysis were used as the primary methods for interpreting the results.

Goals and Scope

The goal of the study was to compare the experimentally demonstrated RCAP concept with the most widely used pyrometallurgical processing option, the Waelz process. The aim of the study was also to assess the issues that need to be addressed in the development of sustainable hydrometallurgical processes for EAFD and other zinc-bearing secondary raw materials within Europe.

The processes were modeled with HSC Sim 9 metallurgical simulation software [28] to obtain the mass and energy balances, which were used as the basis for the life cycle inventories. In addition to inventory analysis, the process simulation was used to aid in the interpretation of the results.

System Boundaries and Functional Unit

The functional unit of the analysis was 1 kg of zinc cathode (assumed commercial purity, 99.995%) from processed EAF dust. The analysis was done gate-to-gate: only the processing of EAF dust to zinc was included within the technical boundary, which is presented in Fig. 1. The refining of the by-products was not included. Instead, a recovery credit was given for the lead sulfate and iron intermediates based on their respective valuable metal content, i.e., the iron and lead products were considered to substitute metals from other sources in the market with a 1:1 substitution ratio. Pre-treatment of the wastewaters by neutralization and hydrolysis was within the system boundary.

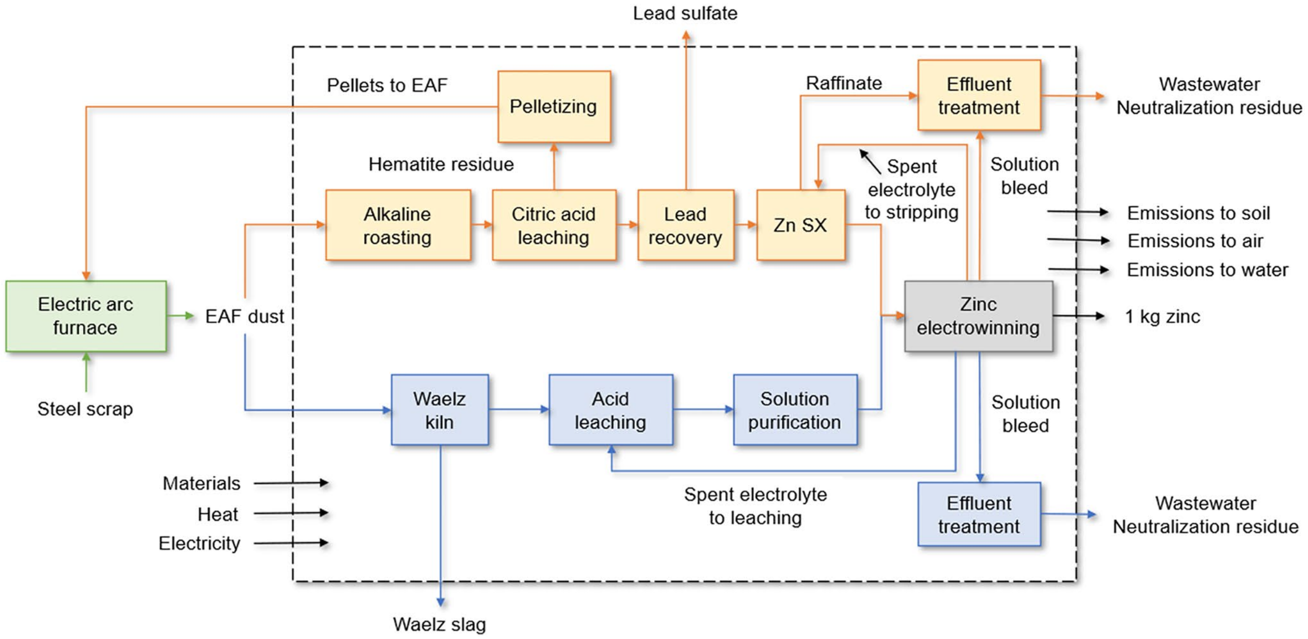

Fig. 1 The technical system boundary (dashed line) of the compared processes: the state-of-the-art *Waelz process* (blue) and *roasting–citric acid process (RCAP)* (orange/yellow) (Color figure online)

Impact Assessment

The impact assessment (LCIA) stage was conducted with GaBi software using the CML 2001 midpoint methodology. The included impact category indicators were global warming potential (GWP, kg CO_2-eq), acidification potential (AP, kg SO_2-eq), freshwater eutrophication potential (EP, kg Phosphate-eq), ozone depletion potential (ODP, kg R11-eq), and photochemical oxidant creation potential (POCP, kg Ethene-eq), as recommended for the metals processing industry by [42].

