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# Comparison of dysprosium production from different resources by life cycle assessment

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## ABSTRACT

Rare earth elements (REEs) are essential for low carbon technologies. Production of dysprosium (Dy) is mostly induced by rare earth magnets demand (approximately (approx.) 95% of total demand). It is almost exclusively supplied by ion adsorption clays (IACs) of Southern China. Other sources, such as bastnaesite/monazite or eudialyte ores, are also conceivable. Bastnaesite/monazite ores usually show low dysprosium contents. So far, hardly any REEs from eudialyte ores have been processed. The Norra Kärr deposit (Sweden) is one of the largest, highest grade, non-Chinese heavy REE deposits in Europe. Almost all studies on environmental effects of REEs production investigate the bastnaesite/monazite route. Recently, a first life cycle assessment (LCA) of IAC in-situ leaching was published. The present study broadens the scope firstly by including additional beneficiation and separation processes and subsequent production of the single metal dysprosium. Secondly, a comparison of the environmental performance of three production routes from different resources, IAC, bastnaesite/monazite and eudialyte is investigated. The results show that the environmental performance based on eudialyte is the best. The results of IAC and bastnaesite/monazite routes are comparable, but only for low amounts of leaching agent for IACs. For all three minerals freshwater ecotoxicity, human toxicity as well as eutrophication marine and freshwater are important environmental effects. In case of IAC marine eutrophication has the largest share due to in-situ leaching. This paper allows for the first time a straight comparison of Dy production based on three different minerals due to a consistent methodological frame, basic assumptions and parameters.

## 1. Introduction

Many rare earth elements (REEs) are used in low carbon technologies, today. Among them, especially the more valuable heavy rare earth elements (HREE, gadolinium – lutetium) are critical resources, including both the supply risks and the vulnerability of a system to a potential supply disruption (Erdmann and Graedel, 2011). Modern electrical generators and motors for hybrid and electric vehicles, wind turbine generators or hard disc drives require dysprosium (Dy). Nearly 95% of the total Dy demand accounts for the use in permanent magnets (Hoenderdaal et al., 2013).

Dysprosium, such as most HREEs, are currently almost exclusively supplied by ion adsorption clays (IACs) located in seven provinces in the south of China, distributed over numerous small deposits (Kanazawa and Kamitani, 2006; Yang et al., 2013). IACs are a result of lateritic weathering, predominantly of granites. Suffering strong chemical and biological weathering REEs were adsorbed mainly on the surface of clay minerals at ion state under warm and moist weather. IAC deposits account for approx. 35% of Chinese REE production (Papangelakis and Moldoveanu, 2014). Together with Chinas biggest REE mine in Bayan Obo (Inner Mongolia) they represent 80% of the world production of REEs (Schüler et al., 2011). In Bayan Obo a bastnaesite/monazite ore is mined with a mining rate of 1.0E + 07 ton per year. This deposit was formed by hydrothermal replacement of the carbonate rocks of sedimentary origin (Kanazawa and Kamitani, 2006). The ore bodies are different zones with iron rich cores. The main zones are riebeckite, fluorite and dolomite (Drew et al., 1990). Mainly light rare earth elements (LREEs) are processed but also a HRE oxide fraction is produced. As a third option to provide Dy a possible European production based on the mineral eudialyte has been proposed. Eudialyte is a rare, cyclosilicate mineral, which is found in 191 deposits (Friedrichs and Meyer, 2017; Mindat.org). Up to now virtually no eudialyte processing for REEs takes place. However, a huge advantage compared to the other RE minerals is the high share of HREEs (up to 50% of total rare earth oxide (TREO)) in combination with low radioactivity.

This study compares the environmental effects caused by the production routes corresponding to these three minerals, Chinese IACs, Chinese bastnaesite/monazite and Swedish eudialyte, focusing on the

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**Table 1**
Main parameters of the process chains compared.

| Parameter | Ion adsorption clay (IAC) | Bastnaesite/Monazite (B/M) | | Eudialyte |
|---|---|---|---|---|
| Location | China, southern provinces | China, Inner Mongolia, Bayan Obo/Baotou | | Sweden, south central, Norra Kärr |
| TREO in crude ore | 0.15% | 6.22% | | 0.59% |
| TREO in concentrate | Not applicable | Bastnaesite concentrate 55.6% | Monazite concentrate 34.1% | 2.7% |
| Beneficiation yield | – | 12.6% Zhang and Edwards (2013) | 6% Zhang and Edwards (2013) | 61.6% |
| Leaching yield | 67.5%[a] | 96.6% | | 96.6% |
| Solvent extraction yield | 97.2% | 97.2% | | 97.2% |
| HREE in REO | 23.9% | 1.2% | | 53.5% |
| Dy content in REO | 2.52% | 0.06% | | 5.26% |
| ton ore/ton REO[b] | approx. 1015 | approx. 220 | | approx. 290 |
| ton ore/ton Dy[b] | 40,873 | 265,000 | | 5472 |
| t ore/t Dy calculated by using economic allocation | 10,846 | 1230 | | 502 |
| $ThO_2$ concentration (ore) | 0.005% | 0.032% | | 0.0026% |
| $U_3O_8$ concentration (ore) | 0.005% | 0.002% | | 0.0018% |

[a] yield of in-situ leaching and precipitation with ammonium bicarbonate.
[b] the values are calculated based on ore concentration and losses along the process chains.

example of Dy. Several studies have approached the topic of environmental consequences related to REE production especially in China in a more general way (Haque et al., 2014; McLellan et al., 2013). For more detailed environmental evaluations of RE processing some life cycle assessment (LCA) studies have been conducted in the last few years (Adibi et al., 2014; Koltun and Tharumarajah, 2014; Sprecher et al., 2014; Zaimes et al., 2015). Almost all of them rely on old data (1990ies) from the Mountain Pass mine (U.S.) or on deduced data for Bayan Obo (Zhao and Navarro, 2014). Additionally, two LCAs considering REO production from IACs have been published recently (Lee and Wen, 2017; Vahidi et al., 2016).

For the first time our approach allows a comparison of the three RE process routes due to consistent frame conditions such as system boundaries, level of detail, inclusion of waste and waste water treatment or allocation procedures. The results are not meant for direct comparison on single processes level (e.g. dry digestion versus roasting), because the type of ores and the ore grades are different at all production sites. Therefore, the entire process chains as described in the following must be taken into account.

## 2. Dysprosium production

The three different minerals demand different processing procedures. From the beginning of the process chain up to the dissolved RE chloride the processing varies quite significantly. While the processing from IACs is described explicitly in the following, a detailed description of the production from bastnaesite/monazite or eudialyte has recently been published by the authors (Schreiber et al., 2016). Mining activities in Bayan Obo representing the bastnaesite/monazite mineral, beneficiation, cracking and separation in Baotou as well as tailings and sludge treatment are considered. As average value for the various ore bodies located in Bayan Obo a total REO content of 6.22% in the bastnaesite/monazite ore with a share of 0.06% Dy is assumed. Based on a prefeasibility study for the Swedish eudialyte deposit in Norra Kärr (GBM, 2015) a REO content of 0.59% in the ore with a share of 5.3% Dy is assumed. Table 1 presents the most important mineral and process parameters of the three process chains. In the following, the process chains for ion adsorption clays and bastnaesite/monazite are abbreviated by IAC and B/M.

The REE concentration and the share of elements in the IACs is calculated following a study analysing four Chinese IAC deposits (Bao and Zhao, 2008). The main components are silica and aluminium oxide. The concentration of HREEs is much higher than in B/M ores. Table 2 shows the assumed compositions for all three minerals.

Although the grade of REEs in IAC ores seems low, the ion state of REEs makes extraction and processing easier. While B/M and eudialyte use a classical mining and beneficiation route, for IACs in-situ leaching has become the major technology since 2011, replacing heap leaching and tank leaching due to its better environmental performance (Yang et al., 2013; Zhao and Navarro, 2014). Therefore, this technology is chosen as representative for the study. An overview of the analysed IAC process chain compared to Dy production from the other two minerals is given in Fig. 1. The conventional mining and beneficiation processes (crushing, grinding, separation and flotation) are missing in case of IAC. In contrast to the other two routes, the IAC process chain starts directly with in-situ leaching by ammonium sulphate. The B/M ore from Bayan Obo requires a roasting process using sulphuric acid and the eudialyte mineral is cracked by a dry digestion using hydrochloric acid (Voßenkaul et al., 2017). The RE sulphates from B/M or IAC are converted to RE carbonates by reaction with ammonium bicarbonate before they are converted to RE chlorides by leaching with hydrochloric acid. Further processing of RE carbonates takes place in the same way for all three routes (Fig. 1).

