Rare Earths from Mines to Metals

Comparing Environmental Impacts from China's Main Production Pathways

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Keywords:

cleaner production
environmental impact
life cycle assessment (LCA)
metallurgy
mining
systems analysis

Supporting information is linked to this article on the *JIE* website

Summary

Over the past decade, China has supplied over 90% of global rare earths, and in doing so bore significant environmental burdens from processing its complex ores. In this study, we used life cycle assessment to quantify environmental impacts for producing 1 kilogram (kg) of 15 rare earth elements from each major production pathway. The scope of assessment included the largest rare earth oxide (REO) production chain in Bayan Obo, as well as lesser known production chains for bastnäsite in Sichuan and in-situ leaching of kaolin clays in the Seven Southern Provinces of China. This was followed by assessing impacts from the three major metal refining processes: molten salt electrolysis, calciothermic reduction, and lanthanothermic reduction. Among 11 impact categories assessed, results were highest for human toxicity that ranged between 13.1 and 50.4 kg 1,4-dichlorobenzene-eq (equivalent)/kg of rare earth metal^{-1}, followed by eutrophication (0.04 to 1.26 kg phosphate-eq/kg of rare earth metal^{-1}), abiotic depletion potential of fossil fuels (592 to 1,857 megajoules per kg of rare earth metal^{-1}), acidification (0.25 to 0.87 kg sulfur dioxide-eq/kg of rare earth metal^{-1}), and global warming (39.1 to 109.6 kg carbon dioxide-eq/kg of rare earth metal^{-1}) potentials. Regionally, impacts in Sichuan were lower across all key impact categories than in Bayan Obo: 32% lower for human toxicity, 67% lower for eutrophication, 58% lower for acidification, and 45% lower for global warming. A scenario analysis between the industry average and best available technologies revealed considerable potential to mitigate impacts across all production chains, particularly by improving waste treatment practices.

Introduction

While often, and sometimes erroneously, grouped together as one material, the rare earths technically consist of 17 different elements, 15 of which are mined for their economic value. They are prized for their unique properties in industries from ceramics (Goonan 2011) to clean energy (Alonso et al. 2012). Unfortunately, its mining and refining also generates considerable environmental harms, especially in China, where more than 90% of global rare earths have been produced over the past decade (USGS 2015). The Chinese government had set restrictions on production and exports in order to curb environmental impacts (Chinese Information Office of the State Council 2012). Their efforts were short lived, however, because a recent World Trade Organization (WTO) (WTO 2014) ruling found those restrictions to be unlawful. This has forced China to more directly, and perhaps more effectively (Wei 2014), mitigate environmental harms through sustainable development of the industry.

Rare earth oxides (REOs) in China are primarily mined in three regions—Bayan Obo in Inner Mongolia, Sichuan, and the Seven Southern Provinces (Southern Provinces). Each

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© 2016 by Yale University
DOI: 10.1111/jiec.12491

Editor managing review: Alexis Laurent

Volume 21, Number 5

region contains different ores with varying compositions of rare earth elements. Whereas Bayan Obo and Sichuan are abundant in light rare earths, the Southern Provinces have a higher concentration of more-valuable heavy rare earths (see table S1-2 in supporting information S1 available on the Journal's website). Unique processes are used to mine, separate, and refine REOs into metals that differ depending on both the geological REO deposit, as well as the desired metal being refined.

Altogether, the production process is fairly energy and materials intensive (Peiro and Mendez 2013; Zaimes et al. 2015) and generates significant quantities of pollution that includes radioactive waste, hydrogen fluoride (HF), and acidic waste water (Hurst 2010; Chinese Ministry of Environmental Protection 2009). So far, life cycle studies on rare earths production in China have all focused solely on the production chain in Bayan Obo (Schüler et al. 2011; Tharumarajah and Koltun 2011; US EPA 2012; Schmidt 2013; Ali 2014; Sprecher et al. 2014; Zaimes et al. 2015; ecoinvent Center 2013). To our knowledge, no studies have been published on REO production in Sichuan or the Southern Provinces. These production chains are significant for two reasons. First, they make up a large portion of the rare earths industry in China: 41% of production quantity in 2013 (Chinese National Development and Reform Commission 2014) and 51% of economic value (Argus Media 2015). It would thus be logical to infer that they also contribute significantly to environmental impacts and would need to be included for any success in developing the industry sustainably. Second, the Bayan Obo production chain only produces light rare earths (lanthanum, cerium [Ce], neodymium, and praseodymium) in significant quantities. Previous life cycle studies are thus applicable for only 4 of the 15 rare earth elements.

In this study, we assessed environmental impacts from producing 1 kilogram (kg) of each of the 15 rare earth metals in China. Our study builds upon previous research in two main ways. The first is the breadth of its scope, expanded from one production chain in Bayan Obo to the three REO production chains and subsequent metal refining processes. Second is the accuracy of our assessment, which draws on a wide range of Chinese and international sources to paint the most complete picture of environmental impacts from the rare earths industry.

Methodology

Scope and System Boundaries

We used life cycle assessment (LCA) to calculate environmental impacts for each of three main rare earth production pathways in China. The goal was to contrast impacts from each pathway using 2013 as the baseline year. Each production chain consists of different processes tailored to the composition of raw ores found in the region. REO production can be broken down generally into the four processes shown in figure 1 from *mining* to *separation*. This is followed by *metal refining*, where we assumed a metal grade of 99.8% for molten salt electrolysis and 99.99% for both metallothermic reduction processes.

Altogether, our study consisted of 15 different functional units corresponding to the different rare earth metals. In this article, we present and discuss LCA results for three elements to represent each of the *metal refining* processes. Results for the remaining metals can be found in sheet S2-7 in supporting information S2 on the Web. In this study, we chose to focus on virgin production. Though recycling would have added another interesting dimension to the study, primary data for those processes were extremely scarce. Further, it only constitutes a small portion of the global supply (1.5%) and is unlikely to change in the near future (Binnemans et al. 2013). Mine exploration upstream of and final goods downstream of rare earth production were also not included.

Data Collection

The life cycle inventory (LCI) was compiled between 2012 and 2014. Primary data were collected for foreground production processes from Chinese and international scientific literature, industry reports, and government surveys, followed by an industry survey to fill any data gaps and harmonize values to the 2013 benchmark. Because of the complexity of rare earths processing along with proprietary barriers in data availability, the process was repeated several times until all data gaps were accounted for.

Data for pollutant discharges were more difficult to obtain because of lack of environmental reporting in China. As in the study by Zaimes and colleagues (2015), we conservatively assumed a baseline for aqueous and gaseous emissions using national discharge standards for the industry (Chinese Ministry of Environmental Protection 2011). We then accounted for exceedances of specific pollutants that are well documented, notably for ammonia (NH_3), chemical oxygen demand, fluoride, HF, sulfuric acid (H_2SO_4), sulfur dioxide (SO_2), and thorium (Chinese Ministry of Environmental Protection 2009; Xu et al. 2008; Yan et al. 2010; Zhou et al. 2012; Zhu et al. 2013). Industry surveys further confirmed that these environmental issues were still prevalent in 2013.