The global warming potential is used to measure the heat trapped in the atmosphere by greenhouse gases (GHGs, such as CO_2, N_2O, CH_4) based on characterization factors assigned for each pollutant. Similarly, the acidification potential is used to quantify the acidifying effect of various emissions (mainly SO_x, NH_3, NO_x, HCl), and the eutrophication potential quantifies nutrient emissions (phosphorus and nitrogen). Hydrochlorofluorocarbons (HCFCs) and chlorofluorocarbons (CFCs) are the main emissions contributing to the ozone depletion potential. The photochemical oxidant creation potential mainly measures emissions that act as precursors to photochemical smog, such as NO_x and volatile organic compounds (VOCs).

Life Cycle Inventory

The inputs and outputs are tabulated in Table 2 for the *Base* EAFD composition for both the *Waelz* and *RCAP*. More detailed data for each of the process steps and the LCI for all the cases are provided in the supplementary material. Relevant background data were obtained from the ecoinvent 3.7 database [43].

The European average electricity generation mix was used for power generation, while heat was assumed to be generated with natural gas. The Waelz kiln was heated by a combination of natural gas and coke, which also acted as the reductant in the process. The consumption of energy as fuels, heat, or electricity was approximately 20.6 MJ in the *RCAP* and 32.9 MJ in the *Waelz process* for 1 kg zinc output.

SX was applied in the citric acid process for the separation of zinc from iron and manganese. The organic phase consisted of D2EHPA diluted by kerosene, but this was not included in the LCI in the absence of background production data for the extractant. Organic losses occur in the process through several mechanisms, which are dependent on many factors including external conditions, equipment, and the system itself [44]. The consumption of make-up organic could therefore not be determined from the simulation alone.

The inventory above gives a simplified estimation of the outflows from the process. This excludes the release of metals as particulates or from the solid wastes. The composition of solid residues and slags was estimated using the simulation to assess their potential hazardousness. Trace metals will also be present in the wastewater and the water in the tailing ponds, and the possible quantities of these metals can be approximated with the simulation.

Table 2 Life cycle inventories for both *RCAP* and *Waelz processes* with *Base* composition (33.4% Zn). The quantities are expressed for the functional unit (/1 kg zinc product)

Roasting–citric acid process			Waelz process		
Input	Quantity	Unit	Input	Input	Unit
EAF dust	3.03	kg	EAF dust	3.07	kg
Electricity	13.91	MJ	Electricity	16.70	MJ
Natural gas	6.72	MJ	Natural gas	8.30	MJ
Water	31.12	kg	Water	6.19	kg
Citric acid	5.30	kg	Coke	0.33	kg
Air	0.77	kg	Air	6.93	kg
Caustic soda	0.84	kg	Silica fluxes	0.18	kg
Sulfuric acid	0.21	kg	Sulfuric acid	0.12	kg
Hydrochloric acid	0.02	kg	Sodium carbonate	0.30	kg
Lime, hydrated	1.13	kg	Lime, hydrated	0.09	kg
			Zinc powder	0.01	kg

Roasting–citric acid process			Waelz process		
Output	Quantity	Unit	Output	Quantity	Unit
Zinc	1.00	kg	Zinc	1.00	kg
Iron pellets	1.42	kg	Waelz slag	1.39	kg
Lead sulfate	0.06	kg	Hydrometallurgical residues	0.61	kg
Wastewater	41.76	kg	Wastewater	8.63	kg
Solid waste	0.05	kg	Carbon dioxide	1.73	kg
Carbon dioxide	0.15	kg	Sulfur dioxide	0.07	kg
Sulfur dioxide	0.06	kg	Sulfate, aqueous	0.24	kg
Sulfate, aqueous	0.09	kg	Chloride, aqueous	0.14	kg
Chloride, aqueous	0.18	kg			

Coke was assumed to be used as the reductant in the Waelz kiln, and the fixed carbon content was assumed to be 90% [45]. The volatile matter in the reductant was assumed not to provide any reducing power to the system. Excess coke not consumed in the process can be recovered from the fine fraction of ground and sieved slag by magnetic separation where carbon is concentrated in the non-magnetic fraction. Coke separation was also included in the simulation as a black box model [38].

Results and Discussion

Additionally, sensitivity analysis was performed for the inputs in the process. The scope of the current work was a technical and environmental analysis of the prospective process compared to the state-of-the-art *Waelz process*; however, the economics were not considered. Observations from the simulation are available in the supplementary material.

Environmental Impacts

Impacts for both investigated processes with a variety of raw material compositions are provided in Fig. 2. It was found that, regardless of the zinc content in the raw material, the impacts for the *RCAP* were almost consistently higher than the *Waelz process*. GWP and POCP were an exception to this using the *Low Zn* (12% Zn) raw material. In the *Waelz process*, the impacts increased steadily with decreasing zinc content, while the situation was more complex for the *RCAP* due to the by-product credits (iron and lead concentrate by-products). The recovery credit had little effect in terms of AP, but in the other categories, the impacts either decreased or began to stabilize with between *Average* and *Low* compositions.