The processes are grouped in the three sections RE concentrate, Dy oxide and Dy metal representing major products along the value chain.

### *2.1. In-situ leaching of IAC and precipitation by ammonium bicarbonate*

Initial to in-situ leaching, injection wells are drilled with an assumed diameter of 0.8 m, a depth of 1.5–3 m and a distance between each leaching hole of 2–3 m (Yang et al., 2013). Then, a leaching solution, using ammonia sulphate as leaching agent, is pumped through

**Table 2**
Composition of rare earth elements of the different resources (%).

| | La | Ce | Pr | Nd | Sm | Eu | Gd | Tb | Dy | Ho | Er | Tm | Yb | Lu | Y |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| IAC | 29.8 | 13.8 | 5.9 | 22.4 | 3.8 | 0.35 | 3.14 | 0.45 | 2.5 | 0.5 | 1.4 | 0.2 | 1.4 | 0.2 | 14.1 |
| B/M | 24.5 | 49.3 | 5.8 | 17.8 | 0.9 | 0.2 | 0.7 | 0.08 | 0.06 | – | – | – | – | – | 0.1 |
| Eudialyte | 8.9 | 20.2 | 2.7 | 11.2 | 3.1 | 0.4 | 3.6 | 0.7 | 5.3 | 1.2 | 3.7 | 0.5 | 3.4 | 0.5 | 34.6 |

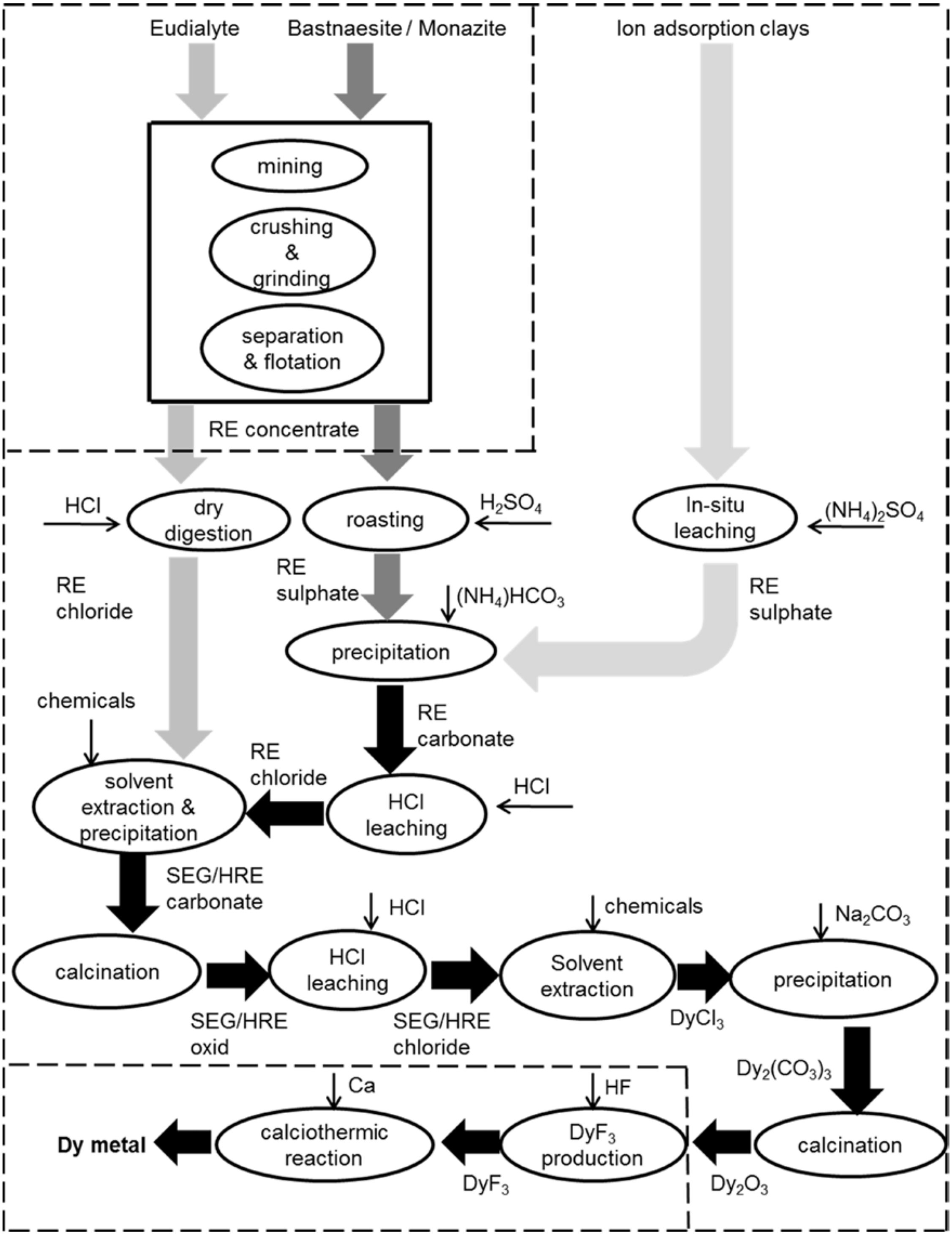

**Fig. 1.** Process chains of Dy production from IAC, B/M and eudialyte.

the wells into the clay layer (Papangelakis and Moldoveanu, 2012). There, an ion exchange takes place replacing RE ions by ammonia ions at the surface of the clay layer. The RE ions dissolute into the leaching solution (Yang et al., 2013). The ammonia sulphate amount necessary is calculated by assuming an average concentration of leaching solution (4%) (Yang et al., 2013) and a REE concentration in the filtrate of 0.5 g/l (Chi and Tian, 2008). The stoichiometric excess corresponds to approx. 60 resulting in a yield of 75% (Papangelakis and Moldoveanu, 2014). This is verified by in-situ leaching experiments using ammonia sulphate (Papangelakis and Moldoveanu, 2013). By addition of a fourfold stoichiometric excess of ammonia bicarbonate the RE carbonates are precipitated. The residual solution is recycled and can be used again for in-situ leaching after pH adjustment to 6 via sulphuric acid (Chi and Tian, 2008). The used ammonia ions are replenished by addition of an appropriate amount of ammonia sulphate. By increasing the pH value of the residual solution to 10 via calcium oxide impurities (e.g. Al, Mg, Si) are separated. The ammonia sulphate necessary is transported 320 km from the port of Guangzhou to the deposits. The energy demand of in-situ leaching is assumed to be 0.45 kWh/t ore deduced from a copper leaching process (Martens et al., 2003).

The amount of ammonia sulphate necessary is highly contentious. Since it is a very sensitive parameter two further estimations are discussed. The above described very likely estimation of 4% ammonium sulphate solution and 0.5 g REE/l filtrate solution represents the high estimation, representing the high amount of ammonium sulphate used. This corresponds to approx. 80.6 kg ammonia sulphate/kg REO. Furthermore, a middle (3% ammonium sulphate solution and 1.25 g REE/l filtrate solution; 24 kg ammonia sulphate/kg REO) and a low estimation (2% ammonium sulphate solution and 2 g REE/l filtrate solution; 9.9 kg ammonia sulphate/kg REO) are calculated also. The low estimation represents a nearly stoichiometric ammonia sulphate amount corresponding with the value of 10.4 kg (in case of their high estimation) stated in Vahidi et al. (2016) and Lee and Wen (2017), recently. However, this 6.1 kg pointed out in Vahidi et al. (2016) seems highly optimistic. Two aspects oppose the low values. Firstly, the high ammonium sulphate contamination of 3500–4000 mg/l in the groundwater is a strong evidence for a high stoichiometric excess during in-situ leaching (Yang et al., 2013). Secondly, approx. 40% of Chinese rare earth output is produced by illegal mining (Packey and Kingsnorth, 2016) with certainly no elaborated process management. The amount of energy, water and drilling mud are not varied for the three estimations. Different ammonia emissions into water and soil occur caused by different amounts of leaching solution. Table 3 shows the main inputs and the ammonia emissions of the first three processes of the IAC process chain. The ammonia emissions range between 2.4–21.7 kg/kg REO depending on the three estimations. The lowest value is similar to the high estimation (3.5 kg/kg REO) in Vahidi et al. (2016) due to the similar value of ammonium sulphate input for in-situ leaching.

