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Review

# Rare earth metals from secondary sources: Review of potential supply from waste and byproducts

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ABSTRACT

Current concerns about lack of diversity in supply of critical metals have spurred research into utilizing domestic sources, particularly from waste streams. Sustainability strategies like urban mining, industrial symbiosis, and the circular economy suggest avenues to realize new supplies of critical metals. In this work we explore the resource and economic potential for extracting rare earth elements (REEs) from industry byproducts (e.g. coal combustion products, red mud) and secondary sources (e.g. waste electronics and light bulbs). Combining materials flow analysis and characterization data, we find that while REE concentrations in waste and byproduct streams are mostly much lower than current REE ores, some secondary sources are richer than ores in high value REEs such as scandium. The quantities of REEs contained in secondary sources could meet current global demand even with low extraction yield rates. Phosphogypsum, coal ash and red mud from aluminum production stand out as promising candidates for recovery due to high concentrations of valuable REEs and sufficient quantities to potentially meet demand. Processes to extract REEs from secondary sources are under development, it is not yet clear which will be profitable at scale and which can be achieved at least environmental impact. This work provides high level guidance on the potential of secondary sources by characterizing quality (concentrations of different rare earths) and quantity (mass of rare earths in global scale wastes and byproducts). This significant first step helps clarify directions for policy and research and development investments.

## 1. Introduction

Rare earth elements (REEs) are ubiquitous in a number of industrial sectors including healthcare, defense, transportation, and information and communication technologies.Their use in electronics and clean energy technologies (e.g. wind turbines, electric vehicles, photovoltaics, energy efficient lighting) has been growing substantially and will likely fuel increased demand in the future. Substitution with other materials is challenging in most applications (Nakamura and Sato 2011). While these elements are present in minerals in a number of geographic areas, currently nearly all of the supply is from China; concerns over this lack of diversity have spurred a variety of research into their criticality. The US, for example, had one rare earth mine (Mountain Pass), which closed in 2015 when MolyCorp declared bankruptcy. Relying solely on imports can create vulnerabilities in the supply chain; for example, China briefly halted exports of rare earth materials in 2010 causing speculation and price volatility.

Sustainability strategies to address such challenges include urban mining, industrial symbiosis, and circular economy approaches. Urban mining is when wastes that typically end up in landfills are instead processed for recovery of contained materials. Industrial symbiosis is the idea that wastes from one industry can be used as inputs to another industry; often co-location makes this an efficient exchange. Circular economy posits that resource recovery can disrupt the currently linear economic system of take (mine), make (manufacture), and dispose of materials. Such strategies can ensure additional environmental incentives in pursuing the extraction of REEs from waste and industrial byproduct materials.

REEs exist in a variety of secondary products. They are present in 1) coal combustion products such as fly ash, bottom ash, and incinerator ash, 2) industrial byproducts like slags, dross, phosphogypsum, and red mud, and in 3) electronic wastes including nickel-metal hydride batteries, hard drives from laptops and desktop computers, cellular phones, and speakers. Many of these byproducts and wastes currently end up in landfills, the REEs effectively lost due to dilution (Du and Graedel 2011, Reck and Graedel 2012, Zimmermann and Gößling-Reisemann 2013).

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Sourcing REEs from secondary sources has the potential to dramatically reduce their environmental burden of extraction (Tharumarajah and Koltun 2011, Weber and Reisman 2012, Koltun and Tharumarajah 2014) as well as provide a domestic supply source, as most countries produce at least some of these waste products.

There are many questions to resolve in order for secondary sources to become real-world sources of REEs. Process development is critical, as each waste stream presents its own challenges in realizing economically feasible extraction. Research groups around the world have been working to develop recycling processes for secondary streams, this literature is reviewed in (Tunsu, Petranikova et al. 2015, Dutta, Kim et al. 2016, Costis, Mueller et al. 2019, Omodara, Pitkäaho et al. 2019, Swain and Mishra 2019). Many approaches are being pursued, including leaching (chemical and bio), pyrometallurgy, ionic extraction, and super-critical extraction. A number of prior works review possible technological paths to recover REEs from wastes and/or byproducts. (Binnemans, Jones et al. 2013) identify REE-containing waste streams and characterize process flows through which REEs might be recovered. Binnemans et al (2015) describe process pathways for industrial residues such as red mud and phosphogypsum (Binnemans, Jones et al. 2015). (Jha, Kumari et al. 2016) and (Xie, Zhang et al. 2014) provide high level analyses of how process types, hydrometallurgical in the former and solvent extraction in the latter, could be used to recover REEs from ores and waste products.

