Life cycle assessment with primary data on heavy rare earth oxides from ion-adsorption clays

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Abstract:

Purpose

Heavy and light rare earth elements (REEs) are critical to clean energy technologies, and thus the environmental impacts from their production are increasingly scrutinized. Most previous LCAs of REE production focus on sites producing light REEs. This research addresses this gap by collecting primary data from sites producing heavy rare earth oxides (HREOs) from ion-adsorption clays, conducting an LCA, and providing open-source life cycle inventory (LCI) datasets of HREO production for the LCA community.

Methods

This study conducts a LCA based on acquired primary data from four mining sites in Jiangxi Province, China. The functional unit is 1 kg of mixed HREOs of 90% purity from ion-adsorption clays using the technology of \textit{in situ} leaching. Previous studies have used the Ecoinvent database, relying mostly European or global life cycle inventories (LCIs). Here, the Chinese Life Cycle Database provided China-specific reference life cycle inventories (LCIs). The Ecoinvent 3 database was also used in this study, however, for electricity generation LCIs used in a scenario analysis evaluating China’s changing grid mix (Wernet et al. 2016). Twelve impact categories were examined using Impact
2002+, USEtox 2.01 and IPCC methods. Results are provided as a bounded range, reflecting low and high estimates based on collected primary data.

**Results and discussion**

Results show 1 kg of mixed HREOs emit 258-408 kg CO$_2$e, and consume 270-443 MJ primary energy. These values fall within the range of previous LCAs that examined both bastnaesite/monazite deposits and ion-adsorption clays using literature values. Other impact categories considered are not similar across studies, however. Differences are due to variability in resource type and quality, technology, and modeling choices, such as reference LCI sources. Mining and extraction contribute most to impacts due to large quantities of chemicals for leaching and precipitation of REOs, and electricity consumption. Among chemicals, ammonium sulfate is the largest contributor to many impact categories. When China’s electricity grid mix change over time is included, environmental impacts for the whole production process can change up to 12%.

**Conclusions**

The primary contributions of this study are the collection and publication of primary data from mining companies in Jiangxi Province, China; the provision of open-source LCI datasets for mixed HREOs from ion-adsorption clays; and a comparison of results between this study and previously published studies. While the scope of this study concludes at the production of mixed HREO, which is a limitation, it provides a foundation for development of LCIs for refined heavy REEs.

**Key words:**

Heavy rare earth elements, HREE, LCA, mining, dysprosium, life cycle inventory
1 Introduction

Clean energy technologies, such as electric vehicles and wind turbines, have increased demand for permanent magnets, and particularly Nd-Fe-B rare earth magnets that are used in electric motors and generators (Alonso et al. 2012; Smith Stegen 2015; U.S. Department of Energy 2010; Van Gosen et al. 2014; Zhou et al. 2017). Nd-Fe-B permanent magnets have become the dominant magnet chemistry because of their significantly stronger magnetic fields than other types of magnets. With the growth in demand for these magnets, the constituent rare earth elements (REEs), such as neodymium and dysprosium, need to be supplied at an increasing rate.

Although many REEs are critically important to sustainable mobility and energy supply, production of REEs incur significant environmental damages, as their widely dispersed locations and low concentrations make them energy intensive and environmentally taxing to mine, extract, and refine (Eriksson and Olsson 2011; Navarro and Zhao 2014; Schüler et al. 2011). Prior reports have shown that REE extraction in China has brought serious environmental and health issues to the surrounding area, such as land depletion, water pollution, air pollution, and human exposure to radioactive materials (Xu et al. 2005; Yang et al. 2013). REEs have primarily been mined in China, with smaller amounts mined in Australia, India, Russia, and the United States (Charles et al. 2013; Du and Graede 2011; Gambogi 2016; Hykawy et al. 2010; Mariano and Mariano 2012). China continues to dominate the global REE supply, accounting for nearly 90% of global mine production in 2014 (Gambogi 2016).

China’s reserves of industrial grade rare earth oxides (REOs) are predominately in three categories, (1) mixed bastnaesite and monazite in Bayan-Obo which contain light REEs (LREEs) in an Iron-Nb-LREE deposit, (2) bastnaesite in Sichuan and Shandong provinces which contain high grade LREE, and (3) ion-adsorption clays in seven provinces of southern China that contain middle REEs (MREEs) and heavy REEs (HREEs) (Gambogi 2016; Schüler et al. 2011; SCIO 2012; Yang et al. 2013; Yongfu 1992). The ion-adsorption clay deposits in southern China are the world’s primary source of heavy REEs (HREEs), accounting for more than 80% of world’s total production of MREEs and HREEs (Chi et al. 2012; Su 2009). The REEs in ion-adsorption ores are adsorbed on the surface of clay minerals with REO concentrations ranging from 0.05–0.2%. Though the grade of ion-adsorption ores seems low, the ion state of REEs in these ores makes extraction and processing easier and more economical than mining from bastnaesite and monazite (Gambogi 2016; SCIO 2012; Yang et al. 2013).