## 2.2. Extraction and metal production

Since the single IAC deposits are small and spread over three provinces (Hunan, Jiangxi, Guizhau) the REE carbonate has to be transported 200 km (averaged distance) by lorry to a central processing plant operated by NFC Southern Rare Earth (Xinfeng) Co., Ltd. in Shaoguan (Province Guangdong) (USGS, 2013). It is also assumed that all chemicals have to be transported 230 km by lorry from the port of Guangzhou to this processing plant in Shaoguan. At first, the RE carbonate precipitate is converted to RE chloride solution by addition of hydrochloric acid and water. Then, a first solvent extraction (SX) process to separate the LREEs (raffinate) is carried out followed by RE carbonate precipitation via adding of soda. Afterwards, a thermal decomposition of the carbonates, leaching with hydrochloric acid and further solvent extractions to separate Dy from the other HREEs follow. The extraction process calculated and the amounts of chemicals used are based on industrial extraction processes of Lynas Corporation Ltd. in Malaysia (DNV, 2010) and are adapted to the REE composition in the IACs. As extraction agents amines and phosphoric acid substituted by organic ligands are used. The total energy demand is estimated to be approx. 6 kWh/kg Dy and allocated to the single extraction steps quantitatively. The subsequent precipitation is carried out using soda again. The resulting Dy carbonates are then calcined in a tunnel furnace at 900 °C. The produced Dy oxide reacts with ammonia and hydrofluoric acid and forms Dy fluoride ($DyF_3$). Finally, the production of Dy metal is carried out by reduction of $DyF_3$ with calcium. The amount of Ca and the composition of slurry is based on literature (Sharma, 1994; Velu and Reddy, 2005).

## 2.3. Treatment of waste, sludge and waste water

Since the groundwater in the mining area is highly polluted by ammonia sulphate (3500–4000 mg/l) (Yang et al., 2013) it is assumed that 91 t ammonia sulphate/t REO remains in soil after termination of mining. Furthermore, it is assumed that the mud produced during drilling of the injection wells as well as emissions occurring during recycling of leaching solution are dumped at the mining site. Due to heavy rainfall in this region the water contained in the sludge including dissolved pollutions is drained into the soil. Particles solubility is mostly assumed to 0.05%, except for silica (1%) and cellulose (100%) contained in the drilling mud.

Due to the lack of information about the waste water treatment at the processing plant in Shaoguan the same treatment processes as of Lynas Corporation Ltd. in Malaysia are assumed (Lynas, 2008) and

**Table 3**
Main inputs of the first three single processes of the IAC process chain related to 1 kg REO.

| Inputs per kg REO | Unit | Mass | Source/comment |
|---|---|---|---|
| In-situ leaching: | | | |
| $(NH_4)_2SO_4$ | kg | 80.6 high | calculation is based on concentration of leaching solution Yang et al. (2013) and REE content Chi and Tian (2008) |
| | kg | 24.0 middle | |
| | kg | 9.9 low | |
| Energy | kWh | 0.463 | analog to copper leaching (Martens et al., 2003) |
| Drilling mud | | | calculation is based on 7000 $m^3$/ha drilling mud (Yang et al., 2013) |
| Water | kg | 35.1 | composition drilling fluid is based on GWE (2017) |
| Bentonite | kg | 0.70 | |
| Cellulose | kg | 0.05 | |
| Water | t | 1.935 | calculation is based on 0.5 g REE concentration per l filtrate Chi and Tian (2008); the rest is water |
| Land Use | $m^2$ | 0.022 | calculation is based on hole diameter, distance between the holes Yang et al. (2013), REE concentration (Bao and Zhao, (2008) and yield Papangelakis and Moldoveanu (2014). |
| Transport | tkm | 0.0257 | 320 km by lorry (transport distance between mining area and Guangzhou) |
| Filtration and precipitation with ammonia bicarbonate: | | | |
| $(NH_4)HCO_3$ | kg | 1.02 | stoichiometric calculation with 25% excess |
| CaO | kg | 0.56 | calculated demand for pH-value adjustment |
| $H_2SO_4$ | kg | 0.20 | calculated demand for pH-value adjustment |
| Energy | kWh | 0.084 | rough estimation, based on solvent extraction |
| Transport | tkm | 0.569 | 320 km by lorry (see in-situ leaching) |
| *HCl leaching* | | | |
| HCl (31%) | kg | 2.13 | calculation analog to DNV (2010) |
| Water | kg | 36.7 | calculation analog to DNV (2010) |
| Transport | tkm | 9.2E-04 | 200 km by lorry for RE carbonate (distance between mining area and processing plant (Shaoguan); 230 km by lorry for HCl (distance between port of Guangzhou and processing plant in Shaoguan) |
| Outputs | | | |
| $NH_4$ | kg | 21.7 high | calculation is based on the amounts of $(NH_4)_2SO_4$ and $(NH_4)HCO_3$ used for in-situ leaching and precipitation assuming that the chemicals will remain in the soil after termination of the mining operation |
| | | 6.2 middle | |
| | | 2.4 low | |

adjusted to the output of each solvent extraction step. Also the leakage rate according to different construction of tailing ponds and storage sites (e.g. liner and drainage systems) are considered differently.

## 3. Methodology

### 3.1. Goal and scope

The goal of this investigation is to compare environmental impacts related to the production of 1 kg Dy (functional unit) from IAC (Southern China), from B/M ore (Bayan Obo), and from an eudialyte deposit (Norra Kärr) using the LCA method (International Organization for Standardization (ISO), 2006a; International Organization for Standardization (ISO), 2006b). Thereby we provide harmonized data for LCA practitioners from already existing IAC and B/M production pathways as well as possible future supply from eudialyte. The specific data for Dy metals are necessary to analyse low carbon technologies such as magnets for wind turbines. Up to now this was not possible because the few existing assessments to Dy ended at REO level.

Data origin for IAC processing is described in chapter 2 and for B/M and eudialyte in Schreiber et al. (2016) and Voßenkaul et al. (2015). Most of upstream process data (e.g. transport, energy supply, auxiliary material) are taken from ecoinvent 3.3 database (Ecoinvent, 2016) and GaBi6.115 (thinkstep, 2012). It has to be kept in mind, that IAC and B/M represent existing practices, while data for eudialyte is taken from a prefeasibility study (GBM, 2015) for mining and up scaled laboratory data for the following processes. The time frame includes data from 2010 up to today for the main process chains.

The selection of impact categories for the Life Cycle Impact Assessment (LCIA) is based on the often used ReCiPe 1.08 (H) midpoint (Goedkoop et al., 2009) as implemented into GaBi6 (thinkstep, 2012) and listed in Table 4. It addresses numerous environmental effects discussed in comparable REE studies. Water depletion as well as land transformation and occupation are excluded from the analysis, because data availability is too poor for these categories. Furthermore, the evaluation of metal depletion is omitted since the ReCiPe method considers only a few metals and has a high overestimation of manganese (Peters and Weil, 2016). No environmental effect is prioritized in its importance, so equal weighing between impact categories is considered.

Although normalisation is no mandatory part of LCA, it helps to gain a better understanding of the contribution of the process chains to the world-wide environmental effects. Each environmental effect is benchmarked against the contribution of a single person to this impact. The so-called 'ReCiPe person equivalent (PE) world' means the average annual contributing of a person to a world-wide environmental impact. So for every impact category the relative contribution due to Dy production is determined.

**Table 4**
Overview of ReCiPe 1.08 midpoint (H) impact categories considered.

| Impact Category | Unit |
|---|---|
| Climate change, incl. biogenic carbon | kg $CO_2$-eqv. |
| Fossil depletion | kg oil-eqv. |
| Freshwater ecotoxicity | kg 1,4-DB-eqv.[a] |
| Freshwater eutrophication | kg P-eqv. |
| Human toxicity | kg 1,4-DB-eqv. |
| Ionising radiation | kg U235-eqv. |
| Marine eutrophication | kg N-eqv. |
| Ozone depletion | kg CFC − 11-eqv.[b] |
| Particulate matter formation | kg $PM_{10}$-eqv. |
| Photochemical oxidant formation | kg NMVOC-eqv.[c] |
| Terrestrial acidification | kg $SO_2$-eqv. |
| Terrestrial ecotoxicity | kg 1,4-DB-eqv. |

[a] 1,4-DB: 1,4-dichlorobenzene.
[b] CFC-11: trichlorofluoromethane.
[c] NMVOC: non-methane volatile organic compound.