To obtain the most accurate representation of China's industries, background processes were modeled using LCA data from the Chinese Life Cycle Database (CLCD) (IKE and Sichuan University 2012). A small number of processes were substituted from the ecoinvent 3.0 database (ecoinvent Center 2013) and European Life Cycle Database (JRC—European Platform on Life Cycle Assessment 2006), where Chinese data were unavailable.

We compiled data for three scenarios to account for large discrepancies in production efficiency and waste treatment. The upper bound models small- to medium-sized enterprises scattered across China, usually with low production capacity and minimal to no waste treatment. At the other extreme, large, high-capacity enterprises are able to adopt latest technologies for production and waste treatment to meet environmental standards (lower bound). The third scenario was developed to capture the industry average, which models the national mean for both production efficiency and waste treatment.

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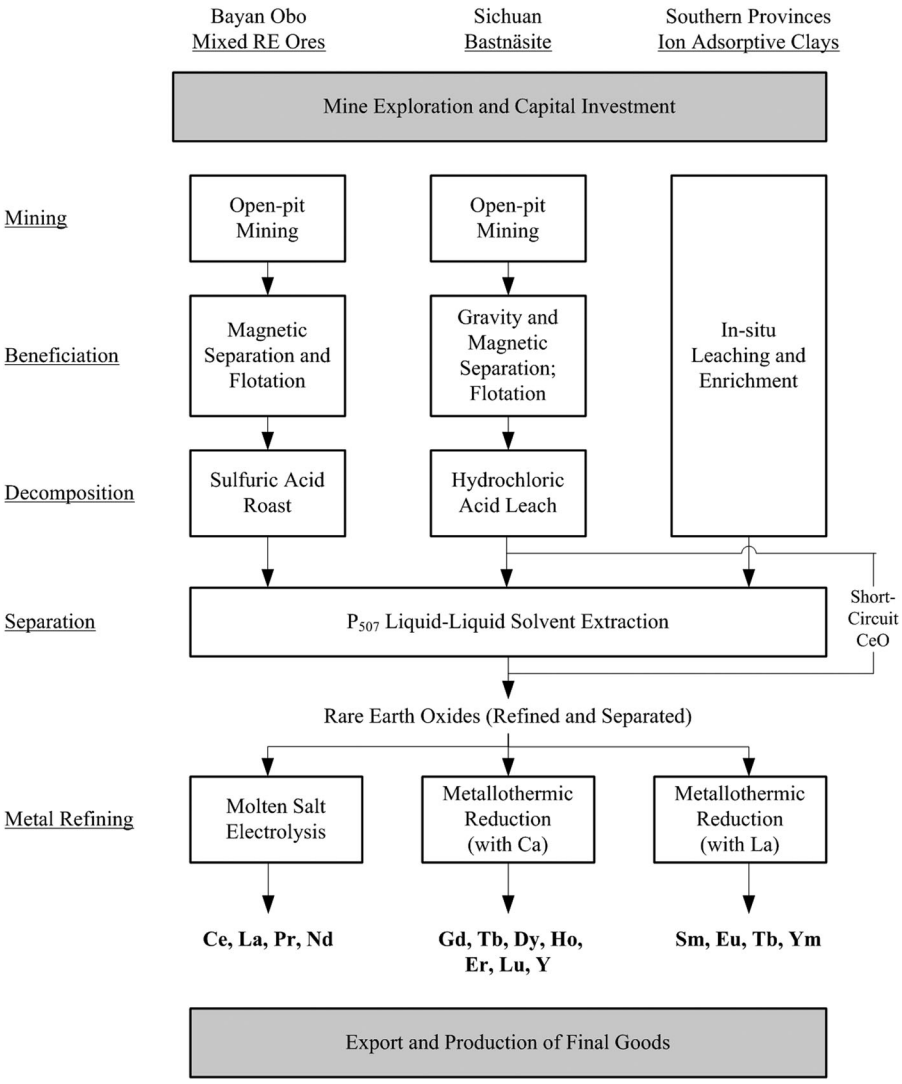

Figure 1 System boundaries for the cradle-to-gate environmental life cycle assessment of rare earths production in China. Each production chain employs a different mining and refining process and produces end products with different compositions of rare earths. Processes shaded in gray are not included within system boundaries. Ca = calcium; Ce = cerium; CeO = cerium oxide; Dy = dysprosium; Er = erbium; Eu = europium; Gd = gadolinium; Ho = holmium; La = lanthanum; Lu = lutetium; Nd = neodymium; Pr = praseodymium; RE = rare earth; Sm = samarium; Tb = terbium; Y = yttrium; Ym = ytterbium.

Life Cycle Impacts Assessment

The LCA model was constructed using eBalance Professional 4.7 (IT & Knowledge for Environment, Sichuan, China). We chose to construct an attributional model because it aligned more with the aims of our research, although we adopted a contributional approach to the scenarios sensitivity analysis. Life cycle environmental impacts were assessed using the CML2002 method (CML 2015). Eleven mid-point impact categories were selected based on suggested LCA practice (Hauschild et al. 2013) and previously identified environmental harms in the industry.

Because of limitations in data collection, primary data were not available to adequately assess the categories *respiratory inorganics* and *land use* and have not been included. Despite several studies highlighting radiation exposure from rare earth mining (Wang 2006; US EPA 2012), CML2002 characterization factors for the category do not include thorium, which is the primary pollutant for radiation exposure from rare earth mining. *Ionizing radiation* is thus also not included in this study. In lieu of these limitations, we provide a breakdown of estimated thorium and uranium emissions from each REO production chain in tables S1-3 to S1-5 in supporting information S1 on the Web. *Abiotic resource depletion of elements*, though included, may be underestimated because of rare earth elements not being taken into account as a characterization factor in the assessment method.

Results for the mid-point categories were then normalized to per capita global impacts for each category during the baseline year of 2000 (as person-years). We note that although there have likely been changes to per capita global impacts between

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2000 and 2013, most updated data were only available for the year 2000 for each of the 11 impact categories, and using this reference still gives some indication of the relative significance of impacts.

The simultaneous production of different REOs (co-mining) posed an allocation problem in our model. We opted for mass allocation of environmental impacts to each REO that is most congruent with the scope of this research. Economic allocation, on the other hand, would skew results with a market element, which would be undesirable given our aim to understand the environmental implications strictly from the production processes alone. To assist in future research, in particular, for economic allocation necessary for LCA studies of downstream products, separate results are provided in tables S1-6 and S1-7 in supporting information S1 on the Web for both REO production and metal production, respectively.