Though life cycle assessment (LCA) has been widely used as an assessment tool for many economically important materials, only a handful of recent studies investigate the environmental impacts of REO/REE production, most of which are based on the largest REE producing mine – the Bayan Obo deposit in China (Koltun and Tharumajarajah 2014; Nuss and Eckelman 2014;
Sprecher et al. 2014; Zaimes et al. 2015), which largely produces LREEs. In general, few studies have focused on the mine production of medium and heavy REOs, though they have more applications and are of higher value than LREEs. In fact, only a few previous LCA studies have characterized the LCA of REOs and REEs produced from ion-adsorption clays and particularly those that are primarily heavy REE bearing. Vahidi et al. (2016) was the first to publish an LCA of REO production from ion-adsorption clays and is most similar in scope to this study, though they relied on literature values for their calculations. Following Vahidi et al., Schulze et al. (2017) also conducted a literature-based LCA of REOs from ion adsorption clays. Zapp et al. (2018) analyzed the production of dysprosium, a heavy REE, from ion adsorption clays and bastnaesite/monazite, and while they did not report results in terms of mixed REO, they modeled the process of mining and extraction from ion adsorption clays as steps in the production process for dysprosium. Other studies have considered the process route for heavy REO and environmental impacts caused from producing heavy REO, but not from a LCA perspective (Liao et al. 2014; Moldoveanu and Papangelakis 2013; Yang et al. 2013; Zou 2012).

The goals of this research are to (1) collect and publish primary data related to the mining of ion-adsorption clays that produce mixed HREOs; (2) develop a new open-source life cycle inventory (LCI) and conduct a LCA to augment the small but growing body of work characterizing the life cycle impacts of REO production from ion adsorption clays and production of heavy REOs; and (3) compare results from this LCA to others published in the literature. This study’s specific contributions to the literature include the provision of primary data from four ion-adsorption clay rare earth mining sites in Jiangxi Province, southern China, and the publication of open-source LCIs that reflect the high and low bounds based on the collected primary data.

2 Methods

This study characterizes the production of mixed REOs from ion-adsorption clays in southern China, with 90% purity. Compositions of individual rare earth minerals in REO differ from ore to ore. Ores from two ion adsorption clays in Jiangxi Providence, the same region from where data were collected for this LCA, are shown in Table 1, and though they might not exactly characterize the particular REOs mined at the sites from which data were collected in this study, they are likely to be similar.\(^1\) The system boundary for the LCA study includes mining using in situ leaching methods, extraction, and calcination. These processes are all conducted on site, and the final product is mixed REOs. Further processes such as solvent extraction and individual metal separation are not included, as they often occur off-site.

\(^1\) The companies that provided the primary data on extraction did not provide the specific composition of the REOs produced at their sites. This information was considered too sensitive and could possibly reveal the identity of the cooperating companies.
The functional unit chosen for this study is 1 kg of heavy REOs at 90% purity. The LCI was developed using primary data for the foreground system collected from four sites in southern China, as described in sections 2.1.2. Background data are from the Chinese life cycle database (CLCD). CLCD is a Chinese national reference LCI database developed by Sichuan University, China and IKE Environmental Technology CO. Ltd, and it sources original data from industry statistics, government publications, academic research and other LCA databases (Liu 2010). The one exception is electricity data provided by Ecoinvent that is used in the scenario analysis conducted in this study (Ecoinvent 2016). These datasets from Ecoinvent are used to model the effects of changing electricity grid composition via scenario analysis.

2.1 Model Development and Data Collection

In bastnaesite/monazite sites, typical processing steps to recover LREEs include mining, physical and chemical beneficiation, extraction, calcination and roasting. For ion adsorption clays, the first three steps are carried out together using leaching technology, but the remaining steps are the same, as shown in Figure 1. Various leaching methods have been used in the past, but now in situ leaching has become the primary technology, replacing heap leaching and pool leaching due to its improved environmental performance (Chi et al. 2012; Yang et al. 2013; Zhao et al. 2001; Zou 2012).

2.1.1 In situ leaching of ion-adsorption clays

The ion-adsorption clay deposit assessed in this study is sparsely distributed throughout seven adjacent provinces in southern China; Jiangxi, Guangdong, Fujian, Zhejiang, Hunan, Guangxi and Yunnan. Figure 1 illustrates a simplified process flow for in situ leaching of ion-adsorption clays. In a typical in situ process, about one-third of the topsoil is removed. Two pipe systems are built, one with ammonium sulfate \((\text{NH}_4)_2\text{SO}_4\) for the ion exchange mechanism and fresh water flushing at the end of mining, and the other for collection of leachate. Leachate is piped to pools built close to the hillside and organized in terraces. After injecting the leaching solution, which generally is 3-5\% \((\text{NH}_4)_2\text{SO}_4\), it takes 150-400 days for leaching to finish, and then fresh water is injected to drive out the remaining REE-bearing solutions (Jun et al. 2011; Yang et al. 2013). The leachate is usually treated with \(\text{NH}_4\text{HCO}_3\) OR \(\text{H}_2\text{C}_2\text{O}_4\) to precipitate REEs (Jun et al. 2011; Zhao et al. 2001). The reaction equation for leaching is described in Eq (1), and precipitation reactions are described in Eq (2) and (3).