**Table 5**
Allocation factors used for products separated during solvent extraction steps.

| Extraction step | Raffinate | Extract | Price in $/kg | Allocation factor in% | | |
|---|---|---|---|---|---|---|
| | | | | IAC | B/M | Eudialyte |
| 1 | $LREEs_{IAC}$ | | 25.47[a] | 66.7 | | |
| | $LREEs_{B/M}$ | | 17.18[a] | | 95.4 | |
| | $LREEs_{Eudialyte}$ | | 21.94[a] | | | 29.5 |
| | | HREE | 40.87[b] | 33.3 | 4.6 | 70.5 |
| 2 | Y/Ho/Lu | | 40.87[b] | 52.4 | 5.6 | 61.1 |
| | | Sm/Tb/ $Dy_{IAC}$ | 72.87[c] | 47.6 | | |
| | | Sm/Tb/ $Dy_{B/M}$ | 36.16[c] | | 94.4 | |
| | | Sm/Tb/ $Dy_{Eudialyte}$ | 105.6[c] | | | 38.9 |
| 3 | Sm/Tb | | 30.41[c] | 35.4 | 82.4 | 21.1 |
| | | Dy | 310.15[d] | 64.6 | 17.6 | 78.9 |

[a] basket price of light rare earths (LREEs) is calculated based on annual output at the different sites and unit prices of La, Ce, Pr, Nd (Asian Metals, 2015).
[b] price of Y/Eu oxide mixture (Asian Metals, 2015) is assumed for HREE mixture and for yttrium/holmium/lutetium carbonate.
[c] the price of samarium/europium/gadolinium oxide mixture (Asian Metals, 2015) is assumed for Sm/Tb/Dy mixture and for samarium/terbium carbonate.
[d] price of dysprosium oxide (Asian Metals, 2015).

Since the production of REE is a multi-output product system the environmental burdens have to be allocated appropriately. In this study, an allocation method based on the mass of produced REEs combined with their market prices (based on an international metals market analysis and pricing index company (Asian Metals, 2015)) are considered, as usual for metals with strongly different prices. As the ores have different compositions, the allocation factor has to be assessed individually for each site. Table 5 shows underlying prices and the resulting allocation factors for the raffiante (separated fraction, which leaves the process chain and is not considered further in this study) and the extract (which is passed to the next extraction step), calculated for each solvent extraction step. Since the share of heavy REE in eudialyte is the highest, the resulting allocation factor for the separated light fraction of REE at the first solvent extraction step (separation of light and heavy rare earth fraction) is the lowest (29.5%). In case of B/M the allocation factor for the extracted HREE fraction is 4.6%, while for the IAC it is 33.3%. At each SX step the allocation factors sum up to 100%.

To consider the quality of the data each single process in the process chains is assigned with a data quality indicator (DQ) between DQ1–DQ5 according to the definition in Table 6, whereat DQ1 is the best quality (measured data) and DQ5 expresses the worst quality (rough estimations). The different data DQ levels represent different data deviations deduced from cost data deviation for mining processes (Bull et al., 2012). In Table 6 deviation levels are given as well as the assignment of the processes to the appropriate DQ levels. During the calculation of impacts, the DQ categories assigned per processes are

**Table 6**
Data quality levels.

| DQ | Definition | Deviation [%] | Example |
|---|---|---|---|
| 1 | measured | −10/+15 | not used |
| 2 | specific references | −15/+20 | Chinese electricity generation mix |
| 3 | calculated, modelled or up scaled | −20/+30 | calcination of carbonates, Chinese ammonium bicarbonate production |
| 4 | estimated data based on similar processes | −30/+50 | most of chemical production and transportation processes |
| 5 | roughly estimated data | −50/+100 | sludge and waste water treatment processes |

**Table 7**
Selected main inputs and outputs per kg Dy.

| | Unit | IAC high | IAC middle | IAC low | B/M[a] | B/M sensitive | Eudialyte[a] |
|---|---|---|---|---|---|---|---|
| Input[b] | | | | | | | |
| Raw ore[c] | kg | 10,846 | 4300 | 2664 | 1226 | 1226 | 1361 |
| Primary energy resources | MJ | 3798 | 2122 | 1688 | 3707 | 2976 | 4075 |
| Transport service, lorry | tkm | 172 | 73 | 50.5 | 15 | 14 | 45 |
| Transport service, rail | tkm | 2.0 | 1.8 | 1.8 | 204 | 204 | – |
| Process water | kg | 157 | 157 | 157 | 136 | 136 | 784 |
| Lubricating oil | g | – | – | – | 38 | 38 | 36 |
| Caustic soda solution (50%) | kg | 18 | 18 | 18 | 16 | 16 | 20 |
| Hydrochloric acid (32%) | kg | 64 | 64 | 64 | 82 | 82 | 310 |
| Sulphuric acid (96%) | kg | 1.1 | 0.4 | 0.3 | 69.9 | 69.9 | 3.1 |
| Soda | kg | 3.3 | 3.3 | 3.2 | 2.4 | 2.4 | 4.3 |
| Oxalic acid | kg | 0.03 | 0.03 | 0.03 | 0.30 | 0.30 | 2.13 |
| Ammonium sulphate, as N | kg | 91.2 | 27.0 | 11.1 | – | – | – |
| Ammonium bicarbonate | kg | 5.5 | 5.5 | 5.5 | 27.9 | 27.9 | – |
| Ammonia | kg | 1.7 | 1.7 | 1.7 | 6.8 | 6.8 | 0.4 |
| Calcium chloride | kg | 0.9 | 0.9 | 0.9 | 0.9 | 0.9 | 0.9 |
| Calcium | kg | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 |
| Hydrogen fluoride | kg | 0.98 | 0.98 | 0.98 | 0.98 | 0.98 | 0.98 |
| Lime | kg | 17 | 15 | 15 | 38 | 38 | 38 |
| Steel | kg | – | – | – | 1.4 | 1.4 | 2.3 |
| Magnesium oxide | kg | 6.7 | 6.7 | 6.7 | 0.9 | 0.9 | 9.4 |
| Sodium phosphate | kg | – | – | – | 55 | – | 2 |
| Sodium silicate | kg | – | – | – | 0.8 | 0.8 | – |
| Phosphoric acid | kg | 0.1 | 0.1 | 0.1 | 0.2 | 0.2 | 0.4 |
| Kerosene | kg | 0.75 | 0.75 | 0.75 | 0.6 | 0.6 | 0.8 |
| Diesel | kg | – | – | – | 1.1 | 1.1 | – |
| Other inorganic chemicals | kg | 7 | 7 | 7 | 73 | 31 | – |
| Other organic chemicals | kg | 0.3 | 0.3 | 0.3 | 0.5 | 0.5 | 0.4 |
| Output[b] | | | | | | | |
| Ammonia (air) | kg | 1.0 | 0.99 | 0.99 | 1.08 | 1.03 | 0.11 |
| HF (air) | kg | 1.3 | 1.3 | 1.3 | 1.3 | 1.3 | 0.054 |
| $CO_2$ (air) | kg | 581 | 351 | 295 | 709 | 549 | 522 |
| CO (air) | kg | 0.9 | 0.6 | 0.5 | 0.7 | 0.7 | 0.6 |
| $SO_2$ (air) | kg | 2.6 | 1.4 | 1.1 | 6.5 | 3.6 | 0.7 |
| $NO_x$ (air) | kg | 1.3 | 0.8 | 0.6 | 2.3 | 1.9 | 1.1 |
| NMVOC (air) | kg | 0.5 | 0.3 | 0.25 | 0.9 | 0.9 | 0.2 |
| Methane (air) | kg | 1.94 | 1.23 | 1.06 | 2.63 | 2.5 | 0.86 |
| Particles into air | kg | 1.9 | 0.8 | 0.7 | 15.8 | 12.7 | 3.6 |
| Radioactive emissions into air | g | 4 | 2 | 1 | 8 | 4 | 0.003 |
| Heavy metal into air | g | 17 | 8 | 6 | 10 | 8 | 2 |
| Heavy metal into water | kg | 0.07 | 0.05 | 0.05 | 0.77 | 0.67 | 0.26 |
| Inorganic emissions into water | kg | 911 | 297 | 145 | 74 | 54 | 30 |
| Ammonium (total N, water) | kg | 167 | 34 | 13 | 0.4 | 14 | $2*10^{-10}$ |
| Organic emissions into water | kg | 0.13 | 0.09 | 0.08 | 0.40 | 0.17 | 0.16 |
| Emissions into salt water | kg | 19 | 14 | 13 | 41 | 21 | 451 |

[a] values are taken from the former publication of the authors (Schreiber et al., 2016).
[b] nputs and outputs of the whole process chains related to 1 kg Dy are calculated using the ecobalance software GaBi6.115.
[c] allocated values; in case of IAC the value is the amount of leaching solution obtained after in-situ leaching.

included in the computation so that consequentially deviations for each impact category follow out of this. DQ1 was not given to any process, because no measurements were conducted at the three production sites. A list of the assigned DQ to each process step can be found in the supplementary material (Table S 24).

