Material flow analysis for identifying rare earth element recycling potentials in the EU-27

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MATERIAL FLOW ANALYSIS FOR IDENTIFYING RARE EARTH ELEMENT RECYCLING POTENTIALS IN THE EU-27

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SUMMARY: Rare earth elements (REEs) are essential for high-techology industrial sectors. This paper presents research on material flow analysis (MFA) applied to REEs in the EU-27. Innovative aspects of this research pertain to (i) considering potential lithospheric stocks of REEs in the EU and (ii) accounting for incomplete and imprecise information in MFA data reconciliation. Results obtained to-date provides a history of EU-27 raw rare earth imports and exports and a methodology for data reconciliation which constitutes an alternative to the classical least-squares method.

1. INTRODUCTION

Material Flow Analysis (MFA) has been used for several decades to study in particular the flows and stocks of susbtances in the anthroposphere (see for example Wolfman, 1965; Ayres, 1989; Baccini and Brunner, 1991). The method has been applied extensively to the cycles of metals, in particular by the group of Th. Graedel at Yale University (e.g. the STAF project), for example to metallic substances such as zinc, iron, steel, lead, etc. In recent years there has been increased interest in the so-called "critical metals" (NRC, 2008; EC, 2010; DOE, 2011), i.e. metallic substances required by certain technologies; e.g. energy or defense technologies and in particular in rare earths. Rare earth elements (REEs) are a group of 17 elements comprising the 15 lanthanides, scandium and yttrium. REEs are identified as "critical" in the sense that they combine high levels of industry requirements with risks in terms of supply. China, who controlled more than 93% of world production in 2010, has been tightening export quotas on rare earths. Criticality is particularly relevant for the so-called "heavy" REEs, e.g., Tb (used primarily in phosphors for low-energy lighting) and Dy (used in permanent magnets), but also for certain lighter elements such as Nd, due to its extensive use in magnets, alloys, etc. For a summary of rare earth applications, see for example Goonan (2011), while Binnemans et al. (2013) provide a synthesis of recycling technologies for the three market sectors which appear to be the most promising in terms of recycling potential; namely permanent magnets, NiMH batteries and phosphors for low energy lighting.

Du and Graedel (2011a, b, c) applied MFA to REEs to identify global (world) flows and stocks. The analysis of global flows and stocks presents the considerable advantage of dealing with a "closed" system. Therefore flows of substances into the anthroposphere can be "constrained" by production from the mining sector, which is relatively well known thanks to data provided in particular by the U.S. Geological Survey (e.g. U.S. Geological Survey, 2012). In this paper we address an "open" system: the economy of the EU-27. In an open system there are flows in and out of the system, all along the value chain. The open character of the system under investigation, combined with the fact that rare earth elements are largely "dispersed" in small quantities in products, makes the application of MFA to this category of substances a relatively difficult task.

The work presented herein was performed as part of an ongoing project (Jan 2012 – Jan 2015) supported by the French National Research Agency (ANR), called ASTER (Systemic Analysis of Rare Earths - Flows and Stocks). At the time of drafting of this paper, the project is at t_0 + 13 months, hence results presented below should be considered as preliminary.

2. METHODOLOGY

2.1 System definition

MFA considers the transfer of substances between processes within a system. As a first step, the system boundary was defined as the EU-27. Processes considered within this system are presented below. As the project unfolds, it is possible that certain processes will be merged for the final MFA.

- Separation. REEs enter the EU-27 in particular as mixtures of rare earth oxides (REOs). Separation processes serve to separate these mixtures into specific rare earth element oxides.
- Fabrication. Once the oxides have been separated, they are processed to produce alloys and compounds with specific chemical and physical characteristics, as required by various applications.
- Manufacture. This process produces the functional units used in the applications; e.g. permanent magnets.
- Applications. In this process, the functional units are included into applications; e.g. permanent magnets in computers, audio-systems, windpower turbines, etc.
- Use. Products fulfill their service life within the Use process.
- Waste collection. Products that have exceeded their service life in the "Use" process enter the waste collection process as end-of-life products.
- Recycling. End-of-life products that have entered the waste collection system may be recycled into various other processes located higher up in the cycle, or exported for re-use.
- Elimination. End-of-life products that are neither re-used nor recycled are eliminated, e.g. by incineration or landfilling.