\[
2(\text{Kaolin})^{3-} + 3(\text{NH}_4)_2\text{SO}_4 \rightarrow 2(\text{Kaolin})^{3-}(\text{NH}_4)_3^+ + \text{RE}_2^{3+}(\text{SO}_4)_3^{2-} \quad (1)
\]

\[
2\text{RE}^{3+} + 3\text{NH}_4\text{HCO}_3 \rightarrow \text{RE}_2(\text{CO}_3)_3 + 3\text{NH}_4^+ + 3\text{H}^+ \quad (2)
\]

\[
2\text{RE}^{3+} + 3\text{H}_2\text{C}_2\text{O}_4 + x\text{H}_2\text{O} \rightarrow \text{RE}_2(\text{C}_2\text{O}_4)_3 \cdot x\text{H}_2\text{O} + 6\text{H}^+ \quad (3)
\]
For industrial operations, ammonium bicarbonate is more often used than oxalic acid, because of its lower cost and lower environmental impact (Chi et al. 2012; Chi et al. 2003; Zou et al. 2014). However, using oxalic acid can lead to higher product purity, and using ammonium bicarbonate may cause additional difficulties in the following dehydration process. In reality, both ammonium bicarbonate and oxalic acid are used depending on different considerations. Other inputs to the process include sulfuric acid (H$_2$SO$_4$) to adjust pH levels in the extraction stage (Jun et al. 2011; Zhao et al. 2001). Electricity is used in mechanical pressing to remove water and for calcination of the precipitated material, which happens at 750-850 °C to get REOs. Fuel oil or coal were often used in calcination in the past, but now the newly built calcination stoves use electricity, rather than oil or coal.

2.1.2 Data collection

A field study was conducted in October 2016 in southern China with the goal of creating an original LCI dataset for heavy REOs to lay the foundation for improved analysis of HREE impacts. Data from four HREE mining sites in Ganzhou city, Jiangxi Province were collected. The data provided by cooperating companies included chemical, water, and energy inputs. Four sets of data (one from each site) were then compiled and reported as a range, rather than a point value. The one exception is for electricity used in extraction, as only one site provided data, thus only a point value is reported. Some data were provided in terms of monetary costs and were converted to quantities using the current market price at the time of data collection. The conversion of monetary costs to mass and energy flows introduces a potential source of uncertainty, due to price variability or volatility. All data were obtained in units of 1 ton REO, and converted to 1 kg REO.

Since the mining happens in China, the CLCD (Liu 2010) was the preferred database for LCI dataset acquisition. As shown in Table 2, LCIs were available for most of the inputs required for heavy REO production; however, an LCI for oxalic acid, though an important input in the extraction stage, was not available in the CLCD or Ecoinvent, which was investigated to fill this gap. Thus, in this study, a surrogate LCI is developed assuming that oxalic acid is produced as the reaction product of coke, H$_2$SO$_4$ and NaOH (Zaimes et al. 2015).

2.1.3 Process Emissions

Process emissions were not available as primary data from producers. Instead, process emissions to air and water were estimated. Estimation of direct process emissions are based on the regulated upper limit in the *Emission Standards of Pollutants from Rare Earth Industry* published by the Ministry of Environmental Protection, the People’s Republic of China (MEP 2011).
These wastewater emissions and air emissions are summarized in Table 3. Previous research has shown that real-world emissions may significantly exceed these upper limits, by as much as a factor of four or five (Yang et al. 2013; Wang 2013). Because of this, a sensitivity analysis is included that tests the effects of increasing on-site process emissions by as much as 500%.

2.2 Life Cycle Impact Assessment

While the primary goal of this analysis is to present a LCI for HREO, an impact assessment is also conducted to assist in interpreting the findings of this study. Impact 2002+ was used to characterize respiratory effects (kg PM$_{2.5}$ eq), non-carcinogenics (kg C$_2$H$_3$Cl eq), carcinogenics (kg C$_2$H$_3$Cl eq), ozone layer depletion (kg CFC$_{11}$ eq), aquatic acidification (kg SO$_2$ eq), terrestrial acidification (kg SO$_2$ eq), eutrophication (kg PO$_4^{3-}$ eq), and photochemical oxidation (kg C$_2$H$_4$ eq); USEtox 2.01 was used to characterize ecotoxicity (CTUh); and 100-year Global Warming Potentials from IPCC (kg CO$_2$ eq) were used to characterize greenhouse gases. Primary energy (MJ), non-renewable energy (MJ) were also calculated. The impact factors were derived from the GaBi Software tool version 8.0 (Thinkstep 2017).

3 Results

The results include a low and a high estimate for all the 12 impact categories investigated. Complete LCIs for the lower and upper bound estimates are provided in the online supplementary material associated with this article. Due to site variation and process conditions, there exist significant differences with regard to environmental impacts. The high estimate is about 140% to 240% of the low estimate, which reflects differences across the four queried sites and the motivation for providing high and low LCIs in lieu of an average.