### 3.2. *Life cycle inventory of Dy production*

In the Life Cycle inventory (LCI) the main energy and material inputs and outputs are summed up. For comparison six process chains are considered, representing the three mineral types and additional variations in process parameters. For IACs, as mentioned in Section 2, the amount of ammonia sulphate is highly contentious, therefore, three different amounts are assumed, resulting in IAC low, middle and high estimations. For B/M processing it is unclear whether the waste water coming from the carbonate precipitation (after roasting) is actually purified by struvite precipitation in Baotou. A scenario without struvite precipitation has been created (B/M sensitive), yielding in a much lower amount of inorganic chemicals (magnesium and phosphorous components) needed. The six cases considered are:

- IAC high, 4% ammonium sulphate solution, 0.5 g REE/l filtrate, 80.6 kg ammonia sulphate/kg REO
- IAC middle, 3% ammonium sulphate solution, 1.25 g REE/l filtrate, 24 kg ammonia sulphate/kg REO
- IAC low, 2% ammonium sulphate solution, 2.0 g REE/l filtrate, 9.9 kg ammonia sulphate/kg REO
- B/M
- B/M sensitive, without struvite precipitation
- Eudialyte

Table 7 shows the main inputs and outputs related to the production of 1 kg Dy unit for the six process chains. The values for the production sites differ regarding the amounts and types of material. For example, the Dy production based on eudialyte uses the highest amount of energy resources, process water, caustic soda and hydrochloric acid due to the highest amount of raw ore mined required for 1 kg Dy. On the other hand most of the emissions released have their highest values for B/M (e.g. radioactive emissions into air, heavy metals and organic emissions into water) due to poor process control as well as hardly existing waste and sludge treatment. Because of the roasting process the facilities for

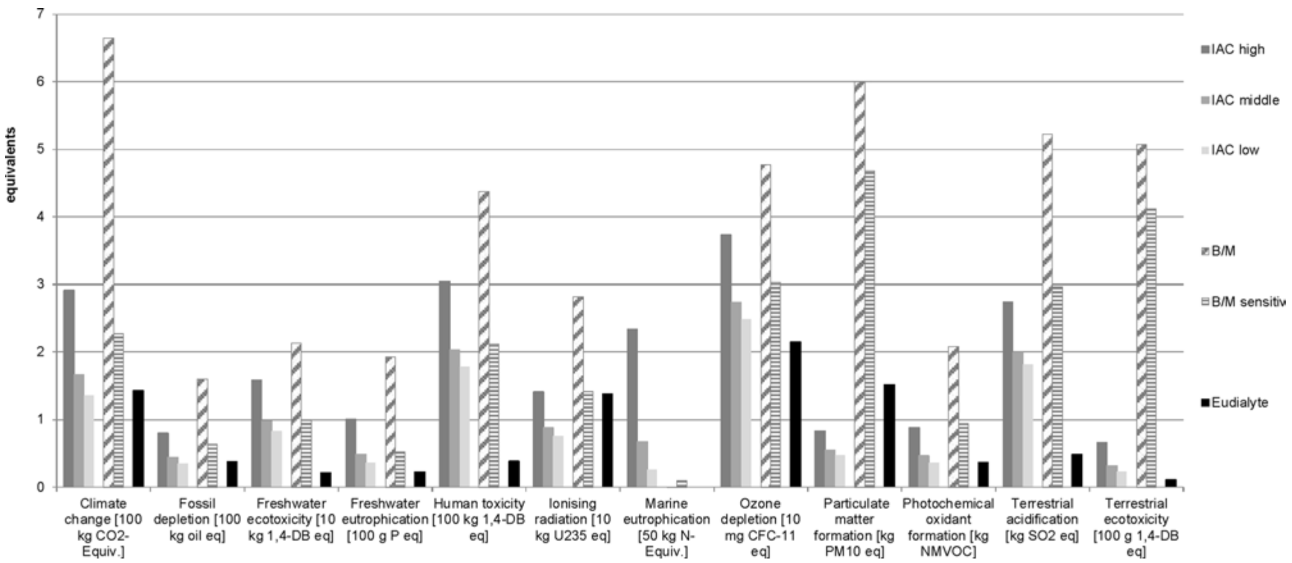

**Fig. 2.** Environmental impacts of process chains.

B/M use high amounts of sulphuric acid. Hydrochloric acid is the main input for the eudialyte process chain due to the dry digestion instead of roasting. In case of IACs almost no acid is used, but ammonium sulphate for in-situ leaching. Because mining, crushing and grinding are missing for IAC, no lubricating oil and steel (abrasion in mills) are considered. For eudialyte processing no ammonium bicarbonate is necessary because a RE chloride solution is obtained after dry digestion which can enter the solvent extraction directly. Hence, the precipitation with ammonium bicarbonate and the subsequent HCl leaching are obsolete. Ammonia, hydrogen fluoride, calcium chloride and calcium are mainly used for the production of $DyF_3$ and the calciothermic reaction to Dy metal. Therefore the amounts of these chemicals are identical for all process chains. A detailed description of inventory data for each process step can be found in the supplementary material.

## 4. Results and discussion

The various environmental impacts during Dy production are summarised in Fig. 2 for all six cases. For each impact category the total equivalents representing the category for the entire process chain are given. The Dy production based on eudialyte is superior in seven out of twelve impact categories (marine and freshwater eutrophication, human toxicity, ozone depletion, terrestrial acidification, freshwater and terrestrial ecotoxicity). In case of climate change, fossil depletion and photochemical oxidant formation the values are comparable to Dy production from IAC low estimation case. Except for marine eutrophication Dy production from B/M has the highest environmental impacts due to the lowest amount of Dy content in the ore. Further reasons are the lower environmental standards presumed for China (MEP, 2011) and the assumed waste water treatment by struvite precipitation using ammonium bicarbonate (following roasting, Fig. 1). The impact of a possible waste water treatment becomes obvious by comparing B/M and B/M sensitive. In case of B/M sensitive (without struvite precipitation) only five out of twelve impacts categories hold the highest values. Due to resulting ammonium emissions into water in case of no waste water treatment, marine eutrophication for B/M sensitive becomes worse than for B/M. This emphasizes the effect that an improvement of marine eutrophication by introduction of waste water treatment results in a considerable increase (sometimes more than twice) in all other environmental categories. However, the highest marine eutrophication values are obtained for all three IAC estimations due to the high amount of ammonium entry into soil and water during in-situ ammonium sulphate leaching. Furthermore, considerable differences due to the different assumed ammonium sulphate amounts can be found for all impact categories. Assuming a high demand of ammonium sulphate as verified in the experiments (IAC high) induces higher effects than for B/M sensitive in eight categories, while the unlikely stoichiometric amount equalises IACs even to the low eudialyte impacts in many cases.

In order to describe the overall environmental effect and understand the contribution to each impact, the different impacts are aggregated. To allow adding of different effects, expressed in different units, each environmental effect is made dimensionless in the normalisation step as described in Section 3.1, by comparing it to the yearly contribution of a single person to this impact. Fig. 3 shows the summarised results for each production chain in terms of normalised impacts in person equivalents related to the production of 1 kg Dy. Thus, the significance of the process chains contribution to the different environmental impacts becomes visible. The impact categories with the highest contributions are freshwater ecotoxicity, human toxicity, eutrophication marine and freshwater for all production chains. For eudialyte and B/M freshwater ecotoxicity is the category with a contribution share of almost 50%, followed by human toxicity (approx. 30%–35%). Due to the high importance of in-situ leaching in case of IAC (high and middle) the marine eutrophication has the largest impact with 70% and 51%, respectively.

Unfortunately, the data quality of these impact categories is rather poor especially for IAC and B/M (mostly DQ4 and DQ5) as indicated in Fig. 4, because these impacts are predominantly based on uncertain Chinese inventory data.

Fig. 4 presents the data qualities for the impact categories of each process chain derived from the number off processes contributing to this impact. Overall, the different impact categories show no consistent pattern regarding data quality. The impacts of the eudialyte based process chains are mostly derived from processes with DQ2 and DQ3 whereas the impacts of (both) B/M process chains strongly originate from processes with DQ5 and the (three) IAC processes are mostly evaluated with DQ4. The results for the European production are associated with a lower deviation than the results of B/M and IAC. Reason is on the one hand experimentally obtained data for eudialyte processing acquired by Aachen University (Stark et al., 2017; Voßenkaul et al., 2017). On the other hand, better matching data are available for European supply processes (e.g. for chemicals) than for China from the databases. For the overall process chains, the following averaged deviations are calculated: IAC −29% to +49%, B/M −39% to +74%, eudialyte −25% to +41%. The corresponding deviations are shown in Fig. 3.