For the *Base* composition, the GWPs were 4.69 kg CO_2-eq for a kilogram of zinc product in the *Waelz process* and 8.67 kg CO_2-eq for the *RCAP* if no recovery credits were given for iron and lead. With the by-product credit, the GWP of *RCAP* decreased to 7.45 kg CO_2-eq. For the *Low* composition, the respective values for the *Waelz* and *RCAP* (iron

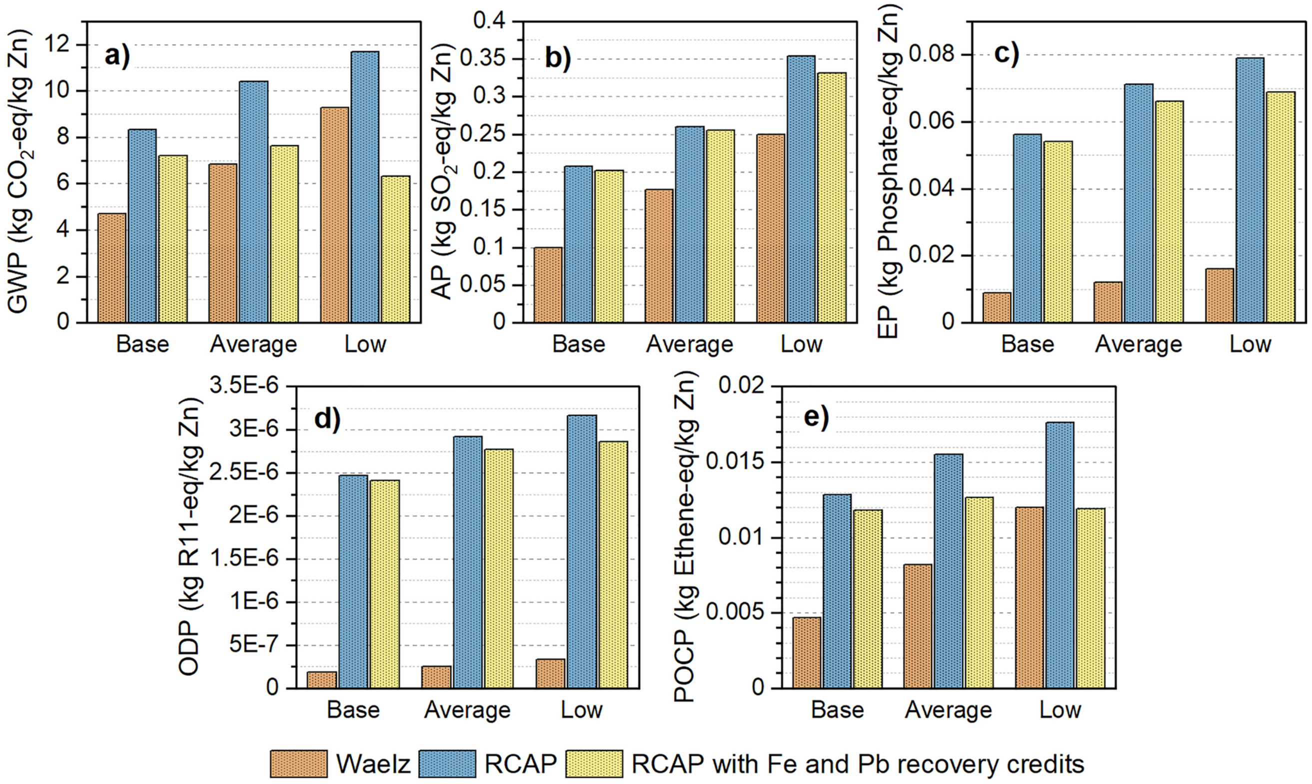

Fig. 2 The impact characterization results for the functional unit (1 kg zinc), values for the *RCAP* are presented with and without the by-product credits. **a** Global warming (kg CO_2-eq). **b** Acidification (kg SO_2-eq). **c** Eutrophication (kg Phosphate-eq). **d** Ozone depletion (kg R11-eq). **e** Photochemical ozone creation (kg Ethene-eq)

and lead un-credited) were 9.72 kg CO_2-eq and 11.94 kg CO_2-eq. The iron and lead recovery credit decreased the GWP value of the *RCAP* down to 6.31 kg CO_2-eq. The differences between the processes decreased in the GWP category; however, the effect was not seen in the AP category due to the environmental profile of iron production. In terms of EP and ODP particularly, the gap remained so large that it is unlikely for the *RCAP* to become the environmentally preferred option in terms of these environmental categories.