Given the variety of waste streams that contain REEs and the multitude of processes that could be used to extract them, researchers, industry, and government face challenges on how to allocate effort and resources to develop different options. Techno-Economic Analysis (TEA) is one approach, it is used to assess the economic profitability of a lab-scale process post industrial scale-up. TEA has been applied to characterize potential economics for a subset of process and waste stream combinations, e.g. using bio-leaching, supercritical $CO_2$, and biosorption for REE extraction from coal ash, mine tailings, ion exchange clays, and fluidized catalytic cracking catalysts (Jin, Park et al. 2017, Das, Gaustad et al. 2018, Thompson, Gupta et al. 2018). While informative, Techno-Economic Analysis is uncertain, labor-intensive, and does not inform the potential future scale of the industry. Additional perspectives are needed.

One useful assessment of waste stream potential is to quantify quality and quantity of desired materials, where quality refers to the concentrations of contained REEs. In mining and recycling there is a general trend that the higher the concentration of a desired substance, the easier it is to extract it (Dahmus and Gutowski 2007, Calvo, Mudd et al. 2016). REE concentrations, collective and for individual elements, thus provides information on potential economic feasibility of recycling. Quantity refers to the total amount of REEs that could be extracted from a waste source. Quantity is important first because there should be enough REE content in a waste stream to satisfy a significant share of demand. Second, quantity relates to economies of scale: It only makes sense to make large investments to research, develop and implement processes that could generate sufficient revenue to offset fixed costs. Note that there are indicators/indices other than quality and quantity that can be used to assess recycling quality, e.g. the resource and technology indices developed by (Sun, Xiao et al. 2016).

The goal of this work is to quantify the global level quality and quantity of REE materials for a wide array of secondary sources, both industrial byproducts (coal ash - fly and bottom, red mud, phosphogypsum, catalysts and polishing powers) and electronic waste (bulbs – linear fluorescent, LED and compactfluorescent, smart phones, NiMH batteries, LCD televisions, desktop computers and earbuds). We include REE concentrations in the earth's crust and ores to provide a baseline for comparison. Our characterization of quality comprehends what is known on the *variability* in REE concentrations. Many secondary sources show large variability, e.g. the total REE concentration in coal ash varies from 26 ppm to 1957 ppm, depending on which coal plant the ash comes from. The economics of extracting REEs from coal ash will thus depend on which source is used (Das et al 2018).

This work goes beyond prior analyses in the breadth of secondary sources analyzed, the geographical scope and the data collection and analysis of variability. In prior work researchers have reported quality of individual resources, e.g.(Zhang, Rezaee et al. 2015) provide an in-depth review of REEs in coal ash. (Swain and Mishra 2019) review concentrations in a number of secondary sources. Quantity (also called recycling potential) has been characterized by (Binnemans, Jones et al. 2013) for magnets, lamp phosphors, and NiMH batteries. (Wang, Gu et al. 2015) assess the potential for indium recycling from waste flatscreen displays in China. However, the utility of characterizing quality and quantity as a guide emerges when different sources can be compared. No prior work has estimated quantity *and* quality across a large cohort of secondary sources. This breadth of coverage enables a preliminary scoping analysis, suggesting promise for a secondary source if it: 1. has relatively high concentrations of rare earths, particularly valuable ones, and 2. contains sufficient rare earths to supply a significant share of demand. This scoping analysis is an important first step to understanding the technical, economic, and environmental viability of REEs from wastes and byproducts.

