Review and new life cycle assessment for rare earth production from bastnäsite, ion adsorption clays and lateritic monazite

Gwendolyn Bailey[a],* P. James Joyce[b], Dieuwertje Schrijvers[c], Rita Schulze[d], Anne Marie Sylvestre[e], Benjamin Sprecher[d], Ehsan Vahidi[f], Wim Dewulf[g], Karel Van Acker[a]

[a] KU Leuven, Department of Materials Engineering, Kasteelpark Arenberg 44, 3001 Leuven, Belgium

[b] KTH Royal Institute of Technology, Department of Sustainable Development, Environmental Science and Engineering (SEED), Stockholm SE 100-44, Sweden

[c] Université de Bordeaux Bâtiment A12, 351 Cours de la Libération, 33405 Talence, France

[d] Leiden University, Department of Industrial Ecology (CML), Einsteinweg 2, 2333 CC Leiden, The Netherlands

[e] Lynas Malaysia SDN BHD PT17212 Jalan Gebeng 3, Kawasan Perindustrian Gebeng, 26080 Kuantan, Pahang, Malaysia

[f] Materials Systems Laboratory, Massachusetts Institute of Technology, 77 Massachusetts Avenue E19-695, Cambridge, MA, 02139, United States

[g] KU Leuven, Center for Industrial Management / Traffic & Infrastructure, Celestijnenlaan 300, 3001 Leuven, Belgium

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HIGHLIGHTS

- The methodological challenges surrounding life cycle assessments for rare earth elements are identified by reviewing 24 publications on life cycle assessment for rare earths.
- The main contributor to the global warming potential of the production of rare earth oxides is solvent extraction.
- Results from a monazite mining site in Mt. Weld, Australia are presented for the first time.
- Two life cycle inventories are presented as the best representation of two of the three main mineralogical routes for rare earth elements.

ABSTRACT

Rare Earth Elements (REEs) are one of the most important--albeit critical--commodities for our green technologies. However, there is a general perception that rare earths are produced using mining and processing techniques that are unsustainable. Life Cycle Assessment (LCA) is the most widely accepted methodology to evaluate the impacts of rare earth oxide (REO) production. This article aims to provide a synthesis of the currently existing LCA studies on REEs using two strategies. Firstly, an overview of published LCA results of REO production. Secondly, a detailed LCA using the best available life cycle inventories (LCIs) in order to: i). evaluate the state-of-the-art LCI for this sector ii). understand better the impacts related to each of the three main production routes and iii). inform a preliminary benchmark for the sector. The analysis of the published LCA results reveal that the three main methodological issues with published LCAs are data gaps, allocation, and waste management. The dominating contributor to the global warming potential of the production of REOs in all three routes is chemical extraction and separation.

1. Introduction

REEs are an essential commodity to the global market. Although accurate production numbers are very difficult to obtain, the current estimation of annual global production is estimated at 170,000 tons (U.S. Geological Survey 2019). In 2017, the EU market consumed 8350 tons of REEs (European Commission 2017).
The global rare earth metals market is expected to grow by 14% to 9 billion USD in 2019 (BusinessWire 2016). The largest market for REEs is the production of permanent magnets, which are used for many low carbon technologies, such as wind turbines and electric vehicles. There is, however, an inherent tension in using REEs for green technologies, because the mining and production of those REEs is associated with critical threats to the environment. For instance, the processes associated with the production of these magnets leads to the production of toxic and hazardous residues (Bailey et al. 2017). Therefore, as clean energy technologies continue and increasingly utilize these elements, it is relevant and timely to form a consensus on the best available practices for determining their environmental impacts.

LCA is the de-facto standard methodology for quantifying the environmental impacts of each stage in a product's life cycle, identifying environmental hotspots and classifying them into impact categories. Attributional LCA has been applied to many REE evaluations, (Sprecher et al. 2014) (Schrijvers 2017) (Koltun and Tharumarajah 2014) (Zaimes et al. 2015) (Schulze et al. 2017). Today, LCA practitioners are faced with many problems when performing an LCA for REEs, such as limited data and allocation issues. Despite early promising results from these LCA investigations, some studies' ability to apply to many different entities has fallen short. For example, in the study by Weng et al. (2016) varying stages in REE production were evaluated and the life cycle impact assessment (LCIA) results are not comparable. It was noted by Pell et al. (2017) that there are challenges associated with comparing different end product(s). Weng et al. (2016) also assumes that two (or more) mines have the same mineral compositions and topology, which is geologically incorrect (Pell et al. 2017).

A state-of-the-art-LCA, which entails recommendations and adaptations made to existing LCA studies, would enable practitioners of LCA or REEs stakeholders to make conclusions more successfully. A cohort of authors on REEs and LCA have joined together to provide a consensus on the best "state-of-the-art" practices for REEs LCA and to elucidate aspects which still need improvement.

Environmental impacts of REEs have often been investigated, but even the well-known databases do not have sufficient data. The ecoinvent LCI database contains one of the first investigations on REOs where environmental impacts of the Bayan Obo pathway were examined (Althaus et al.,2007). The ecoinvent database contains five process steps: mining, beneficiation, roasting, cracking (chemical treatment) and solvent extraction (Primas 2017a) (Primas 2017b). There are also several GaBi Thinkstep flows and process datasets related to REO production. Further, numerous LCA investigations have been conducted using different production systems and methodologies (Nuss and Eckelman 2014, Sprecher et al. 2014, Lee and Wen 2017). We consider reanalysis of inventories available in LCA investigations with one set of system boundaries, one modelling requirement for allocation, and one set of impact assessment methods a good way of reviewing LCAs, and overcoming the incomparability resulting from discordant methodological choices.