Table 4 compares the results of this study to results from other published papers on REO production (Sprecher et al. 2014; Vahidi et al. 2016; Zaimes et al. 2015). For energy and GWP, the results of this study (which are reported as high and low values), fall within the range of results reported for the three other studies, and this is similarly true for estimates of respiratory effects. However differences are evident in estimates of non-carcinogenics, carcinogenics, and ozone depletion. The LCI database used in this study is CLCD which represents the average Chinese market, while other studies used different versions of Ecoinvent (Wernet et al. 2016) which is mostly focused on global and European contexts. Because of different reference LCI databases and differences in the characterization methods used, it is perhaps not unexpected that some impact categories show significant differences in their findings. With respect to different reference LCI databases, the assumed region of
production is important; the LCIs for chemicals produced in China, as modeled in this study and which reflect the source of chemicals used at Chinese mining and extraction sites, are very different from the global or European average LCIs which are used in other studies. Finally, REOs of different levels of purity were analyzed by these studies.

To further analyze the differences across all impact categories, breakdown of the 12 categories by process stage and input types are presented in Tables 5 and 6, respectively. Table 5 presents the contribution of life cycle impact by stages for the production of 1 kg of heavy REO. Figure 2 plots the percent contribution consistent with Table 5. On-site emissions contribute to four categories; eutrophication potential, terrestrial acidification, aquatic acidification and respiratory effects. Table 6 presents contribution of life cycle impact by input type for the production of 1 kg of heavy REOs. Estimates of different input types are provided for each impact category, as well as their percent contribution to the whole process in Figure 3.

4 Discussion

For most of the impact categories, the mining and extraction stage are the top two contributors. The mining stage includes chemicals used in equation (1), and also includes electricity for pumping the solution in and out of the soil. The extraction process includes chemicals used for the leaching and the precipitation processes, represented by equations (2) and (3). The difference between the lower range and higher range for a particular stage is the result of different amounts of chemicals used per kg REO produced.

Ammonium sulfate is the largest contributor to nearly all 12 impact categories, especially in the higher range case (Table 6). The same finding was noted in Vahidi et al. (2016). Ammonium bicarbonate influences terrestrial acidification and eutrophication heavily. Oxalic acid contributes largely to ecotoxicity, and its contributions are quite significant in other categories as well. Because there is no LCI for oxalic acid, different measures were taken by researchers to develop surrogates, including use of an LCI for citric acid (Vahidi et al. 2016) or developing a surrogate based on reactants that produce oxalic acid (Zaimes et al. 2015), as was done in this study. The development of an LCI for oxalic acid would reduce model uncertainties.

Producers actually have some flexibility in their use of chemicals, and in particular ammonium sulfate. Leaching time and ammonium sulfate usage are inversely correlated, meaning that producers can reduce ammonium sulfate usage and achieve the same yields by increasing leaching time. Thus a sensitivity analysis was conducted to explore the sensitivity of impact results to reducing chemical inputs to the production process. Figure 3 illustrates these results and shows that a number of impact categories, namely human toxicity (non-carcinogenic and carcinogenic), aquatic acidification, global warming, and ozone
depletion are highly sensitive to ammonium sulfate use (a 10% decrease in ammonium sulfate decreases these impact categories by about 5% or more). Ecotoxicity is highly sensitive to the use of oxalic acid (a 10% decrease in use leads to more than a 6% decrease in ecotoxicity), and both eutrophication and terrestrial acidification are highly sensitive to ammonium bicarbonate use. The effect of reducing sulfuric acid use was also tested, but resulted in much lower sensitivity; a 10% decrease in use never reduced the impact of any category by even 1%, and so is not shown in Figure 4.

A source of significant uncertainty in this LCA is the level of pollution occurring on-site because of a lack of primary data for these flows. In the absence of primary data, in-situ leaching operations were assumed to pollute at the maximum regulatory limits. Unfortunately, as indicated previously, studies have shown that some sites have exceeded pollution limits by up to 500%. To examine the potential effect of non-compliance, a sensitivity analysis was conducted assuming that on-site emissions standards were exceeded by a factor of five (500%). The most sensitive impact category result was respiratory effects, where exceedance of standards by a factor of five led to an increase of 169% (about 3.4% change in respiratory effects for every 10% change in on-site emissions). The second most sensitive impact category was eutrophication, where an exceedance by a factor of five led to an increase of 44% (less than a 0.5% change for every 10% increase in on-site emissions). Thus while on-site emissions are highly uncertain, life cycle impacts are generally more sensitive to the quantity of chemical inputs, than on-site pollution. Of course, in reality these two choices may be correlated (not independent), so in fact reducing chemical inputs could reduce on-site emissions (i.e. reducing ammonium sulfate use could also reduce eutrophying emissions from the site), thus increasing the benefits of reduced chemical inputs.

For the low range case, electricity is the largest contributor to primary energy consumption and is second only to ammonium sulfate for GWP. China’s electricity grid is undergoing rapid change and development. Scenario analysis is used to provide a dynamic picture of the effect of electricity grid mix changes on the results of this LCA. The baseline year is 2015, and future scenarios for 2020 and 2030 are taken from literature (Chu 2015). Electricity generation in China relies heavily on burning hard coal, and some on hydropower. Gradually, the grid mix in China will shift away from the dominance of coal, and more renewables and natural gas shall be used in the future (Chu 2015). Note that Table 7 presents the shares of current and future electricity grid mix in China, not the generation amount. Reduction in the share of hard coal, for example, does not mean reduced consumption, but rather that as generating capacity grows, an increased share of new sources are derived from natural gas and renewables. All electricity production LCI data are selected from province-specific datasets available in Ecoinvent.
(Wernet et al. 2016), except for nuclear power. Currently there is no nuclear power reactor in Jiangxi Province, thus the reference LCI for electricity generation from nuclear power is taken from reactors from an adjacent province.