## 2. Methods

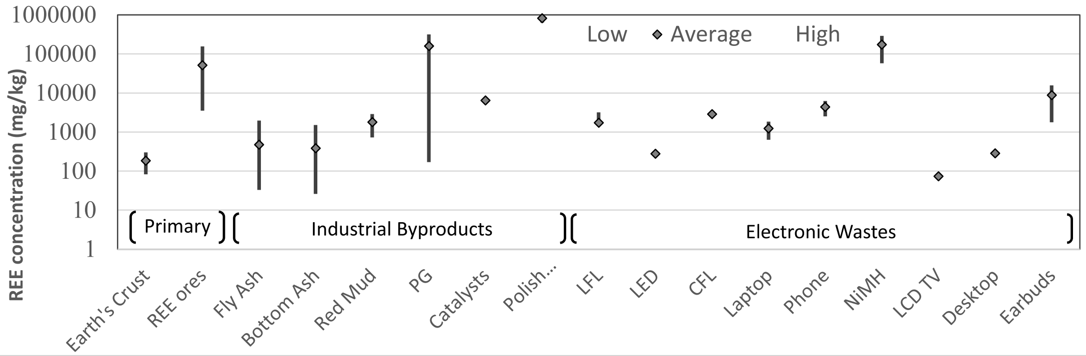
### 2.1. Characterizing quality

Characterizing quality involves quantifying the concentration of REEs in primary and secondary sources. For many streams, such as ore and coal ash, REE concentration varies considerably by location, thus a full scale data search was undertaken to identify publicly available sources for each stream. Supplemental Information (SI) Table 1SI1 shows the minimum, average, and maximum crustal abundance values from eight studies. Table SI2 is data from the U.S. Geological Survey on rare earth ore grade at mines located in the United States (Mountain Pass and Green Cove), China (Byan Obo, Nangang, Weishan, and Jiangua), Australia (Mount Weld, North Capel, and Stradbroke), and Revda in Russia. The minimum, average, and maximum values from these mines were also used for comparison although it should be noted that the maximums are not a physically relevant number (ie. no individual mine would demonstrate all of the maximum values). Coal ash combustion wastes have been the source of much research and therefore a significant amount of primary data was collected for this byproduct source; Table SI3 summarizes this primary data. No municipal solid waste combustion sources were included in this analysis. Data was then gathered from a variety of sources for industrial byproducts and electronic wastes; these sources include industry reports, lab-scale research on extraction from these sources, product information that is available publicly, and other research literature. Table SI4 shows the minimum and maximum reported values for four industrial byproducts containing rare earth elements:bauxite processing residue most often referred to as red mud, phosphogypsyum, spent fuel cracking catalysts, and spent polishing powders. In cases where multiple values were not available in the reported literature, the average is not reported in the results, merely the maximum and minimum values. Table SI5 shows rare earth element content for a variety of electronic wastes including light bulbs (LFL, CFL, and LED), laptop computers, desktop computers, smart phones, and ear buds (headphones). Tables SI1-SI5 list all of the primary sources which are also included in the caption of Fig. 1. In Section 3.1 the concentrations of the secondary sources compared to primary sources are analyzed, first looking at the raw magnitude of contained total rare earth elements and then comparing to average ore grades. Eq. 1 shows how this comparison was made to clarify when some secondary sources, while potentially lower in overall REE concentration, may have potential to be a concentrated sources of particular elements.

**Table 1**

Average rare earth element concentration in waste/byproduct compared to ore average (see Equation (1)).Values above one indicate that the element is more concentrated in the waste/byproduct than in ore.Blue, yellow and green blocks call attention to those elements with high concentrations, pink for low concentrations. These were calculated using the raw data shown in Fig. 1 but calculated for individual elements; the raw data is from the fifty references cited there.

| Fraction to Ore | Ce | Dy | Er | Eu | Gd | Ho | La | Lu | Nd | Pr | Sm | Sc | Tb | Tm | Y | Yb | TREE |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Fly Ash | 0.0 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 | 0.0 | 2.9 | 0.0 | 0.7 | 0.1 | 0.0 | 0.0 |
| Bottom Ash | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 | 0.0 | 1.1 | 0.0 | 0.9 | 0.1 | 0.0 | 0.0 |
| Red Mud | 0.0 | 0.2 | 0.2 | 0.6 | 0.1 | 0.4 | 0.0 | 0.7 | 0.0 | 0.0 | 0.0 | 11.6 | 1.1 | 4.2 | 0.3 | 0.1 | 0.0 |
| PG | 0.0 | 0.1 | 0.2 | 0.6 | 0.1 | 0.3 | 0.0 | 0.6 | 0.0 | 0.0 | 0.0 | 8.6 | 1.1 | 3.9 | 0.3 | 0.1 | 0.0 |
| Catalyst | 3.2 | 0.1 | 0.1 | 0.3 | 0.1 | 0.1 | 3.4 | 0.0 | 3.2 | 3.4 | 0.1 | 0.5 | 0.1 | 0.5 | 0.2 | 0.0 | 3.1 |
| Polishing | 3.3 | 0.2 | 0.2 | 0.6 | 0.2 | 0.2 | 3.6 | 0.0 | 6.4 | 6.8 | 0.3 | 0.8 | 0.1 | 0.7 | 0.3 | 0.0 | 3.2 |
| LFL | - | - | - | 1.1 | - | - | - | - | - | - | - | - | 2.1 | - | 3.4 | - | 0.0 |
| LED | - | - | - | - | - | - | - | - | - | - | - | - | - | - | 0.4 | - | 0.0 |
| CFL | - | - | - | 1.3 | - | - | - | - | - | - | - | - | 2.4 | - | 3.9 | - | 0.1 |
| Laptops | - | 0.1 | - | - | - | - | - | - | 0.1 | 0.0 | - | - | - | - | - | - | 0.0 |
| Smart phones | 0.0 | - | - | - | - | - | - | - | 0.3 | 0.4 | - | - | 0.4 | - | - | - | 0.1 |
| NiMH batteries | 2.8 | - | - | - | - | - | 5.9 | - | 2.5 | 2.2 | - | - | - | - | 0.3 | - | 3.4 |
| LCD | 0.0 | - | - | 0.0 | - | - | 0.0 | - | - | - | - | - | 0.0 | - | 0.1 | - | 0.0 |
| Desktop | 0.0 | 0.1 | - | 0.0 | - | - | 0.0 | - | 0.0 | 0.0 | - | - | 0.0 | - | 0.0 | - | 0.0 |
| Earbuds | - | 3.5 | - | - | - | - | - | - | 0.8 | 0.6 | - | - | - | - | - | - | 0.2 |
| Less than 50% | < 1X | | 1X to 3X | | | 3X to 6X | | | 6X to 12X | | | | | | | | |