Life Cycle Inventory

The LCI is first described individually for the three REO production chains, followed by a section on metal refining. Aggregated modeling data are also included for the industry average REO production in tabular form (Tables 1, 2, and 3). A more in-depth description of the processes, LCI, and modeling data used for the entire study (including metal refining) is available in section 1 in supporting information S1 on the Web.

In particular, electricity production was modeled using background processes found in the CLCD for each region in China. Energy production in Bayan Obo was modeled using the process for northwest China, whereas for Sichuan and the Southern Provinces, we used energy production in southern China. Given that metal production is not regionally sensitive, we used the background process for average electricity production across the whole country. Throughout this section, we use industry-standard nomenclature of REO equivalents to describe the grade of rare earths in its various forms (rare earth chlorides, carbonates, oxides, etc.) before it becomes a metal.

Bayan Obo

Ore Excavation

Raw ores in Bayan Obo consist of mixed REO-bearing bastnäsite and monazite that are excavated by open-pit mining. We modeled the process using background data from the CLCD for *iron ore, open-pit mining* (IKE and Sichuan University 2012), modified to account for the composition of minerals. Ores are mined for both its rare earths (5% to 7%) and iron (28% to 45%) content (Ding et al. 2013; Xu and Che 2007). Using economic allocation, 81.5% of environmental impacts from excavation were allocated to rare earths.

Beneficiation

Ores are then treated by beneficiation to physically separate REOs from iron and other gangue material. Peiro and Mendez (2013) estimate that 60 megajoules (MJ) of energy are required to grind 1 tonne (t) of raw ore. We conservatively adopted an 80% efficiency factor and allocated impacts as in the mining process above. Production of hydroxyamic acid collectors H_{205} and H_{102} used for flotation could not be found in any life cycle database and were thus modeled using national production standards (Office of the Chinese Development Planning Commitee for Rare Earths 2000).

Decomposition: Sulfuric Acid Roast

Beneficiated oxides are further refined by REO decomposition. For our model, we adopted the H_2SO_4 roasting technique used for 90% of REO produced in the region (Zhao et al. 2013), followed by primary liquid-liquid extraction using D2EHPA (or P_{204}) (Xie et al. 2014; Hou 2005) to separate light and heavy rare earths. We used the unallocated ecoinvent process *heat production, at hard coal industrial furnace* (ecoinvent Center 2010) to model roasting, substituting Chinese data for upstream processes (IKE and Sichuan University 2012).

When roasted, fluoride-bearing gangue emits HF. In lieu of extremely high concentrations reported by the Chinese Ministry of Environmental Protection (2009), we calculated a value of 264 grams (g) of HF/kg REO^{-1} for the upper bound. For the average, we assumed 99% removal efficiency by spray towers (2.5 g of HF/kg REO^{-1}), and for the lower bound, the effluent meets national discharge standards (0.45 g of HF/kg REO^{-1}). The final product from H_2SO_4 roasting is a mixed rare earth chloride (95% REO). An ammonium chloride by-product is also produced, which we modeled as negative environmental impacts.

Rare Earth Oxide Separation

The last process in REO production is the separation of individual elements (still as REO). The description below for P_{507} liquid-liquid solvent extraction is also applicable for separation in Sichuan and the Southern Provinces, although a unique LCI was conducted in each to account for process nuances.

Separation consumes a large amount of ammonia for P_{507} saponification and hydrochloric acid (HCl) that hosts the reaction. For the lower bound, we adopted efficient practices based on fuzzy linkage extraction, which reduces ammonium and HCl consumption by up to 66% and 33%, respectively (Han 2013). Following separation, rare earths are precipitated with ammonium bicarbonate at high temperatures to produce a relatively pure REO (>99.5% REO) (Xie et al. 2014).

Ammonia in the effluent can reach concentrations of 5 g per liter $(L)^{-1}$ (Wang 2006; Yu et al. 2008), which we adopted for the upper bound. Fortunately, recent developments (Zhou et al. 2012; Zhu et al. 2013) have led to high recycling efficiencies (as ammonium chloride): 35% for the average and 75% for the lower-bound scenarios. We again modeled these as negative impacts. We assumed the remaining NH_3 is treated using crystallization evaporation to reach 750 milligrams (mg)/L^{-1} for the industry average.

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Table 1 Life cycle inventory for average rare earth oxide production in Bayan Obo

Unit process	*Unit*	*Industry average*	*Reference*
Inputs: technosphere to product system			
Fe-Nb-RE ores (6% REO), raw from open-pit mining	kg	33.93	Calculated
Sulfuric acid (modified), substituted for hydroxyamic acid collectors H_10_2 and H_20_5	kg	0.07	(Luo et al. 2002; Huang et al. 2012; Office of Chinese Development Planning Committee for Rare Earths 2000; Li and Zeng 2003)
Sodium silicate, ratio 3.4	kg	0.15	(Luo et al. 2002; Huang et al. 2012)
Ammonia, Chinese average	kg	1.22	(Luo et al. 2002; Huang et al. 2012; Yu et al. 2008; Xu et al. 2008; Chinese Ministry of Environmental Protection, 2009)
Sulfuric acid, Chinese average	kg	3.86	Industry survey
Phosphoric acid (modified), substituted for P204 and P507 extractant	kg	0.01	(Chinese Ministry of Environmental Protection 2009; Hou 2005; Sichuan Rare Earth 2007)
Kerosene, typical decompression	kg	0.04	(Sichuan Rare Earth 2007)
Hydrochloric acid (31%)	kg	8.46	(Han 2013; Chinese Ministry of Industry and Information Technology 2014; Chinese Ministry of Environmental Protection 2009)
Magnesium oxide, Chinese average	kg	0.55	Industry survey
Lime, Chinese average	kg	2.43	Calculated
Iron pellet production, substituted for iron flakes	kg	0.13	Industry survey
Ammonium bicarbonate, carbonization method	kg	1.60	Industry survey
Sodium carbonate, Chinese average	kg	1.05	Industry survey
Steam, 1.6 MPa at 204°C[a]	MJ	4.43	Industry survey
Charcoal production (modified), substituted for powdered activated carbon	kg	0.041	Industry survey
Electricity, grid from northwest China	kWh	6.99	(Peiro and Mendez 2013)
Coal, for heating	MJ	56.66	(Chinese Ministry of Environmental Protection 2009); industry survey
Water, desalinated	kg	306.21	(Chinese Ministry of Environmental Protection 2009, 2011)
Outputs: product system to technosphere			
Concentrated REO (100%), separated	kg	1	Functional unit
Ammonium chloride, Chinese average	kg	1.85	Calculated
Outputs: product system to environment (key parameters)			
Ammonia (water, unspecified)	kg	0.171	(Chinese Ministry of Environmental Protection 2009, 2011; Xu et al. 2008; Zhou et al. 2012; Zhu et al. 2013; Yan et al. 2010)
Thorium (water, unspecified)	kg	1.01×10^{-5}	(Chinese Ministry of Environmental Protection 2009)
Hydrogen fluoride (air, unspecified)	kg	0.003	(Chinese Ministry of Environmental Protection 2009)
Sulfur dioxide (air, unspecified)	kg	0.112	(Chinese Ministry of Environmental Protection 2009)
Sulfuric Acid (air, unspecified)	kg	0.017	(Chinese Ministry of Environmental Protection 2009)
Thorium (air, unspecified)	kg	7.6×10^{-5}	(Chinese Ministry of Environmental Protection 2009)
Carbon dioxide (air, unspecified)	kg	2.040	Calculated

Note: Fe-Nb-RE = iron-niobium-rhenium; REO = rare earth oxide; MPa = megapascals; kg = kilograms; MJ = megajoules; kWh = kilowatt-hours.

