

Life Cycle Assessment studies of rare earths production - findings from a systematic review

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Highlights

- Most LCA studies analyze REE production from Bayan Obo and ion adsorption clays
- Life cycle inventories are different despite same process chains
- Different life cycle inventories result in similar environmental impacts
- Treatment of tailings, sludge, and waste as well as radioactivity is often neglected
- The review explains reasons for over- and underestimated environmental impacts

Graphical abstract

Abstract

Rare earth elements (REEs) are one of the most important elements used for transformation of the fossil era into a decarbonized future. REEs are essential for wind, electric and hybrid vehicles, and low-energy lighting. However, there is a general understanding that REEs come along with multiple environmental problems during their extraction and processing.

Life cycle assessment (LCA) is a well-established method for a holistic evaluation of environmental effects of a product system considering the entire life cycle. This paper reviews LCA studies for determining the environmental impacts of rare earth oxide (REO) production from Bayan Obo and ion adsorption clays (IAC) in China, and shows why some studies lead to over- and underestimated results.

We found out that current LCA studies of REE production provide a good overall understanding of the underlying process chains, which are mainly located in China. However, life cycle inventories (LCI) appear often not complete. Several lack accuracy, consistency, or transparency. Hence, resulting environmental impacts are subject to great uncertainty. This applies in particular to radioactivity and the handling of wastewater and slurry in tailing ponds, which have often been neglected.

This article reviews 35 studies to identify suitable LCAs for comparison. The assessment covers the world's largest REO production facility, located in Bayan Obo, as well as in-situ leaching of IACs in the Southern Provinces of China. A total of 12 studies are selected, 8 for Bayan Obo and IACs each. The LCIs of these studies are reviewed in detail. The effects of over- and underestimated LCIs on the life cycle impact assessment (LCIA) are investigated.

The partly controversial results of existing LCAs are analyzed thoroughly and discussed. Our results show that an increased consistency in LCA studies on REO production is needed.

1. Introduction

Rare earths include the chemical elements of the third subgroup of the periodic table (with the exception of actinium) and the lanthanides, a total of 17 elements. Rare earth elements (REEs) have gained enormous economic, public, and scientific interest in recent years. They are often used in key technologies for sustainable mobility and energy supply due to their outstanding functionality in a wide range of applications. REEs can be found in systems like energy saving lamps, electric vehicles, batteries or wind turbines. Today approx. 25% of REEs are used for catalysts and magnets each, 13% for glass and ceramics, 12% for polishing, 8% for batteries and metallurgy each, and 2% for phosphors (Goodenough et al., 2017). In 2015 the utilization of the total REEs was divided between the individual REEs by the following order: 39.5% cerium, 26.4% lanthanum, 19.9% neodymium, 7.1% yttrium, 4.1% praseodymium, 1.1% gadolinium, 0.7% dysprosium, and 1.2% other REEs (Goodenough et al., 2017). In 2019, 210,000 t of rare earth oxide (REO) equivalents were produced in total, including 132,000 t in China, 26,000 t in the U.S., 22,000 t in Myanmar, and 21,000 t in Australia (Gambogi, 2020). Besides 53.4% of neodymium and praseodymium from Bayan Obo, 46.6% REOs came from ion adsorption clays (IACs) including illegal mining (Langkau and Erdmann, 2020). According to (Geng et al., 2020), approx. 59% of REOs came from Bayan Obo (China) in 2016. The Bayan Obo mine is located in Inner Mongolia (Autonomous Region in North China) and is the world's most important production site for REEs and niobium as well as the largest iron ore mine in China.

REE demand growth up to 2026 is likely to be linked mainly to the use of NdFeB magnets (permanent magnet made from an alloy of neodymium, iron, and boron) particularly for hybrid and electric vehicles as well as wind turbines, and in erbium-doped glass fiber for

communications (Goodenough et al., 2017). Many studies have identified REEs as some of the most critical materials nowadays and in the future (Ray Moss et al., 2013), in terms of supply risk and, with the exception of erbium and lanthanum, also in terms of their economic importance for the European Union (European Commission, 2017).

The scientific literature evaluated different aspects like RE resources and availability, political implications regarding the high geographic concentration of RE production in China (Alonso et al., 2012) or material flow analysis (Peiró et al., 2013; Guyonnet et al., 2015; Watari et al., 2020). Furthermore, economic, social, and sustainable challenges, constraints, and opportunities were highlighted in various papers (Wübbeke, 2013; Golev et al., 2014; Haque et al., 2014; McLellan et al., 2014; Packey and Kingsnorth, 2016; Fernandez, 2017). In addition, several Life Cycle Assessment (LCA) studies are available that focus on the environmental effects of REE production based on primary and secondary resources. This review focuses on LCA studies of primary REE production, as this is associated with high environmental impacts.

LCA is standardized according to ISO 14040 and 14044 (ISO 14040:2006, 2016; ISO 14044:2006, 2016). Despite these LCA standards, in practice, several LCA studies on REO production are available, that lead to a wide range of environmental impacts. Reasons for the variations are settings of different system boundaries and parameters, incomplete inventories, different or incorrect allocations, choices of impact assessment methods or simply calculation errors. Consequently, decision-makers in industry and politics feel insecure, and even the credibility of LCA is impaired. Therefore, a consistent determination of the environmental performance of REO production is needed. Due to the large discrepancies in LCA literature and their importance for a sound assessment of REO production, we offer a clarifying perspective. In this paper, we show why some LCA studies led to over- and underestimated environmental impacts for REO production and their effects on the conclusions drawn.

This paper reviews LCA studies on primary REO production with the aim of identifying those suitable for a systematic comparison. The proceeding is described in the method chapter 2.

An initial screening shows that only studies focusing on Bayan Obo and IACs are suitable, as they are available in a sufficient number (Chapter 3). The selected LCA studies are scrutinized to an in-depth analysis (Chapter 4 for Bayan Obo; Chapter 5 for IACs), initially focusing on life cycle inventory (LCI). Since not all LCI data are provided equally, they are standardized for comparison.

Finally, the influence of the most important inputs and outputs on individual impact categories as well as on the overall environmental performance are discussed. Furthermore, special attention is paid to radioactivity released during REO production, because many studies completely neglect this important aspect so far.

2. Methods

For the literature search, we used "web of science" (www.webofknowledge.com), the "scopus" platform (www.scopus.com), and "sciencedirect" (www.sciencedirect.com). We considered the terms "life cycle assessment", "life cycle analysis", "life cycle inventory", "life cycle impact", "life cycle impact assessment", "environmental impacts", "environmental burdens" in combination with "rare earth", "rare earth elements", "rare earth oxides", "rare earth metal", "rare earth product", "rare earth supply", "rare earth deposit", "light rare earth", "heavy rare earth", "neodymium", "dysprosium", "praseodymium", "NdFeB permanent magnet", "bastnäsite", "Bayan Obo", "ion-adsorption deposit", "ion-adsorption rare earth resources" and "ion adsorption clay". LCA studies on REO production from secondary resources such as mining waste, tailings, and magnet scrap were not included.

Following the study selection (Chapter 3), short descriptions of the process chains for Bayan Obo (Chapter 4) and IACs (Chapter 5) are given. The LCIs of the studies were subject of a detailed analysis. From raw ore mining to separation of individual REOs, the data were compared along the Bayan Obo process chain. Since separation by solvent extraction (SX) and calcination to individual REOs are identical for both deposit types, the IACs are only

discussed in detail regarding in-situ leaching, precipitation, and calcination to mixed REOs. For SX, only data sources were compiled and a few general statements were made.

LCI data were not always given in the same way in all studies. Some studies related data to 1 kg REO, others to 1 kg neodymium or dysprosium. A few studies summarized inputs and outputs for individual processes. In order to compare the different studies, we assigned the inputs and outputs to 1 kg of REO under consideration of e.g. ore compositions, RE concentrations in ore, concentrates, and chlorides as well as allocation factors. If calculation errors were detected, some LCIs were recalculated exemplarily. Inputs with minor impact on the results that were not considered in all studies, such as transport, land requirements, and expenditures for the construction of facilities, buildings, etc. were not compared. Only emissions that were included in several studies were listed in the inventory tables. The complete lists of emissions can be found in the cited studies. For some processes (e.g. mining or roasting), the studies used various energy sources such as coal, natural gas, oil, and electricity. In order to compare these different energy sources, the given values were converted to primary energy (PE) using PE factors (Deutscher Bundestag, 2017). For Chinese electricity, a PE factor of 3.3 was determined based on its electricity mix (International Energy Agency (IEA), 2016).

After the detailed analysis of the LCIs, the influence of the inventory data on the life cycle impact assessment (LCIA) results was examined. The studies used different LCIA methods, which differ on the one hand in number and selection of the individual impact categories and on the other hand in methods for quantifying the same impact categories. For example, the results of Acidification potential (AP) were given in kg SO₂ eq. for the LCIA methods ReCiPe and CML and in mol H⁺ eq. for ILCD and TRACI. Therefore, a direct comparison was often not possible. Only the same impact categories calculated by the same method with the same units can be compared reasonably. This only applies to Global warming potential (GWP) and Ozone depletion potential (ODP). Moreover, Human toxicity potential (HTP), Eutrophication potential (EP), Photochemical ozone creation potential (POCP), Fossil depletion (FD), Particulate

matter formation (PM), Ionizing potential (IR), Ecotoxicity potential (ET), and Freshwater aquatic ecotoxicity (FAETP) were considered in the studies. Since Althaus et al. (2007b) only provided LCI results, we calculated the LCIA ourselves.

3. Selection of LCA studies

In the last 15 years the following 35 important studies based on primary resources were cited most, due to their quality (Althaus et al., 2007b; Bouorakima, 2011; Peiró et al., 2013; Koltun and Tharumarajah, 2014; Navarro and Zhao, 2014; Nuss and Eckelman, 2014; Sprecher et al., 2014; Zaimes et al., 2015; Browning et al., 2016; Jin et al., 2016; Schreiber et al., 2016; Vahidi et al., 2016; Vahidi and Zhao, 2016; Weng et al., 2016; Ikhlayel, 2017; Lee and Wen, 2017; Schulze et al., 2017; Vahidi and Zhao, 2017; Arshi et al., 2018; Chen et al., 2018; Lee and Wen, 2018; Lima et al., 2018; Marx et al., 2018; Vahidi and Zhao, 2018; Zapp et al., 2018; Adibi et al., 2019; Deng and Kendall, 2019; Pell et al., 2019; Zhang et al., 2019; Bailey et al., 2020; Fernandes et al., 2020; Koltun and Klymenko, 2020; Langkau and Erdmann, 2020; Schreiber et al., 2020; Bailey et al., 2021). Only two of them (Althaus et al., 2007b; Bouorakima, 2011) were published more than 10 years ago. It is remarkable that 27 of 35 studies were published in the last 5 years. This illustrates the increased importance and attention given to REEs. It is also noteworthy that 25 studies examined Bayan Obo. Five studies exclusively investigated REE production outside China, namely from a monazite-rich niobium deposit in Brazil (Lima et al., 2018; Fernandes et al., 2020), from Australian monazite mineral sands (Browning et al., 2016; Koltun and Klymenko, 2020), and from the Bear Lodge project in Wyoming, USA (Pell et al., 2019). Chen et al. (2018) considered the bastnäsite deposit in Sichuan and used data from Arshi et al. (2018) and Lee and Wen (2017). Therefore, these six studies were not considered because they were not comparable to any other. There were not enough studies available to make an adequate comparison for these deposits.

In the following, all 35 studies are briefly described with the aim of selecting appropriate Bayan Obo and IAC studies for a fair comparison regarding LCI and LCIA results. Major differences between the studies regarding system boundaries, functional units, data sources, and level of detail are discussed.

3.1 Bayan Obo studies

Due to the high importance of the Bayan Obo mine for the world's REO production, it is no surprise that 25 of the 35 studies examined Bayan Obo, and sometimes other deposits additionally. However, not every study gathered primary LCI data from Bayan Obo. Seventeen of the 25 Bayan Obo studies were not suitable for comparison. The reasons were manifold, e.g. missing LCI data (Bouorakima, 2011; Weng et al., 2016; Ikhlayel, 2017), fragmentary LCI data (Peiró et al., 2013; Koltun and Tharumarajah, 2014) or LCI data that exclusively relied on already published data (Navarro and Zhao, 2014; Nuss and Eckelman, 2014; Jin et al., 2016; Adibi et al., 2019; Langkau and Erdmann, 2020; Bailey et al., 2021). For example, Langkau and Erdmann (2020) used data from Lee and Wen (2017) for Bayan Obo, data from Marx et al. (2018) for Mountain Pass and Mount Weld and data from Zapp et al. (2018) for Norra Kärr. Four studies were excluded because only parts of the process chain were examined, namely SX and molten salt electrolysis (Vahidi and Zhao, 2016; 2017; 2018; Schreiber et al., 2020). Two of our previous studies (Schreiber et al., 2016; Zapp et al., 2018) were not considered for Bayan Obo, since they focused on the production of neodymium and dysprosium from eudialyte mineral located in the Norra Kärr mine in Sweden on the one hand and on IAC on the other.