| | Earth's Crust | REE ores | Fly Ash | Bottom Ash | Red Mud | PG | Catalysts | Polishing | LFL | LED | CFL | Laptop | Phone | NiMH | LCD TV | Desktop | Earbuds |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Low | 83 | 3508 | 33 | 26 | 723 | 169 | 5320 | 638812 | 1500 | | | 635 | 2527 | 58000 | | | 1767 |
| Average | 185 | 50950 | 475 | 381 | 1796 | 159467 | 6444 | 819406 | 1720 | 278 | 2853 | 1238 | 4387 | 173610 | 73 | 285 | 8741 |
| High | 299 | 155100 | 1957 | 1512 | 2870 | 316766 | 7567 | 1000000 | 3200 | | | 1840 | 6247 | 289220 | | | 15714 |

**Fig. 1.** Total rare earth element (REE) content (in ppm or mg/kg) for ores and potential secondary sources (log scale): Earth's crust (Kleber and Love 1963, Jackson and Christiansen 1993, Sabot and Maestro 1995, Wedepohl 1995, Gunn 2014), REE ores (Orris and Grauch 2002, Long, Van Gosen et al. 2012, Mariano and Mariano 2012), fly and bottom ash from coal combustion (Zhang, Yamasaki et al. 2001, Smolka-Danielowska 2010, Seredin and Dai 2012, Całus-Moszko and Białecka 2013, Hower, Dai et al. 2013, Mayfield and Lewis 2013, Całus Moszko, Białecka et al. 2016), Industrial Byproductsred mud from bauxite mining (Ochsenkühn-Petropulu, Lyberopulu et al. 1996, Binnemans, Pontikes et al. 2013, Qu and Lian 2013, Borra, Pontikes et al. 2015, Liu and Li 2015), fuel cracking catalysts from petroleum refining (Gerber, Frye et al. 1999), powders from polishing processes (Ozaki, Machida et al. 1999, Kato, Yoshioka et al. 2000, Xu and Peng 2009),phosphogypsum labelled as PG (Rutherford, Dudas et al. 1995, Todorovsky, Terziev et al. 1997, Santos, Mazzilli et al. 2006, Binnemans, Pontikes et al. 2013, Binnemans, Jones et al. 2015), electronic waste including Linear Fluorescent Lamps (LFL) (Ku, Setlur et al. 2015, Osram 2017), Light Emitting Diodes (LED) (Ku, Setlur et al. 2015), Compact Fluorescent Lamps (CFLs) (Ku, Setlur et al. 2015), laptops (Buchert, Manhart et al. 2012), smart phones (Navazo, Méndez et al. 2014, Szamałek and Galos 2016), nickel metal hydride (NiMH) batteries (Müller and Friedrich 2006, Yang, Zhang et al. 2014, Shin, Shin et al. 2015, Lin, Huang et al. 2016, Meshram, Pandey et al. 2016), LCD TVs (Hobohm and Kuchta, 2000; Buchert, Schüler et al. 2009, Yang, Zhang et al. 2014), desktop computers (Buchert, Schüler et al. 2009, Ueberschaar and Rotter 2015) and earbuds (Hobohm and Kuchta, 2000).

$$\frac{Secondary\ Source\ Elemental\ Concentration\ (ppm)}{Primary\ Source\ Elemental\ Concentration\ (ppm)} = A \qquad \begin{matrix} A \geq 1\ more\ than\ ore \\ A < 1\ less\ than\ ore \end{matrix} \tag{1}$$

### 2.2. Characterizing quantity

The amount of REE present in any single secondary or byproduct material source is not relevant if the amount of that material recovered is trivial compared to local or global REE demand. To analyze this, first, data on global REE demand was synthesized from multiple sources over available years (Supplemental Information Table SI6). Total global REO production by country is published each year, however, demand for individual rare earth compounds is challenging to find publicly available and therefore data from multiple years was averaged to estimate the total global demand for individual rare earth elements (Table SI7). In some cases, a stoichiometric multiplier was used to convert from rare earth oxide equivalents to rare earth element amounts (units are noted in each Table). For the industrial byproducts, data on global annual amount of each byproduct was synthesized and the most recent year available was used (Table SI8). For secondary sources (electronics and light bulbs), sales estimates for previous years were combined with data on lifespan and average product weight to estimate waste amounts (Table SI8 and SI9).