The results suggest that the *RCAP* could have some benefits in terms of total GHG emissions (GWP) and smog precursors (POCP) for low-zinc dusts. A very low zinc content ($< 10\%$) in steelmaking dusts could also render the process feasible in terms of AP, but this was not seen with the dust compositions studied. The hotspot analysis of the *RCAP* (Fig. 3) shows that the benefits are mainly due to the recovery of iron, which is GHG-intensive in the case of primary ores. The *RCAP* also only had lower impacts than the *Waelz process* in the GWP category when the zinc content in the raw material was 10%, or presumably also below that. Such low zinc contents are more typical for basic oxygen steelmaking than electric arc furnace dusts [46], and this would limit the feasible feed materials to low volumes. It should also be observed that the decay of citric acid to carbon dioxide and water was not accounted for in the model, which results in underpredicted GWP values. It was also observed that the roasting and leaching steps covered most of the impacts in the *RCAP*, where the composition had only a small effect on the relative contributions of the subprocesses.

The same analysis was performed for the *Waelz process*, Fig. 4. In the *Waelz process,* the pyrometallurgical units were the main contributor in the investigated impact categories regardless of the EAFD composition, apart from the ODP and EP of *Base* composition. In the case of *Base* composition, EW and pyrometallurgical units contributed to the ODP and EP equally (approximately 40% of the total impacts for both units). The contribution of the hydrometallurgical units, including EW, decreased with decreasing zinc content in the raw material while the contribution of the pyrometallurgical units increased with a similar trend in all impact categories. For example, the share of the pyrometallurgical units with *Base* composition in the EP category was 38.6% and 51.2% with *Low* composition. This was because of the decreased throughput to the hydrometallurgical refining in the *Waelz process* (*Low* Zn) and the fact that more of the reductant was consumed in reducing iron in the slag rather than the zinc product.

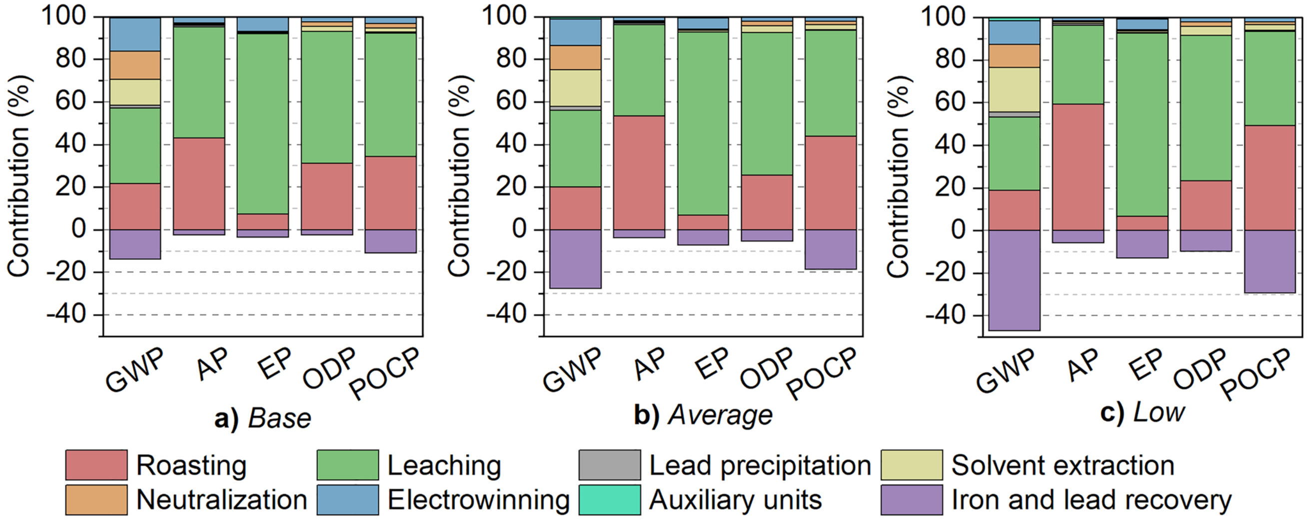

Fig. 3 Contributions of units in the *RCAP* to the total impacts (%) with all the raw material compositions used; "Auxiliary units" refers to general electricity consumption. **a** *Base* composition. **b** *Average* composition. **c** *Low* composition