This led to eight studies being suitable for comparison (Table 1). The first study on REOs produced at Bayan Obo was conducted by ecoinvent - a not-for-profit association founded by several institutes of the ETH Zurich and Agroscope - which examined RE concentrates and REOs (Althaus et al., 2007b). This study served as LCI database for many other LCA studies (Navarro and Zhao, 2014; Nuss and Eckelman, 2014; Adibi et al., 2019). However, from today's perspective it has considerable limitations which have already been discussed in detail

by Navarro and Zhao (2014) and Marx et al. (2018). Since then, research has progressed. In the recent ecoinvent 3.7 version, which was released in September 2020, new datasets for Chinese REO production from Bayan Obo, Sichuan, and IAC were provided by the Swiss Federal Laboratories for Materials Science and Technology (EMPA - Eidgenössische Materialprüfungs- und Forschungsanstalt) (Kakkos, 2020). The datasets consider data taken from Arshi et al. (2018), Sprecher et al. (2014) and Lee and Wen (2017). As the three original articles are analyzed in detail in this review, it is not necessary to discuss the RE datasets of ecoinvent 3.7 additionally. Only at two points, we interpret the ecoinvent 3.7 data.

In 2014, Sprecher et al. (2014) conducted detailed LCIs related to 1 kg REO, 1 kg neodymium, and 1 kg NdFeB magnet. Due to data uncertainty, the authors considered three scenarios: a baseline scenario that represents the current state of the industry, a high-tech scenario that assumes best available technologies and a low-tech scenario. The main differences between the scenarios are efficiencies of various processes along the production chain and differing emission control systems. For the comparison, we used the baseline scenario because Sprecher et al. (2014) provided LCI data only for this scenario. They used many ecoinvent data (Althaus et al., 2007b) as well as extended and adapted data for RE concentrate and neodymium oxide production. In recent years, Sprecher's study has often been used as basis for other REE studies (Navarro and Zhao, 2014; Jin et al., 2016; Bailey et al., 2020).

The LCA study by Zaimes et al. (2015) provided results related to 1 kg of light, medium, and heavy REOs, respectively, as well as to 1 kg of neodymium oxide. The authors used the ecoinvent database and Chinese scientific literature for calculation of input data like energy and chemicals. An older MEP document was used for calculation of emissions (Chinese Ministry of Environmental Protection, 2009). In addition, Zaimes et al. (2015) compared mass-based, market-based and exogetic-based allocation schemes for the individual REEs. Finally, the authors showed the life cycle carbon footprint and primary energy consumption for REO production against other common metals like steel, cobalt and aluminum.

Another detailed study was presented by Lee and Wen (2017). The study considered 15 different functional units corresponding to different RE metals. The assessment included three main RE production sites in China: REO production in Bayan Obo, Sichuan, and IACs. Primary data were collected for foreground production processes from Chinese and further international scientific literature, industry reports, government and industry surveys. The authors assumed a baseline for aqueous and gaseous emissions using national discharge standards for industry from the Chinese Ministry of Environmental Protection (2011). For the documentation of exceedances of specific pollutants the authors used an older MEP document (Chinese Ministry of Environmental Protection, 2009). Lee and Wen (2017) compiled data for three scenarios to account for large discrepancies in production efficiency and waste treatment. The upper bound includes small- to medium-sized enterprises scattered across China, usually with low production capacity and minimal to low waste treatment. The lower bound contains large, high-capacity enterprises, which are able to adopt latest technologies for production and waste treatment to meet environmental standards. The third scenario was developed to capture the industry average, which models the national mean for both production efficiency and waste treatment. For comparison, we used the average scenario.

Building on Lee and Wen (2017), the same authors published a follow up study that focused on harmonized LCIs for both legal and illegal Chinese RE production from Bayan Obo, Sichuan, and Southern Provinces (Lee and Wen, 2018). For the Bayan Obo pathway, the authors harmonized data about material inputs and pollutant discharges of five studies (Sprecher et al., 2014; Zaimes et al., 2015; Lee and Wen, 2017; Vahidi and Zhao, 2017; Zapp et al., 2018). For the IAC pathway, they used the LCI from Vahidi et al. (2016), Vahidi and Zhao (2017), Lee and Wen (2017), Schulze et al. (2017), and Zapp et al. (2018). For legal production, the authors harmonized data by calculating the average from the previous studies. For unregulated REO production, they conservatively assumed worst-case values that yielded highest environmental impacts. In addition, they adjusted the efficiency of recovery, collection, energy and recycling to reflect legal and illegal production. Furthermore, Lee and Wen (2018) developed a total of 30 scenarios to quantify impact changes caused by projected annual

production output from legal and illegal mines, policy and technology development trends. For our comparison, we used the LCI datasets for legal REO production from Bayan Obo and IAC in combination with the base case scenario of business as usual production technologies in 2015 from Lee and Wen (2018). We did not include the datasets for illegal (unregulated) REO production because they are not comparable to the other studies. For completeness, we included the data for illegal REO production in the supporting information (Table S3 and S4).

Arshi et al. (2018) compiled LCIs based on representative production pathways in China using data on facility level, covering Bayan Obo as well as IACs. The study built on existing scientific literature and Chinese facility reports. Detailed material and energy flows for each process step were compiled to create an interlinked Excel-based model that allows users to analyze different production pathways for REOs, RE metals, and NdFeB magnets.

We performed a LCA study to quantify the environmental impacts of neodymium and praseodymium oxide production from Bayan Obo, Mount Weld (Australia), and Mountain Pass (USA) deposits (Marx et al., 2018). Furthermore, subsequent metal and NdFeB magnet production were considered.

Bailey et al. (2020) provided general LCIs of bastnäsite (Bayan Obo), monazite (Mount Weld), and IACs (Southern Provinces) by bringing together and reanalyzing published LCIs.

3.2 IAC studies

For comparison of IACs, we selected 8 studies (Table 1). In addition to four Bayan Obo studies, which also include LCIs for IACs (Lee and Wen, 2017; Arshi et al., 2018; Lee and Wen, 2018; Bailey et al., 2020), four further IAC studies were appropriate for comparison (Vahidi et al., 2016; Schulze et al., 2017; Zapp et al., 2018; Deng and Kendall, 2019). Zhang et al. (2019) used data from Vahidi et al. (2016) and Schulze et al. (2017) and was therefore not considered.

The study by Vahidi et al. (2016) was the first LCA on IACs. The authors gathered material and energy flows from Chinese literature and verified them through personal interviews with Chinese experts working in the RE industry. They also used emission limits set by MEP

(Chinese Ministry of Environmental Protection, 2009) as the best case scenario. The process chain ends at mixed REOs. SX and metal production were not included in their analysis. Nevertheless, this study provided a comprehensive inventory and served as a basis for further studies.

In Schulze et al. (2017), the in-situ leaching data were largely adopted from Vahidi et al. (2016). However, the system boundaries were broader and included the separation of RE concentrate into individual REOs using SX.

In a previous study of ours (Zapp et al., 2018), we assessed the environmental impacts of REOs, dysprosium oxide, and dysprosium metal produced from Chinese IACs. Special attention was paid to the treatment of sludge, waste, and wastewater.

Deng and Kendall (2019) gathered primary data from sites producing heavy rare earth oxides (HREOs) from IACs during a field study in southern China. The authors collected data from four HREE mining sites in Ganzhou (Jiangxi Province) to create two original LCI datasets for HREOs representing a low-tech and a high-tech scenario. The compiled datasets, which include in-situ leaching, extraction, and calcination, were provided as open-source LCI for the LCA community. Further processes such as SX and individual metal separation were not included.

In the following, the selected studies are cited only with the name of the first author and, if necessary, with the year in order to improve readability: Althaus (Althaus et al., 2007b), Arshi (Arshi et al., 2018), Bailey (Bailey et al., 2020), Deng (Deng and Kendall, 2019), Lee 2017 (Lee and Wen, 2017), Lee 2018 (Lee and Wen, 2018), Marx (Marx et al., 2018), Schulze (Schulze et al., 2017), Sprecher (Sprecher et al., 2014), Vahidi 2016 (Vahidi et al., 2016), Zaimes (Zaimes et al., 2015), Zapp (Zapp et al., 2018). It should also be noted that some studies were published by the same group of authors. Thus, Marx, Schreiber and Zapp, as well as Bailey, Schulze and Sprecher, and also Arshi, Vahidi and Zhao each form a group. This note is often very helpful in interpreting the data.

311

312 Table 1 gives an overview of the 12 studies that are suitable for comparison of the Bayan Obo
313 bastnäsite/monazite pathway and those for an IAC comparison.

314

315 Table 1

316 Main features of the selected LCA studies for comparison.

317

318 **4. Review of the Bayan Obo studies**

319 **4.1 Process chain (Bayan Obo)**

320 The ore is mined by open pit mining using conventional surface mining techniques such as
321 drilling and blasting (Fig. 1). It has a high iron content, which is separated by magnetic
322 separation after crushing and grinding. The remaining tailings run through a froth flotation,
323 where the REE containing minerals are concentrated by separation of gangue. The number of
324 flotation and purification stages as well as the magnetic separation process vary. During
325 subsequent roasting using sulfuric acid (H_2SO_4), which is applied to 90% of the Bayan Obo
326 ore (Zaimes, Lee 2017), the RE concentrate is converted into an easily soluble RE sulfate. The
327 RE sulfate is then separated from remaining gangue and secondary elements by leaching. In
328 a second hydrometallurgical process step, various chemicals precipitate the REEs. By adding
329 hydrochloric acid (HCl), the RE sulfate is converted into a RE chloride solution which is
330 required for the following SX. To produce individual REEs multiple stages of mixers and settlers
331 and different types of extracting agents are utilized in the SX, mostly based on phosphoric acid
332 (P507, P204). Up to 300 separation steps may be required to obtain individual REEs with a
333 purity over 99% (Vahidi 2017). Subsequent, the separated REEs are precipitated as RE
334 oxalates or RE carbonates by adding oxalic acid or ammonium bicarbonate (NH_4HCO_3). REOs
335 are produced by calcination of the RE oxalates or RE carbonates at temperatures of approx.

900°C under the release of CO₂. Finally, individual RE metals can be produced from REOs by different techniques such as molten salt electrolysis, calciothermic reduction, and metallothermic reduction (Vahidi 2018). However, the metal reduction step is not subject of this review, as it has already been examined in detail in another publication by Schreiber et al. (2020) and therefore no additional findings are expected.

Fig. 1

Schematic representation of RE production at Bayan Obo.

For a better understanding, not each individual process of all studies is shown in *Fig. 1* and *Fig. 3* since some processes (e.g. pH adjustment, special leaching steps and recycling options of chemicals) are only described in one study.

4.2 Life Cycle Inventory of REO production (Bayan Obo)

4.2.1. Mining

Because no primary mining data from Bayan Obo were available, LCI data in the studies (Sprecher, Arshi, Marx, Bailey (Classen et al., 2007)) are based on different data sources either from iron mining or in Althaus from phosphate rock mining. Main inputs are energy and blasting agent (Table 2).

With regard to the demand of blasting agent, a large difference can be recognized between Zaimes and the other studies. In our opinion, Zaimes assigned an excessive amount of blasting agent (76 kg) to 1 kg of REO. He used the amount of crude ore (76 kg) instead the amount of blasting agent (0.0002 kg) as required by theecoinvent process “RER blasting”, which he used to model the mining process.