Note that because there is currently no extractive activity in the EU-27 with respect to REEs, no process is identified here as "Mine": all raw materials (see below) are imported into the EU-27. However, an additional process, identified as "Lithosphere" was included in the analysis, because it constitutes a stock, albeit unused to-date. This is one of the innovative aspects of the proposed MFA, since traditionally, the lithosphere is considered as a source of flux, but rarely as a stock (MFAs typically calculate variations of stock in the lithosphere; e.g. van Beers et al., 2005).

2.2 Disaggregating rare earth compound imports and exports

The ore mined from the ground to produce REEs is primarily extracted from alkaline igneous rocks and carbonatites, such as bastnäsite, a fluorocarbonate mineral with a rare earth content of approximately 70% rare earth oxide primarily composed of Ce, La, Pr and Nd (Jordens et al., 2013). While the giant Bayan Obo deposit, in northern China, extracts primarily bastnäsite, in southern China, ion-exchange clay in lateritic deposits are mined, that are particularly rich in heavy REEs (Tb, Dy, …).

To estimate the global flows of individual REEs into world economy, Du & Graedel (2011a) rely in particular on data from the USGS regarding mine production and on the proportions of individial REEs in the mined ores. For the study of flows and stocks in an open system such as the EU-27, a different approach must be used. First, we consider the statistics of imports and exports of rare earth compounds in and out of the EU-27, based on EUROSTAT (2013) data for the relevant product HS codes. HS is the harmonized commodity description and coding system of tariff nomenclature, an internationally standardized system of names and numbers for classifying traded products, developed and maintained by the World Customs Organization (WCO). The HS codes considered for the flows of "raw" rare earth materials into and out of the EU-27 system, are the same as those considered by Öko-Institut (2011) and are summarized in Table 1.

Next we convert the flows of compounds to flows of rare earth oxides (REOs), assuming an average REO conversion factor as shown in Table 1. For the metals (the first two HS codes in Table 1), the estimated REO conversion factor (1.2) is based directly on the ratios of atomic weights of rare earth oxides compared to pure metals (see also Hedrick, 2004). Cerium compounds (HS code 284610) include a variety of products, where carbonates (with a REO content of approx. 50%) represent an important proportion. An average value of 0.75 was selected. For code 284690, compounds include primarily oxides, fluorides, phosphates, etc., with relatively low proportions of carbonates. A REO conversion factor of 0.90 was selected (Rhodia-Solvay, pers. comm.).

Table 1. HS codes for raw rare earth product imports and exports

HS code	Description in EUROSTAT	Estimated REO conversion factor
28053010	Intermixtures or interalloys of rare-earth metals, Scandium and Yttrium	1.2
28053090	Rare-earth metals, Scandium and Yttrium (Excl. intermixtures or interalloys)	1.2
28461000	Cerium compounds	0.75
28469000	Compounds, inorganic or organic, of rare-earth metals, of Yttrium or of Scandium or of mixtures of these metals (Excl. Cerium)	0.90

Further to estimating imports/exports of REOs in the EU-27, they were disaggregated into individual rare earth oxides using the method presented in Goonan (2011). Knowing the relative REO consumption of each market sector and considering the proportions of REEs in the applications manufactured by these sectors, the REO totals obtained from the previous step were decomposed into individual oxides. A distribution of world market sector REO consumption is found in Lynas (2010), while REE requirements by application are found in Table 1 of Jordens et al. (2013; also from Lynas, 2010).

Application of the method to the 2007 global mine production compares well (Figure 1) with values presented in Du and Graedel (2011a). It should be stressed that, given the uncertainty with respect to the definition of products containing rare earths (note the lack of specificity of descriptions in Table 1) and also the dispersion of these susbtances in products, a material flow approach to REEs can only achieve order of magnitude comparisons. The disaggregation approach based on Goonan (2011) is felt to be well suited to this objective.