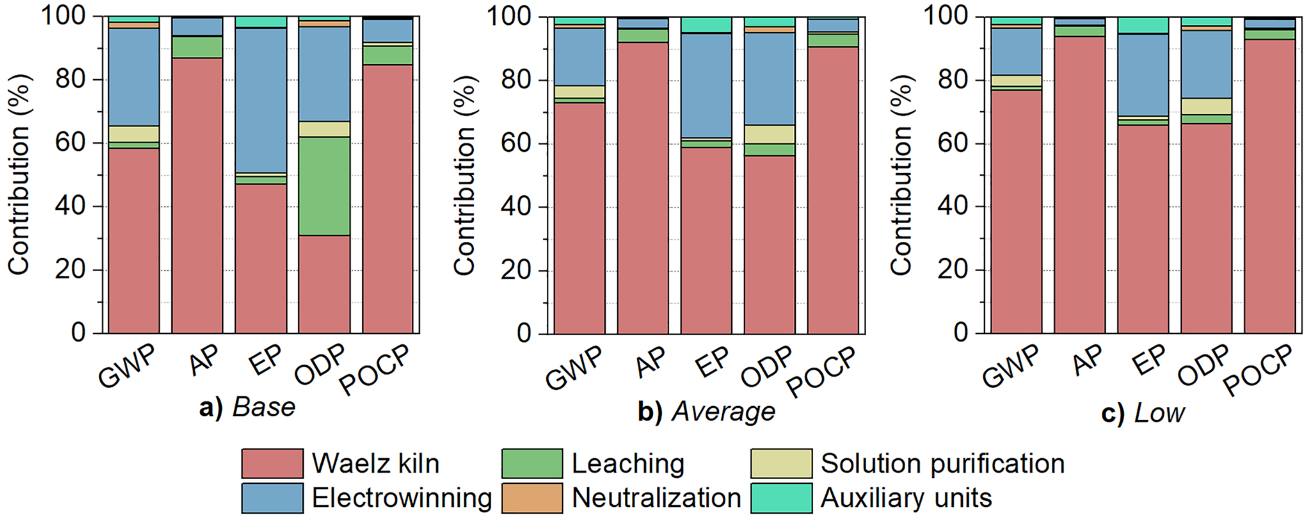

Fig. 4 Contributions of units in the *Waelz process* to the total impacts (%) with all the raw material compositions used; "Auxiliary units" refers to general electricity consumption. **a** *Base* composition. **b** *Average* composition. **c** *Low* composition

The input-specific contribution analysis (Fig. 5) shows that the impacts of the *RCAP* are largely due to the production of the lixiviant (38% GHGs), roasting chemical (caustic soda, 15% of GHGs), and neutralizing chemicals (lime, 14% GHGs). In the *Waelz process*, energy consumption and direct carbon and sulfur dioxide emissions prevailed. In terms of GWP alone, approximately 25% of the emissions occurred directly due to the use of coke and natural gas in the process, whereas electricity production accounted for 42% of GHGs. The differences between the raw material compositions were extremely subtle, and the breakdown for the other compositions is available in the supplementary material.

The differences between the methodologies and system boundaries generally make the results of different LCA studies not directly comparable. However, the findings of the current work can be compared with the other existing studies on EAFD treatment. Most of the studies have found that the alternative processes outperform Waelz process either environmentally or in terms of energy efficiency: the results of Viklund-White [22] indicated that the DC furnace process and EZINEX resulted in lower GHG emissions than the Waelz process, which in turn was better than steel scrap dezincing. The DC furnace consumed less coke than the Waelz process but is more electricity intensive, which makes the use of clean energy more critical: an observation also made

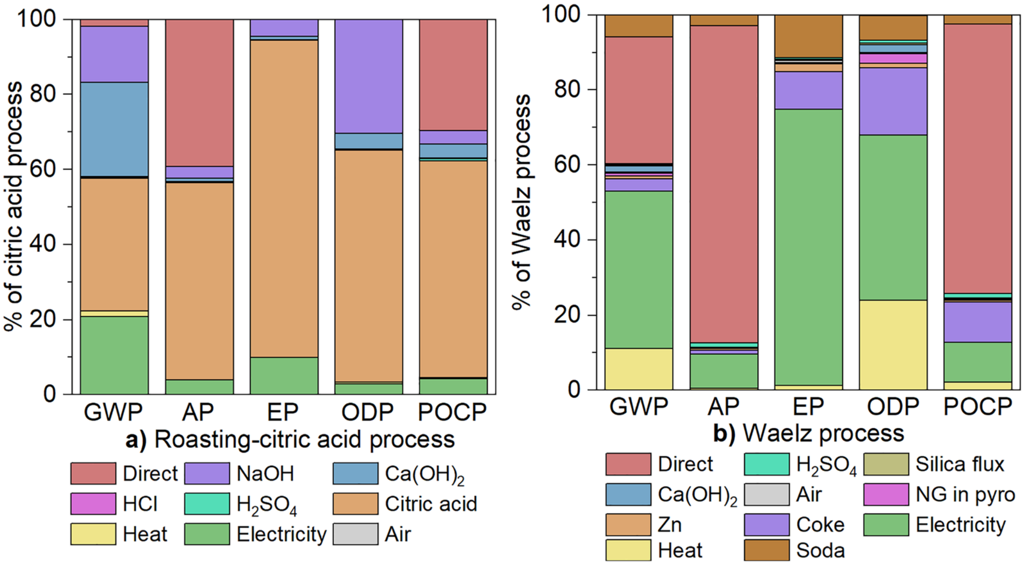

Fig. 5 Contribution analysis of individual inputs and direct emissions to the impacts with *Base* composition. **a** *RCAP*, **b** *Waelz process*. "NG in pyro" refers to the extraction of the natural gas burned in the Waelz kiln