Sprecher assumed a high PE input of 1.5 MJ/kg ore with reference to Norgate and Haque (2010). However, since a PE input of 150 MJ/t ore (excluding crushing, transport to port, etc.) was given in Norgate and Haque (2010), we suspected a conversion error (factor 10). The same applies to Arshi and Bailey, who used the energy value from Sprecher. Due to this conversion error, the energy required for mining is the highest compared to the total energy required for the entire process chain (78%) in the study by Sprecher. It is also high for Bailey (35%) and Arshi (37%). Marx used the value for iron mining in Classen et al. (2007) and adjusted it to the lower stripping rate (kg moved rock per kg mined ore) of Bayan Obo. The energy value in Norgate and Haque (2010) is about a factor 10 higher than in Classen et al. (2007) due to the assumed high PE demand for hauling (0.102 MJ/kg ore). Zaines presented the lowest energy requirement of 0.754 MJ/kg REO. This is due to the low energy requirement for ore transport (4.3E-05 MJ/kg ore), which is significantly lower than, for example, that of a mine truck in the GaBi database from thinkstep (thinkstep, 2019). GaBi mine truck data is often used by the LCA community, for which an energy requirement of 0.019 MJ/kg rock is specified.

Although the dust emissions given in all studies are based on the same ecoinvent process “iron mine operation, crude ore, 46% Fe (GLO)”, they differ. Reasons are the different ore inputs/t REO (Table 2, Table 3) as well as different stripping rates, which were only considered by Marx taking into account the conditions of Bayan Obo. It should be mentioned that the dust emissions stated in the aforementioned ecoinvent process were originally based on personal information of the European Aluminum Association (EAA) concerning bauxite mining.

Lee 2017 and Lee 2018 did not provide exclusive mining data, although those were considered and modeled with data from the Chinese Life Cycle Database (CLCD). As CLCD has no open access, the values cannot be verified.

4.2.2. Beneficiation

The process sequences of beneficiation including crushing, grinding, magnetic separation, and froth flotation were presented very similar in all studies. Nevertheless, there are large differences regarding type and quantity of chemicals, energy consumption, and emissions (Table 2) due to various data sources. For example, data are based on stoichiometric calculations, Environmental Assessment Reports of Chinese RE companies, and threshold values by MEP. In addition, different ore and RE concentrate inputs per kg REO (Table 3) were used depending on REO concentration in ore and concentrate as well as recovery rates.

Although Arshi and Bailey took the mining data from Sprecher, different LCIs were determined. Reason is that Bailey adopted Sprecher's assumptions (REO concentration in ore (4.1%) and concentrate (61%), 50% recovery rate) but Lee's 2017 ore input of 15 kg ore per kg RE concentrate in contrast to Sprecher and Arshi who required 30 kg ore (Table 3). However, Lee's 2017 ore input is based on a REO concentration of 7% in ore and 50% in RE concentrate. Since Bailey assumed also Lee's 2017 recovery rate of 50%, the ore input is too low by a factor of two. A similar problem occurred for the newest ecoinvent 3.7 RE datasets (Kakkos, 2020). Kakkos (2020) used Sprecher's mining data and in the following Arshi's data for beneficiation and separation. However, Kakkos (2020) did not take into account that Sprecher assumed a 61% RE concentrate, while Arshi assumed a 50% RE concentrate. Therefore, data regarding Bayan Obo mining (e.g. blasting agent, diesel, mine infrastructure, recultivation area) were miscalculated for the RE datasets in ecoinvent 3.7.

In the ecoinvent process "Rare earth concentrate 70%, from bastnäs site, at beneficiation (CN)" provided by Althaus, the sum of flotation chemicals (1.2 kg/kg REO) is the largest. Reason is an additional HCl leaching process during flotation. The HCl accounts for 82% of the total flotation chemicals. However, the flotation process used in Althaus is based on a pure bastnäs site ore (Molycorp Process (Gupta and Krishnamurthy, 2005)) and is not representative for Bayan Obo. The sum of flotation chemicals assumed in other studies is between 0.11 and 0.32 kg/kg REO (with the exception of Sprecher) and accounts for only a small share on the total chemical consumption of the entire process chain.

Some studies estimated a need for steam during beneficiation. In Althaus, the steam demand is based on the Molycorp process, which involves steam conditioning of the ore before flotation. Sprecher's steam demand is based on Althaus. Lee 2017 did not specify the intended purpose for steam. It can be assumed that steam will be used to dry the concentrate. The extremely low steam input in Bailey (4.8×10^{-11} kg) cannot be explained, although Lee 2017 was given as reference.

Althaus reported the lowest electricity demand (0.139 kWh/kg REO) and Zaines the highest (2.03 kWh/kg REO). The low value in Althaus is based on an energy requirement of 0.016 kWh/kg ore. Accordingly, 0.311 kWh/kg of concentrate are required presuming an ore input of 19.44 kg/kg of concentrate. However, Althaus stated 0.088 kWh/kg concentrate for incomprehensible reasons. Thus, a corrected energy demand is 0.494 kWh/kg REO, which is still lower than in the other studies. Althaus' value is based on phosphate rock, whereas Marx and Zaines calculated the electricity for crushing and grinding on the basis of rock hardness. Besides several data sources, the different ore input (15 to 40 kg ore/kg concentrate, Table 3) also contributes to the differences in energy demand.

During beneficiation, considerable quantities of tailings are generated, which are discharged into an open tailing pond (Qifan et al., 2010; Huang et al., 2014). They might be released into the environment by infiltration or erosion. The tailings include finely ground ore and flotation chemicals. Marx's study is the only one that assumed an infiltration corresponding to the Bayan Obo ore composition based on Castor and Hedrick (2006). However, due to a lack of data, the infiltration rate could only be roughly estimated. The other studies used threshold values by MEP (Zaines, Lee 2017, Lee 2018, Bailey), emissions based on theecoinvent process "iron ore 65% Fe, at beneficiation (GLO)" (Sprecher), data from an Environmental Assessment report of a Chinese RE facility (Arshi), and data from the Environmental Protection Agency (EPA) (Environmental Protection Agency, 1998) (Althaus), which are not specifically adapted to Bayan Obo.

Data on dust exposure are available in 5 of 8 studies (Lee 2017, Arshi, Lee 2018, Marx, Bailey). The values vary between 0.0008 (Lee 2017) and 0.33 kg dust/kg REO (Marx). The low values in Lee 2017 and Bailey were adopted from MEP for mining and probably do not include dust generated by crushing. Arshi's low dust emission is based on data from a Chinese company and cannot be comprehended. Marx calculated the dust emissions during crushing. They are based on dust emissions for metal ore processing and are adapted to the rock hardness of the Bayan Obo deposit.

4.2.3. Roasting and hydrometallurgical processes

The very low energy demand for roasting in Althaus (1.06 MJ/kg REO) is based on a value for phosphate rock (Landbank, 1994). Other energy inputs for roasting vary between 8 (Zaimes) and 68.4 MJ/kg REO (Lee 2017). The reasons are again various data sources. Lee 2017 used an industry survey. Zaimes and Marx calculated the thermal energy to 8 and 12.2 MJ/kg REO, respectively. Marx calculation is based on ASPEN modeling considering heat capacities. Zaimes did not provide an explanation. In Marx, an additional energy input of 2.7 MJ/kg REO is given for turning of the rotary kiln (Krüger et al., 2002). Arshi and Bailey used Sprecher's energy value, which is based on a Chinese reference stating 6.59 MJ/kg roasted RE concentrate. The different energy values of Bailey and Arshi in contrast to that of Sprecher are due to different amounts of roasted concentrate/kg REO, which varies between 1.98 (Bailey), 3.44 (Sprecher), and 3.86 kg (Arshi).

With the exception of Althaus, the differences for the required H_2SO_4 are not large. Althaus assumed a stoichiometric excess of only 5%. Sprecher, on the other hand, stated the highest amount of H_2SO_4 (5.32 kg/REO), assuming that only 64% of the H_2SO_4 used reacts. Although Bailey adopted Sprecher's data for the roasting process, the H_2SO_4 required for the entire process chain is lower. The reason for this is that Bailey switched between the data from Lee 2017 and Sprecher without considering the different input amounts of RE compounds such as

RE chloride. The amounts of H_2SO_4 used in Lee 2017, Arshi, and Marx are based on Chinese references and do not differ significantly.

Almost all studies considered emissions caused by the decomposition of the RE concentrate (CO_2 , hydrogen fluoride (HF)) and by the H_2SO_4 excess (SO_2), but no specific plant data were used in any study. There is a large difference between the highest and the lowest SO_2 emissions (529 g/kg REO in Zaimes and 0.0875 g/kg REO in Arshi). Arshi supposed an advanced pollution control and H_2SO_4 recycling. However, these assumptions only applies to large facilities, which do not reflect the Chinese industrial average. The other studies presented SO_2 emissions between 0.9 (Althaus) and 130 g/kg REO (Lee 2018). A low stoichiometric excess of H_2SO_4 can explain the low value in Althaus. Lee's 2017 and Marx's values also differ significantly, although both studies refer to MEP, but to different MEP versions. Lee 2017 used MEP data from 2009 (Chinese Ministry of Environmental Protection, 2009) which are based on unfiltered exhaust gas that have been reduced by a factor of 5 taking into account an exhaust gas treatment. Marx used MEP data from 2011 (Chinese Ministry of Environmental Protection, 2011) with significantly lower threshold values.

The CO_2 emissions for roasting differ between 83 (Zaimes) and 594 g/kg REO (Lee 2017). Althaus stated an even higher value (855 g), but this cannot be taken into account, as this value also includes calcination. Since Zaimes reported the lowest CO_2 and highest SO_2 emissions, we suspect a mix-up of both values, especially since the high SO_2 value (529 g) would match the CO_2 values of the other studies. Although Marx indicated the highest total CO_2 emissions (823 g/kg REO), the stoichiometric calculated CO_2 emissions caused by roasting (204 g) are the lowest. The remaining 619 g CO_2 emissions/kg REO are caused by combustion of natural gas in the rotary kiln. The other studies only considered direct CO_2 emissions caused by roasting of the RE carbonates with H_2SO_4 . The energy-related CO_2 emissions caused by combustion of gas, coal or oil are included in the combustion processes and are not reported in Table 2 separately.

With the exception of Marx, all studies indicated HF emissions, which also show large differences from 0.0048 g (Arshi), over 5.4 g (Althaus) to 84 g (Sprecher) per kg REO. Sprecher and Althaus calculated the HF emissions based on the HF quantities produced during roasting and the efficiencies of exhaust gas scrubbing. The produced HF quantities during roasting range from 108 g (Althaus) to 280 g (Sprecher) per kg REO without exhaust gas scrubbing. Sprecher referred to a value from a Chinese reference (81.6 g HF/kg $\text{RE}_2(\text{SO}_4)_3$). Althaus used a fluorine concentration of 6.4 to 9.8% in the RE concentrate, which is based on a study of RE resources for the Mount Weld deposit in Australia, to calculate the HF emissions. However, the differences in HF emissions (factor 16) result mainly from the different efficiencies of exhaust gas scrubbing, which are 70% for Sprecher and 95% for Althaus. The low HF emissions in Arshi are probably caused by the use of a new type of pollution control system. Since this system is not described in the paper, it is not comprehensible how Arshi has calculated the HF emissions. In any case, the value does not reflect the Chinese industrial average. Marx stated fluoride emissions based on an emission list in MEP. However, it could be that the MEP list actually refers to HF instead of fluorides.

The sludge resulting from exhaust gas scrubbing was only considered in Marx, but with large uncertainties regarding the infiltration rate and the share of dissolved solids. Sprecher and Althaus used theecoinvent process "disposal, sulphidic tailings, off site" for the disposal of excess H_2SO_4 (1.73 for Sprecher, 1.97 kg/kg REO for Althaus). This assumption is incorrect, as this is not sulfidic but sulfatic waste. An analysis of Bayan Obo tailings has identified less than 2% sulfides in form of pyrite (Li et al., 2016), which were already separated during beneficiation and before roasting. In the new ecoinvent 3.7 dataset "REO production, from REO concentrate, 50% REO, CN-NM" (Kakkos, 2020), sulfidic tailings were also assumed (based on Sprecher). Furthermore, Kakkos (2020) calculated 5.81 kg tailing output/kg REO, although he used only 2.49 kg H_2SO_4 input/kg REO for roasting. This seems to be a mix-up, because Sprecher used approx. 5 kg H_2SO_4 input and 1.73 kg tailings as output (Table 2). Therefore, it can be assumed that ET and HTP impacts are overestimated in the ecoinvent 3.7 dataset.

After roasting, a water leaching process is carried out to remove accompanying elements. The studies differ with regard to the chemicals being used (MgO , CaCO_3 , FeCl_3 , H_2SO_4). Since their quantity is small compared to the chemicals required for HCl leaching and precipitation the water leaching chemicals are mostly neglected (Althaus, Arshi, Specher, Zaimes). Various precipitation chemicals e.g. caustic soda, FeCl_3 or NH_4HCO_3 are used in the subsequent hydrometallurgical precipitation processes. Precipitation products can be RE carbonates, RE hydroxides or RE double salts. Lee 2017 assumed liquid-liquid extraction with P204 and saponification with ammonia. Due to different data sources used in the studies, six different hydrometallurgical procedures are applied (Fig. 1, blue box) so that the precipitation cannot be compared in detail.