## 3. Results

### 3.1. Quality of REEs from secondary sources: Comparingconcentrations

To compare the quality of REEs in secondary and byproduct sources, first one must understand the quality of REEs from traditional extraction sources. Rare earth elements occur naturally in the Earth’s crust with cerium, yttrium, neodymium, and lanthanum being the most plentiful; several geochemistry studies investigating REE crustal abundance were compiled. The most abundant, cerium, is present at an average of 53 ppm in the Earth’s crust. For economic extraction of REE materials, monzanite and bastanite ores are targeted for their higher concentrations. While nearly 98% of all global REE production resides in China, these ores are also present in the United States, Australia, Russia, and elsewhere. Several mines are compared for their rare earth ore grades including Mount Weld, North Capel, and North Stradbroke Island in Australia; Mountain Pass and Green Cove Springs in the United States; Bayan Obo, Nangang, WeiShan, and JiangHua in China; and Revda in the Lovozero district of Russia. Total rare earth ore grades (TREO) range from 16% for Mount Weld in Australia, 8% for Mountain Pass in the US, to 6% for Bayan Obo in China. Following crustal abundance, cerium is present in ore with the highest average concentration at 24,000 mg/kg, followed by lanthanum at 13,000 mg/kg. However, the next highest average ore concentrations material are for dysprosium at 7,960 mg/kg and praseodymium at 2,330 mg/kg (Fig. 1).

The aim of this work is to explore other sources of rare earth materials and compare to the currently economically extractable sources (i.e. Chinese montzanite and bastanite ores mentioned above). One such source that has been studied extensively are combustion byproducts with a focus on coal ash. Coal ash is a byproduct of the combustion process in coal power plants and has two main components: fly ash and bottom ash. Other combustion byproducts containing REEs include waste incinerator ash. Much compositional characterization work has been conducted to determine the quantity of valuable elements within these combustion waste sources. On average, bottom and fly ash have similar amounts of each of the rare earth elements with the most being cerium, neodymium, lanthanum, and yttrium.

#### 3.1.1. Total REE concentration ranges

Fig. 2 shows ranges in total REE concentrations (mg REE contained/kg of medium, or parts per million) in natural environments and various human-induced secondary flows. Results show significant variability in the total rare earth element content both within and between flows; most ranges cover orders of magnitude. One key trend is that currently extracted ore grades, with average 5% total rare earth content and running as high as 15%, have far higher concentrations than most of the secondary sources, excepting polishing powders, NiMH batteries and some phosphogypsum sources. In contrast, fly and bottom ash have lower concentrations, ranging from 0.9-1.3% total rare earth content. Concentration is important in the economics of extraction: Cost generally increases with decreasing concentration (Grübler 2003). Factors other than concentration also play a role, however, including internal factors such as capital expenditures, labor costs, and utilities and external factors like regulation, transportation, and commodity market volatility. It is important to note that the economic value depends on the mix of REEs (and other valuable materials) contained.

Another potential source of rare earth elements is recycling from the wide array of end-of-life products in which they are present. Table 4 in the Supplemental Information details some of the key products containing REEs; some of these uses are dissipative in nature and therefore

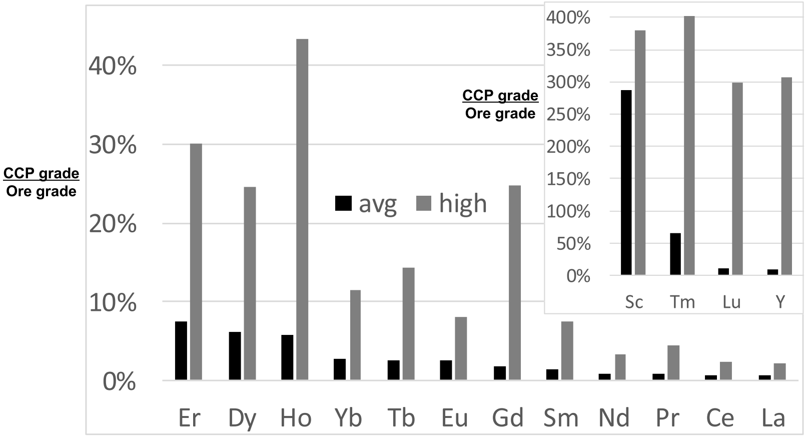


**Fig. 2.** Average and highest value (high) concentrations of select rare earth elements in coal combustion products (CCP) compared to average ore grades

recovery is not an option, such as steel metallurgy, fluid cracking catalysts, glass additives, and polishing powders. However, many products like magnets, batteries, auto catalysts, and phosphors in displays and lighting are applications where recycling may be economically viable. Feasibility will depend on the composition of REEs in the products, how challenging collection and sorting of those products may be, and the viability of recycling technologies to extract REEs at a high yield. Fig. 1 shows that both phosphogypsum and polishing powders contain significantly more total rare earth content compared to the ores from which they are mined while both red mud and fuel cracking catalysts contain several orders of magnitude less. One can see that among these industrial byproducts, phosphogypsyum has the highest degree of uncertainty in REE content.