by the author of the study. In the current work, Waelz process and zinc electrowinning were observed to be electricity intensive and thus greatly benefit from decarbonization of the energy mix.

Nakajima et al. [24] predicted considerable energy and GHG savings from their "lime addition and magnetic separation" (LAMS) process, but the resolution of the LCI was too poor to make any further judgements about the alternative process. Exergy analysis methodology, which can be used alongside LCA to measure process performance, showed RHF and in-process separation in the EAF to be more exergy efficient than the Waelz process [23]. In the case of Nakajima et al. [24] and the current work, any benefits from the alternative process are largely reliant on useful co-products. The contamination of the iron product by impurities poorly tolerated in steel processes, such as sulfur, could therefore easily negate any of the observable benefits.

Overall, the comparison shows that the lixiviant consumption in the *RCAP* is the most critical factor that renders the process unfeasible in comparison with the Waelz kiln process. The most obvious challenge is the high consumption of the lixiviant, which cannot be completely avoided due to the decomposition of citric acid in the process. Citric acid provides selectivity over mineral acids in the leaching process [2], which enables high iron recovery. Nevertheless, further optimization of the system is possible. The roasting step, for instance, has not been experimentally optimized for caustic soda consumption or temperature, and the effects of process variables in roasting have not been investigated.

While many organic lixiviants are non-toxic, the results would seem to indicate that organic acid-based hydrometallurgical processes are not necessarily environmentally benign, and a thorough assessment is necessary to prove the sustainability of the process. The natural degradation of organic lixiviants is often seen as proof of their environmental soundness [47], but their production impacts may often be overlooked. Furthermore, the ability to regenerate or reuse organic acids has scarcely been considered in the experimental literature with only few exceptions. Metal removal from citrate liquors to regenerate the chemicals has been applied in wastewater treatment from soil washing [48], but this is unlikely to be feasible or effective in concentrated leaching solutions. Precipitation of the leaching reagent has been applied for sodium glutamate [18]. Folens et al. [49] successfully recycled citric acid leachate by pH adjustment for several leaching cycles after electrochemical lead sulfate recovery despite observing some citric acid oxidation. Studying the decomposition of citric acid and the reuse of the acidic raffinate from SX in calcine leaching could, based on the contribution analyses, be critical in improving the feasibility of the process.

Sensitivity Analysis

Although the results appear clear, sensitivity analysis was performed for a number of selected process parameters to assess their effect on the overall results. The studied variables and parameters were selected based on the contribution analysis, e.g., the impacts of the *RCAP* were primarily from chemical (caustic soda, acid, lime) consumption, whereas the use of fossil fuels and electricity was more significant for the *Waelz process*.

With the *RCAP*, the studied variables were caustic soda consumption in roasting, solid/liquid ratio in leaching, free acid content in the final PLS, and energy consumption in EW. In contrast to the chemically intensive *RCAP*, the impacts from the *Waelz process* were mainly from fuel consumption, either directly or indirectly. The selected variables and parameters for the *Waelz process* were the iron reduction degree in the Waelz kiln, excess coke feed to the kiln, power consumption in the kiln and the electrostatic filter, and energy consumption in EW. The values were changed by ± 20% from the baseline to assess the response.

The consumption of caustic soda in the roasting step is theoretically 4 mols of NaOH for 1 mol of zinc ferrite, i.e., 153 kg for 1 tonne of *Base* composition EAFD. Twice the stoichiometric amount of caustic soda was assumed to be fed to the roasting step, which was used as the baseline in the model and in the sensitivity analysis. The solid/liquid ratio in citric acid leaching was 100 g/L, and the baseline free acid concentration for the *Base* composition was 0.8 M. In EW, 2.8 kWh of electricity was assumed to be consumed in the deposition of 1 kg zinc on the cathode. It should be observed that the typical range for zinc EW power consumption is 2.6–2.8 kWh/kg Zn when the anodic reaction is oxygen evolution [40], indicating that + 20% and − 20% are not realistic. The analysis was, however, conducted uniformly for all the investigated parameters.