The amount of HCl added to produce RE chlorides required for SX varies between 0.45 (Althaus) and 2.46 kg/kg REO (Lee 2017). If only a small stoichiometric excess is expected (Althaus) or HCl recycling is taken into account (Arshi), the calculated HCl quantities are rather small. Sprecher and Marx calculated HCl amounts that are close to Lee's 2017 industry data.

Only two studies indicated emissions to water, which are based on MEP (Zaimes, Lee 2017). Some studies are aware of the wastewater problem, but do not provide information due to lack of data (Sprecher, Arshi). Marx considered sludge produced during leaching and wastewater treatment by including seeped sludge components. However, the infiltration rates and solubility of the solids contained in the sludge are only roughly estimated.

4.2.4. Solvent extraction and calcination

SX mainly uses HCl and extraction agents dissolved in kerosene. The total of the extraction agents used is compared, since they are chemically similar. Except for Althaus, the demand varies between 0.005 (Lee 2017, Bailey) and 0.125 kg/kg REO (Sprecher). Althaus assumed a significantly higher demand of extraction agents (1.0 kg/kg REO). Althaus equated the amount of waste generated to the amount of extraction solution. Based on data for solvent

waste (0.006 to 6.1 kg solvent waste/kg REO) given in EPA (Environmental Protection Agency, 1998), Althaus assumed an average for waste of 3 kg/kg REO. As the share of kerosene in the extraction solution is approx. 68%, he divided it into 2 kg kerosene and 1 kg extracting agent/kg REO. Sprecher calculated the demand of extraction agent to 2.5 kg extraction agent/kg REO and assumed an actual consumption of only 5% (0.125 kg/kg REO). However, Sprecher assumed the high kerosene demand from Althaus, which is too high in relation to Sprecher's low amount of extraction agent. Zaimes did not specify extraction chemicals.

The amount of used HCl varies. There is a clear difference between calculated values (0.28 to 0.97 kg/kg REO) in Zaimes, Althaus, and Sprecher as well as values based on industrial figures (6.0 to 6.96 kg/kg REO) in Lee 2017 and Marx. The basic data in Marx were adopted from a technical report of a Malaysian REE facility (DNV, 2010). Arshi's HCl demand is also based on industrial figures. However, the low requirement of 1.31 kg/kg REO is not comprehensible because the information from the Chinese plant (2.32 kg/kg RE chloride solution) would actually result in about 4 kg HCl/kg REO for Arshi's study. The industrial figures suggest that the industry operates with a high stoichiometric excess and that the values calculated in Zaimes, Althaus, and Sprecher do not reflect actual consumption.

Arshi's energy requirement for extraction is 18 times higher than in other studies and accounts for 25% of the total energy demand. However, an explanation was not given. We assume that additional energy is needed to recover RE chloride by evaporation from an aqueous RE chloride solution obtained after SX and HCl stripping according to a process scheme of Vahidi and Zhao (2017). The recovered RE chloride is again fed to the SX.

The precipitation of REEs is carried out as RE oxalate or RE carbonate. The amount of precipitation chemicals varies between 0.435 (Sprecher) and 1.6 kg/kg REO (Lee 2017, Bailey). Again, studies based on industry figures show the highest demand. In Zaimes, NaOH, H₂SO₄, and coke were given instead of oxalic acid. Due to the missing reference to the used production process of oxalic acid, the amount of oxalic acid cannot be determined and compared to the other studies. However, the very large amount of NaOH stands out.

571 Another striking feature is the low energy requirement for calcination in Althaus und Sprecher.
572 The data in Althaus, which is also used by Sprecher, is based on the production of phosphate
573 rock from phosphate ore (Althaus et al., 2007a), where after crushing and sieving, the
574 phosphate ore is calcined to remove impurities. Althaus indicated an energy requirement of
575 0.34 MJ/kg feed. This basic value is used for calcination of the RE oxalate given in Landbank
576 (1994). This energy requirement is significantly lower than that in other studies, for example,
577 20 and 30 times lower than in Marx and Zaines. Marx's data is based on ASPEN modeling
578 and Zaines' data is based on Chinese literature data. Another aspect that questions Althaus'
579 energy demand is that all other studies indicate a significantly lower energy requirement for
580 roasting than for calcination. This suggests an underestimated energy requirement for
581 calcination by Althaus and Sprecher.

582 The quantities of CO₂ emissions during calcination are different. Marx and Arshi considered
583 not only emissions from the decomposition of carbonates, but also the CO₂ emissions from the
584 combustion of natural gas. The other studies only considered direct CO₂ emissions caused by
585 decomposition of carbonates. Althaus and Sprecher used the ecoinvent process "heavy oil
586 burned in industrial furnace" and Lee and Bailey used "electricity, grid from northwest China"
587 to heat the furnace. The CO₂ emissions related to these processes are already included in the
588 ecoinvent processes and therefore not explicitly visible in Table 2 as direct process emissions.

589 Other emissions into air and water are based on EPA (Althaus, Sprecher) or MEP (Lee, Bailey).
590 The use of the MEP emission list sometimes leads to emissions that should not occur during
591 the production of REO from Bayan Obo. An example are high zinc emissions during SX in Lee
592 2017, which are also used by Bailey. However, an analysis of 8 samples of Bayan Obo ore
593 has shown less than 350 ppm zinc in 6 out of the 8 samples and less than 1500 ppm in the
594 other two samples (Drew et al., 1990). Sprecher used Althaus' emissions for SX, which also
595 contain emissions from roasting. This results in a double counting of SO₂, HF, and CO₂ for SX
596 in Sprecher because he calculated these emissions of the roasting process additionally.

Only Marx roughly estimated emissions from wastewater treatment that are discharged into the tailing pond and seep away (Qifan et al., 2010).

Table 2

Main inputs and outputs per kg REO for mining, beneficiation, roasting/hydrometallurgy and SX/calcination.

Table 3 summarizes key inventory data within the studies for better comparability.

Table 3

Comparison of RE compounds, chemicals, and energy considered in the process chain.

4.2.5. Discourse on radioactivity in Bayan Obo

Radioactivity is an environmental problem that arises during the extraction of REEs, but was not considered sufficiently in most LCA studies. However, the thorium activity in the Bayan Obo ore is particularly extensive. The thorium oxide (ThO_2) concentration is reported between 0.032 (Qifan et al., 2010) and 0.04% (International Atomic Energy Agency, 2011). The radioactivity of 1 g of ThO_2 is 3,566 Bq (International Atomic Energy Agency, 2011). Marx calculated a total Th^{232} activity of $1.3\text{E}13$ Bq of the mined ore assuming an annual mining rate of $1.0\text{E}07$ t ore and an average ThO_2 concentration of 0.036%. Radioactivity can be found along the entire process chain, mainly in tailings and leaching sludge (96 to 98%), in dust (0.1 to 0.5%), and in wastewater from wet process methods (0.6 to 2%) (Qifan et al., 2010). The tailings and the wastewater are discharged into the tailing pond. The leaching sludge is discharged into the Radioactive Storage Facility (RSF). Together, tailing pond and RSF include

more than 99% of the total radioactivity. Assuming operation in accordance with regulations, only a small fraction of radioactive substances should be dissolved in water and seeped away. Therefore, the radioactive emissions, which are caused by dissolving and infiltration, are low in comparison to the deposited radioactivity. An accidental release of radioactivity from the tailing pond and the RSF e.g. due to a dike breach is not subject of an LCA but of a risk analysis.

The radioactivity into air is quantified in different ways. Zaimes and Arshi, for example, made only general statements about radioactivity. Lee 2017 stated thorium emissions based on MEP values, which do not represent the Bayan Obo ore. Marx, Althaus, Sprecher, and Bailey estimated radioactive emissions into air based on dust emissions and Th232 concentration in the ore. Marx calculated 590 Bq/kg REO. This value fits well with assumptions by Qifan et al. (2010), when (1) 1.3×10^{13} Bq/year related to ore mining (calculated by Marx), (2) 0.5% of the total radioactivity emitted as dust (stated by Qifan et al. (2010) and (3) 100 kg ore/kg REO requirement (see Table 3, Marx), would amount to 600 Bq/kg REO.

The main reasons for the large radioactivity range between Althaus, Sprecher, and Bailey are incorrect interpretations of data sources or conversion errors (Table 4). This results in strongly overestimated or underestimated values of radioactivity in Sprecher's and Bailey's studies as well as in Althaus' study, respectively. Althaus used the dust quantity of an iron mine (20 g/kg iron concentrate) as a basis, but did not take into account the significantly higher ore input required for REO concentrate production (about 12 times higher) in contrast to iron concentrate production. Sprecher erroneously used Althaus' activity for beneficiation as an activity for mining (Table 4). Bailey, in turn, used Sprecher's radioactivity without verification. In addition, Sprecher assumed very low dust emissions of 1.3 kg dust/t REO based on an unusually low total dust quantity of 61.8 t for 46,000 t REO production in 2008 t (Schüler et al., 2011). Unfortunately, Sprecher incorrectly converted the amount of dust from kg REO to kg concentrate. He multiplied the dust quantity per kg REO by 0.61 (assuming 61% REO concentration in the concentrate) instead of dividing it by the quantity of concentrate per kg

649 REO (3.81 kg concentrate/kg REO). As a result, he obtained a higher dust amount/kg
650 concentrate (2.1 kg dust/kg concentrate) than per kg REO (1.3 kg dust/kg REO). The correct
651 value is 0.34 kg dust/kg concentrate.

652 In Table 4 we present the original assumptions and resultant radioactivity found in the studies
653 and, where possible, the corrected values. Looking at the original published radioactivity the
654 values vary between 80.5 and 5800 Bq/kg REO. After correction, these values converge
655 between 134 and 939 Bq/kg REO, which is still a wide range.

656

657 Table 4

658 Data on radioactive emissions into air.

659

660 Although 99% radioactivity is stored in the tailings and RSF some radioactivity is released into
661 water by leakage which is considered differently in the studies. Althaus and Marx considered
662 infiltration rates and solubility of solids. Althaus used effluent rates by EPA (Environmental
663 Protection Agency, 1998), from which he calculated the fraction of suspended solids and
664 assumed the same radioactivity for them as for dust during mining. Sprecher calculated an
665 activity of 1000 Bq/kg tailing on the basis of IAEA data (International Atomic Energy Agency,
666 2011). Furthermore, Sprecher assumed that only 1 kg of tailings are produced per kg RE
667 concentrate, even though 30 kg of ore per kg concentrate are needed. Most of the ore is
668 discharged into the tailing pond together with the used water and flotation chemicals, so that
669 considerably more than 1 kg of tailings/kg concentrate is produced. Qifan et al. (2010) reported
670 65.5 kg tailings/kg REO. In addition, Sprecher assumed that the total activity of the tailings is
671 emitted into water, although only a fraction is released into the environment as mentioned
672 above (Qifan et al., 2010). As a result, Sprecher calculated a higher radioactivity (3810 Bq/kg
673 REO in water), although he assumed a lower tailing amount than Marx (73 Bq/kg REO in
674 water). Bailey, in turn, used Sprecher's values without verification.

675

676 **4.2.6. Handling of data uncertainty in the Bayan Obo studies**

677 None of the studies performed a statistical procedure to determine data uncertainties in detail.
678 The most common reason is missing information about uncertainties of a particular input or
679 output derived from the only available source. This is often the case due to the sparse data
680 availability in the field of Chinese REO production. However, the studies are aware of the large
681 data uncertainties and deal with them differently. Only two Bayan Obo studies (Althaus, Marx)
682 directly addressed data uncertainties. Some studies used scenarios to represent the range of
683 data or the different industry standards (Sprecher, Lee 2017, Lee 2018). Two studies
684 performed sensitivity analyses (Bailey, Arshi) by performing input deviation of +/-10% for
685 selected inputs.

686 Althaus used the pedigree matrix approach to quantify the standard deviation. Data sources
687 are evaluated based on six characteristics: reliability, completeness, temporal correlation,
688 technological correlation, geographic correlation, further technological correlation. Each
689 characteristic is divided into quality levels with a score between 1 (best) and 5 (worst). From
690 this, the standard deviation for a lognormal distribution was calculated for each input and
691 output. However, it should be noted that no study that used Althaus' ecoinvent inventory data
692 applied these standard deviations for further LCIA assessment.