Figure 5 shows how the change in electricity mix will change heavy REO production for all impact categories. For most of the impact categories, changes compared to the baseline year are negative. This means that the proportional decrease in reliance on coal, and increased reliance on renewables and natural gas, lead to reduced impacts. However, for ozone depletion and ecotoxicity categories, the results are positive, which means for these two categories the change in electricity mix increases emissions. For ozone depletion, solar power, natural gas, and nuclear all have much higher estimates (about five, 10 and 20 times) than that of coal, thus promoting their use leads to higher ODP estimates. For ecotoxicity, natural gas has significantly higher impacts per unit of generation compared to coal power, and to a lesser extent, solar power and wind power also have higher ecotoxicity impacts.

Depending on the portion electricity takes over the entire life cycle process for each impact category (see Table 6), the influences of electricity grid mix change varies. For example, the change in electricity grid has little influence on non-carcinogenics category, as electricity is a small contributor to this category (see Fig 3).

According to statistics by the China electricity council (2016 China electricity industry statistics), about 65% of all electricity in 2016 was generated by coal-fired power plants. The future picture for China’s electricity grid mix is surely to shift toward renewable power and away from dominance of coal. Among all renewables, hydro power plays a pivotal role in the generation mix, accounting for about 19% of all electricity generation in 2015 and 2016 (2016 China electricity industry statistics). The potential to utilize more hydro power may be constrained in the short run, however there is still great potential for solar and wind. Currently, in Inner Mongolia, governments and companies are working in collaboration to allow for greater absorption of wind electricity into local grids; while in Jiangxi Province, subsidies are provided by both state and provincial levels to fuel the growth of solar installations (Chu 2015). Thus, it is expected that the future picture for electricity grid mix in China will be “cleaner”, which will lead to less energy consumption and environmental pollutants for rare earth industry.
Conclusion

This study has attempted to provide two original contributions to the existing body of work characterizing the life cycle impacts of HREE production; the first is the collection of primary data from ion-adsorption clay mining sites in Ganzhou, Jiangxi province China to characterize this increasingly important source of HREEs, and the second is the provision of open-source reference LCI datasets for mixed HREOs for the LCA research and practitioner community. Rather than providing an average LCI, both the open-source LCI and the LCA results are presented as low and high estimates. The provision of high and low estimates is important, as high estimates are about 140% to 240% of the low estimates. While the scope of this study concludes at the production of mixed HREO, which is a limitation, it provides a foundation for future studies that seek to develop LCIs of refined HREEs.

In terms of process stages, mining and extraction are the two largest contributors to most impact categories. In terms of inputs to the production system, ammonia sulfate is a very significant contributor to all impact categories, ammonia bicarbonate is the top contributor to terrestrial acidification and eutrophication potential, and oxalic acid dominates the ecotoxicity category. Based on sensitivity analysis conducted on the use of chemical inputs, LCA results are quite sensitive to the quantity used for all these chemicals, and especially ammonia sulfate. Adopting practices that reduce chemical usage, such decreasing ammonia sulfate use and increasing contact time, could be an opportunity to reduce the environmental impacts of heavy REO mining and extraction.

The results from this study were compared with published studies, including two papers on REO production from bastnaesite/monazite, and one from ion-adsorption clays based on literature data. Comparisons showed that some environmental impacts for in situ leaching of ion-adsorption clays fall into the same range of bastnaesite/monazite processing, for example, energy use, global warming and respiratory effects. However, considering the different databases used by the compared studies, the comparison should be interpreted with care. In addition, changes to China’s electricity grid mix in the future will lead to changes in the environmental impacts of producing REO as well. With shifts away from hard coal towards the use of natural gas and renewables, most of the estimates in LCIA categories are lower.