Fig. 1 also shows REE concentrations for a variety of electronic products, batteries, and various bulbs for lighting including light-emitting diodes (LEDs), linear fluorescent lamps (LFLs), and compact fluorescent lamps (CFLs). On average, with the exception of nickel metal hydride (NiMH) batteries, these products contain less rare earth elements by concentration compared to ores. NiMH batteries contain roughly an order of magnitude more.

The range of concentrations found is important. In the most extreme case, phosphogypsum, REE concentrations vary by three orders of magnitude depending on the source. The profitability of a given process to extract REEs thus depends on which source is used. Process development should account for this variability.

#### 3.1.2. Individual REE content

The price and market for different REEs varies widely. For example, global production and price of scandium in 2016 was 10-15 metric tons and 6,700$/kg respectively, while 5,000 tons of yttrium was produced with average price $3/kg (Gambogi 2018). It is thus important to distinguish individual concentrations of REEs. Also, while total rare earth content of a secondary source may be lower than ore grades, this is, as will be seen, not true in general for individual REEs. For most elements, combustion byproducts contain significantly more REE than their crustal abundance and for some elements nearly on par with average ore grades. In fact, concentrations in coal ash of some elements like gadolinium, samarium, and scandium are higher than ore grade. Fig. 2 shows the percentage of individual elements contained in fly ash (average content and high content) compared to the average ore grade content. A majority of the elements contain less than 10% the content on average compared to ores and less than 30% even for the high content values of fly ash, specifically Er, Dy, Ho, Yb, Tb, Eu, Gd, and Sm are all less than 40% of what ore contains even for the high content values and Nd, Pr, Ce, and La are all less than 5% of what ore typically contains. However, the average content of scandium, currently the most expensive rare earth element with a price of approximately $6,700 $/kg, is almost 3X more in coal ash than ores on average, and nearly 4X for the high-content fly ashes. When comparing the high values in ash to average content in ores, lutetium, yttrium, and thuliumare also more concentrated at approximately 3X, 3X and 130X respectively. As these ores are more expensive, it may suggest economic viability to be achievable for certain extraction processes.

Table 1 shows the ratio of average rare earth element concentration in a waste/byproduct stream compared to global ore average. Ratios comparing high concentration appear in the supporting information Table SI10. While red mud has less rare earth content in total compared to ores, it has concentrated amounts of scandium (11X average ore), thulium (4X), and just slightly more terbium (1.1X). As scandium is by far the most valuable REE in current commodity markets, and both thulium and terbium are quite expensive, this could motivate red mud as a potential source. Similary, for phosphogypsum, a byproduct of the fertilizer production process that combines sulfuric acid with phosphate ores, concentrations of scandium (8.6X), thulium (3.9X), and terbium (1.1X) are greater than in REE ores. Spent polishing powders on average have approximately 6 times more concentrated cerium and lanthanum than REE ores and 6 times more neodymium and praseodymium.

Despite having total REE concentrations much lower than ore, some electronic waste has high concentrations of specific rare earth elements. LFL and CFL bulbs contain 3 times the amount of yttrium, 2 times the amount of terbium, and 1.5X the amount of europium compared to REE ores. Nickel metal hydride batteries contain 6X more lanthanum, and 2-3X more neodymium and praseodymium. Average earbuds contains 3.5X more dysprosium compared to ores.

Looking at the high content values in secondary materials compared to ores shows even more potential. . This richer subset of secondary material streams shows even higher concenrations, and thus potential for recovery, of select REEs. For example, spent polishing powders could have up to 32X more cerium, 21X more lanthanum, 17X more praseodymium, and 15X more neodymium. Nickel metal hydride batteries could contain 9X lanthanum, nearly 5X more cerium, and over 4X of neodymium and praseodymium compared to average ore grades. Fly and bottom ash have very concentrated amounts of lutetium, scandium, and thulium. Looking at maximum values, light bulbs have high concentrations of europium (1.3X for CFL), terbium (2X+ for both LFL and CFL), and yttrium (about 4X for both LFL and CFL).