693 In order to evaluate the validity of results in our previous study (Marx), we accounted for data
694 uncertainty by assigning a data quality indicator to each single input of all processes based on
695 the classification system of the American Association of Cost Estimation applied for the Mining
696 and Processing Industry. From this, we calculated an average value for the data quality of each
697 individual process. These values were used in the calculation of the LCIA by Gabi software
698 and error bars were given for all impact categories. The worst data quality was described for
699 ET, HTP and EP. GWP, FD, PM and POCP achieved a higher data quality. In general, the
700 data quality for the Bayan Obo pathway is very poor and lies between -30% and +60%

deviation. Differences between various REO production pathways lay beyond the deviations assumed.

Lee 2017 and Sprecher compiled data for different scenarios to account for large discrepancies in production efficiency and waste treatment as described in chapter 3.1. The scenarios in Lee 2018 represent different future options instead of the range of specific data.

Zaimes used point estimates due to data limitations. The authors captured uncertainty about impacts by using Monte Carlo technique to randomly sample from the statistical distributions for environmental impact. It remains unclear which distribution function was used. The error bars present the 2.5th and 97.5th percentile for PE and GWP for the production of 1 kg REO (light, medium, heavy and neodymium oxide). They look very similar, because the same distribution function might have been used.

Arshi carried out sensitivity analyses with focus on global warming. Various inputs such as electricity, HCl, and citric acid were varied by +/- 10%. The sensitivity analyses are not related to REO but to NdFeB magnet. The results have shown that the variations of inputs do not result in a drastic change in environmental impacts. However, the assumed deviations of 10% are very small compared to the deviations of the values between the studies shown in this review.

Bailey performed a sensitivity analysis for 57 energy-related inputs. Additionally, a sensitivity analysis was performed for the SX, since it emerged as the most relevant process at Bailey's study. The +/- 10% variation was used for all inputs. The electricity consumption during SX causes differences up to 1.77% across the impact categories. Ammonia emissions are more sensitive with a change of +/- 7.75% for EP. The +/- 10% variation of HCl used for SX results in a 6% increase/decrease for ODP.

Table 5

Statistical parameters for assessing uncertainty in LCI data of Bayan Obo.

727

728 As a quick overview of which inputs have the largest uncertainties across the studies
729 considered, Table 5 shows some meaningful statistical measures. We only evaluated inputs
730 and outputs that were considered by at least six of the eight studies. In addition to Table 5,
731 Figure S1 shows the associated boxplots disclosing the outliers. Besides the common
732 standard deviation (SD) and coefficient of variation (CV), Table 5 also shows the quartile
733 coefficient of dispersion (QCD). Latter is less sensitive to outliers than the CV, which occur
734 frequently in the data sets considered. The QCD is highest for primary energy for mining, water
735 for roasting and especially kerosene and extraction agents for SX. This indicates input data
736 with a very large dispersion. In the case of kerosene, there are two very high values that are
737 close to each other. Therefore, the second highest value is included in the upper quartile.
738 However, since the median is very small due to all the other very low values and the QCD is
739 normalized using the median, the QCD becomes very high. In contrast, the outlier in blasting
740 agent for mining (Zaimes' study), which is about 1000 to 30,000 times larger than the other
741 values, does not have a large effect on the QCD. This indicates that the blasting agent inputs
742 of the other studies do not disperse very much. The same applies for other processes that
743 have one or two outliers in the inputs (e.g. PE and H₂SO₄ for roasting, inorganic flotation
744 agents). In general, the QCD is hardly affected when one outlier is large and another is small.
745 Overall, it can be concluded that the data quality for most inputs is acceptable, for some even
746 satisfactory. However, outliers cannot be ignored, as they are responsible for some erroneous
747 results in the LCIA (Chapter 4.3). Though, several outliers and errors in the inventory can
748 unintentionally compensate each other.

749 Looking at some outputs that are considered in an appropriate number of studies the QCD for
750 sulfur dioxide (roasting) is the largest, followed by hydrogen fluoride (roasting). This means
751 that the uncertainty is high for acidification and human toxicity, which are particularly caused
752 by these emissions. The data uncertainty for carbon dioxide (roasting) and particulates
753 (mining) is acceptable. The low QCD for CO₂ emissions (calcination) can be explained by the

fact that most studies calculate them stoichiometrically for the calcination of RE oxalates and RE carbonates. Thus, the resulting GWP is robust.

4.3 Life cycle impact assessment of REO production (Bayan Obo)

Goal of this chapter is to identify the most important differences and uncertainties of crucial parameters in the LCIs and to link those directly to the affected LCIA. However, comparability of LCIA results shown in Fig. 4 is limited due to different LCIA methods, as already mentioned in the chapter “methods”. All LCIA results are shown in Table S1 and S3 (supplementary material). It should also be mentioned that we calculated Althaus’ LCIA data, as he provided only the LCI in the ecoinvent database. For this, we rebuilt Althaus’ original process “rare earth oxide production from bastnasite” in the GaBi software and evaluated it with two LCIA methods for comparison (Table S1).

Using the mining process as an example, the influence of strongly different inventory data on the LCIA results will be discussed. Fig. 2 presents the environmental impacts of 1 kg of mined raw ore of the original studies by Sprecher, Zaimes, and Marx as well as revised inventories of Sprecher’s and Zaimes’ studies. The highest impacts for mining are caused by the exorbitant use of blasting agent in Zaimes’ study as well as by the overestimated energy demand in Sprecher’s study. To calculate the revised impacts we assumed 0.125 MJ energy demand/kg ore instead of 1.25 MJ in Sprecher’s LCI. Further, we reduced the amount of blasting agent from 1 kg/kg ore assumed in Zaimes’ LCI to a common value of 5E-04 kg/kg ore. Using both revised data, we created new LCIs for Sprecher and Zaimes (GaBi software, version 10, (thinkstep, 2019)) and evaluated them with ReCiPe 2016. Since the original impacts in Zaimes’ study are extremely high, a second scale had to be inserted in Fig. 2 (right side).

Fig. 2

Environmental impacts of 1 kg of mined raw ore. The right scale presents the original numbers in Zaimes' study (light blue striped bar).

The results shown in Fig. 2 demonstrate that the impacts become more comparable after the LCIs have been adjusted. For example, GWP and FD are reduced by 88%, POCP by 82% and AP by 78% after adjustment of Sprecher's LCI. In case of Zaimes, all impacts of 1 kg of mined ore are reduced by even more than 99%.

In the following, the influence of the LCI data on the overall LCIA results is discussed. The 2000 times higher use of blasting agent for mining in Zaimes' study compared to other studies is clearly reflected in the results of the overall process chain also. Mining contributes to more than 50% in seven of 10 impact categories and even more than 90% in two categories. This is in contradiction to the results of the other studies in which the chemical requirements for the hydrometallurgical processes determine the results. In Marx's and Arshi's studies, mining only contributes significantly to those impact categories that take dust into account. For example, mining contributes to nearly 40% for PM in Marx's study. Dust emissions for mining and beneficiation range from 0.032 (Althaus) to 0.57 kg/kg REO (Marx), which results in a 15 times higher PM in Marx' study compared to Althaus.

The sum of chemicals used along the overall process chain varies between 7.5 (Arshi) and 50.6 kg (Zaimes) per kg REO (Table 3). Zaimes stated that 86% of total chemical consumption is needed for the upstream processes of oxalic acid production (43.35 kg NaOH/kg REO, source: supporting information of Zaimes: Table S21 'LCI for roasting'), which seems very high. When looking at the GWP, however, this high amount of NaOH is not reflected. The GWP would have to be approx. 7 times higher when using 43.35 kg NaOH (54 kg CO₂ eq/kg REO instead of 8 kg CO₂ eq/kg REO stated in the study). Taking the 8 kg CO₂ eq/kg REO, Zaimes can only have calculated the impacts with a maximum of 1.2 kg NaOH. Reason might be a mix-up since identical figures are given for both the NaOH requirement (43.35 kg/kg REO) and

the energy requirement for calcination (43.35 MJ/kg REO) are given. It is very likely that the given value for precipitation chemicals is not correct (Table 3, Zaimes) and the sum of chemicals should be about 7 kg/kg REO instead of the reported 50.6 kg.

As the mass fraction of flotation chemicals is small (0.6 to 5.8%) compared to the total chemicals requirement, the differences described in chapter 4.2.2 have only a marginal effect on the LCIA results. HCl, H₂SO₄, and the precipitation chemicals have the highest impacts. The largest variations are found for the HCl demand, which varies between 0.97 (Zaimes) and 8.95 kg (Marx) per kg REO. As the production of 1 kg HCl causes a GWP of 0.852 kg CO₂ eq, a difference of 6.8 kg CO₂ eq. between the highest and lowest HCl demand should be expected. However, this difference is not reflected in the LCIA of Marx's and Zaimes' studies. In a further example, Althaus shows 2/3 the GWP of Marx's (Table S1), although Marx's HCl demand is five times higher and additionally the energy demand is eight times higher (Table 3) so the GWP difference should be much more pronounced. One reason for the low difference is the high demand for extraction agent in Althaus' study (see 4.2.4), which is up to 200 times higher than in other studies. Additionally, the assumptions that the extraction solution (3 kg/kg REO) is incinerated after use and that the sulfate waste from roasting and precipitation is deposited (using the Ecoinvent process "disposal, sulfidic tailings, off-site (GLO)") result in a GWP of 7.8 kg/kg REO, which accounts for 49% of the total GWP. Without these incineration and disposal processes, the GWP would be 5.1 kg/kg REO. This example shows again that a single overestimated input (extraction agent requirement and its subsequent treatment) can strongly influence the overall result.

In addition to the inputs, emissions show the greatest variations in the LCIs. The emissions differ both in quantity and in composition. Often they do not reflect the real composition of waste or tailings. Especially the HF emissions into air during roasting show large differences depending on different exhaust gas scrubbing efficiencies and data sources. Variations in emissions generated by SX (e.g. inorganic and organic chemicals), however, do not affect the results at all due to missing characterization factors in the LCIA method. Arshi has already

addressed this problem. Therefore, Arshi did not indicate any other emissions apart from ammonia.

Sprecher and Bailey calculated high HTP and ET impacts. The disposal of excess H_2SO_4 as sulfidic tailings instead of sulfatic waste and especially the high HF emissions caused by low efficiency of exhaust gas scrubbing during roasting are the reasons in Sprecher's and therefore also in Bailey's study. Additionally, Bailey assumed high zinc emissions during SX, which is also responsible for the high HTP and ET. This assumption is astonishing because the concentration of zinc in the Bayan Obo ore is small (Drew et al., 1990; Castor and Hedrick, 2006) and heavy metals contained in the ore are almost completely separated before SX. In addition, Bailey made a conversion error (factor 1000) when assessing the impacts of the zinc emissions. Specifying emissions of 300 mg Zn/kg REO and using the characterization factor for $\text{ET}_{\text{freshwater}}$ of 38,000 $\text{CTU}_e/\text{kg Zn}$ (ILCD PEF v1.09) would result in approx. 11 instead of 11,700 $\text{CTU}_h/\text{kg REO}$ as stated by Bailey (Table S1). The same applies for $\text{HTP}_{\text{non-cancer}}$. The correct value should be $3.9\text{E-}07$ instead of $3.9\text{E-}04$ $\text{CTU}_h/\text{kg REO}$. This results in a highly overestimated ET and $\text{HTP}_{\text{non-cancer}}$, dominated to 99% by SX and zinc emissions as the main contributor. This result clearly contradicts those in all other studies.

The impacts (Fig. 4, Table S1 and S3) can only be compared to a limited extent, due to different LCIA methods. Only GWP and ODP are equally considered in all studies. The high GWP in Lee 2017 (Table S1) and Lee 2018 (Table S3) is caused by high energy consumption for roasting and separation as well as the high HCl demand. AP and EP are also highest for Lee. This is most probably due to the amount of chemicals used and the energy requirements. Both are in the upper range compared to other studies. Lee 2017 also stated high SO_2 emissions during roasting which contribute to AP.

Regarding IR, all studies except Marx's did not consider Th232 emissions caused by dust during mining and beneficiation in the LCIA's, because no characterization factor for Th232 are implemented in the models used. In order to integrate the radioactive load, Marx converted the Th232 emissions into U238. This procedure was described in detail in the supporting

information in Marx. Upstream processes of energy and chemical supply, especially the nuclear energy for electricity generation cause the IR reported by Althaus, Sprecher, and Bailey.