The world production of permanent magnets is highly dependent on Chinese supply of REEs. However, local environmental issues related to the extraction of REEs have motivated Chinese policy makers to restrict the export of REEs (Mancheri et al. 2019). The Chinese Information Office of the State Council argued that export quotas would enable the Chinese government to better control and limit the environmental impacts of REEs production which could signify the relevance of environmental impacts for their global accessibility (China State Council 2012).

This work (1) reviews and evaluates 24 LCA studies of REE production, discussing issues associated specifically with LCI incompatibility of REEs, (2) re-implements the highest quality case-study inventory data from these previous studies in a relatively consistent manner, (3) adds a novel primary (company-specific) dataset on monazite production route. We expect that using the best available data to produce an up-to-date dataset, along with a new life cycle assessment with an appropriate allocation procedure and radioactive waste modelling procedure, will be crucial for future high quality evaluations of downstream products containing REEs.

2. Methodology

2.1 Overview of Published Impacts

A better understanding of REE LCAs including their assumptions and affectations could improve the usage and the acceptability of the data. From the studies selected for further review, impact assessments related to the production processes (from mining to solvent extraction) have been determined. Moreover, there are multiple types of mines and deposits that contain monazite and/or bastnäsite, including carbonatites and mineral sands, which could result in very different kinds of deposits and process flowsheets. These might have alternative processing and LCA outcomes, which are not directly comparable.

2.2 Model Calculation

The model is based on three different mineralogical routes which are not process routes but a combination of a specific ore with a typical accompanying process. The mineralogical route and process route are listed below for each route:

Route 1 (R1) = Bastnäsite ore (Bayan Obo) + sulfuric roasting

Route 2 (R2) = Monazite ore (Mount Weld) + sulfuric roasting

Route 3 (R3) = Ionic ore (Southern Provinces) + ammonium sulfate leaching

The main steps for the calculation of the environmental impacts of the representative model are:

- Selection of the most representative processes and routes in terms of real-life practices.
    - We conducted a structured data search to look for all possible sources for LCA of REEs. The search for electronically available literature was run via Web of Sciences (Thomson Reuters, New York, NY), Scopus (Elsevier, Amsterdam, the Netherlands), and Google scholar (Google, Mountain View, CA). In these three portals, we searched for articles, conference proceedings and scientific reports containing keywords "life cycle assessment" and "rare earth elements." The LCAs outlined in Table 1 were selected because they provided some inventory data for the foreground processes for the most important production routes and had similar system boundaries.

- Definition of key assumptions according to the goal and scope of the study
    - By bringing together and reanalyzing published LCIs, we aim to evaluate REE production and provide a consensus on the state-of-the-art combination, which is not possible by looking at individual studies. In contrast to the original studies, the LCA studies within this reanalysis use one set of modelling choices. These choices were made after first formulating the goal and scope of the state-of-the-art dataset. The intended application of the study is the development of the first draft of the state-of-the-art LCA for the REE sector.

- Collection of primary data and adaptation of inventory data
    - Ideally, primary data, meaning data collected/measured directly by a company, would be used for all stages, but in practice, sometimes only secondary data are available for some processes to be modelled. For each set of foreground data (that is, data for processes which could be influenced by the decision maker) extracted from the literature, we reanalyzed the environmental impact to equivalent functional units: the cradle-to-gate production of 1 kg of separated REO. The reanalysis was executed in GaBi (Thinkstep) using background (that is, data which refers to the background system, which cannot be influenced by the decision maker) inventory data mainly from Ecoinvent (ecoinvent Center, St. Gallen, Switzerland) and where there were data gaps in the Ecoinvent database, then data from Thinkstep were used.

- Calculation of the environmental impacts, adopting the International Reference Life Cycle Data System /Product Environmental Footprint (ILCD/PEF) recommendation version 1.09 methodology (Marc-Andree Wolf 2012).
    - During the development of the PEF framework, an impact assessment methodology was chosen for its benchmarking qualities (European Commission 2013). We use the same ILCD methodology as it contains the "state-of-the-art" impact assessment methods.

- Additional calculation of impacts resulting from Naturally Occurring Radioactive Materials (NORM)
    - Because the ILCD/PEF impact category for ionizing radiation (IR) initially aimed to reflect impacts from the nuclear fuel cycle, the potential impacts resulting from increased exposure to naturally occurring radioactive materials (NORM) remain unassessed (Joyce et al. 2017). Emissions of NORM radionuclides not included in the ILCD/PEF IR impact category, particularly ^{232}Thorium which is a common emission from the processing of bastnäsite and monazite ores, may be of significance, and therefore should be assessed in this context. Hence, in addition to the ILCD/PEF impact categories, the human health impact category for NORM exposure from Goronovski et al. (2018) was applied in this study.

- The cradle-to-gate nature of the study means that the LCI does not include the life cycle stages beyond the 'gate', such as distribution to users of REO, the manufacture of downstream products, their use and their end-of-life management. Instead, the data cover the life cycle stages prior to the 'gate', comprising mining, beneficiation, leaching, extraction, and REO product finishing.