In the Waelz process, 90% of the iron reduces to metallic iron, while the rest remains as wüstite (FeO), so the + 20% value was limited to 100% metallization. The coke feed to the furnace was assumed to be 200 kg for 1 tonne of feed, and the power consumption of the kiln was estimated at 107 kWh for a tonne of feed [50] and approximately 0.2 kWh for 1 m^3 treated gas [51]. The results are provided in Table 3.

Table 3 Sensitivity analysis results for the *RCAP* and *Waelz processes* by changing selected parameters or variables by 20%, *Base* composition

	GWP	AP	EP	ODP	POCP
RCAP					
NaOH use in roasting (mol/mol $ZnFe_2O_4$)					
+ 20%	+ 6.9%	+ 4.5%	+ 7.1%	+ 10.1%	+ 4.9%
− 20%	− 7.8%	− 4.5%	− 7.6%	− 10.9%	− 5.6%
Solid/liquid ratio (g/L)					
+ 20%	− 0.8%	0%	− 0.5%	− 0.8%	− 0.4%
− 20%	+ 0.1%	0%	+ 0.2%	+ 0.4%	+ 0.1%
Free acid in leaching (mol)					
+ 20%	+ 4.6%	+ 3.5%	+ 5.5%	+ 3.9%	+ 3.9%
− 20%	− 5.4%	− 3.5%	− 6.1%	− 5.1%	− 4.6%
EW energy consumption (kWh)					
+ 20%	+ 2.6%	+ 0.5%	+ 0.9%	+ 0.4%	+ 0.2%
− 20%	− 3.6%	− 0.5%	− 1.9%	− 1.2%	− 1.0%
Waelz process					
Iron metallization degree (%)					
+ 20%	+ 1.0%	0%	+ 1.1%	+ 1.0%	+ 0.2%
− 20%	− 3.3%	0%	− 0.2%	− 0.5%	− 0.2%
Coke load in the furnace (kg/t furnace burden)					
+ 20%	+ 1.4%	0%	+ 1.5%	+ 1.6%	+ 4.3%
− 20%	− 3.1%	0%	− 3.2%	− 5.2%	− 4.3%
Furnace and ESP power consumption (kWh)					
+ 20%	+ 4.7%	+ 1.0%	+ 6.8%	+ 4.1%	+ 0.6%
− 20%	− 5.7%	− 0.7%	− 5.6%	− 3.1%	− 0.8%
EW energy consumption (kWh)					
+ 20%	+ 2.3%	+ 1.0%	+ 9.0%	+ 5.7%	+ 1.3%
− 20%	− 4.9%	− 3.0%	− 5.6%	− 4.7%	− 1.3%

It can be seen in the *RCAP* that the excessive feed of caustic soda had, by far, the largest effects on the citric process. In addition to affecting the caustic soda input, the excess feed increased the acid consumption in leaching and lime consumption in the final neutralization, explaining the large response. In contrast, the solid content in the slurry was largely meaningless when the acid feed was controlled by acid consumption in leaching rather than the starting concentration. The impacts were likewise sensitive to changes in the amount of free acid after the leaching step, albeit the response was not as large as the excess caustic soda feed, which affected several inputs in the model. The analysis shows that the optimization of caustic soda consumption in the roasting step is critical for the process.

In contrast to the *RCAP*, the responses to model variations were more moderate in the *Waelz process*, overall. Variations in the electricity consumption seemed to have a greater effect than controlling either the coke feed or the reduction of iron, which was the main side reaction affecting the consumption of coke. Renewable or low-carbon electricity would also have a significantly positive effect on the impacts based on the results, although fossil fuels are still consumed in the process as a reductant and the primary source for heat. The use of biomass-based reductants or bio-coke has been considered as an alternative to fossil petroleum or metallurgical coke, since the process requires no mechanical strength from coke [52].

Limitations

There are several data gaps and assumptions that may affect the results in this study. The results are dependent on the quality of the LCI data. The used simulation-based method has the strength that it is not reliant on reported mass and energy balances and the data are internally consistent and detailed. Tsadilis and Korevaar [53], for instance, demonstrated that data from process simulations of emerging processes can be highly accurate. It is still noteworthy that the assumptions made in the simulation and the selected system boundaries affect the results of the environmental assessment.

Electricity consumption of the processes was estimated based on the mass balance by approximating the dimensions of the main equipment including furnaces, reactors, filters, thickeners, and electrolysis, which is a clear source of error. All the auxiliary equipment was included as a constant, which is why the power consumption estimates are indicative at best. Heat losses in the hydrometallurgical circuits were not modeled either, but the effect is probably similar between the models since electrowinning was the only endothermal step in the hydrometallurgical stages. Heat recovery in the pyrometallurgical steps was likewise excluded.