### 3.2. Quantity of REEs in secondary sources: Comparing annual flows

The compositional content of REEs in waste and secondary products is suggestive of the future economics of recycling. Another important aspect is the total magnitude of REEs contained in these waste streams. If REE content is high but waste stream volume is low, there may not be enough total REE to achieve economies of scale in recycling. Also, if waste and secondary sources are to replace primary production, total available REE in these streams must be comparable to demand. The REE contained in waste stream depends on a variety of dynamic variables including: yearly production, product lifespan, collection rates, and processing yields. We collect data from around the world pertaining to the total quantities generated in waste and secondary streams and when such data is unavailable, we estimate using sales and lifespan data. We then compare total REE quantities in each waste/byproduct stream with global demand for that metal.

Results for coal ash, phosphogypsum, red mud and NiMH batteries are shown in Fig. 3, additional results for other waste/byproduct streams are in the supplemental information. The large volume of many waste materials would provide more than enough REEs to meet global demand, even with low yields. As shown in Fig. 3, considering REEs only from coal combustion products and their average content, annual production of such wastes could supply more than 5X the global demand of yttrium, samarium, gadolinium, and erbium; and provide over 1,500X the demand of scandium. Red mud wastes from the bauxite mining and aluminum production process could supply 5X or more of yttrium, gadolinium, and terbium with significantly higher amounts of terbium and scandium. Phosphogypsum byproducts could provide 75X demand or more of scandium, praseodymium, neodymium, lanthanum, and cerium. As materials like scandium would have increased usage in aluminum alloys for both aerospace and automotive applications if prices were to come down; enabling such secondary sources could have large ramifications for the materials industry. While in comparison, NiMH batteries are much smaller in magnitude in regards to weight produced, they could still provide 3.6X the global demand for lanthanum, 2X the demand for cerium, 1.5X for neodymium, and 1.1X demand for praseodymium.These heavy industries (phosphogypsum, coal ash, red mud) could produce orders of magnitude higher quantities of REEs than end-of-life electronics products, due mainly to volumes produced.

It should be noted that these are global totals compared to global demand; availability is a function of geographic location, affluence, and population demographics. Assessing the potential to meet domestic demand thereby requires nuanced geospatial data; some work has been done for specific sources like coal ash (Beretta, Costa et al. 2010),

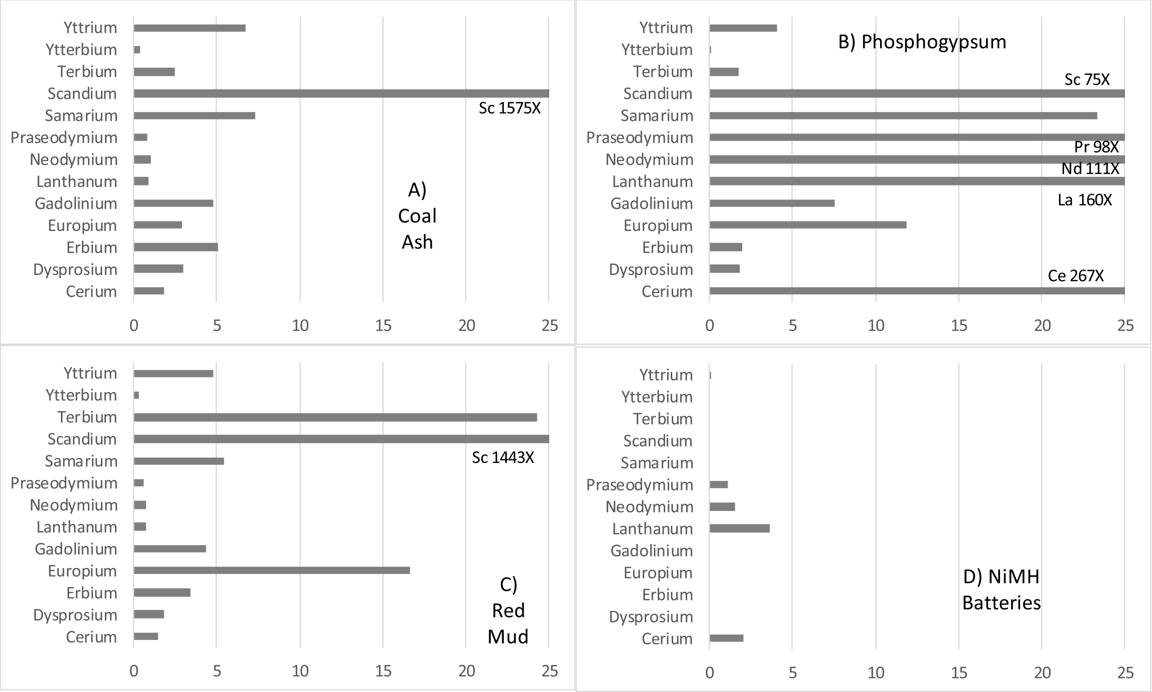


**Fig. 3.** Ratio of potential global supply and demand of REEs for select secondary sources. Potential supply is based on annual waste quantities (tons), REE concentrations, and assuming 100% collection and yield. A value of 1 implies global demand could just be met. Sources: see Fig. 1 and Supplemental Information.