Sichuan

Mining and beneficiating raw ores in Sichuan are principally the same as in Bayan Obo. The major difference is that REOs are found in the region only as bastnäsite (as opposed to mixed ores in Bayan Obo). This makes it easier to beneficiate, significantly affecting the model in two ways, namely, a higher REO recovery efficiency (85%) and a higher concentration (70% REO) in the

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final product (Jordens et al. 2013; Li and Zeng 2003; Huang et al. 2012). REO separation is also the same in Sichuan as in other production chains. We therefore focus the remainder of this section on the unique decomposition process.

Decomposition: Hydrochloric Acid Leach

Beneficiated REOs are first calcined at temperatures up to 600ºC in order to oxidize carbonaceous material (Wang et al. 2013; Sichuan Rare Earth 2007). This is followed by initial HCl leaching, alkaline treatment, and second HCl leaching to produce a relatively pure rare earth chloride (95% REO).

Altogether, HCl leaching in Sichuan is able to completely separate and recover the majority of cerium oxide (CeO) in a short process. For our model, we assumed the entire quantity of Ce (50% cerium dioxide [CeO_2]/REO) is produced here as CeO_2 with a grade of 98% REO (Wang et al. 2013). Foreground carbon dioxide CO_2 emissions were calculated from chemical reactions of calcining beneficiated ores.

While the process produces significant quantities of HCl in waste water and gas, we assumed both to be neutralized before being discharged to the environment (Chinese Ministry of Environmental Protection 2009). We also assumed a small quantity of HF released to air attributed to reactions with fluoride gangue, conservatively assuming similar concentrations as in Bayan Obo (25 mg per meter $[m]^{-3}$) for the upper bound and average scenarios.

Southern Provinces

Rare earths are found naturally in kaolin clays across seven Southern Provinces that include Fujian, Guangdong, Guangxi, Hunan, Jiangxi, Yunnan, and Zhejiang. As in Bayan Obo and Sichuan, P_{507} liquid-liquid solvent extraction is also used to separate REOs and is thus not discussed here.

In-Situ Leaching

Concentration of REO in kaolin clays is extremely low (0.03% to 0.5%) (Yang et al. 2013; Ding and Deng 2013). Unlike in Sichuan and Bayan Obo, however, REOs are adsorbed to the surface of the clays, making them easier to extract. For our model, we adopted the in-situ leaching method of extraction, which has been promoted as the environmentally friendly option replacing traditional methods such as heap or pool leaching (Chinese Information Office of the State Council 2012; Yang et al. 2013).

One study reported that approximately 450 cubic meters per day^{-1} of leaching solution (Li 2011) containing between 2% and 5% ammonium sulfate are injected into the ground. The solution leaches rare earths and is collected through an underground collection system. Collection efficiencies vary considerably (Ding and Deng 2013; Zhu et al. 2011; Li et al. 2012); we assumed 50% and 85% for the upper- and lower-bound scenarios, respectively, with an industry average of 70%. Leaching solution not collected is essentially discharged directly to the soil environment. Of the quantity collected, fortunately a large portion can be recycled. We calculated ammonium sulfate consumption and recycling by applying results of a case study by Li (2011) for the different collection efficiencies. Because of the relatively high concentration of REO in the collected solution, it only needs to be precipitated once using ammonium bicarbonate to produce a high-grade mixed rare earth carbonate (92% REO).

A considerable amount of NH_3 is discharged throughout the process. By far, the main source is loss of ammonium sulfate attributed to low collection efficiencies (Table 2). For this model, we assumed ammonium sulfate in solution readily decomposes into H_2SO_4 and NH_3, the latter of which has been reported in much higher concentrations in surface and ground waters around mine sites (Yang et al. 2013). Waste water from ammonium bicarbonate precipitation also has a high concentration of NH_3; we assumed no treatment for the upper bound ($4{,}000\ mg/L^{-1}$) (Li et al. 2010) and NH_3 removal by stripping (Yan et al. 2010) for the average scenario, capable of reducing concentrations to $300\ mg/L^{-1}$.

Metal Refining

Unlike for REO production, rare earth metal refining is not unique to any region. Rather, the process is dependent on the rare earth element being refined and the desired grade. In this section, we provide a brief description of each process, whereas specific data and calculations can be found in section 1.6 in supporting information S1 and sheets S2-4 to S2-6 in supporting information S2 on the Web.

Molten salt electrolysis is most common for light rare earth metals lanthanum, Ce, praseodymium, and neodymium and accounts for 80% to 90% of rare earth metal production in China (Pang et al. 2011). In the process, REOs are electrolyzed in a fluoride salt medium to produce rare earth metals (99.8% purity) (Huang 2014). Process inputs are well documented in Chinese literature (Pang et al. 2011). Discharge of HF was found to be most environmentally significant, with an upper bound value of 45 g of HF/kg REO^{-1} based on the chemical conversion of rare earth fluoride (REF_3) expended in the process. For the average, we assumed treatment similar to that in Bayan Obo, yielding a specific discharge of 0.6 g of HF/kg REO^{-1}.

Metallothermic reduction produces the purest rare earth metals (99.99% assumed in this study) and is most common for heavy rare earths. Two different reduction processes are generally used. Calciothermic reduction using a calcium reductant is used for gadolinium (Gd), terbium, dysprosium, holmium, erbium, lutetium, and yttrium. In the process, rare earth oxides are first converted to REF_3 by direct fluorination (Liu and Li 2013). Based on industry surveys, we assumed 2 to 3 times molar excess of HF to drive the reaction, which also results in significant discharges of HF in waste gas. The REF_3 is then reduced with calcium metal under high temperature to produce rare earth metal. Based on literature (Gupta and Krishnamurthy 2005) and industry surveys, we assumed a 10% to 50% molar excess of calcium to drive the process. Because no background processes for calcium metal were available, we modeled calcium

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Table 2 Life cycle inventory for average rare earth oxide production in Sichuan province