5. Review of the IAC studies

5.1 Process chain (IAC)

For IAC extraction, different types of leaching methods are applied. Because of the severe environmental consequences of heap and tank/pool leaching, in-situ leaching is the dominating technology nowadays. Therefore, all selected studies focused on in-situ leaching. For the preparation of the deposit, injection wells are drilled into the ground and a pipeline system is built. A leaching solution using ammonia sulfate ($(\text{NH}_4)_2\text{SO}_4$) as leaching agent is pumped through the wells into the clay layer. The RE ions on the clay minerals surface are exchanged with ammonium (NH_4^+) ions and diffuse in the solution, forming the RE leachate. The NH_4^+ ions used are replaced by addition of $(\text{NH}_4)_2\text{SO}_4$ before the leaching solution is recirculated into the clay deposit. The process is repeated until the REE content becomes too low.

The IACs include some elements co-existing with REE such as aluminum, calcium, iron, and magnesium. The separation of these accompanying elements is achieved by adjusting the pH value. The REEs are recovered from the leachate by precipitation, predominantly with NH_4HCO_3 as precipitation agent to obtain RE carbonates. After precipitation, mixed RE carbonates are transported to a central extraction plant. In case of Vahidi 2016, Arshi, Schulze, and Bailey it is assumed that RE mixed oxides are formed by an additional calcination step. After precipitation HCl leaching is carried out with both the mixed oxides and the mixed carbonates. The separation into individual RE oxalates and RE carbonates is carried out by SX. Finally, the oxides are produced by calcination. A simplified illustration of the process chain is given in Fig. 3.

Fig. 3

Schematic representation of IAC production in the Southern Provinces.

5.2 Life Cycle Inventory of REO production (IAC)

5.2.1 In-situ leaching

The environmental performance of in-situ leaching is significantly influenced by the quantity of $(\text{NH}_4)_2\text{SO}_4$ used as leaching agent in the leaching solution. This quantity depends on the $(\text{NH}_4)_2\text{SO}_4$ concentration in the leaching solution, the RE recovery efficiency as well as a potential recycling rate (*Table 6*). Usually, the $(\text{NH}_4)_2\text{SO}_4$ concentration varies between 1 to 5%, Vahidi 2016 indicating 1 to 2% and Yang et al. (2013) 3 to 5%. RE recovery efficiency is between 40 to 85%, with an industry average of 70%, indicated by Lee 2017. Moldoveanu and Papangelakis (2013) even achieved 75 to 90% under laboratory conditions. Reuse of $(\text{NH}_4)_2\text{SO}_4$ is only considered in Schulze's study. Additionally, the in-situ leaching is influenced by specific deposit conditions such as REE concentration in the IACs as well as hydrological and mineralogical properties (Chi and Tian, 2008). In the scientific literature, the REE concentration of the IAC deposits in the Chinese southern provinces ranges from 0.03 to 0.15% (Yang et al., 2013), over 0.05 to 0.2% (Vahidi 2016) up to 0.05 to 0.5% (Moldoveanu and Papangelakis, 2013).

To cover uncertainties of these most relevant parameters, all studies, except Arshi and Bailey, analyzed several scenarios (*Table 6*). Though, the diverse assumptions and inputs vary depending on the data sources used. In almost all studies, the data are based on Chinese scientific literature (Vahidi 2016, Deng, Lee 2017, Zapp), Chinese plant reports (Vahidi 2016, Deng), or personal information by Chinese experts (Lee 2017). Deng, Lee 2017, Vahidi 2016, and Zapp presented their own calculated data based on the literature. Deng used data from four Chinese industrial plants. The lowest value of each of the four plants was used for the low scenario and the highest value for the high scenario. No mean values were calculated. Schulze

also presented a scenario with 69% reuse of the leaching solution at other mining sites. It should be noted that Vahidi's 2016 data refer to 1 kg of mixed REOs without the SX procedure. All other studies refer to 1 kg of separated individual REOs including the SX (Table 6).

The quantity of $(\text{NH}_4)_2\text{SO}_4$ varies between 2 (Schulze, low, with reuse) and 80 kg (Zapp, high) per kg REO. As no detailed information was given on the calculation in almost all studies, the amount of $(\text{NH}_4)_2\text{SO}_4$ cannot be verified in most cases. Only Lee 2017 and Zapp provided information on how the quantities were calculated for the different scenarios (Table 6). Lee 2017 calculated the $(\text{NH}_4)_2\text{SO}_4$ quantities of his scenarios starting from a Chinese study that indicates 6.7 kg $(\text{NH}_4)_2\text{SO}_4$ /kg REO when the recovery efficiency is 88%. Zapp's low scenario represents a nearly stoichiometric $(\text{NH}_4)_2\text{SO}_4$ quantity (9.9 kg/REO) as also stated in Vahidi's 2016 high estimate (10.4 kg/kg REO). The low estimates of Vahidi 2016, Schulze, and Deng show even lower $(\text{NH}_4)_2\text{SO}_4$ quantities. However, according to Moldoveanu and Papangelakis (2013), a stoichiometric excess of at least 6.6 times is necessary to achieve a RE recovery rate of 80%. In addition, the high $(\text{NH}_4)_2\text{SO}_4$ loading of 3500 to 4000 mg/l in the groundwater is also a strong evidence for a high stoichiometric excess during in-situ leaching (Yang et al., 2013), contradicting the assumptions made for the scenarios with near stoichiometric $(\text{NH}_4)_2\text{SO}_4$ quantities.

The electricity consumption varies between 0.5 (Vahidi 2016, low) and 7 kWh/kg REO (Lee 2018) due to different data sources. In Zapp, the electricity demand of 0.55 kWh/kg REO was based on an in-situ leaching process for copper (Martens et al., 2003). The other studies used Chinese references or data from Chinese plants. The energy demand of Vahidi's 2016 high scenario is 10 times higher than that of the low scenario, likely due to the altitude of the mining zone and thus the energy required pumping water. Lee 2018 reported the highest electricity demand although he stated that he used the average electricity value of in-situ leaching by Zapp, Vahidi 2016, Lee 2017, and Schulze. Reason is that Lee 2018 included Schulze's energy requirement for an additional calcination step resulting in a highly overestimated energy demand for Schulze's in-situ leaching (14.5 instead of 6 kWh/kg REO). In addition to local

conditions, the large difference could also be due to an additional energy requirement for mechanical pressing, which is probably taken into account in the studies with higher energy requirements. For example, mechanical pressing as preparation for calcination is included in Vahidi's 2016 scheme of the system boundary. Since the energy data were taken from secondary sources, no further information on the calculation was available.

All studies analyzed the preliminaries for the IAC deposits in different ways. As the preliminaries only make a small contribution to the overall result, we did not compare the different options.

The emissions released during in-situ leaching result from ion exchange of REE by NH_4^+ , from leakages and mainly from the disposal of the leaching solution. Zapp assumed for all scenarios that the leaching solution remains in the soil and seeps away after the deposit is closed. The other studies did not provide any information about the handling of the leaching solution. However, the NH_4^+ and SO_4^{2-} emissions stated in the studies clearly exceed the amounts, which are induced by ion exchange and leakage, of approx. 0.6 to 1.1 kg/kg REO (Vahidi 2016). Furthermore, the high $(\text{NH}_4)_2\text{SO}_4$ concentration in groundwater of 3500 to 4000 mg/l indicated by Yang et al. (2013) shows that the solution probably remains in the soil or is released on site as wastewater. In the low scenarios of Vahidi 2016 and Lee 2017, a wastewater treatment was considered, which reduces emissions by more than two and three times compared to the high scenarios. The lowest emissions were reported in the low scenario with reuse of leaching solution in Schulzes' study. In this scenario, the leaching solution is pumped out, transported, and reused at another site. The sum of NH_4^+ and SO_4^{2-} emissions in the high scenarios vary between 14.1 (Arshi) and 79.6 kg (Zapp) per kg REO. Reason is the different $(\text{NH}_4)_2\text{SO}_4$ concentration of the leaching solution. Deng only reported emissions from the MEP list, which, however, contain neither NH_4^+ nor SO_4^{2-} . In Arshi and Vahidi 2016 (high scenario), the sum of NH_4^+ and SO_4^{2-} emissions into water and soil is up to 1.9 kg/kg REO higher than the input of $(\text{NH}_4)_2\text{SO}_4$. Reason is that H_2SO_4 used for pH adjustment causes additional SO_4^{2-} emissions. The NH_4^+ emissions in Schulze's study cannot be comprehended

since on the one hand the NH_4^+ emissions into water of the low scenario with reuse are higher than for the low scenario without reuse and on the other hand the NH_4^+ input (from $(\text{NH}_4)_2\text{SO}_4$) is lower than the NH_4^+ output.

5.2.2 Precipitation

The precipitation of REEs from the leaching solution can be done using oxalic acid as well as NH_4HCO_3 . Although precipitation using oxalic acid results in a higher product quality, on an industrial scale NH_4HCO_3 is mainly used for cost and environmental reasons (Vahidi 2016, Deng). For this reason, five of eight studies evaluated NH_4HCO_3 precipitation. The NH_4HCO_3 quantities vary between 1.02 (Zapp) and 6.0 kg (Deng, high scenario) per kg REO. The number by Zapp is based on a fourfold stoichiometric excess, but contains a calculation error in the reaction equation by factor three. The correct NH_4HCO_3 quantity for Zapp should be 3.06 kg/kg REO, matching the other values given in Table 6. Schulze calculated own data for NH_4HCO_3 quantity. Bailey used an average number of Schulze's values. In Chi and Tian (2008), a quantity of NH_4HCO_3 of 2.5 to 3.0 times the REO weight was discussed to form crystalline RE carbonate. According to this, about 3 kg of NH_4HCO_3 would be required, which correspond to the NH_4HCO_3 consumption in Lee 2017, Schulze, Bailey, and Zapp (corrected number). Vahidi 2016, Arshi, and Deng considered a mix of both precipitation options. As the share of precipitation with oxalic acid and NH_4HCO_3 is not specified, the quantities of precipitating agent cannot be determined. Thus, a comparison of the NH_4HCO_3 demand with the other studies was not possible.

The removal of accompanying elements such as Al, Mg, and Si from the leaching solution is achieved by increasing the pH value and was only considered in Zapp. To increase the pH value from five to 10, 0.56 kg lime/kg REO needs to be added. The significantly higher lime quantity indicated by Lee 2017 in the middle and low scenarios is not used for removal of accompanying elements, but for purification of wastewater. The subsequent addition of H_2SO_4 after precipitation was considered in all studies either for pH adjustment or in case of Lee 2017

to produce $(\text{NH}_4)_2\text{SO}_4$ from ammonia leftover after precipitation. The H_2SO_4 amount varies between 0.2 (Zapp, Deng, low scenario) und 1.7 kg (Lee 2017) per kg REO. In Zapp's study, the H_2SO_4 was calculated based on the assumption that the pH value needs to be reduced from 10 to five. In Vahidi 2016 and Deng, the H_2SO_4 quantities are based on Chinese scientific literature and facility data.

Emissions resulting from purification were only considered in Zapp, Schulze, and Lee 2017. In Zapp, they are based on the accompanying elements in the leaching solution mentioned by Chi and Tian (2008) and considered as emissions into water according to their solubility. The amounts of these metals dissolved in water are small, ranging from 0.0003 to 0.03 g/kg REO. Lee 2017 reported emissions based on MEP. However, the emissions listed there do not reflect the composition of the IACs. Schulze only stated an emission of 3.4 to 5.65 g Al/kg REO, assuming that 5% of the Al ions contained in the IAC are emitted.

The ThO_2 content in the IACs is about 0.005% (International Atomic Energy Agency, 2011) resulting in a Th^{232} radioactivity below 0.2 Bq/g. In contrast to open pit mining at Bayan Obo, where the entire radioactivity is contained in the mined ore, the radioactivity remains in the deposit after in-situ leaching. Therefore, the radioactive emissions were not considered in the inventories of the IAC studies.

Table 6

Main inputs and outputs per kg REO for in-situ leaching and RE carbonate precipitation for the low (l), middle (m) and high (h) scenarios.