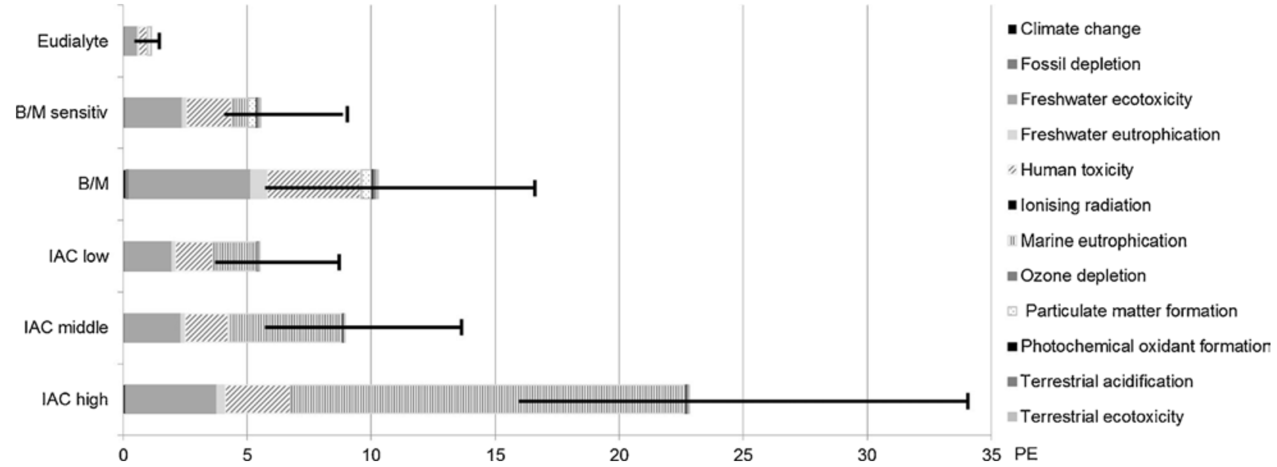

**Fig. 3.** Normalised impacts of process chains in person equivalents per kg Dy with deviation.

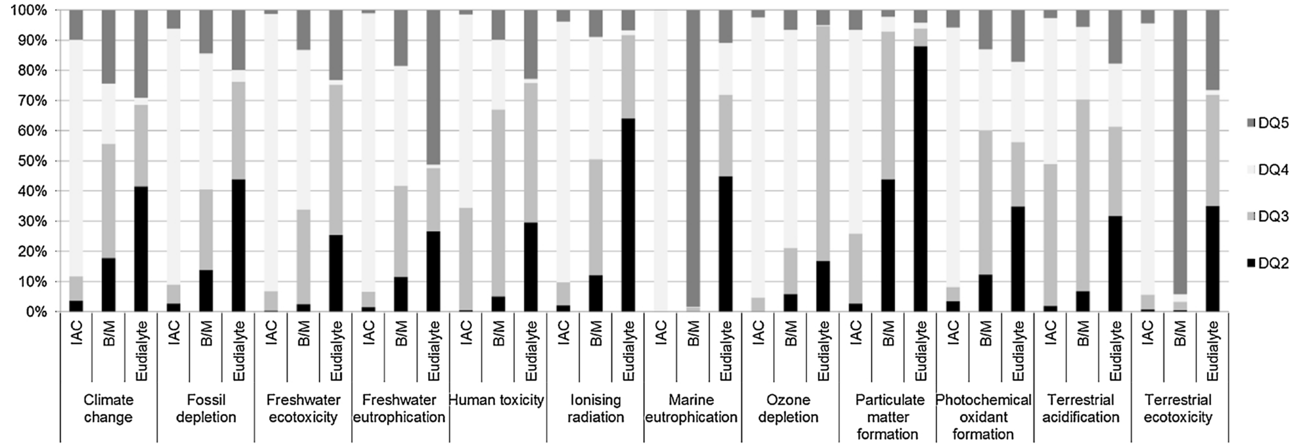

**Fig. 4.** Data qualities of the environmental impacts selected.

To assess which part of the process chain contributes to which environmental effect the processes are grouped into three sections (RE concentrate, Dy oxide, Dy metal) representing major products in the value chain as indicated in Fig. 1. The section “RE concentrate” includes all processes from mining to flotation and “Dy metal” contains the last two processes of the process chain, namely $DyF_3$ production and calciothermic reaction. The section “Dy oxide” comprises all processes between “RE concentrate” and “Dy metal”. The IAC process chains directly start with the section “Dy oxide”, because no concentrate is produced as intermediate. In Fig. 5 contributions of the three blocks are shown for selected impact categories. The share of the “Dy oxide” section is most often the highest due to the high number of processes associated with a variety of chemicals. In case of human toxicity the section “Dy metal” is also visible because of hydrogen fluoride emissions from $DyF_3$ production. Particulate matter is mostly caused by dust emissions from crushing and grinding. Hence, the section “RE concentrate” is the highest for both B/M and eudialyte.

Beside the main production processes shown in Fig. 1 also supply of operating materials or energy and waste or waste water treatment are considered in an LCA. While producers can influence the production technology of the main chain directly, their potential of direct intervention in the supply chains is limited. Exemplarily, in Fig. 6 the processes are distinguished between main RE production processes and upstream (supply of chemicals, energy and transport) or downstream (waste water treatment) processes for each mineral type. For IACs the high estimation case and for B/M the sensitive scenario are considered. Chemicals needed for the various beneficiations and separation steps dominate most impact categories especially in the IAC and eudialyte process chains. The share of energy supply as well as transport and infrastructure rarely exceeds 10%, except for ionising radiation of eudialyte processing (approx. 45%). Reason is the high share of nuclear power in the Swedish electricity grid mix. The high share of the main process chain (98%) on the total marine eutrophication in case of B/M sensitive is caused by ammonium emissions during precipitation with ammonium bicarbonate. In case of eudialyte 37% on the total freshwater eutrophication are caused by phosphate emissions during waste water treatment after flotation. Apart from that the share of waste water treatment processes is mostly < 5%. The contributions of the main processes are often insignificant. Only for marine eutrophication (IACs) and particulate matters (B/M and eudialyte) the direct REE beneficiation processes are crucial. The eutrophication is caused by ammonium emissions during in-situ leaching and the particulate matters result from dust during mining, crushing and grinding.

LCA identifies those processes along the complete production chain, which contribute most to the different impacts. Using the example of IACs (high estimation), the process chain is separated into single processes (Fig. 7). The in-situ leaching with ammonium sulphate is the main contributor to almost all impact categories with exception of terrestrial acidification. It must be noted, that 78% − 95.5% of the total in-situ leaching impacts are generated during the upstream ammonium sulphate production. Only marine eutrophication is caused by emissions of in-situ leaching to 99.9% at the mineral deposit directly. The high acidification value is mainly caused by ammonia emissions into air during $DyF_3$ production. A slightly smaller proportion arises from sulphur dioxide emissions during ammonium sulphate production. Second ranked are often the solvent extraction processes except for human