The *RCAP* was simulated from only partly optimized laboratory scale batch experiments, which were scaled up in the simulation. Kinetics are likely to improve when scaling up to continuous industrial processes, which were not taken into account in the models. The harmful emissions from the roaster are uncertain. Carbon, sulfur, and halogens were assumed to oxidize to carbon and sulfur dioxide and chlorine gases, of which chlorine was scrubbed with water from the gas. Fluorine was not considered in the analyses, but it was assumed to enrich in the same fractions as chlorine due to their similar behavior. Further work on the removal of fluorine and chlorine from the roaster gas is necessary from a safety standpoint.

The Waelz process and the hydrometallurgical refining of crude zinc oxide were simulated based on process phenomena and conditions reported by the industry. The model did not consider particulate or VOC emissions that are likely created in the pyrometallurgical process, which could be reflected particularly on the POCP values. The hydrometallurgical process used in the simulation did not perfectly represent typical zinc EW processes, where secondary zinc residues are integrated to sphalerite concentrate processing.

The current analysis assumes no valorization of the Waelz slag, and no credits were calculated. The partial substitution of natural aggregates with the slag has, however, been suggested possible without negative effects on physical properties or leachability [37]. Muñoz et al. [54] studied the environmental aspects of Waelz slag use in bricks and estimated that the overall benefits are limited, and Waelz slag bricks performed worse than standard bricks in human toxicity and acidification categories due to the presence of sulfur and halogens in the slag. Studies from steel slag valorization are more extensive, and the results vary between applications. Gao et al. [55], for instance, determined that steel slag aggregates result in higher CO_2 emissions than andesite aggregates in road base layer construction, while Anastasiou et al. [56] predicted that significant environmental load reductions are possible by incorporating EAF slag aggregates in special concretes. The use of the slags would, however, decrease the impacts from landfilling [54], although further investigation is needed to determine the environmental significance in specific applications.

Conclusions

In this study, a comparative techno-environmental assessment was performed to critically evaluate the performance of a novel hydrometallurgical roasting–citric acid process (*RCAP*) for the treatment of electric arc furnace dust waste against the *Waelz process,* using process simulation as the basis for the life cycle assessment. The strength of early process evaluation is that it can help identify focus points for further research, which was also demonstrated in this study. A holistic view of the entire flowsheet is necessary to develop truly sustainable processes.

The global warming potential of the processes at the highest investigated zinc content ($>30\%$) was 7.5 kg CO_2-eq for 1 kg Zn for the *RCAP* and 5.0 kg CO_2-eq in the *Waelz process*. With the lowest zinc content ($\sim 10\%$), the respective values were 6.3 kg CO_2-eq and 9.7 kg CO_2-eq. The environmental footprints of the two processes were highly dependent on the zinc content in the treated EAFD. In the *Waelz process*, the impacts increased consistently when the zinc content decreased, mainly because fuels and reductants were consumed in converting the iron oxides into disposable slag. Iron recovery decreased the impacts in the *RCAP,* particularly in the GWP category, when the zinc content of the raw material was low ($<10\%$), indicating that the process could be environmentally viable in the treatment of iron residues with low zinc content. This may not, however, be economical while landfilling remains the most cost-effective method to dispose of iron-rich residues.

The *RCAP* surpassed the *Waelz process* only in terms of GWP at the lowest zinc content, mainly due to the high consumption of chemicals. Citric acid consumption was consistently the main problem in the process, which was due to the decomposition of the lixiviant in the process, preventing its reuse, and the acid-consuming alkali in the raw material. The *RCAP* was, however, more tolerant to the presence of halogens in the feed material than the *Waelz process*, where chloride was observed to accumulate easily in the EW solution even with an additional halogen washing step.

Opportunities for decreasing the environmental burden of the *RCAP* were suggested based on the sensitivity analysis. Reducing caustic soda consumption in the roasting step appears to be the most impactful measure for decreasing the environmental impacts of the process. Therefore, the focus should be aimed at the optimization of the roasting stage, which would decrease not only caustic soda consumption in roasting but also acid consumption in leaching. The scrubbing of the roasted calcine with dilute mineral acid solutions to remove easily soluble alkali and unreacted caustic soda could also possibly decrease the excess consumption of citric acid in the process. Studying the decomposition behavior and the reuse of citric acid in the leaching step would be necessary to decrease the impacts of the process.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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Authors and Affiliations

Marja Rinne[1] · **Petteri Halli**[1] · **Jari Aromaa**[1] · **Mari Lundström**[1]

✉ Mari Lundström
mari.lundstrom@aalto.fi

[1] Department of Chemical and Metallurgical Engineering,
School of Chemical Engineering, Aalto University,
Vuorimiehentie 2 K, P.O. Box 16200, 00076 Aalto, Finland