Unit process	*Unit*	*Industry average*	*Reference*
Inputs: technosphere to product system			
Iron ore, open-pit mining substituted for bastnäsite-containing ores (5% REO)	kg	26.24	Calculated
Sulfuric acid (modified), substituted for hydroxyamic acid collectors H_10_2 and H_20_5	kg	0.047	(Office of the Chinese Development Planning Committee for Rare Earths 2000; Luo et al. 2002; Li and Zeng 2003; Huang et al. 2012)
Sodium silicate, ratio 3.4	kg	0.039	(Luo et al. 2002; Li and Zeng 2003; Huang et al. 2012)
Steam, 1.6 MPa at 204°C[b]	MJ	3.68	Industry Survey
Hydrochloric acid (31%)	kg	3.23	(Sichuan Rare Earth 2007; Wang et al. 2013; Han 2013)
Sodium hydroxide (30%), Chinese average	kg	1.75	(Sichuan Rare Earth 2007; Wang et al. 2013)
Calcium chloride, without water	kg	0.05	(Sichuan Rare Earth 2007; Wang et al. 2013)
Phosphoric acid (modified), substituted for P507 extractant	kg	0.002	(Sichuan Rare Earth 2007)
Ammonia, Chinese average	kg	0.32	(Sichuan Rare Earth 2007; Chinese Ministry of Industry and Information Technology 2014)
Kerosene, typical decompression	kg	0.01	(Sichuan Rare Earth 2007)
Ammonium bicarbonate, carbonization method	kg	0.89	(Sichuan Rare Earth 2007)
Sodium sulfide production[a]	kg	0.05	(Wang et al. 2013)
Sodium carbonate, Chinese average	kg	0.19	(Wang et al. 2013)
Charcoal production (modified), substituted for powdered activated carbon[a]	kg	0.02	Industry Survey
Electricity, grid from Southern China	kWh	478.77	(Sichuan Rare Earth 2007; Peiro and Mendez 2013)
Coal, for heating	MJ	82.88	(Sichuan Rare Earth 2007; Wang et al. 2013)
Water, desalinated	kg	229.80	(Sichuan Rare Earth 2007; Chinese Ministry of Environmental Protection 2009; Chinese Ministry of Environmental Protection 2011; Wang et al. 2013)
Outputs: product system to technosphere			
Concentrated REO (100%), separated	kg	1	Functional unit
Calcium fluoride	kg	0.032	(Sichuan Rare Earth 2007)
Ammonium chloride, Chinese average	kg	0.295	Calculated
Outputs: product system to environment (key parameters)			
Fluoride (water, unspecified)	kg	0.002	(Chinese Ministry of Environmental Protection 2009)
Ammonia (water, unspecified)	kg	0.029	(Chinese Ministry of Environmental Protection 2009; Zhou et al. 2012; Zhu et al. 2013; Yu et al. 2008)
Thorium (water, unspecified)	kg	7×10^{-6}	(Chinese Ministry of Environmental Protection 2009)
Hydrogen fluoride (air, unspecified)	kg	5.97×10^{-4}	(Chinese Ministry of Environmental Protection 2009)
Thorium (air, unspecified)	kg	0.079	(Chinese Ministry of Environmental Protection 2009)
Carbon dioxide (air, unspecified)	kg	1.283	Calculated

Note: Data for production inputs were confirmed with industry experts. Processes marked with [a] were modeled using background data from the ecoinvent Database and marked with [b] from the European Life Cycle Database. All other processes were modeled using data from the Chinese Life Cycle Database. REO = rare earth oxide; MPa = megapascals; kg = kilograms; MJ = megajoules; kWh = kilowatt-hours.

production using a similar reduction process involving an aluminum reductant.

For several rare earth metals that are more volatile, calciothermic reduction cannot be used. Instead, oxides of samarium (Sm), europium, thulium, and ytterbium are reduced with lanthanum metal at temperatures of 1,300 to 1,400°C under vacuum. The metals volatilize, are collected, and then condensed at temperatures of 300 to 400°C (Chinese Ministry of Environmental Protection 2009). Based on industry surveys, we calculated lanthanum consumption as a molar excess of 5% to 10%, produced by a molten salt electrolysis as described above. Lanthanum oxide is produced as a by-product that was recycled internally in our model.

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Table 3 Life cycle inventory for average rare earth oxide production in the Seven Southern Provinces (in-situ leaching)

Unit process	*Unit*	*Industry average*	*Reference*
Inputs: technosphere to product system			
Ammonium sulfate	kg	9.07	(Li 2011)
Sulfuric acid, Chinese average	kg	1.16	Calculated
Ammonium bicarbonate, carbonization method	kg	4.78	(Li 2011)
Lime, Chinese average	kg	3.37	Calculated
Hydrochloric acid (31%)	kg	10	Industry Survey
Phosphoric acid (modified), substituted for P507 extractant	kg	0.004	(Hou 2005)
Ammonia, Chinese average	kg	11	(Chinese Ministry of Industry and Information Technology 2014; Chinese Ministry of Environmental Protection 2009)
Kerosene, typical decompression	kg	0.01	Industry Survey
Sodium carbonate, Chinese average	kg	1.05	Industry Survey
Charcoal production, modified[a]		0.04	Industry Survey
Water, distilled	kg	893.80	(Li 2011; Chinese Ministry of Environmental Protection 2011)
Electricity, grid from southern China	kWh	12.36	(Li 2011)
Onshore well production, oil/gas[a]	m	0.00005	(Li 2011)
Outputs: product system to technosphere			
Concentrated REO (100%), separated	kg	1	Functional unit
Ammonium chloride, Chinese average	kg	1.08	Calculated
Outputs: product system to environment (key parameters)			
Ammonia (water, unspecified)	kg	0.219	(Chinese Ministry of Environmental Protection 2009, 2011; Zhou et al. 2012; Zhu et al. 2013; Yan et al. 2010)
Thorium (water, unspecified)	kg	3.2×10^{-7}	(Chinese Ministry of Environmental Protection 2009)
Sulfur dioxide (air, unspecified)	kg	0.029	(Chinese Ministry of Environmental Protection 2009)
Thorium (air, unspecified)	kg	3.2×10^{-4}	(Chinese Ministry of Environmental Protection 2009)
Carbon dioxide (air, unspecified)	kg	1.235	Calculated
Ammonium sulfate (soil, unspecified)	kg	7.50	Calculated

Note: Data for production inputs were confirmed with industry experts. Processes marked with [a] were modeled using background data from the ecoinvent Database. All other processes were modeled using data from the Chinese Life Cycle Database.
REO = rare earth oxide; kg = kilograms; kWh = kilowatt-hours; m = meters.

Both metallothermic reduction processes consume large quantities of electricity (12 to 15 kilowatt-hours [kWh]/kg of rare earth metal $[RE]^{-1}$). Data were scarce in Chinese literature; data collection thus consisted primarily of industry surveys.

Life Cycle Assessment Results

We calculated results for a total of 45 unique production pathways combining REO production with metals refining. Results were similar within each metal refining processes. For example, results for molten salt electrolysis of Ce and neodymium differed by a maximum of 1.18% for any given impact category. The one exception was for yttrium production, where impacts differed from other calcium-reduced metals by up to 41%. Rather than list results for all pathways, we present three representative elements that are most commonly produced under each metal refining process: Ce for molten salt electrolysis, Gd for calciothermic reduction, and Sm for lanthanothermic reduction.