3. Results

3.1. Selection of the most representative processes and routes in terms of real-life practices.

3.1.1 Literature Review

A majority of the studies on REEs follow a specific production route starting from one (and sometimes two) mineral deposit types. Although there are more than 200 known rare earth-containing minerals, the economically viable production sources are mostly limited to monazite, bastnäsite-Ce, and rare earth-containing clays (Arshi, Vahidi, & Zhao, 2018). Our literature review concentrated on three specific deposits - or deposits types for ion adsorption clays -- chosen to illustrate three major current mines. The most important features and findings of the above studies are summarized below:

3.1.2 R1: Bastnäsite

Sprecher et al. (2014) applied the LCA methodology to evaluate Nd-Fe-B magnets in hard disk drives. The LCI is one of the most complete to date and is filled with data from literature, calculations, and expert interviews. Lee and Wen (2017) focused on the environmental impacts of fifteen REEs produced in China. The interesting aspect of this study is the detailed look into the differing metal refining processes. These processes include molten salt electrolysis, calciothermic reduction, and lanthanothermic reduction. A majority of the data used for this study comes from the Chinese Ministry of Protection and directly from Chinese industry surveys. Where appropriate, the primary data of Lee and Wen (2017) have been incorporated into our state-of-the-art LCA.

3.1.3 R2: Monazite

Koltun and Tharumarajah (2014) evaluated impacts for heavy, medium, and light REOs via a monazite and bastnäsite processing route. Niobium co-production during beneficiation was also taken into account. Marx et al. (2018) assessed the environmental impacts from the Mt. Weld mine in Australia which produces monazite bearing ore. Most recently, Arshi et al. (2018) analyzed two different magnet production processes of two independent Chinese facilities using neodymium (Nd) and praseodymium (Pr) from monazite/ bastnäsite deposits in Bayan Obo. There are two LCA studies which present results from monazite deposits. Sala and Bieda (2018) provided life cycle inventory information for separation process only but was published after the literature review was conducted and not considered for the assessment. (Browning et al. 2016) assessed monazite coming from Indian mineral sands but no LCI was published.

3.1.4 R3: Ion Adsorption Clay Deposits

Vahidi et al. (2016) evaluated the production of 1 ton of REOs from ion adsorption clay deposits. Schulze et al. (2017) evaluated the impacts from the ion adsorption route and included the solvent extraction step. Compared to the open pit mining route from Sprecher et al. (2014), both studies concluded that less environmental impacts are incurred in REOs production from ion adsorption clay deposits. However, the terrestrial and aquatic ecotoxicity ratings are higher. The aquatic ecotoxicity impacts are due to the large amount of effluent and heavy metals laced wastewater produced during the in-situ leaching. Sprecher et al. (2017) made the point that the environmental impact of illegal REE production (which mostly occurs with ionic clay deposits) is significantly larger compared to legal production methods. This discrepancy is not taken into account in LCA studies.

3.1.5 REE processing and other production routes

A recent study was also carried out by Vahidi and Zhao (2016) and further expanded in another investigation Vahidi and Zhao (2017) to evaluate the impact of the solvent extraction process and the characterization of chemical reagents. The provision of a new Chinese organic solvent dataset was one of the highlights included in this study. In general there is a lack of detailed, quantitative and comparative studies on chemical solvent options for REEs in LCA. Ikhlayel (2017) focused on REEs and how they compare with other metals, such as precious and base metals. The author concluded that precious metals have the highest impact but REEs environmental impacts are not insignificant and REE production is much more impactful than base metals production. Schreiber et al. (2016) focused on other, less common, REE mineral types such as eudialyte ores.

3.2. Definition of key assumptions according to the goal and scope of the study

3.2.1. Goal and scope of reviewed datasets

Twenty-four recent publications were selected for the construction of the state-of-the-art dataset, as they represent the most common production routes and they have similar scopes. The goal and scopes of the reviewed literature are summarized in Table 1. In this study, the REE production system was analyzed from mining up to and including solvent extraction which contains product finishing processes such as precipitation and calcination (Figure 1). The other categories assessed in the literature review were mineral type, radioactivity, waste treatment, and co-mining. For mineral type, we noted the mineralogy accounted for in each study, which makes comparison difficult. For radioactivity, we marked with an X those studies which exemplified radioactivity in the LCI (if provided). For waste, we distinguished those LCIs which quantified waste treatment flows. For co-mining, since all REE are co-products of each other, this column is meant to highlight the co-products in addition to REE. According to the LCIs available we marked those which took into account co-mined products (apart from REE) into their study.

Figure 1. Processing steps of REEs from mining up to solvent extraction. 1) Mining of REE from host material 2) Physical removal from host material 3) Decomposition of beneficiated ores (sometimes referred to as cracking) 4) Transformation into a REE chlorides 5) Separation of individual REOs

Table 1. Input data available in literature sources, *Referring to the processes depicted in Figure 1.

Geographical scope	System Boundary	Processes	Mineral type	Radioactivity	Waste treatment	Comining	Reference
China	Ore to 70% REO	1,2,3,4,5	Bastnäsite	X	X by products modeled as waste		(Primas 2017a, b)
Global	Market for Nd oxide	1,2,3,4,5	Bastnäsite				(Bourgault 2011)
China	Ore to Nd oxide	1,2,3,4,5	Bastnäsite /Monazite	X		X	(Sprecher et al. 2014)
China	Ore to 80% REO	4,5	Ion adsorption			X	(Schulze et al. 2017)
Norway	Ore to Nd/Dy	1,2,3,4,5	Eudialyte and Bastnäsite			X	(Schreiber et al. 2016)