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| REO         | Bastnaesite | Monazite | Ion adsorption clay |
|-------------|-------------|----------|---------------------|
|             | Longnan,Jiangxi | Xinfeng,Jiangxi |
| **LREE**    |             |          |                     |
| La₂O₃       | 27.00       | 23.35    | 2.10                | -                   |
| CeO₂        | 50.00       | 45.69    | 1.00                | -                   |
| Pr₂O₃       | 5.00        | 4.16     | 1.10                | -                   |
| Nd₂O₃       | 15.00       | 15.72    | 5.10                | 17.00               |
| Sm₂O₃       | 1.10        | 3.05     | 3.20                | -                   |
| Eu₂O₃       | 0.20        | 0.10     | 0.30                | -                   |
| **HREE**    |             |          |                     |
| Gd₂O₃       | 0.40        | 2.03     | 2.69                | 5.90                |
| Tb₂O₇       | -           | 0.10     | 1.13                | 0.60                |
| Dy₂O₃       | -           | 1.02     | 7.48                | 3.70                |
| Ho₂O₃       | -           | 0.10     | 1.60                | -                   |
| Er₂O₃       | 1.00        | 0.51     | 4.26                | 2.40                |
| Tm₂O₃       | -           | 0.51     | 0.60                | -                   |
| Yb₂O₃       | -           | 0.51     | 3.34                | -                   |
| Lu₂O₃       | -           | 0.10     | 0.47                | 0.90                |
| Y₂O₃        | 0.30        | 3.05     | 62.90               | 24.00               |
| Stage                  | Input                | Use                  | Lower range | Higher range | Unit       | CLCD LCI database                          |
|-----------------------|----------------------|----------------------|-------------|--------------|------------|--------------------------------------------|
| Mining/In situ leaching | Ammonia sulfate      | ion exchange         | 4           | 10           | kg/kg-REO  | Ammonium sulfate, production average (CN), aggregate | CLCD-China-Public 0.8.1 2012 |
|                       | Electricity          | injection            | 4.3         | 4.3          | kWhr/kg-REO| Central China mix power, at grid (CN), aggregate | CLCD-China-Public 0.8.1 2012 |
| Extraction            | Coke                 | produce oxalic acid  | 1.02        | 1.28         | kg/kg-REO  | Coke, production average (CN), aggregate | CLCD-China-Public 0.8.1 2012 |
|                       | H$_2$SO$_4$          | produce oxalic acid  | 1.9         | 2.38         | kg/kg-REO  | Sulfuric acid, market average, 98% (CN), aggregate | CLCD-China-Public 0.8.1 2012 |
|                       | NaOH                 | produce oxalic acid  | 1.84        | 2.3          | kg/kg-REO  | Sodium hydroxide, 100% NaOH (CN), aggregate | CLCD-China-Public 0.8.1 2012 |
|                       | Ammonia bicarbonate  | precipitation        | 3.3         | 6            | kg/kg-REO  | Ammonium bicarbonate, production average (CN), aggregate | CLCD-China-Public 0.8.1 2012 |
|                       | H$_2$SO$_4$          | adjust pH level      | 0.2         | 1            | kg/kg-REO  | Sulfuric acid, market average, 98% (CN), aggregate | CLCD-China-Public 0.8.1 2012 |
| Calcination           | Electricity          | heating              | 4.5         | 4.5          | kWhr/kg-REO| Central China mix power, at grid (CN), aggregate | CLCD-China-Public 0.8.1 2012 |
| Emission Type   | Total/Limit | Emissions   |
|-----------------|-------------|-------------|
| **Water**       |             |             |
| Suspended solids| 100 mg/L    | 0.002500 kg/kg REO |
| Fluorides       | 10 mg/L     | 0.000250 kg/kg REO |
| Ammonia         | 50 mg/L     | 0.003053 kg/kg REO |
| Total nitrogen  | 70 mg/L     | 0.004275 kg/kg REO |
| **Air**         | mg/m³       | kg/kg REO   |
| SO₂             | 300 mg/m³   | 0.007500 kg/kg REO |
| H₂SO₄ mist      | 85 mg/m³    | 0.002020 kg/kg REO |
| Dust            | 47 mg/m³    | 0.001375 kg/kg REO |
| Fluoride        | 20 mg/m³    | 0.000500 kg/kg REO |
| Chlorine        | 20 mg/m³    | 0.000600 kg/kg REO |
| Hydrogen chloride| 90 mg/m³  | 0.002500 kg/kg REO |
| Impact Category       | Current study                                                                 | Vahidi et al.                                                                 | Zaines et al.                                                                 | Sprecher et al.                                                                 |
|----------------------|-------------------------------------------------------------------------------|-------------------------------------------------------------------------------|-------------------------------------------------------------------------------|-------------------------------------------------------------------------------|
|                      | **Functional unit**                                                           | **1 kg of REO from ion-adsorption clays, 90% purity**                         | **1 kg of REO from ion-adsorption clays, 90-92% purity**                      | **1 kg of REO from bastnasite/monazite, 99% purity**                          |
|                      | **LCI Database**                                                              | CLCD                                                                          | Ecoinvent 3                                                                   | Ecoinvent 3                                                                   | Ecoinvent 2.2                                                                 |
|                      | **LCIA method**                                                              | Impact 2002+, USEtox 2.01 and IPCC                                           | TRACI/ ILCD                                                                   | TRACI                                                                         | CML 2001                                                                     |
| Primary energy       | MJ                                                                            | 269.67                                                                        | 442.60                                                                        | 255–388                                                                      | 578.79                                                                       | 174–232                                                                      |
| Non-renewable energy | MJ                                                                            | 247.66                                                                        | 408.00                                                                        |                                                                                |                                                                               |                                                                               |
| Global warming       | kg CO₂ eq                                                                     | 18.80                                                                         | 33.11                                                                         | 20.9–35.5                                                                    | 35.27                                                                        | 12–16                                                                         |
| Respiratory effect   | kg PM₂.₅ eq                                                                  | 9.94E-02                                                                      | 1.46E-01                                                                      | 1.80E-01                                                                     |                                                                               |                                                                               |
| Non carcinogenics    | kg C₂H₅Cl eq                                                                  | 7.14E-03                                                                      | 1.73E-02                                                                      | 1.29E-05                                                                     |                                                                               |                                                                               |
| Carcinogenics        | kg C₂H₅Cl eq                                                                  | 1.00E-03                                                                      | 1.83E-03                                                                      | 1.39E-06–2.21E-06                                                           | 3.11E-06                                                                     |                                                                               |
| Ozone depletion      | kg CFC₁₁ eq                                                                   | 3.45E-08                                                                      | 6.11E-08                                                                      | 2.43E-06–3.21E-06                                                           | 1.97E-05                                                                     | 2.5E-06–3.0E-06                                                              |
| Ecotoxicity          | CTUe                                                                           | 2.54E-03                                                                      | 3.61E-03                                                                      |                                                                               |                                                                               |                                                                               |
| Aquatic acidification| kg SO₂ eq                                                                     | 1.73E-01                                                                      | 3.53E-01                                                                      |                                                                               |                                                                               |                                                                               |
| Terrestrial acidification| kg SO₂ eq                                                                     | 7.06E-01                                                                      | 1.25E+00                                                                      |                                                                               |                                                                               |                                                                               |
| Eutrophication potential | kg PO₄³- eq                                                                  | 7.71E-03                                                                      | 1.10E-02                                                                      |                                                                               |                                                                               |                                                                               |
| Photochemical oxidation| kg C₂H₄ eq                                                                   | 3.18E-03                                                                      | 5.19E-03                                                                      |                                                                               |                                                                               |                                                                               |
Table 5. Impact assessment results of producing 1 kg of heavy REO by stages