Table 4 shows LCA results across the different metal refining and REO production processes. Disaggregated results for REO production can be found in table S5 in the supporting information on the Web. It should be noted here that in a strict LCA sense, raw results should not be compared directly given that functional units are not the same. Rather, we make a careful comparison of results in the discussion section.

Normalized Results

Normalized results (figure 2) show that among the 11 impact categories assessed, human toxicity potential was highest for production in Bayan Obo and Sichuan, whereas in the Southern Provinces, eutrophication was highest. This was followed by abiotic resource depletion of fossil fuels (fossil fuel depletion), acidification, and global warming potentials. The remaining six impact categories were negligible in comparison and are thus not shown in figure 2. Abiotic resource depletion of elements was underestimated in this study because of rare earths not

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being included as a characterization factor in the CML2002 assessment method.

Upper- and lower-bound scenarios are represented by error bars (figure 2), highlighting extreme cases especially in the adoption of waste treatment technologies. Upper-bound results for acidification (in Bayan Obo), eutrophication (in the Southern Provinces), and human toxicity (all regions) were much higher than the average, corresponding to direct discharges of H_2SO_4 and SO_2, NH_3, and HF, respectively, without any waste treatment. Upper-bound results for human toxicity all extend well past the limits of the graph. Its inclusion in graphical form would render the rest of figure 2 unreadable. Instead, data labels have been used to indicate their scope.

Contributions and Scenarios Sensitivity Analysis

A contributions analysis is shown graphically for Ce production using REO from each region (figure 3). The contributions profile is similar for Gd and Sm and thus not shown here. Because of the way data were collected during the LCI, results are aggregated for the *decomposition* and *separation* processes in Sichuan (under *separation*).

Results show that for production in Bayan Obo and Sichuan, impacts were primarily concentrated in three processes—*decomposition*, *separation*, and *metal refining*—where, not surprisingly, energy and materials consumption as well as waste pollutant discharges were highest. One explanation for the relatively lower environmental impacts in Sichuan (table 4) is that *mining* and *beneficiation* in the region can more effectively recover REO (beneficiated REO have a concentration of 70% in Sichuan as opposed to 50% in Bayan Obo), relieving some stress from the more impactful downstream *decomposition* and *separation* processes. For this reason, *mining*, *beneficiation*, and *metal refining* constitute a larger proportion of impacts in the region.

For production in the Southern Provinces, *mining* by in-situ leaching also contributes considerably to LCA results along with *separation* and *metal refining*. This is expected, however, given that in-situ leaching plays the role of *mining*, *beneficiation*, and *decomposition* to produce a high-grade mixed rare earth oxide. In in-situ leaching, impacts are primarily driven by consumption and subsequent discharge of ammonium sulfate. Across production in all three regions, *transportation* was negligible to overall impacts.

LCA results were sensitive to several parameters in the model, as evidenced by the difference between the industry average and lower-bound results; in particular, were emissions of key substances: HF, H_2SO_4, SO_2, and NH_3. Foreground emissions of HF contributed most to human toxicity potential, prevalent in *decomposition* (in Bayan Obo and Sichuan), as well as *metal refining*. An increase in removal efficiency during waste treatment from ~98% for the industry average to 99.6% in the lower bound (Chinese Ministry of Environmental Protection 2009) reduced human toxicity potential by 47% for cerium production in Bayan Obo, 31% in Sichuan, and 29% in the Southern Provinces. HF emissions from calciothermic reduction were found to be even more sensitive. A small

Table 4 Life cycle environmental impacts from cradle-to-gate production of select rare earth metals

		Ce, molten salt electrolysis			*Gd, calciothermic reduction*			*Sm, lanthanothermic reduction*		
Impact category	*Unit*	*BO*	*S*	*SP*	*BO*	*S*	*SP*	*BO*	*S*	*SP*
ADP, Elements	kg Sb-eq	1.4E−4	4.5E−5	3.5E−4	2.4E−4	1.4E−4	4.5E−4	1.8E−4	8.1E−5	3.9E−4
ADP, fossil fuels	MJ	1130	592	1294	1671	1112	1841	1588	1037	1757
Acidification	kg SO_2-eq	0.706	0.320	0.770	1.001	0.601	1.067	0.944	0.548	1.009
Eco-toxicity, freshwater	kg 1,4-DCB-eq	0.864	0.330	0.558	1.299	0.744	0.982	1.112	0.565	0.799
Eco-toxicity, terrestrial	kg 1,4-DCB-eq	0.017	0.025	0.006	0.018	0.027	0.007	0.022	0.030	0.011
Eutrophication	kg PO_4-eq	0.123	0.043	1.160	0.144	0.062	1.222	0.160	0.079	1.223
Global warming	kg CO_2-eq	76	41	82	111	75	117	106	70	112
Human toxicity	kg 1,4-DCB-eq	24	16	13	50	42	39	30	22	18
Ozone depletion	kg CFC-11-eq	1.9E−7	1.7E−7	2.2E−7	2.5E−7	2.3E−7	2.8E−7	2.6E−7	2.4E−7	2.8E−7
Photochemical oxidation	kg ethylene-eq.	0.020	0.009	0.018	0.027	0.016	0.025	0.027	0.016	0.025
Water use	kg H_2O	988	739	3162	1166	908	3422	1295	1041	3521

Note: Ce = cerium; Gd = gadolinium; Sm = samarium; BO = Bayan Obo; S = Sichuan; SP = Southern Provinces; ADP = abiotic depletion potential; kg = kilograms; Sb = antimony; MJ = megajoules; eq = equivalent; SO_2 = sulfur dioxide; PO_4 = phosphate; DCB = dichlorobenzene; CO_2 = carbon dioxide; CFC = chlorofluorocarbon.

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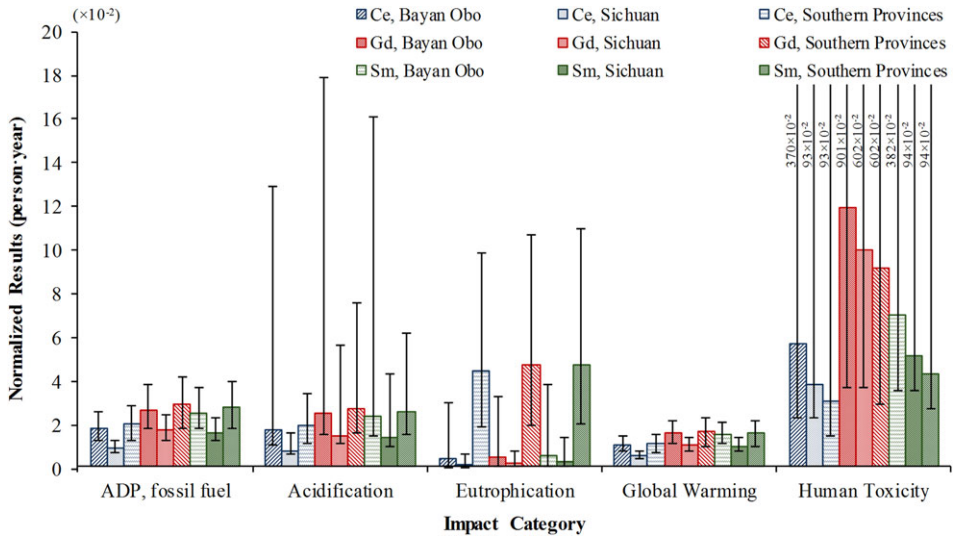

Figure 2 Normalized results to global impacts in the baseline year 2000 for each impact category. Lower- and upper-bound impacts are indicated with error bars. ADP = abiotic depletion potential; Ce = cerium; Gd = gadolinium; Sm = samarium.