China	Ore to REO	4,5	Ion adsorption				(Vahidi et al. 2016)
Global	Ore to REO	1,2,3,4,5	Bastnäsite			X	(Nuss and Eckelman 2014)
China	Ore to REO	1,2,3,4,5	Bastnäsite and Monazite			X	(Koltun and Tharumarajah 2014)
China	Ore to REO	1,2,3	Bastnäsite /Monazite			X	(Zaimes et al. 2015)
China	Ore	1,2,3,4,5	Bastnäsite /Monazite	X			(Zhou et al. 2016)
China	Recycled Nd-Fe-B	1,2,3,4,5	N/A				(Jin et al. 2016)
China	REE leachate to REO	4,5	N/A				(E. Vahidi, Zhao, F., 2017)
	Ore to RE metals	1,2,3,4,5	N/A				(Ikhlayel 2017)
China, Canada, Australia	Ore to selected REOs	1,2,3	Bastnäsite			X	(Weng et al. 2013)
China	Ore to RE metals	1,2,3,4,5	Bastnäsite	X	X Internal recycling	X	(Lee and Wen 2017)
China	Ore to REO	1,2,3,4,5	Bastnäsite ,Monazite, Ion adsorption			X	(Weng et al. 2016)
China and US	Ore to REO	1,2,3,4,5	Bastnäsite ,Monazite, Ion adsorption				(Navarro and Zhao 2014)
China	Ore to REO	4,5	N/A				(Vahidi et al. 2016)

Boxes where no X appears indicates that there was no LCI data available for this process element, however it does not mean that it was not considered in the study.

From this table, one can conclude that many of the system descriptions have a limited scope, especially related to radioactivity and waste. REEs are mostly produced as by-products of other (host) metals, such as iron, titanium, zirconium, and thorium (Elshkaki and Graedel 2014). Co-production and allocation are not always reflected in the LCI provided by these LCA studies. That is, it is not always clear which allocation choices were made. Some of the inconsistencies in the comparison of projects is due to the difference in system boundaries such as the study by Vahidi and Zhao (2017) which evaluates only the solvent extraction stage. But other inconsistencies—such as missing details or documented waste treatment or allocation methods—mean that the LCA is not formed with a rigorous enough methodology to draw truly meaningful conclusions. The 24 literature sources did not reveal any cohesion on the following three aspects: data availability, allocation, and waste. For instance, many of these studies took different approaches with regards to allocation and waste modelling and all show different levels of data quality/detail.

Other areas in which the reviewed literature is lacking are the intermediate flows for the removal from REEs concentrate from gangue material in the beneficiation process and the combination of chemicals used for solvent extraction. Moreover, for the beneficiation and solvent extraction steps, several stages are required to get a concentrated or high purity product. It is important to highlight that the processing steps applied for ore type 1 and ore type 2 are similar but for ore type 3 the process steps are different. Beneficiation does not take place for production from ion-adsorption clays, for example. Modelling the average solvent extraction (SX) process is difficult because the number of steps to extract each individual REO is not readily known.

3.2.2 Data gaps and data quality

Ideally, primary data would be used for all stages, but in practice, sometimes only secondary data will be available for some processes to be modelled. Indeed, since the LCA approach aims to model reality, LCA must simplify where necessary. Also, depending on the goal of the study, there may be no need to spend much time or energy to collect primary data for processes that are not relevant to the specified goal. We considered important to collect primary data on the most relevant life cycle stage(s) for REEs identified by our literature review: solvent extraction.

Table 1 showed that there are many data gaps. The strategy for resolving the data gap problem is to complement missing information with company-specific data. The data coming from these documents are given priority as they are considered as high-quality data. The authors applied aspects of the pedigree matrix approach brought forth by Weidema and Wesnæs (1996) to determine the quality of the data. This data-filling strategy implies that, for studies that do not give information on the most relevant life cycle stages or on co-mining then these inventory data are estimated based on LCIs using data from the REE industry.

The most complete dataset is R2 (monazite). For this route, we have collected primary data for all the activities of the foreground system from Lynas, the largest commercial source for REEs outside of China. Secondary data were used for background processes. Special attention was given to the data quality of processes that will influence hotspots.

3.2.3 Allocation

Primary REOs are always produced in co-production; often of other minerals, and always as co-product of each other. This makes several steps within the production route of REEs multifunctional. In order to identify the LCI of an individual REO, an allocation procedure must be applied (in attributional LCA studies – see next paragraph).

3.2.3.1 Overview of allocation in reviewed papers

The allocation methods for REE LCIAs range from mass (Lee and Wen 2017) to economic (Sprecher et al. 2014, Schreiber et al. 2016, Schulze et al. 2017). The fact that all papers apply a partitioning method and none of the studies proposes to model the co-production by substitution implies that all studies apply an attributional approach (Schrijvers, Loubet, & Sonnemann, 2016). In this section, we discuss which allocation approach is considered most appropriate in an attributional study on REEs.

3.2.3.2 Allocation in an attributional LCA of REEs

According to International Standard Organization (ISO) 14044, allocation should be applied according to the following hierarchy: 1) subdivision or system expansion, 2) partitioning reflecting underlying physical relationships, 3) partitioning reflecting other relationships, e.g. economic value (ISO, 2006). System expansion could refer to a) the inclusion of the additional function of the co-products in the functional unit or b) the modeling of substitution effects (Heijungs, 2013). Option a) is not desirable, as the purpose of this paper is to provide the inventory for the individual REOs. Option b) is considered to be only appropriate in a consequential LCA (Schrijvers et al., 2016), and is therefore not further discussed in this section.

A detailed discussion on the application of the ISO allocation hierarchy to the production of REOs is provided in the (Supporting Information). The allocation problem of the solvent extraction process could be reduced by subdivision, but only when the groups of REEs can be traded on the market in unseparated form and detailed process data are available. The latter is however a limiting factor, due to the confidentiality of industrial separation techniques. If subdivision is not possible, economic partitioning is recommended using the allocation procedure "Allocation at the point of substitution", as introduced in version 3 of the ecoinvent database (Weidema 2013).