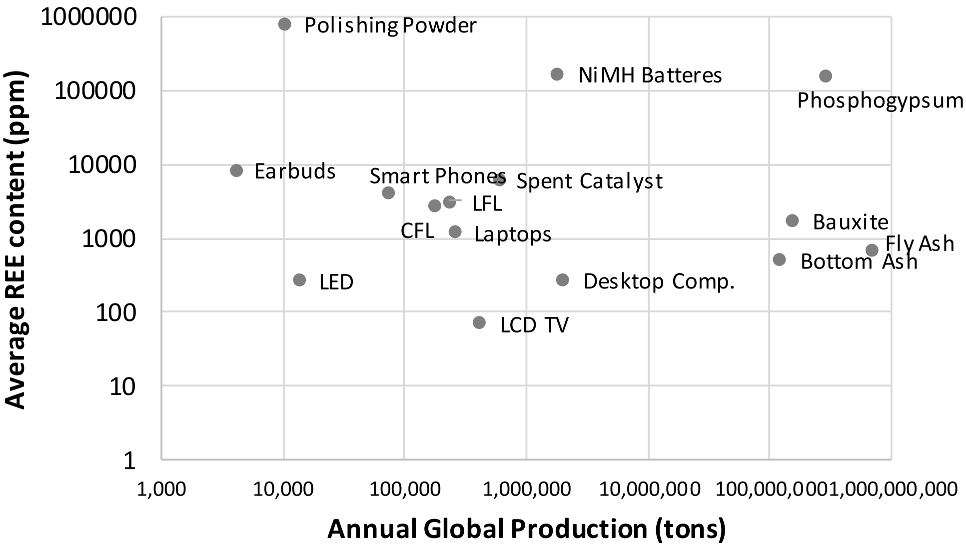


**Fig. 4.** Average Rare Earth Element (REE) concentration and total global flow for a variety of industrial and consumer waste streams (log scales). A combination of high concentration and large material flow suggests higher potential for economic and significant recovery of REEs.

photovoltaics (Goe, Gaustad et al. 2015) or electronic wastes (Tong and Wang 2004, Bahers and Kim 2018), but has not been compared comprehensively. Additionally, there is likely to be mismatch between where secondary materials are collected and available (particularly for electronics) and where manufacturing that demands a supply of rare earth metals is currently happening.

It is useful to combine measures of both quality (REE concentration) and quantity (total mass of waste/byproduct stream). A stream with both high quality and content suggests higher economic and strategic potential to recover REEs. Results are shown in Fig. 4. Phosphogypsum, red mud, and coal ash stand out as large mass streams with high concentrations. Nickel metal hydride batteries, while lower in magnitude, may also be a promising potential source.

## 4. Conclusion

This work showed the quantity and quality of rare earth elements contained in a variety of byproduct and waste materials; this builds a foundation for understanding the broader sustainability potential for these streams. Implementing sustainability strategies such as urban mining, industrial symbiosis, or other circular economy approaches requires a deep understanding of the quantity and quality of these potential sources as laid out in the work. It is important to note that there are still many additional factors that will determine the feasibility of sourcing REEs from any of these potential products. Processing routes will be key; a variety of combinations of processes may be employed including leaching, hydrometallurgy, pyrometallurgy, electrowinning, supercritical extraction, etc. The variety of sources (eg. red mud, laptops), processes (listed previously), and targeted outputs (specifics REEs, base metals, etc.) indicate that a combinatorial approach to understanding techno-economic and environmental impacts is essential (Das, Gaustad et al. 2018).

The wide ranges shown in these results speak to the high variability of concentration information; the quality of composition can be geospatially sensitive (for ores and industrial byproducts from production of minerals) and/or lifespan sensitive (or electronic wastes). The quantity and quality perspective suggests promising streams to research further are coal ash (for scandium in particular), phosphogypsum and other byproducts, and nickel-metal hydride batteries.

Each secondary source has a variety of processing routes to consider; each of these routes results in differing yields, costs, and revenues. Most processing and extraction approaches for these sources are still in the early stages of development. Extraction of rare earth metals from red mud is one of the furthest along with pilot scale plants currently being industrialized. Sustainably sourcing rare earth metals will be essential to many clean energy and energy efficiency technologies. This work informs scoping on promising paths; research, development and investments is needed to bring rare earth elements extraction from secondary sources to fruition.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Supplementary materials

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