| Impact Category            | Unit  | Direct emission | Upstream emissions by stages (lower range) | Upstream emissions by stages (higher range) |
|----------------------------|-------|-----------------|-------------------------------------------|---------------------------------|
|                            |       |                 | Mining | Extraction | Calcination | Mining | Extraction | Calcination |
| Primary energy             | MJ    | 117.51          | 110.49 | 41.67      |            | 234.05 | 166.90      | 41.67       |
| Non-renewable energy       | MJ    | 104.68          | 107.73 | 35.25      |            | 211.18 | 161.58      | 35.25       |
| GWP<sub>100</sub>          | kg CO<sub>2</sub>eq | 0.00 | 10.44 | 5.37 | 2.99 | 21.83 | 8.29 | 2.99 |
| Respiratory effect         | kg PM<sub>2.5</sub>eq | 4.92E-02 | 2.98E-02 | 1.55E-02 | 4.94E-03 | 6.74E-02 | 2.40E-02 | 4.94E-03 |
| Non-carcinogens            | kg C<sub>2</sub>H<sub>5</sub>Cleq | 0.00 | 5.41E-03 | 1.65E-03 | 8.44E-05 | 1.34E-02 | 3.85E-03 | 8.44E-05 |
| Carcinogens                | kg C<sub>2</sub>H<sub>5</sub>Cleq | 0.00 | 4.29E-04 | 5.16E-04 | 5.39E-05 | 9.96E-04 | 7.84E-04 | 5.39E-05 |
| Ozone depletion            | kg CFC-11eq | 0.00 | 1.50E-08 | 1.65E-08 | 2.94E-09 | 3.33E-08 | 2.49E-08 | 2.94E-09 |
| Ecotoxicity                | CTUe  | 0.00 | 3.80E-04 | 2.05E-03 | 1.10E-04 | 7.92E-04 | 2.71E-03 | 1.10E-04 |
| Aquatic Acidification      | kg SO<sub>2</sub>eq | 1.08E-02 | 9.57E-02 | 5.63E-02 | 1.06E-02 | 2.24E-01 | 1.08E-01 | 1.06E-02 |
| Terrestrial acidification  | kg SO<sub>2</sub>eq | 6.13E-02 | 2.43E-01 | 3.45E-01 | 5.64E-02 | 5.26E-01 | 6.06E-01 | 5.64E-02 |
| Eutrophication potential   | kg PO<sub>4</sub>³eq | 1.13E-03 | 7.77E-03 | 1.13E-03 | 9.59E-03 | 1.35E-02 | 1.13E-03 |
| Photochemical oxidation     | kg C<sub>2</sub>H<sub>4</sub> eq | 0.00 | 1.21E-03 | 1.62E-03 | 3.47E-04 | 2.52E-03 | 2.33E-03 | 3.47E-04 |
Table 6. Impact assessment results of producing 1 kg of heavy REO by input types