We used economic allocation for R1 and R3 since there was no precise information on the individual processing steps (particularly for solvent extraction). We did not apply any allocation for R2 since enough data was provided to show that there were no co-products produced. We recognize that the prices of REEs are volatile, therefore, we used the price based on the 10 year average (2008-2018) (Santero and Hendry 2016). For R1, there are two processes which contain two outputs, thus two processes in which allocation factors must be calculated. The first is the beneficiation process. We estimate that in 1 kg of crude ore 27% of the value is represented by iron ore while 73% is represented by the REO content. The price of 62% iron ore is 80 EUR/mt, so 30% iron ore would be ± 40 EUR/mt. Using pure REO Asian Metal (2017) prices we calculated that the REO content of 1 kg of crude ore is ± 1 EUR/kg.

For R1 and R3, allocation factors were calculated for the solvent extraction process. For R1, the solvent extraction step combined values of REOs results in a total price of 17.35 EUR/kg of REO along with 0.02 EUR/kg of ammonium chloride. For R3, the combined prices of the REOs after the solvent extraction stages amount to 125.08 EUR/kg REO mix. To perform economic allocation, one needs the average price of each individual REO, and the amount of the separated product based on the modelled process chain (kg). The calculated allocation factors for these routes can be found in the (Supporting Information, Table S1 and Table S2).

3.2.4 Waste

In many regions where REE processing occurs the environment has suffered from contamination through insufficient waste management, notably radioactive contamination caused by thorium and its daughter radionuclides (Li et al. 2012) (Wu et al. 2011). Moreover, there is almost no information regarding what happens to the wastewater and tailings at Bayan Obo and other Chinese sites. The "Pollutant Discharge Standards for the Rare Earth Industry" replace the general emission standards and set stricter limits for pollutants in water and air for the total discharge of waste water, waste air, thorium, and uranium (China Rare Earth 2011). According to this set of standards, nitrogen in the form of ammonia pollution was originally limited to 15 mg per liter of wastewater. However, this regulation was loosened to 20 mg/L in the draft of 2009, and to 25 mg/L in the final standards. If we take the general discharge standards for actual practice, we can calculate the ammonia emissions based on what is *allowed* to be emitted but not on what is actually emitted. Therefore, it is not the best solution to use these reports for emission values as in Lee and Wen (2017).

Firstly, we will address the issue of non-radioactive waste. Secondly, the next section will be devoted to radioactive waste and the modeling challenges and later in the paper we will include the results from a naturally occurring radioactive materials (NORM) assessment. The main problem with waste in REE LCAs is that there is limited information on the inputs and outputs of waste streams, since oftentimes the residue transportation and treatment is handled by a third party. Many LCA calculations only account for the removal of the extracted material and energy and emissions. In general, REE LCAs do not include actual environmental measurements, such as water contamination. Regarding tailings waste, except for the radioactivity of uranium and thorium, the potential waste emissions would be generally comparable to a typical hard rock mine (Weber and Reisman 2013). The non-radioactive waste can be found in large tailings dams. Researchers warned against the possible collapse of the tailings dam at Baotou, the largest REEs mining site in China (He 2010). Even if the dam does not fail, the hazardous tailings could come into contact with the environment (Wübbeke 2013).

For R1 and R3 we are not aware of high-quality data representing the waste treatment, and therefore did not include it. For R2, we included waste gases treatment and modelled the treatment of minerals processing waste based on the current processes performed by our primary data source. The full explanation of the justifications for our recommended state-of-the-art LCIs are presented in the (Supporting Information). A brief summary of assumptions made in the LCA study regarding inclusions and exclusions are shown in the (Supporting Information, Table S3).

3.3 Collection of primary data and adaptation of inventory data

The 24 inventories of the reviewed studies with the ecoinvent process used and quantified for the reanalysis can be found in the (Supporting Information). The location considered for all processes in R1 and R3 is China, and therefore the electricity mix is the average Chinese electricity mix. For R2, we assumed electricity use was generated from the rest-of-world electricity grid mix. There is a sensitivity analysis provided for grid mix in the Supporting

Information, Section For more details on the elaboration of this state-of-the-art LCI, see the (Supporting Information) excel sheet which presents the data used in each modelling approach.

3.4 Calculation of the environmental impacts

Representative model impact assessment

The functional unit of this study is the production of 1 kg of the basket REO from each corresponding mineralogical and processing route. The reference flow is 1 kg of mix of the separated REO product. In this context, it is important to mention that the separated REO is from the 3 different geologies and processing routes. Another important distinction to make is that after solvent extraction REOs have different compositions, and the outputs are therefore not directly comparable.

The ILCD methodology (Marc-Andree Wolf 2012) characterization factors (v1.09) have been applied. The results indicate that, for the majority of the impact categories, the typology of ore with the greatest environmental burden is bastnäsite. It should be noted that the LCIA results indicate relative effects only and do not predict actual impacts. Individual results of the LCA are further discussed in the (Supporting Information). The environmental profile of REO production shows that all the production stages contribute to impacts with the main contribution made by the solvent extraction stages (Figure 2). It should be noted that the chemical reagents in the solvent extraction processes are not recycled since the authors were not provided information this data.