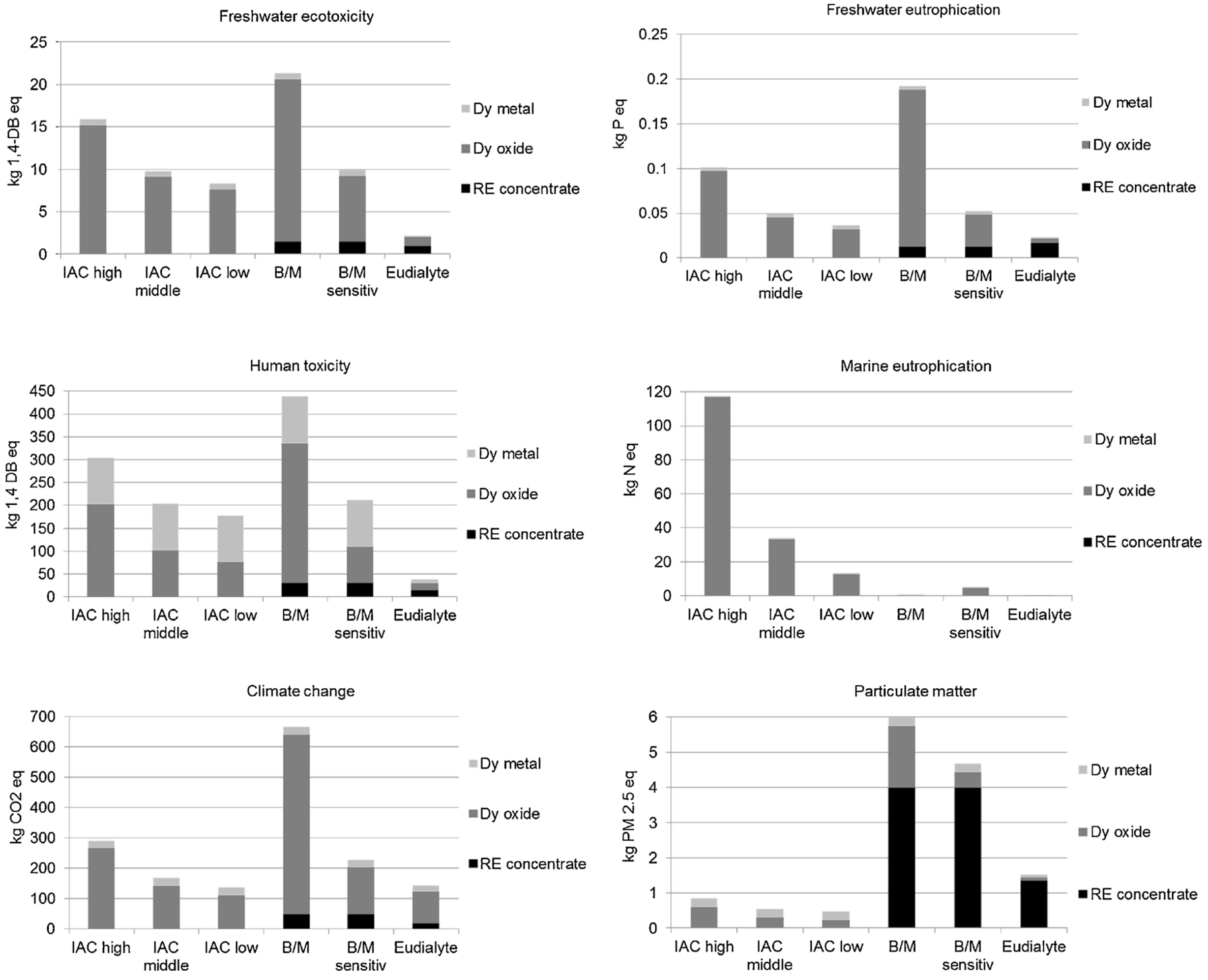

**Fig. 5.** Contribution of process chain sections to selected impact categories.

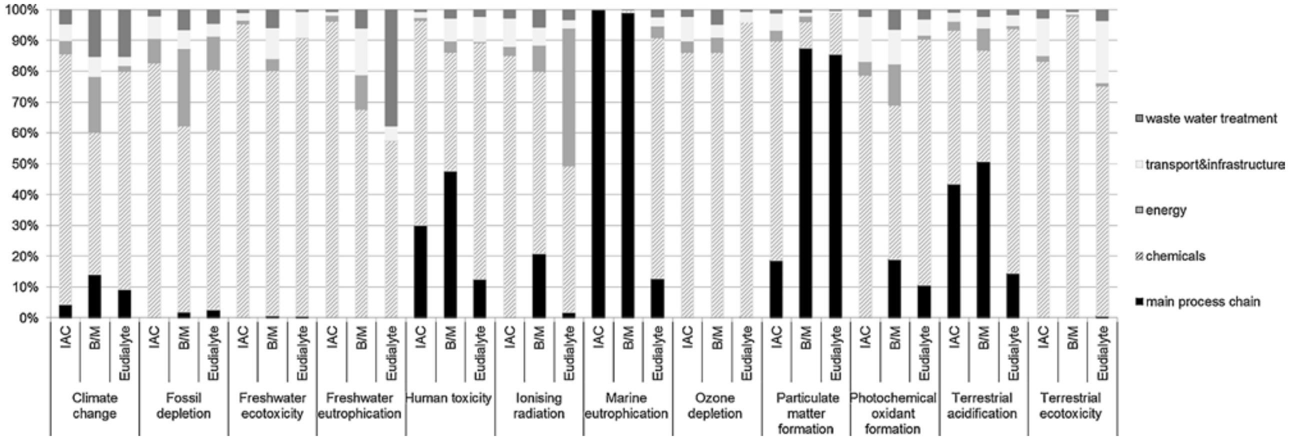

**Fig. 6.** Share of main process chain and up/downstream processes on the total environmental impacts.

toxicity, acidification and particulate matter. In case of human toxicity and particulate matter $DyF_3$ production has a higher share due to hydrogen fluoride and ammonia emissions into air, respectively. All other single processes have only minor importance.

## 5. Comparison with other LCA studies

To classify the results obtained into the existing discussion about environmental effects caused by REE production they have to be adapted in a comparable form. Therefore, the LCIA is also performed by TRACI (Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts) developed by the United States Environmental Protection Agency (EPA, 2001). Additionally, they are adjusted to the production of 1 kg REO (Table 8) to be comparable to IAC values published by Vahidi et al. (2016).

The lowest and highest environmental impacts for the production of 1 kg REO from IAC are achieved by the low and high IAC estimation cases of this study, respectively. Usually, the values of Vahidi et al.

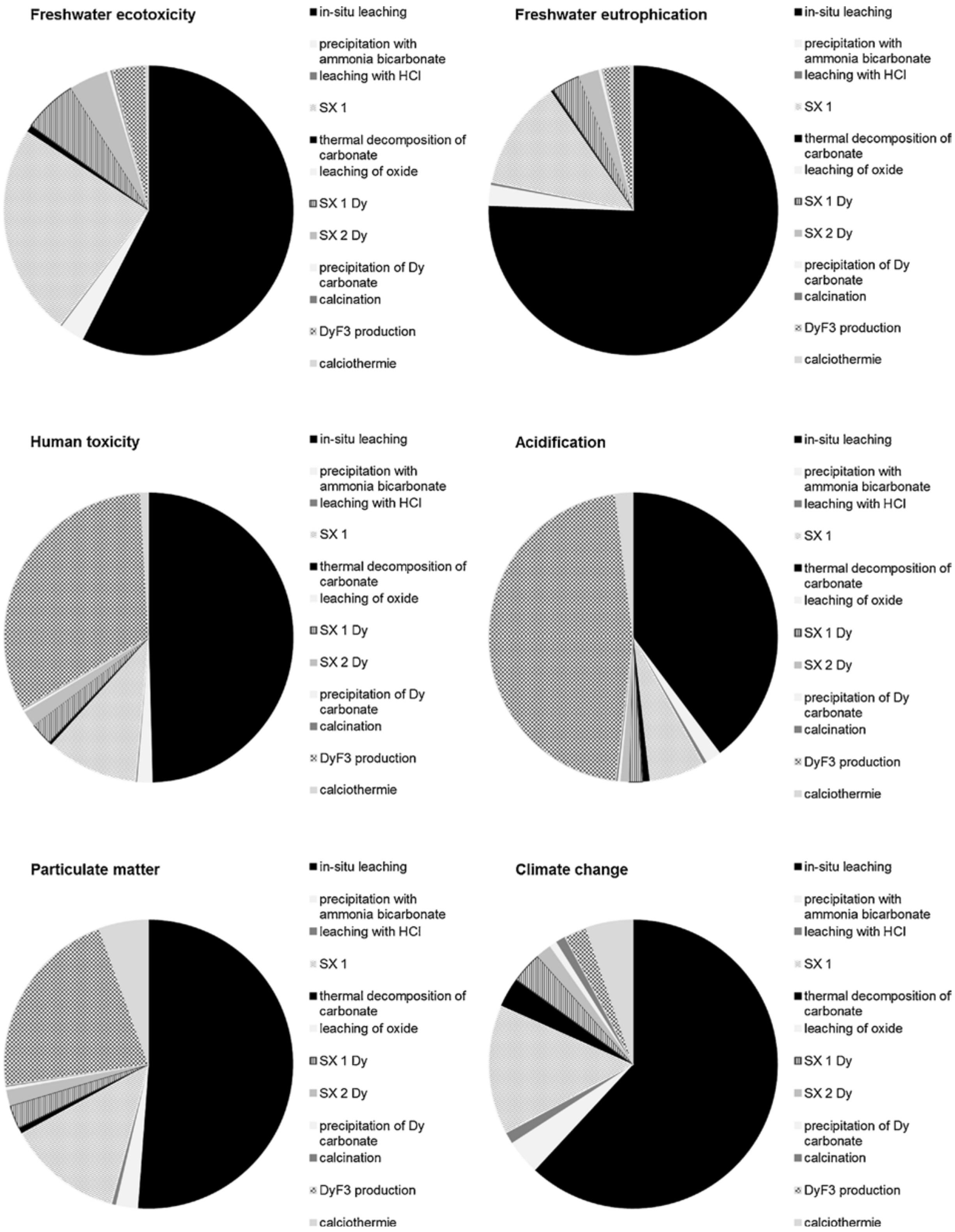

**Fig. 7.** Relative share of single processes on the total environmental impacts of IAC process chain.

(2016) show the same order of magnitude and lie in between. Only eutrophication values of this study are considerably higher due to the higher amounts of ammonium sulphate assumed for in-situ leaching. When comparing the environmental impacts related to either 1 kg REO (third and fourth column of Table 8) or 1 kg $Dy_2O_3$ (fifth column of Table 8), latter are considerably higher. Reasons are additional processes necessary for further separation of mixed REO into single metal oxides and the resulting allocation between each RE oxide.