| Impact Category     | Unit     | ammonium sulfate | ammonium bicarbonate | sulfuric acid | oxalic acid | electricity |
|---------------------|----------|------------------|-----------------------|--------------|------------|-------------|
|                     |          | lower range      | higher range          | lower range  | higher range| lower range  | higher range|          |
| Primary energy      | MJ       | 77.69            | 194.23                | 42.00        | 76.37      | 1.31        | 6.55        | 67.18      | 83.97      | 81.49       |
| Non-renewable energy| MJ       | 71.00            | 177.49                | 40.98        | 74.51      | 0.97        | 4.84        | 65.78      | 82.23      | 68.94       |
|                     | GWP 100  | 7.59             | 18.98                 | 2.26         | 4.10       | 0.08        | 0.41        | 3.03       | 3.78       | 5.84        |
| Respiratory effect  | kg PM$_{2.5}$ eq | 2.51E-02    | 6.27E-02              | 6.51E-03     | 1.18E-02   | 2.53E-04    | 1.26E-03    | 8.70E-03   | 1.09E-02   | 9.65E-03    |
| Non carcinogens     | kg C$_3$H$_2$Cl eq | 5.33E-03    | 1.33E-02              | 9.93E-04     | 1.81E-03   | 3.27E-04    | 1.64E-03    | 3.30E-04   | 4.12E-04   | 1.65E-04    |
| Carcinogens         | kg C$_3$H$_2$Cl eq | 3.78E-04    | 9.45E-04              | 2.00E-04     | 3.64E-04   | 6.50E-06    | 3.25E-05    | 3.10E-04   | 3.87E-04   | 1.05E-04    |
| Ozone depletion     | kg CFC$_{11}$ eq | 1.22E-08    | 3.05E-08              | 5.48E-09     | 9.95E-09   | 3.10E-10    | 1.55E-09    | 1.07E-08   | 1.34E-08   | 5.75E-09    |
| Ecotoxicity         | CTUe     | 2.75E-04        | 6.87E-04              | 2.06E-04     | 3.75E-04   | 8.49E-06    | 4.24E-05    | 1.83E-03   | 2.29E-03   | 2.16E-04    |
| Aquatic acidification| kg SO$_2$ eq | 8.56E-02    | 2.14E-01              | 3.93E-02     | 7.14E-02   | 4.06E-03    | 2.03E-02    | 1.30E-02   | 1.63E-02   | 2.07E-02    |
| Terrestrial acidification | kg SO$_2$ eq | 1.89E-01    | 4.72E-01              | 2.88E-01     | 5.23E-01   | 2.84E-03    | 1.42E-02    | 5.51E-02   | 6.88E-02   | 1.10E-01    |
| Eutrophication potential | kg PO$_4^3-$ eq | 3.40E-03   | 8.51E-03              | 6.61E-03     | 1.20E-02   | 2.02E-05    | 1.01E-04    | 1.14E-03   | 1.42E-03   | 2.21E-03    |
| Photochemical oxidation | kg C$_3$H$_4$ eq | 8.74E-04   | 2.19E-03              | 4.11E-04     | 7.48E-04   | 1.78E-05    | 8.90E-05    | 1.19E-03   | 1.49E-03   | 6.78E-04    |
Table 7. Current and future electricity grid mix in China (in percentage)

| Source          | Baseline (2015) % | Scenario 1 (2020) % | Scenario 2 (2030) % |
|-----------------|-------------------|---------------------|---------------------|
| Hard coal       | 68.00             | 59.73               | 50.88               |
| Solar           | 0.68              | 2.40                | 3.98                |
| Hydro power     | 19.90             | 18.93               | 18.23               |
| Natural gas     | 5.10              | 5.28                | 7.78                |
| Wind            | 3.30              | 9.44                | 12.62               |
| Nuclear power   | 3.02              | 4.21                | 6.51                |
Fig. 1 Processing route for REO from ion-adsorption clay, southern China
Fig. 2 Contribution of life cycle impacts by process stage for producing 1 kg of heavy REO; (a) lower bound, (b) higher bound (PE – Primary Energy, NRE – nonrenewable energy, GWP – global warming, 100-year, RE – respiratory effect, NCAR – human toxicity, non-carcinogenic, CAR – human toxicity, carcinogenic, OD – ozone depletion, ECO – ecotoxicity, AA – aquatic acidification, TA – terrestrial acidification, EP – eutrophication potential, PO – photochemical oxidation)
Fig. 3 Contribution of life cycle impacts by inputs for producing 1 kg of heavy REO; (a) lower bound, (b) higher bound

(PE – Primary Energy, NRE – nonrenewable energy, GWP – global warming, 100-year, RE – respiratory effect, NCAR – human toxicity, non-carcinogenic, CAR – human toxicity, carcinogenic, OD – ozone depletion, ECO – ecotoxicity, AA – aquatic acidification, TA – terrestrial acidification, EP – eutrophication potential, PO – photochemical oxidation)
Fig. 4 Sensitivity of impact category results to a 10% decrease in process chemical use (PE – Primary Energy, NRE – nonrenewable energy, GWP – global warming, 100-year, RE – respiratory effect, NCAR – human toxicity, non-carcinogenic, CAR – human toxicity, carcinogenic, OD – ozone depletion, ECO – ecotoxicity, AA – aquatic acidification, TA – terrestrial acidification, EP – eutrophication potential, PO – photochemical oxidation)
Fig. 5 Percent change in impact category results for producing 1 kg of heavy REO by change in electricity grid mix (PE – Primary Energy, NRE – nonrenewable energy, GWP – global warming, 100-year, RE – respiratory effect, NCAR – human toxicity, non-carcinogenic, CAR – human toxicity, carcinogenic, OD – ozone depletion, ECO – ecotoxicity, AA – aquatic acidification, TA – terrestrial acidification, EP – eutrophication potential, PO – photochemical oxidation)