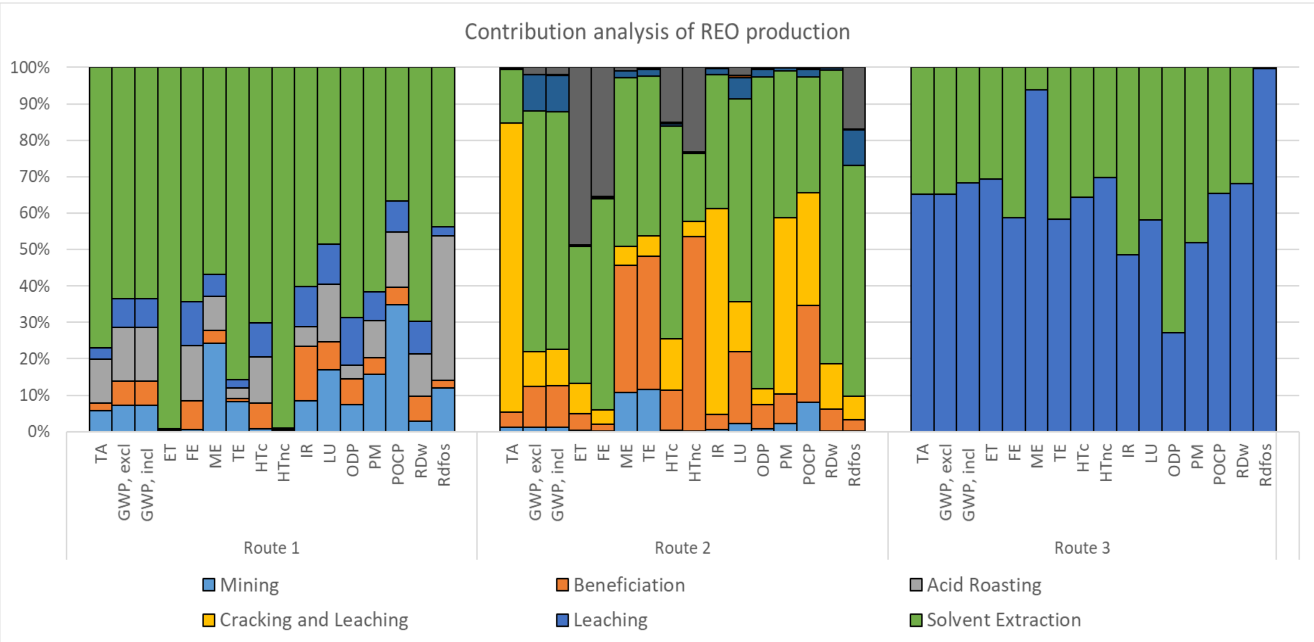

Figure 2. Contribution analysis of all three routes reveals that solvent extraction is one of the most impactful production steps. *The impact categories are: climate change (GWP), stratospheric ozone depletion (ODP), human toxicity, cancer and non-cancer effects (HTc and HTnc), particulate matter (PM), ionizing radiation (IR), photochemical ozone formation (POFP), terrestrial acidification (TA), terrestrial eutrophication (TE), freshwater eutrophication (FE), marine eutrophication (ME). Freshwater ecotoxicity (ET), land use (LU), and resource depletion for water (RDw) and minerals, metals and fossils (RDfos).*

Normalization is applied in the relation to the 2010 EU-27 population persons equivalent (Marc-Andree Wolf 2012). By using these normalized results, the significance of each impact in a population-based context can be evaluated and relevant impacts for comparative assessment can be identified. The application of these normalization factors, however, increases the uncertainty of the whole assessment, given that the determination of these factors is complex and lack of data can result in erroneous values (Bisinella et al. 2016).

The impact categories with the highest overall scores as persons equivalent are human toxicity with carcinogenic effects (HTc), freshwater ecotoxicity (ET) and depletion of abiotic resources (RDfos). Regarding global warming potential (GWP), R3 has lower impacts than R1 and R2. R1 has the largest impacts overall because the electricity mix is predominantly Chinese and the consumption is substantially higher than R3. Although R2 is similar to R1, R2 shows lower overall impacts due to the reduced flow of untreated waste. R3 impacts are related to resource depletion in the mining and leaching stages, due to the fact that ionic ore deposits have a high production of heavy REOs. Moreover, REEs contain the same characterization factor for RDfos in the ILCD methodology, except for yttrium, which has a different factor (EC-JRC 2013). It should be noted that ion adsorption clays supply almost all of the world's heavy REEs. The other two mines: Bayan Obo and Mt. Weld are mostly light REEs deposits. The concentrations of heavy REEs in ore deposits are lower than light REEs (their absolute crystal abundance are much lower).

For HTc, R1 has high impacts due to the emissions of HF during the solvent extraction process. The major impacts for ME are connected with R3 due to the chemicals leached during the in-situ leaching process. ET is dominated by R1. ET is higher in R1 due to the SX process where all heavy metals go to waste water. The emission of zinc to water, for example, has a highly relevant contribution. This is partially due to the high characterization factor assigned to zinc. Figure 3 shows the normalized impact scores for all three routes and impact categories. The primary scale is for R2 and the secondary scale is for R1 and R3.