(~1.6%) increase in HF removal efficiency for Gd production in the Southern provinces, for example, reduced human toxicity potential by 70%.

Similarly, higher removal efficiencies of H_2SO_4 from 99% to 99.7% and SO_2 from 80% to 90% can reduce acidification potential in Bayan Obo by 22%. For NH_3, best available technologies for waste treatment (Zhou et al. 2012; Zhu et al. 2013) reduced eutrophication potential in the industry average by 68% for Ce production in Bayan Obo, 33% in Sichuan, and 7% in the Southern Provinces, compared to the industry average.

Rather, eutrophication potential in the Southern Provinces is primarily sourced from ammonium sulfate lost directly to soil during the *mining* phase. Those losses alone contributed to 85% of eutrophication potential in the region. Results were thus very sensitive to collection of leaching solution. An increase from 70% in the average scenario to 85% in the lower bound decreased eutrophication potential by 46%. Given that collected solution can be recycled, it also translated into savings of 3.77 kg ammonium sulfate per kg of REO^{-1} consumed, or –48% from the average to lower bound, reducing abiotic resource depletion of fossil fuels by 13%, acidification by 17%, human toxicity by 5%, and global warming potential by 14%.

Energy consumption was strongly linked to impact categories abiotic resource depletion of fossil fuels and global warming potential. For example, it contributed to 38% of abiotic resource depletion of fossil fuels and 37% of global warming in Bayan Obo. Results were not very sensitive to changes in energy consumption, however, between average and lower bound scenarios because of the fact that there was little variation in energy consumption between the two. Rather, sensitivity of results for abiotic resource depletion of fossil fuels and global warming potentials were brought about from a culmination of background processes, particularly magnesium oxide in Bayan Obo and ammonium sulfate in the Southern Provinces.

Discussion

A Deliberate Comparison between Production Chains

Impacts are difficult to compare given nonuniformity of functional units and the co-mining of a basket of different elements during REO production. We thus provide a careful and deliberate discussion to compare impacts between regions and processes, given their implications on industry policies.

LCA results for REO production in Bayan Obo and Sichuan are easiest to compare, given their similar composition of rare earths (table S1-2 in the supporting information on the Web). Between the two, life cycle environmental impacts were lower in Sichuan for 10 of 11 impact categories, including the five key categories: abiotic resource depletion of fossil fuels (–48%), acidification (–55%), eutrophication (–65%), global warming (–46%), and human toxicity (–32%). For some categories, impacts from the industry average in Sichuan were even lower than lower-bound impacts in Bayan Obo.

This suggests that the most effective measure for addressing environmental impacts may not be to adopt cleaner technologies (more discussed below), but rather to shift mining altogether from Bayan Obo to Sichuan. Of course, there are other factors to consider. Bayan Obo has the largest reserves of rare earths in China and currently commands the highest production quantity. Mining in the region is thus largely viewed as an inevitability. Further, its ores are also co-mined for iron. Given the glut in Chinese steel production (Chinese Information Office of the State Council 2015), however, this may be an opportune time to halt mining in the region. One solution could be to temporarily shift REO production to Sichuan while developments are made to cleaner production technologies in Bayan Obo. Such developments include research into low-temperature H_2SO_4 roasting (Zhao et al. 2013) and alkaline treatment as an alternative REO decomposition method (Du et al. 2010). This would help mitigate environmental

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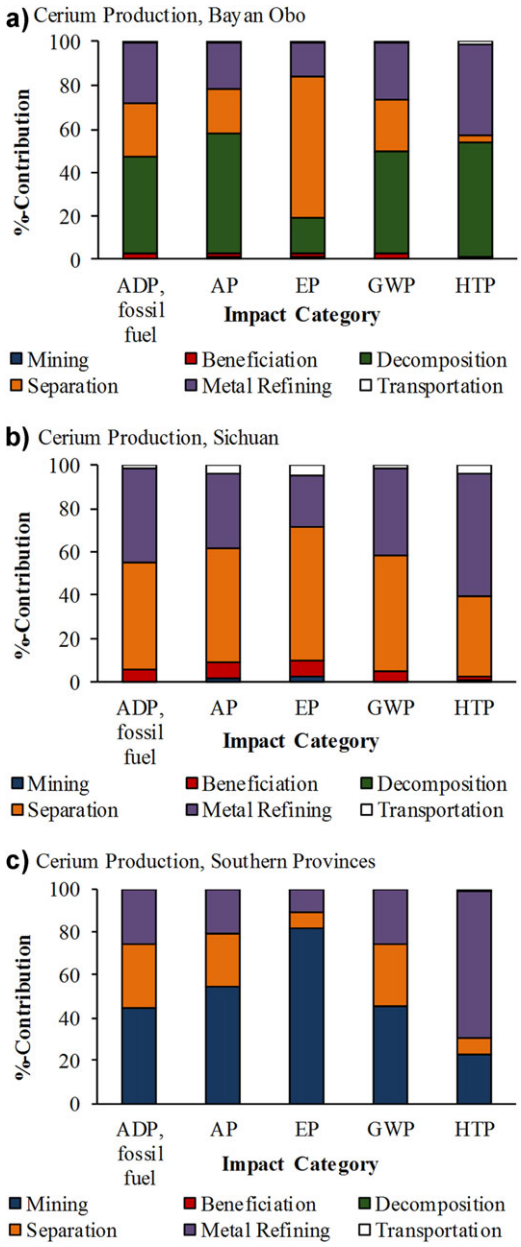

Figure 3 Contributions to environmental impacts from the three main rare earths production chains. Impacts from REO decomposition and separation in Sichuan have been aggregated and shown here as separation. ADP = abiotic depletion potential; AP = acidification potential; EP = eutrophication potential; GWP = global warming potential; HTP = human toxicity potential.

impacts both in the short term, as well as secure a sustainable supply (both economically and environmentally) of rare earths going forward.