5.2.3 Calcination to mixed REOs

Arshi, Bailey, Schulze, and Vahidi 2016 included an additional calcination step forming mixed REOs after $(\text{NH}_4)\text{HCO}_3$ precipitation and before SX (green colored box in Fig. 3). The primary

energy demand varies between 0.31 (Vahidi 2016, low) and 68.16 MJ (Arshi) per kg REO. Vahidi's energy demand is very low for an assumed calcination temperature of 750 - 800°C. Since no further information was available, Vahidi's low primary energy could not be comprehended. Since Bailey's energy value of 0.41 MJ/kg REO is based on Vahidi's values, it is also assumed that the energy requirement is too low. Schulze's energy value is based on discussions with industry experts and is about 100 times higher than those of Vahidi 2016 and Bailey. Arshi's energy value is about twice that of Schulze. Arshi used the same process for the calcination of mixed REOs (Fig. 3) as for the calcination of individual REOs after SX. In both processes, RE carbonates are converted to REOs. Schulze also uses the same energy requirement for both processes. The energy requirement in Arshi and Schulze is comparable to that for calcination at the Bayan Obo pathway (Table 2). CO₂ emissions range roughly from 0.5 (Vahidi, low) to 3 kg CO₂/kg REO (Arshi), with the latter additionally accounting CO₂ emissions from natural gas combustion.

5.2.4 Solvent extraction and calcination

SX using phosphoric acid based extraction agents like P507 and P204 as well as calcination are applied for IACs, analogous to that in Bayan Obo. Thus, these issues are not discussed again. In general, the production of RE chlorides from IACs up to SX shows lower environmental impacts in comparison with those of the Bayan Obo route. Therefore, the share of SX in the whole REE production is significantly higher for the IAC route than that for the Bayan Obo pathway (Vahidi and Zhao, 2017). Similar to Bayan Obo, bulk chemicals such as HCl, NaOH, and oxalic acid are the main cause of environmental impacts due to the considerable amount of materials and energy inputs required for their production. The largest differences between the SX in the Bayan Obo and IAC routes are based on different amounts of chemicals needed to separate the individual REEs. This in turn is due to the different concentrations of REEs in bastnäsite/monazite (Bayan Obo) and IACs, as well as the share of separated individual light and heavy REEs.

As mentioned before, Vahidi 2016 and Deng did not considered SX and the subsequent calcination. Lee 2017 used the same SX and calcination processes for IAC as for Bayan Obo. Schulze used data for SX from a Chinese literature source and for the extractant values from (Vahidi and Zhao, 2016). Arshi used the values for SX from Vahidi and Zhao (2017). Although Arshi said that after precipitation the individual RE precipitates (RE carbonates or RE oxalates) must be roasted to pure REO, the corresponding LCI data are lacking. He probably forgot to include the final calcination step to individual REOs because he already considered an additional calcination to mixed REOs before SX (see chapter 5.2.3). Thus, Arshi's GWP value is too low. Bailey used the SX model from Schulze, but surprisingly the values from Vahidi and Zhao (2017). Zapp adapted the SX scheme for Bayan Obo to the IAC composition. LCI data of SX and calcination (gray colored box in Fig. 3) are not considered in the LCI table (Table 6).

5.2.5 Handling of data uncertainty in the IAC studies

As in the BO studies, no statistical procedure was performed to determine data uncertainties in the IAC studies. To cover uncertainties of the most relevant inputs, all studies, except Arshi and Bailey, analyzed several scenarios described in chapter 5.2.1. Schulze did not specify data uncertainties in her study.

Arshi and Bailey carried out sensitivity analyses for several inputs. Bailey tested the influence of various inputs such as oxalic acid, NaOH and extraction agent P204. The +/- 10% variation of oxalic acid results in a difference up to 1.85% across the impact categories. The NaOH variation results in a 3% increase/decrease of ODP. The extraction agent is not very sensitive for all impact categories. Arshi varied inputs such as electricity, $(\text{NH}_4)_2\text{SO}_4$ and citric acid by +/- 10%. The +/- 10% variation of the inputs results in a 3.2%, 2.3%, and 1.7% increase/decrease of GWP for electricity, $(\text{NH}_4)_2\text{SO}_4$ and citric acid, respectively, related to 1 kg NdFeB magnet.

In our previous IAC study (Zapp), we chose the same approach to assess data quality as in our Bayan Obo study (Marx), as described in Chapter 4.2.6.

Deng performed a sensitivity analysis to address data uncertainty regarding oxalic acid, $(\text{NH}_4)_2\text{SO}_4$, and NH_4HCO_3 by a +/-10% variation. A 10% decrease in $(\text{NH}_4)_2\text{SO}_4$ decreases the impacts HTP, AP, GWP, and ODP by about 5 – 8%. A 10% decrease in use of oxalic acid leads to more than a 6% decrease in ET. EP and AP are highly sensitive to NH_4HCO_3 use. To examine the potential effect of non-compliance, a sensitivity analysis was conducted assuming that on-site emission standards were exceeded by a factor of five (500%). The most sensitive impact categories were respiratory effects (+169%) and EP (+44%). Deng stated that the impact categories are generally more sensitive to the quantity of chemical inputs, than on-site pollution. Unfortunately, Deng did not consider NH_4^+ emissions.

Table 7

Statistical parameters for assessing uncertainty in LCI data of IACs.

Table 7 shows the statistical parameters used to assess data quality for IACs. The QCDs for the inputs of the IAC process chain are lower than those for the inputs of the Bayan Obo process chain. This indicates a lower dispersion of the inputs between the studies. The reason for this is that, in the case of the IACs, the authors of the studies have already reduced uncertainties by creating scenarios for the main inputs ($(\text{NH}_4)_2\text{SO}_4$, NH_4HCO_3). Data in Table 7 are based on the input values of the "high" scenarios whenever possible, otherwise on non-specified scenario. For the "middle" and "low" scenarios as well as for the outputs not enough data are available to conduct meaningful statistical evaluations.

5.3 Life cycle impact assessment of REO production (IAC)

1094 All LCIA results for the whole process chain, including SX and calcination, are shown in Table
1095 S2 and S4 (Lee 2018).

1096 The use of $(\text{NH}_4)_2\text{SO}_4$ during in-situ leaching has the most significant influence on the overall
1097 results. Zapp's scenarios confirmed this statement, where only the $(\text{NH}_4)_2\text{SO}_4$ demand is
1098 varied. The increase in $(\text{NH}_4)_2\text{SO}_4$ demand from approx. 9 (Zapp, low scenario) to 81 kg/kg
1099 REO (Zapp, high scenario) results in a significant increase of all impact categories, with the
1100 highest increase for EP (factor nine) (*Fig. 4*, Table S2). The impact category $\text{EP}_{\text{marine}}$ is
1101 significantly affected by NH_4^+ emissions, while the increase in other impact categories is due
1102 to the production of $(\text{NH}_4)_2\text{SO}_4$ (upstream process).

1103 The different choice of LCIA methods used in the studies makes it not only difficult to compare
1104 the results, but also leads to strange effects. For example, Arshi stated the highest AP in
1105 contrast to all other studies, although he used the input data from Vahidi 2016. The reason is
1106 that NH_4^+ emissions into water contributing to AP are considered only in TRACI 2.1 and not in
1107 TRACI 2.0 used by Vahidi 2016. Furthermore, Zapp erroneously marked the NH_4^+ emission as
1108 "ammonium (total N)" instead of "ammonium, ion". However, "ammonium (total N)" does not
1109 contribute to AP. This is the reason why Zapp showed low AP.

1110 The EP value in Bailey is much lower than in the other studies. Bailey used Schulze's NH_4^+
1111 value, but marked them erroneously as "aluminum emissions into freshwater". Since Al
1112 emissions have no impact on EP, a contribution of NH_4^+ to EP is missing in Bailey's study.

1113 In order to compare Zapp's results with those by Vahidi 2016, we performed an additional LCIA
1114 for 1 kg mixed REO without SX based on Zapp's data (Table S2, Zapp TRACI 2.1). Although
1115 the $(\text{NH}_4)_2\text{SO}_4$ requirement of 10.4 kg in Vahidi 2016 is significantly lower than that of Zapp
1116 with 80.6 kg/kg REO (high scenario), there is little difference in impacts. As the upstream
1117 production of $(\text{NH}_4)_2\text{SO}_4$ has a clear influence on the result, the differences in impacts should
1118 be higher. Vahidi 2016 and Zapp used the same ecoinvent process "ammonium sulfate, as N"
1119 for the production of $(\text{NH}_4)_2\text{SO}_4$. This process refers only to the N content in the $(\text{NH}_4)_2\text{SO}_4$,
1120 which amounts to 21.2%. Therefore, when using the ecoinvent process "ammonium sulfate,

as N", the intended quantity of $(\text{NH}_4)_2\text{SO}_4$ must be reduced by 78.8%. While Zapp proceeded correctly, Vahidi 2016 erroneously used the total amount of $(\text{NH}_4)_2\text{SO}_4$ instead of 21.2% of 10.4 kg. This results in an inadvertently higher input of $(\text{NH}_4)_2\text{SO}_4$ of 49 instead of 10.4 kg/kg REO used by Vahidi 2016 for in-situ leaching modeling. In addition, Vahidi 2016 assumed oxalic acid instead of $(\text{NH}_4)\text{HCO}_3$ for precipitation, which has higher impacts during upstream production. Furthermore, Vahidi's 2016 energy demand was higher. These three facts caused that most impacts of Vahidi 2016 and Zapp were hardly different, although the $(\text{NH}_4)_2\text{SO}_4$ input varied strongly.

Only, Zapp's $\text{EP}_{\text{marine}}$ is about 6 times higher than Vahidi's 2016 due to the higher $(\text{NH}_4)_2\text{SO}_4$ amount. In addition to the already mentioned incorrect designation of NH_4^+ emissions ("ammonium (total N)" instead of "ammonium, ion"), the NH_4^+ emissions were not scaled to nitrogen as required by the use of "ammonium (total N)". Therefore, the $\text{EP}_{\text{marine}}$ is 22% too high in each scenario in Zapp's study (Table S2 shows the wrong and corrected $\text{EP}_{\text{marine}}$ values). Deng, Schulze, Lee 2017, and Lee 2018 did not publish $\text{EP}_{\text{marine}}$ because there are no $\text{EP}_{\text{marine}}$ characterization factors for NH_4^+ emissions in the LCIA methods used (e.g. CML 2001, IMPACT2002+). The low $\text{EP}_{\text{freshwater}}$ in Dengs' study can be explained by missing NH_4^+ emissions. Deng only published NH_3 emissions, which also do not contribute to $\text{EP}_{\text{freshwater}}$ in IMPACT2000+.

Deng's results refer to mixed REOs without SX, but are not comparable due to the mix of LCIA methods used (Table S2). Deng used processes from CLCD. Vahidi 2016 and Zapp used ecoinvent processes. Remarkable is the low ET (factor 1.0E-05), which cannot be explained by the inputs, because they are similar to Vahidi's. Deng stated that oxalic acid contributes largely to ET, and its contributions are quite significant in other categories as well. Deng used a surrogate for oxalic acid based on Zaimes assuming that oxalic acid is produced as the reaction product of coke, H_2SO_4 , and, NaOH. As mentioned above (chapter 4.2.4) the calculation of the amount of oxalic acid, however, cannot be comprehended due to missing process description.

1148

1149 **6. Results and discussion**

1150 The review identified an overall of 35 LCA studies that assess the environmental impact of
1151 REE production. Of those, 25 studies provided information about REO production from Bayan
1152 Obo and/or from IAC in the Southern Provinces of China. The quality of the inventory data is
1153 one of the most important factors for reliable results. Therefore, the limited amount of original
1154 LCI data underlying the reviewed studies is noteworthy. We identified only 12 studies that
1155 provided sufficient information to evaluate the environmental impacts of REO production. Of
1156 those, six studies relied exclusively on own primary LCIs (Althaus, Deng, Lee 2017, Marx,
1157 Zaimes, Zapp), while the other six reused these LCIs either entirely (Bailey, Lee 2018) or
1158 partially, amending them with own original data (Sprecher, Schulze, Vahidi 2016, Arshi). Thus,
1159 the amount of original LCI data is weak, with only a few publications providing the LCIs for
1160 other studies. Still, the variation is very high, concerning different key assumptions regarding
1161 RE raw ore concentration and composition, yields of beneficiation as well as hydro- and
1162 pyrometallurgical processes, exhaust gas cleaning efficiencies, and waste treatment.
1163 Sometimes it remains unclear why some inputs and outputs are particularly high or low. After
1164 the extensive and detailed review of all studies, we most recommend the studies of Lee 2017,
1165 Arshi and Marx for the use of LCI for REO production in Bayan Obo. Lee 2017 had the
1166 opportunity to use Chinese company data and interviewed Chinese RE experts. On the output
1167 side, however, the Lee 2017 study is not convincing because only MEP data were used. This
1168 is where Marx's study scores. In Marx's study, particular effort was made to model
1169 environmental exposures from tailings and sludge including radioactivity from wastewater
1170 leakage. In Arshi's study, a transparent and user editable Excel model was created, which is
1171 particularly commendable. For IACs, we recommend the studies by Vahidi 2016, Lee 2017
1172 and Zapp despite all the shortcomings described. Vahidi's 2016 advantage is that he used data
1173 from Chinese literature verified through personal interviews of Chinese experts in the RE

1174 industry. Like Lee 2017, the Vahidi's 2016 study is less convincing for outputs than for inputs
1175 because only MEP data were used.