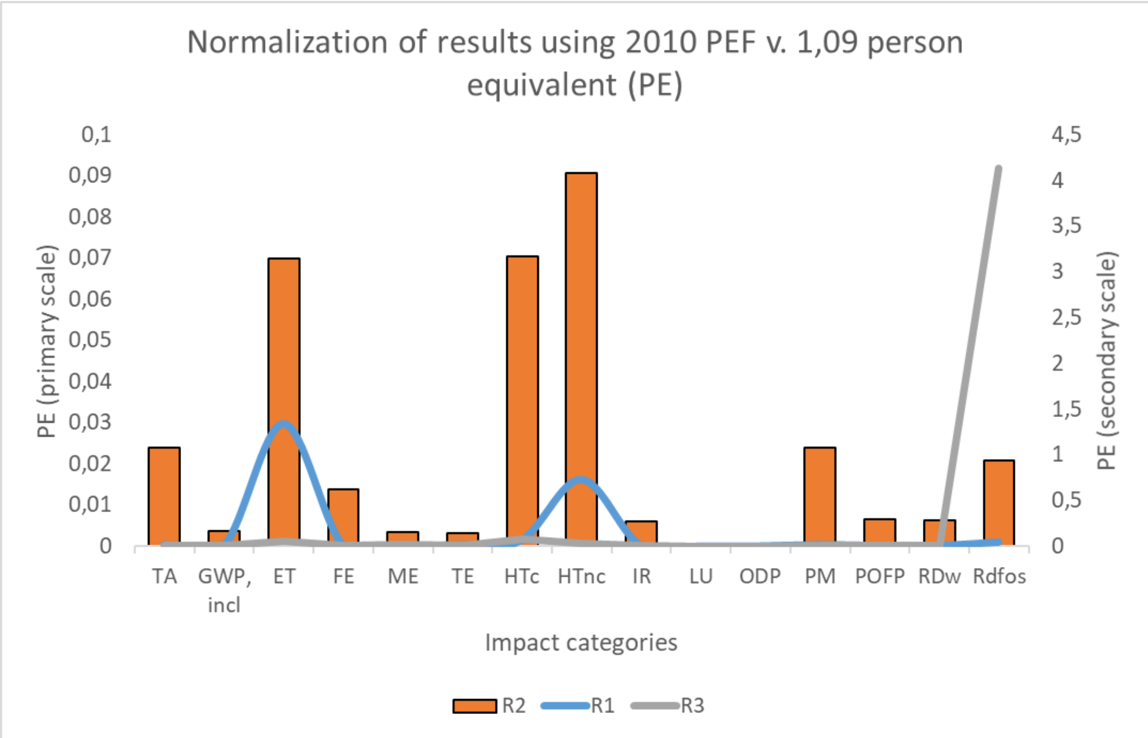

Figure 3. Normalized result scores of the three routes. *Functional unit refers to 1 kg of REO mix. Results are given in Persons Equivalents (PE), meaning one average person in the EU27. The primary scale refers to R2 and the secondary scale refers to R1 and R3.*

The graphs illustrating the differing routes process' contribution to the impacts can be found in the (Supporting Information, Section 2.4). The detailed list of processes and substances with the highest contributing impacts is also reported in the (Supporting Information, Tables S9-19).

Naturally occurring radioactive materials (NORM)

The results of the LCIA using the Human Health NORM exposure characterization method (IRn) were calculated at midpoint and endpoint, however only the endpoint results are shown. R1 has a greater NORM impact per kg REO than R2 and R3. This is primarily due to airborne emission of ^{232}Thorium from mining. t's a radiation dose unit meant for the entire population. For impact to humans, there is a collective radiation dose, Sieverts (Sv) for a given inventory flow (kBq). These can be summed to provide the midpoint indicator for humans, with the unit of man. Sv (a unit, used to represent collective dose for the entire population).

The endpoint indicator for both the NORM impact category of Goronovski et al. (2018) and the artificial nuclide ionizing radiation impact category of Frischknect (2000) recommended in ILCD PEF v 1.09 are measured in Disability Adjusted Life Years (DALY). Comparing the results of the two impact categories (Figure 4) shows that the ionizing radiation impact from natural radionuclides is 6 and 1.25 times greater than that from artificial radionuclides for R1 and R3 respectively. This suggests that the uncharacterized impacts from natural radionuclides are important to include. For R2, the ionizing radiation impact from artificial sources is greater than from natural sources.

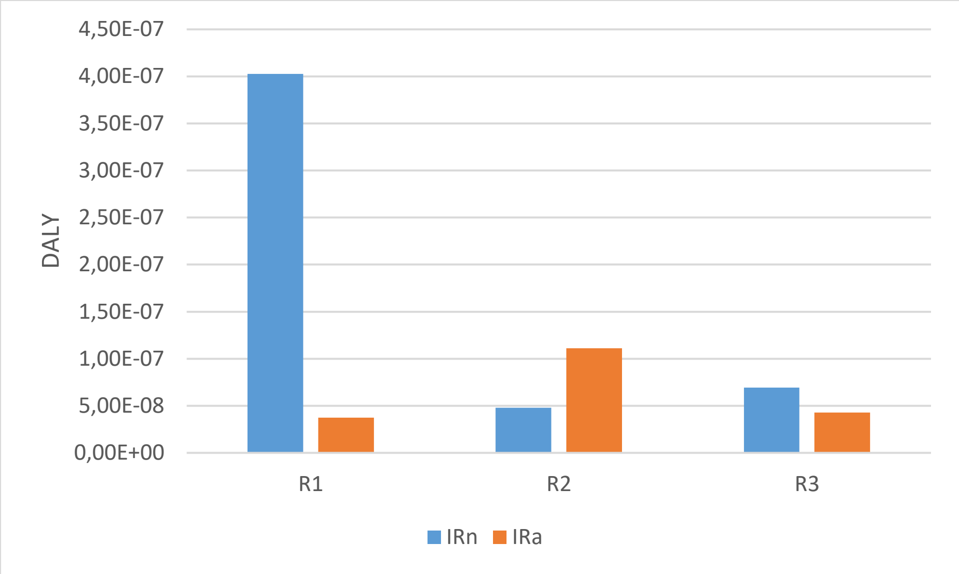

Figure 4. Endpoint results for ionizing radiation using the NORM impact category (IRn) and ILCD/PEF impact category for artificial radionuclides (IRa)

4 Sensitivity Analysis of Solvent Extraction life cycle impact results to different parameters

Because we identified solvent extraction process as the most relevant process in each of our routes, a sensitivity analysis is necessary for identifying parameters that should be known accurately before drawing conclusions (Saltelli et al. 2008). For the solvent extraction process of each route, variants of the LCI dataset were modified by -/+10%. The baseline dataset was used as a reference for the change in results. The changes in the baseline dataset sometimes result in a big change in the results. In R1, a case in point is the electricity consumption during the SX and the ammonia emissions. The electricity consumption causes differences between 0 and 1.77% across the impact categories. The ammonia emissions main impact is eutrophication. For this category, the results are extremely sensitive to the ammonia emission estimates, with a change of +/-7.75% between the -10% and +10% LCI estimate.