Production in the Southern Provinces is more difficult to compare with other regions because of its unique composition of heavy rare earths. Given its importance in industry and to global supply, we assume there is little to no likelihood that production would be curbed because of environmental concerns. Thus, we instead shift our attention to how rare earths are mined. Whereas in-situ leaching has long been viewed as the environmentally friendly option, we echo recent concerns over the process (Yang et al. 2013), citing environment and human health damages assessed in this study. Significant acidification, eutrophication, and global warming potentials largely stem from the direct loss of ammonium sulfate solution to soil. Traditional methods, such as pool or heap leaching, could potentially mitigate some of these impacts through better-controlled leaching techniques, in exchange for large-scale surface excavation. This trade-off between environmental discharges and ecological damages would be an interesting topic for future research.

For metal refining, results expectedly show that there is indeed an environmental trade-off in producing higher-grade rare earths through metallothermic reduction as opposed to molten salt electrolysis, primarily attributed to higher consumption of electricity and key influent materials: rare earth fluoride, calcium, and lanthanum. Fortunately, the industry has, and is continuing to shift toward more widespread use of molten salt electrolysis, with ongoing research into its application for metals such as dysprosium and Gd alloys (Pang et al. 2011). If the technique is able to meet the technical requirements of most commercial applications as has been done with light rare earths, there would be few remaining barriers to more widespread use of molten salt electrolysis and thus lower environmental impacts.

Shifting the Industry Average to the Lower Bound

The lower-bound scenario can be seen as an indicator of ideal sustainable practices in the industry and shows significant potential to mitigate impacts: for Ce production, impacts are 21% to 38% lower for abiotic resource depletion potential (fossil fuels), 21% to 42% for acidification, 56% to 82% for eutrophication, 20% to 38% for global warming, and 40% to 60% for human toxicity. Unfortunately, only a few select enterprises have adopted said practices, which leaves an opportunity gap to move the rest of the industry in that direction.

Our sensitivity analysis shows that waste treatment is key in mitigating life cycle environmental impacts, particularly for HF, SO_2, and NH_3. Increasing the industry average (mean) on removal efficiency, for example, from 98.6% to 99% for treatment of HF, largely rests in small- to medium-size enterprises with little waste treatment capabilities. Although waste treatment technologies exist, they are often too costly to implement. In 2013, an estimated 80% of enterprises did not meet standards largely attributed to 70% higher production costs (Wübbeke 2013).

The Chinese government's approach has thus been to consolidate the many small- to medium-sized enterprises into a few megacorporations (Chinese Information Office of the State Council 2012). Theoretically, it would solve several immediate problems: By reducing the number of enterprises, the government could more easily enforce environmental standards; larger enterprises have more capital and financial flexibility to invest in costly waste treatment technologies; and batch treatment of large quantities of waste water and gas are usually more cost-effective. The undertaking is large and has already had

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both successes and failures (Wübbeke 2013). If done properly, however, it could fundamentally improve the industry's ability to address environmental problems.

Other key areas for improvement includes increasing collection efficiencies for ammonium sulfate leaching solution in the Southern Provinces. Apart from reverting to traditional methods as described above, new technologies and better planning have potential to increase efficiencies. Back-up collection systems certainly help (Zhu et al. 2011), but these systems are normally applicable to turn a good collection system (~80% collection efficiency) into a great collection system (90%). The bigger problem is with very low collection efficiencies (<50%). In these cases, more regulation is required to ensure better knowledge of local hydrogeology before commencing operations.

Energy consumption, and thus global warming potential, had the lowest variability of any impact category between the industry average and lower-bound results. For the purpose of mitigating global warming potential, research and development in this area should not be a priority. Even the highest carbon polluting rare earth metal (109 kg CO_2 equivalent [CO_2-eq]/kg Gd^{-1} in the Southern Provinces) ranks in the lower-middle stratum compared to global warming potential of other metals production (Nuss and Eckelman 2014). Coupled with its low production quantity (1,040 t of Gd in 2013), absolute discharges of greenhouse gases (1.23×10^8 kg CO_2-eq) from Gd production were miniscule compared to heavy-hitting industries such as cement (1.14×10^{12} kg CO_2-eq in 2010) (Wen et al. 2015).

Conclusions

An extensive LCA was conducted for rare earths production in China, contrasting environmental impacts from each of the main production pathways. In particular, the LCI included novel data for the two production chains in Sichuan and the Southern Provinces that are often ignored.

Results showed significant impacts of human toxicity, eutrophication, abiotic resource depletion of fossil fuels, acidification, and global warming potentials. For production of metals in Bayan Obo and Sichuan, impacts were concentrated in the oxide and metal refining processes, whereas for the relatively new in-situ leaching technique used in the Southern Provinces, both mining and refining contributed significantly to overall results. A sensitivity analysis of the lower bound showed significant opportunities to mitigate environmental damages.

Some structural changes have already been initiated through the Chinese government's consolidation of the rare earths producing enterprises, which, if done properly, would put the industry in a better position to meet environmental standards. Aside from waiting for the results of consolidation to complete later this year, we identify key opportunities in industrial policy to bring about changes.

For one, the five key impact categories were 32% to 67% lower for the REO production chain in Sichuan than Bayan Obo. Given that the composition of REO is also similar between the two regions, shifting a portion of production from the latter to the former would reduce impacts overall. Second, more regulation in the Southern Provinces is necessary, particularly regarding studies of local hydrogeology, before undertaking in-situ mining operations. This would significantly mitigate losses of ammonium sulfate during mining, facilitate higher internal recycling rates, and reduce demand for virgin ammonium sulfate. Last, improved waste treatment can most effectively reduce impacts for human toxicity (reduction of 31% to 70%), eutrophication (7% to 68%), and acidification (22% in Bayan Obo), depending on the production chain. Promoting better treatment practices and enforcing national discharge standards is vital to sustainability within the industry.

Acknowledgments

The authors are grateful for financial support from the National Natural Science Fund for Outstanding Young Scholars of China (71522011) and the Common Wealth Program on Environmental Protection (No. 200809062). Additional thanks goes to Dr. Zhiqi Long and Dr. Xiaowei Huang (Beijing Research Institute for Non-ferrous Metals) and Dr. An Wen Zhang (Chinese Society for Rare Earths) for sharing their expertise on the rare earth industry and to Huifang Li of Tsinghua University for her assistance in research.

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Supporting Information

Supporting information is linked to this article on the *JIE* website:

Supporting Information S1: This supporting information includes detailed descriptions of life cycle inventory for each rare earth production chain, life cycle impacts assessment, and results comparison.

Supporting Information S2: This supporting information contains information on Life Cycle Inventory - Bayan Obo, Life Cycle Inventory - Sichuan, Life Cycle Inventory - Seven Southern Provinces, Life Cycle Inventory - Molten Salt Electrolysis of Cerium, Life Cycle Inventory - Calciothermic Reduction of Gadoliunium, Life Cycle Inventory - Lanthanothermic Reduction of Samarium, LCA Results.

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