1176 An adequate LCI should be based largely on original industry data and should reflect the state-
1177 of-the-art of the different production facilities in relation to the total annual REO production.
1178 Until such a LCI for REO production is established, all existing results should be viewed with
1179 caution. In general it can be said that in the LCI most of the input data are much better known
1180 than outputs. If no measured data are available, material quantities can be derived
1181 stoichiometrically and energy demands can be calculated. Despite the unclear verification of
1182 the different studies it can be stated, that uncertainty in beneficiation (e.g. flotation) has a
1183 smaller effect than those in SX or leaching. This is mainly due to the large quantities of
1184 chemicals (4.9 (Arshi) to 14.8 kg/kg REO (Marx)) used for SX and leaching, whose upstream
1185 production is associated with many environmental impacts. In the case of outputs, there is
1186 often a lack of data on the efficiency of exhaust gas cleaning, wastewater treatment, and waste
1187 disposal. Radioactive sludge and tailings stored in open storage facilities are hardly
1188 considered. Only one study roughly estimated the emissions caused by infiltration. Others
1189 used emissions from the MEP or EPA lists, but these are not specific to Bayan Obo or IACs.
1190 Damage caused by dam bursts, such as in Hungary 2010 (red mud dump) or in Brazil 2019
1191 (mine effluent from an iron ore mine) are accidents and are per se not considered in an LCA.
1192 However, incidents associated with the production of REOs can result in the release of large
1193 quantities of openly deposited radioactive sludge or sludge contaminated with heavy metals,
1194 leading to significant environmental impacts. Therefore, a combination with an additional risk
1195 analysis would widen the knowledge of truly potential environmental impacts caused by REO
1196 production.

1197 Illegal facilities, which account for more than 40% of Chinese RE production from IACs and do
1198 not comply with any environmental protection standards (Packey and Kingsnorth, 2016), were
1199 not considered in the studies except for Lee 2018. Therefore, the results of Lee 2018 are
1200 presented in two additional tables (Table S2 and S4) since the results of the illegal REO

1201 production are not comparable to the other studies. In general, the results paint a too optimistic
1202 picture. The high $(\text{NH}_4)_2\text{SO}_4$ demand and NH_4^+ emissions reported in Zapp's middle and high
1203 scenario for IACs are not based on facility data but were calculated on the basis of literature
1204 (Chi and Tian, 2008; Yang et al., 2013). Whether this data comes close to that of illegal mines
1205 remains to be seen. Decision maker have to keep in mind, that today's overall RE production
1206 in China is by all means worse than the results presented by the studies (except for the results
1207 obtained for illegal production in Lee 2018), due to the high share of illegal activities (in Bayan
1208 Obo and Southern Provinces). On the other hand, Chinese authorities have started to push
1209 illegal mining and metal refining back.

1210 Next major findings for the Bayan Obo and IAC routes are discussed separately before a
1211 comparison between the two routes will be made where possible.

1212 Looking only at the total environmental impacts of REO production at Bayan Obo (Fig. 4, Table
1213 S1), the studies do not suggest major differences, as many values are quite comparable.
1214 However, if the underlying LCIs are also taken into account (Table 2, Table 3), a completely
1215 different picture emerges. Reason is an unintentionally compensation of effects by various
1216 differences. The following three examples will illustrate this. The low HCl and H_2SO_4 demand
1217 and the very low energy demand for calcination in Althaus' study are not reflected in the LCIA,
1218 because they are compensated by the high demand for extraction agents. The significantly
1219 lower need for chemicals in Zaimes' study compared to the other studies is not visible in the
1220 LCIA, since the assumed need for blasting agent counterbalances the results. The high GWP
1221 for mining in Sprecher is also not visible in the overall result. Reasons are the low HCl demand
1222 compared to the other studies which otherwise requires a lot of electricity for production and
1223 the low energy demand for calcination.

1224 Although the RE extraction procedure from IACs is described almost identically in the studies,
1225 there are clear differences in LCIs (Table 6) and consequently in LCIA (Table S2). The largest
1226 influence on the LCIA results has the use of $(\text{NH}_4)_2\text{SO}_4$ due to its significantly higher demand
1227 compared to other chemicals. The amount of $(\text{NH}_4)_2\text{SO}_4$ shows the highest variations between

the studies. Since the $(\text{NH}_4)_2\text{SO}_4$ amount depends on several factors, the figures are hard to follow. No representative data of the actual RE extraction from Chinese IACs are available. Data is largely based on environmental reports and on a few individual Chinese facilities and thus represents the $(\text{NH}_4)_2\text{SO}_4$ range of only legal RE production sites. Lee 2018 also presents only worst-case estimates for illegal production, but no site-specific data. Because almost no studies give information on the production capacities of the individual low, middle, and high IAC scenarios, no realistic picture of the IAC mining industry can be given. Only Lee 2018 estimated the environmental impacts of even illegal REO production in the future using scenarios. Deng's data, while based on four Chinese facilities, do not reflect the overall situation of RE extraction from IACs, as he also did not consider illegal production. Therefore, Deng's claim to provide a state-of-the-art LCI of REO production by IACs for the LCA community cannot be fulfilled completely. Lee 2017 quoted an industry survey without citing the source. Perhaps he referred to an expert survey that is supposed to reflect the Chinese industry average of REO production. Again, Lee 2017 did not give information on how many tons of REO are produced by old and new plants.

Beyond the much discussed LCI, the review showed that also the incorrect use of flow names during modeling in the LCA software (see discussion about NH_4^+ emissions ("ammonium (total N)" instead of "ammonium, ion"), aluminum emissions ("aluminum emissions into freshwater" instead of "ammonium, ion"), and the incorrect use of theecoinvent process "ammonium sulfate, as N") can have a large influence on the environmental impacts.

This review showed that AP, EP, HTP, ET, and GWP are the mostly relevant environmental impacts for REO production. For the IAC route, $(\text{NH}_4)_2\text{SO}_4$ emissions to water during in-situ leaching contribute mostly to AP and EP. The upstream impacts of producing $(\text{NH}_4)_2\text{SO}_4$ and NH_4HCO_3 have also significant environmental burdens in many impact categories. SX uses several chemicals and therefore accounts for a high share of many impacts especially for the Bayan Obo route.

Considering the Bayan Obo studies, we found a GWP of 14 to 47 kg CO₂ eq. and an AP of approx. 0.07 – 0.7 kg SO₂ eq. ODP varies between approx. 3E-06 and 7E-06 kg CFC-11 eq. for all studies except for Lee 2017, who specifies 9.5E-08 kg CFC-11 eq. For HTP we found values ranging from 9 – 140 kg 1,4-DCB eq. and for EP_{freshwater} between 0.004 – 0.09 kg P eq. Due to Bailey's error regarding zinc emissions, the range for ET is particularly wide, going from 30, over 70 and 90 up to 11,700 CTU_e (Fig. 4, Table S1). Looking at the LCIA results for illegal REO production at Bayan Obo estimated by Lee 2018 (Table S3), the impacts increase by a factor of 1.5 to 2.5. For AP and EP, the increase rises to almost a factor of 6 due to the high SO₂ emissions during roasting, which also increase by a factor of 6.

Assessing the IAC studies related to individual REOs we found a GWP of approx. 25 to 67 kg CO₂ eq. and an AP of approx. 0.2 – 5 kg SO₂ eq. HTP varies between 1.7 and 57 kg 1,4-DCB eq. For EP_{marine} we found values ranging from 0.5 – 22 N eq. Again, the range for ET is particularly wide, going from 57, up to 428 CTU_e (Fig. 4, Table S2). Considering the IAC studies related to mixed REOs without SX we found lower impacts in most cases. For example, GWP ranges only between 9 and 38 kg CO₂ eq. and AP varies between 0.05 – 0.35 kg SO₂ eq. However, the range for ET is even wider, from 0.003 to 450 CTU_e. Looking also at the LCIA results for illegal REO production based on IACs estimated by Lee 2018 (Table S4), all impacts increase by a factor of 1.5 to 2.5.

Fig. 4

Environmental impacts of 1 kg REO produced at Bayan Obo deposit and from IACs obtained by the selected studies subdivided by different LCIA methods used.

Not all studies analyzed both REO production pathways, making it difficult to compare the Bayan Obo and IAC route. They can be compared well in the case of Marx's, Lee's 2017, Bailey's, and Arshi's studies because these four studies examined both pathways. REO

production based on IACs, compared to that based on the Bayan Obo deposit, performed worst in all impact categories in Arshi's study, in six of seven in Marx's/Zapp's studies, in four of eight in Lee's 2017 study, and in three of seven impact categories in Bailey's study. Among these, all studies see higher impacts for IAC in GWP, ODP, and EP. For AP, three studies except Bailey's see disadvantages for IAC. The higher AP for Bayan Obo that Bailey found can be explained by the high SO₂ emissions caused during roasting that she used from Sprecher's study. The POCP values are quite similar for both REO pathways. ET_{freshwater} is higher for Bayan Obo in Lee's 2017 and Bailey's study in contrast to Marx's due to the assumed zinc emissions during SX. However, these emissions are questionable, as already discussed in detail. The same applies for HTP. In the light of the discussion regarding switching between data from Lee 2017 and Sprecher without consideration of different assumptions (Bayan Obo), the striking zinc emissions (Bayan Obo) as well as aluminum instead of NH₄⁺ emissions (IAC), the results of Bailey's study, which was intended to evaluate the state-of-the-art LCI for REO production, should be carefully reviewed before further use.

7. Conclusions

There is still a matter of controversy concerning the environmental impacts of REO production. For various reasons, there is a wide spread in results from different LCAs. Some of which are addressed in this review. Besides the age of two studies, however, the large spread in results can be seen in the more recent studies reviewed in this paper.

None of the studies was flawless and considered all necessary inputs and outputs adequately. Some studies used different scenarios to compensate for missing data, while others derived data from analog processes or surrogate materials. Some errors have a large effect on all impact categories, while others have a minimal effect on one or two impact categories. Multiple errors made in a study can outweigh each other and subsequently make the environmental impacts appear to fit. Therefore, transparency is an essential prerequisite. Own approaches

should be compared with those of others. In the case of striking results it must be checked exactly, which inventory data caused these impacts. When results contradicted the results of all other studies, it often remained open why it was not scrutinized. Sometimes it should be argued rather cautiously to avoid wrong conclusions. For the users of the studies it is important to know all assumptions in detail to be able to assess the results. Therefore, the entire inventory should be made available and properly documented.

Based on the considered studies, this review wanted to clarify whether a conclusive evaluation of the environmental performance of RE production at Bayan Obo and by IACs can actually be conducted. We conclude that this has not been fully achieved yet. Although some studies considered different scenarios that reflect the different state-of-the-art, it is not possible to derive an overall assessment of the environmental impact of REO production in China. On the one hand, this would require determining what share of total REO production the scenarios account for. On the other hand, the environmental impact of illegal mining would also have to be included in the overall assessment. Only Lee 2018 considered the latter through worst-case estimates. A comparison with other studies is not possible.

To strengthen the reliability of LCA and to help decision makers to evaluate environmental consequences associated with the increasing use of REEs transparency of data and assumptions is eminent. Since the majority of REEs are produced in China, this poses a special challenge in terms of data availability. Many studies in recent years show the great interest in this topic and have already highlighted problems in REO production. Nevertheless, there is not yet a complete picture of the environmental impacts associated with REO production.

In order to increase transparency, more detailed and comprehensible descriptions of the inventory data are required. On the other hand, the provision of inventory data on the level of individual processes is helpful to build up cumulative knowledge about REO production. Despite the considerable variability in results and the limited current data availability that have been discussed, however, some of the existing LCAs provide useful insights into REO

production and a good understanding of their environmental impacts. Thus, this review succeeds in reducing confusion and uncertainty regarding variability of the results.

Declaration of competing interest

We acknowledge that the submission declaration of "Science of the total environment" journal has been compiled with. We also confirm that all necessary permissions have been obtained. The authors declare that there is no conflict of interest regarding the publication of this article.

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