For R1, when comparing the +/-10% LCI estimates against the baseline results for the solvent extraction for the production of hydrochloric acid (HCl), the impact assessment of ozone depletion results roughly in a 6% increase/decrease. The LCI estimates for other chemicals in this process do not entail a large variation, and therefore are not sensitive. For R2, the HCl values show similar variation with a +10/-10% increase/decrease. However, with the oxalic acid and P204 values, there is a not a noticeable impact (<1%) on the results. The influence of oxalic acid on the environmental impacts of R3 was also tested and it resulted in difference between 0 and 1.85% across the impact categories.

For R3, a +/-10% variant was modelled for the production of sodium hydroxide. The results show that the impact category of ozone depletion potential varies +/3 % depending on the amount of sodium hydroxide added/removed. Surprisingly, in all three routes, the P204 values are not very sensitive in the climate change category (only about 0.11% change in results with a +10% increase) and are even less sensitive in other categories.

4 Discussion

Of the three processing routes explored, our novel non-Chinese monazite dataset (R2) contains the most complete and up-to-date LCI currently available. However, this route produces a small fraction of the world supply. The main production routes take place in China, either from bastnäsite , or ionic clays (modelled in R1 and R3, respectively). Primary data for Chinese production processes are not readily available, negatively affecting the relative quality of the LCAs for R1 and R3. Another aspect which complicates our REE LCA study is the REE balance problem (Binnemans and Jones 2015). That is, the need for one REE, such as neodymium for rare earth permanent magnets will result in an over-supply of the other (usually light) REEs. Thus, the imbalance of modeling an operation which produces profitable REEs and other REEs which are oftentimes stockpiled or disposed results in an additional potential complication. Today there is no consensus on how to tackle this in LCA.

The contribution analysis allowed us to be able to make some conclusions regarding the 3 aspects contributing to the state-of-the-art: allocation, waste treatment and data gaps. The contribution analysis highlights that the solvent extraction process is one of the most important in all three routes, therefore particular attention should be focused on obtaining accurate data in this step. A complete separation of REOs requires more mixer-settler steps and therefore higher materials and energy consumption, hence bigger environmental impacts. In regard to allocation for multi-output processes, the solvent extraction step is one of the highest contributing steps therefore using system expansion when possible is essential for getting close to the correct results. For waste treatment, the contribution analysis tells us that the waste treatment processes have an influence on the toxicity categories. R2 model has a superior data quality and we recommend using this route as an example when modeling REOs if no other source is available for a light REE mine.

Not all impact categories are equally representative for the production of metals due to controversial assumptions (Eurometaux 2014). LCAs on metal and mining products should at least report the following impact categories (Santero and Hendry 2016):

- Global warming potential
- Acidification potential
- Eutrophication potential
- Photochemical oxidant creation potential
- Ozone depletion potential.

Given the limitations of the characterization models for each of these toxicity categories such as lack of CFs for metals speciation and essentiality, comparative assertions should not rely on toxicity results from USEtox or other toxicity models (PE International 2014).

Another limitation is the lack of data on illegal procurement. The ion adsorption clays are subject to illegal mining since taking the leachate from the ground or ground water requires little effort (Packey and Kingsnorth 2016). These types of mines were at one point estimated to produce approximately 40 percent of the total global production of heavy REOs (Packey and Kingsnorth 2016). There is no data on illegal mining and processing suitable for calculating LCAs. There is however plenty of anecdotal evidence of the extreme environmental consequences. One New York Times investigation wrote that, "The gangs have terrorized villagers who dare to complain about the many tons of sulfuric acid and other chemicals being dumped into streambeds during the processing of ore. Illegal REE mining and chemical runoff have poisoned thousands of acres of prime farmland, according to the government of Guangdong Province, and have been blamed for many illnesses" (Bradsher 2010).What is not measured is not managed, so we don't have information on how the illegal mining and processing is done (or even if it is done differently).

5 Conclusion

The main focus of this article was the discussion of the state-of-the-art of LCI for REEs. The overall results show that the production of REOs from bastnäsite ores (R1) has the highest environmental impacts. R1 and R2 (production from bastnäsite/monazite ores) are two routes that could be considered similar, but the results differ from each other in terms of how the wastes are managed (no life cycle information on Chinese waste management system). Each of the three routes differ in mineralogy which could be an important factor for environmental management of the sites. Furthermore, the variability of the ore structure and composition as well as the chemical reagent management has an influence on the impacts of REE processing and thus is not easily compared.

We identified the important life cycle stage in all three routes: solvent extraction. Therefore, we can focus on better data collection for these processes. But since the collection of this life cycle data is resource intensive and costly for many industries, there are little incentives to update this state-of-the-art. An industry association could help collect life cycle data to continue developing work on a sector benchmark. Of the ten metals commodities associations in Europe, eight have already conducted LCAs (Santero and Hendry 2016). It is recommended to build upon these LCA studies to create an environmental footprint representing the REE sector. A REE industry footprint could help the stakeholders make informed choices about our future green technologies.

Conflict of interest: It should be made clear that one of the co-authors is an employee of Lynas, the company whose mine and processing operations are modelled in one of the three examples presented.

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