For the B/M ore from Bayan Obo the results are compared to those from Zaimes et al. (2015) economically allocated and related to 1 kg $Dy_2O_3$. The values assessed by Zaimes et al. (2015) are in the same range mostly. Though, it is noticeable that the acidification and photochemical oxidant formation values from Zaimes et al. (2015) are considerably higher. Vahidi et al. (2016) explains this by release of

**Table 8**
Comparison of selected impacts (using TRACI) relating to 1 kg REO from IAC and 1 kg $Dy_2O_3$ from B/M.

| Category | Unit | 1 kg REO (IAC) current study | | 1 kg REO (IAC) Vahidi et al. (2016) | | 1 kg $Dy_2O_3$ (IAC) current study | | 1 kg $Dy_2O_3$ (B/M) current study | | 1 kg $Dy_2O_3$ (B/M) Zaimes et al. (2015) |
|---|---|---|---|---|---|---|---|---|---|---|
| | | low | high | low | high | low | high | B/M sensitive | B/M | |
| climate change | kg $CO_2$ eq | 13 | 52 | 20.9 | 35.5 | 84.7 | 200 | 153 | 479 | 367 |
| acidification | $H^+$ mol eq | 3.4 | 16.6 | – | – | 20.8 | 59.8 | 69 | 163 | 1520 |
| | kg $SO_2$ eq | 0.065 | 0.32 | 0.17 | 0.29 | 0.40 | 1.15 | 1.32 | 3.12 | – |
| eutrophication | kg N eq | 3.25 | 29.4 | 0.3 | 2.87 | 9.70 | 86.7 | 3.91 | 1.58 | 2.90 |
| ozone depletion | $10^{-5}$ kg CFC − 11 eq[a] | 0.15 | 0.5 | 0.24 | 0.3 | 1.95 | 3.1 | 2.5 | 4.2 | 6.5 |
| photochemical oxidant formation | kg $O_3$ eq | 0.7 | 2.9 | 1.6 | 2.6 | 4.3 | 11 | 14 | 30 | 632 |
| human toxicity$_{cancer}$ | $10^{-5}$ CTU[b] | 0.005 | 0.3 | 0.14 | 0.2 | 0.46 | 1.2 | 0.75 | 7.4 | 3.0 |
| human toxicity$_{noncancer}$ | $10^{-5}$ CTU[b] | 0.5 | 2.8 | 1.0 | 1.7 | 0.31 | 9.9 | 4.2 | 15.4 | 8.6 |
| ecotoxicity | CTUe | 80 | 491 | 279 | 449 | 540 | 1753 | 712 | 2550 | 493 |
| particulate matter formation | kg $PM_{10}$ eq | 0.034 | 0.15 | – | – | 0.286 | 0.638 | 6.26 | 7.63 | 2.61 |
| | kg $PM_{2.5}$ eq | 0.015 | 0.07 | 0.026 | 0.045 | 0.13 | 0.29 | 2.86 | 3.58 | – |
| energy | $MJ_{surplus}$ | 17 | 69 | 13 | 18 | 94 | 246 | 186 | 384 | – |
| cumulated energy demand | MJ | 171 | 706 | 255 | 388 | 1058 | 2633 | 2019 | 5490 | 5064 |

[a] CFC-11: trichlorofluoromethane.
[b] CTU: comparative toxic unit.

sulphur dioxide from the roasting process with sulphuric acid used for B/M processing. The supporting information of Zaimes et al. (2015), however, show that more than 95% of acidification and photochemical oxidant formation comes from mining. In our opinion Zaimes et al. (2015) have assigned an excessive amount of blasting agent to 1 kg REO. We suppose that Zaimes et al. (2015) used the amount of crude ore (76 kg) instead of the quantity of blasting agent (0.0002 kg) as required by the ecoinvent process “RER blasting”.

In summary, the results of available studies for both IAC and B/M processing are comparable, with a few exceptions. In case of IAC for the two available LCA studies (current study and Vahidi et al. (2016)) the assumed amount of ammonium sulphate used as leaching agent varies mostly. As long as no primary data from China's deposits are available, no better estimations can be obtained. The same applies to data concerning waste (tailing) and waste water treatment. Those few data available are very rough estimations.

## 6. Conclusion

This life cycle assessment compares for the first time the environmental impacts of Dy production from IAC, B/M and eudialyte by generating a consistent framework for the description of the production processes. The best environmental performance is given for the production from eudialyte. The impacts from IAC and B/M routes are on a comparable level within the scope of data quality. For all three mineral types the impact categories with the highest contribution are freshwater ecotoxicity, human toxicity as well as eutrophication marine and freshwater. The demand for and supply of chemicals, though different for each mineral route, has the biggest impact on most environmental effects. While ammonium sulphate is used for in-situ leaching in case of IACs, hydrochloric acid is the main input into dry digestion for the eudialyte process chain and sulphuric acid is the main input into the roasting process for B/M. Energy supply along with transportation processes play only a minor role. The contribution of the direct REE processing is most often insignificant for all three mineral types. Only for marine eutrophication (IAC and B/M, by ammonium emissions during in-situ leaching and ammonium bicarbonate precipitation) and particulate matters (B/M and eudialyte, by dust from mining, crushing and grinding) the direct REE beneficiation and separation processes are crucial.

In case of IAC the marine eutrophication is the most important environmental effect due to ammonium emissions during in-situ leaching with ammonium sulphate. The ammonium sulphate amount dominates all other impacts also. However, 78%–95.5% of the in-situ leaching impacts (except marine eutrophication) are generated during upstream ammonium sulphate production. As the precise amount of ammonium sulphate necessary is not known and at the same time it is a highly critical parameter, three cases were considered representing 2%, 3% and 4% leaching solutions. The choice of this parameter alone varies the overall outcome by the factor of 4. In case of the most likely 4% solution, the overall environmental effects exceed those of the other mineral routes by far, almost exclusively due to the marine eutrophication effect. Whereas the extremely optimistic assumption of lower ammonium sulphate demand results in levels comparable to B/M sensitive. As long as no primary data is published, this parameter remains highly debatable. As a consequence the advice for producers must be to keep the amount of ammonium sulphate as small as possible.

For B/M a highly sensitive parameter is the use of waste water treatment using struvite. As ammonium emissions have been identified as a major concern during Chinese RE production a waste water treatment seems appropriate. Again, no primary data about treatment technologies at the processing site exists and only general statements about environmental improvements can be found. However, as the results show, the choice of the treatment process introduced can also cause other environmental effects to worsen. Hence an overall assessment is useful to avoid unwanted trade-offs.

The results for eudialyte describe a mainly hypothetical process chain, which technical feasibility has been shown on laboratory scale. No overall processes optimisation or any chemical reuse options are considered so far. Therefore, the already good environmental performance could be further improved.

Results support findings of earlier studies each for a different mineral route. However the consistent system boundaries from mining to the Dy metal, economic allocation on the same price basis, inclusion of the same upstream and downstream processes as well as the same impact assessment methodology allows the direct ranking of production routes. From an environmental point of view Dy production from a mineral with a high HREE content is favourable. However, the recent past has shown that economic and also social conditions outrank environmental concerns. Molycorp, Inc. suspended production at the only REE mine in the United States (Mountain Pass Mine, California) due to bankruptcy in 2015 although the pollution control has been further improved since the year 2000 (California Regional Water Quality Control Board 2010; EPA, 2012). The reason for closure was the lost conflict against Chinese pricing policy. Although the Chinese Ministry of Environmental Protection (MEP) has already made several attempts to set stricter maximum amounts for pollutants in water and air and for the total discharge of waste water, waste air, thorium, and uranium, the environmental conditions are hardly affected (Wübbeke, 2013). According to estimates, 80% of enterprises will fail to meet these standards because the implementation could raise production costs by 70% (Lee and Wen, 2017; Wübbeke, 2013). The horrendous pollution by illegal mining activities, however, will persist (Packey and Kingsnorth, 2016).

## Acknowledgement

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## Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.resconrec.2017.12.006.

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