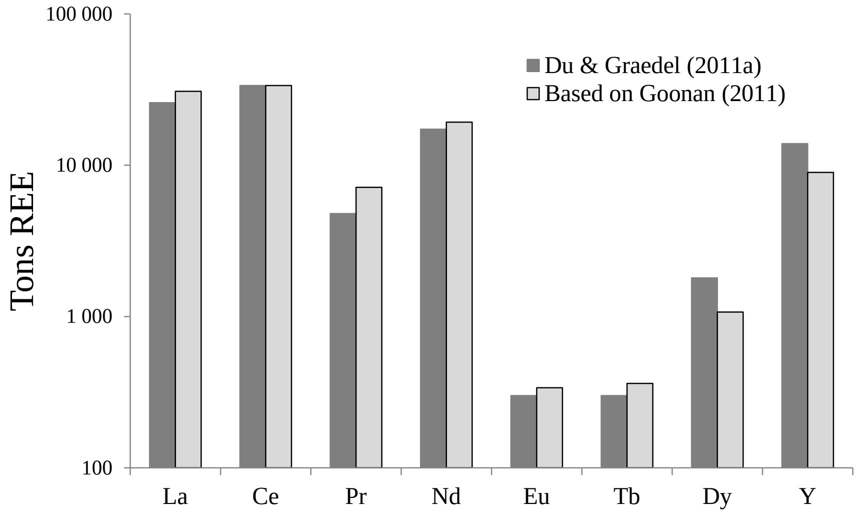

Figure 1. Global mine production (year 2007) of specific REEs, from Du & Graedel (2011a) and based on the disaggregation method of Goonan (2011).

2.3 Lithospheric stocks

As mentioned in the introduction, one of the innovative aspects of the ASTER project is to take into account the geologic potential of the system under investigation. For the analysis, EU-27 was extended to continental Europe, to include the Baltic Shield and Greenland, where numerous occurrences are known, in relation with alkaline intrusions, carbonatites and pegmatites. Based on a global geological and metallogenic synthesis, over 270 REE occurrences and deposits were classified with respect to typology and potential (Charles et al., 2013). The Baltic Shield and southern Greenland clearly represent the strongest potential: for example the Kringlerne intrusion is estimated to hold on the order of 20 Mt REO. Should such deposits enter the production phase, the question of rare earth criticality would be significantly altered.

2.4 Reconciling MFAs

An additional innovative aspect of the ASTER project with respect to MFA, pertains to data reconciliation in presence of imprecise/incomplete information. When investigating flows and stocks of REEs (or other susbtances), one is typically confronted with such information. Yet current approaches for treating uncertainty in MFAs generally rely on a classical probabilistic approach, whereby uncertain parameters are represented as normal probability distributions, with an average value and a standard deviation. Data reconciliation is typically performed using the least-squares method, by minimizing a sum of squares of differences between estimated and reconciled flows, weighted by their respected standard deviations (see e.g. the STAN tool; Brunner & Rechberger, 2004). While this approach presents the advantage of being very "robust" (it will generally find a solution), it also presents some shortcomings that are described below.

As part of the ASTER project, a reconciliation methodology has been developed, whereby uncertain parameters are not represented by single probability distributions, but rather as "nested-intervals" (i.e., "fuzzy numbers"; Dubois and Prade, 1988; Dubois and Guyonnet, 2011). The reconciliation method consists in identifiying the areas of compatibility between flows and stocks, considering all mass balances and constraints on values (Dubois et al., 2012a; Dubois et al., in preparation).

3. RESULTS TO DATE

3.1 Data mining and disaggregation

A history of EU-27 imports and exports of REOs is presented in Figure 2. With respect to the initial EUROSTAT data, we verify that the quantities for the four HS code products in Table 1, sum up to 23 013 tons in 2008; i.e., the value presented in Öko-Institut (2011). It is reminded that the quantities presented in Figure 2 result from converting products to oxides, using the conversion factors in Table 1.

Note that a correction was brought to EUROSTAT data for the special case of Austria. This country has decided, for reasons of confidentiality, to no longer disclose its imports/exports of rare earths, as from 2009. We therefore assumed the same values for Austria in 2009, 2010 and 2011 as in 2008, the last year for which data are reported.

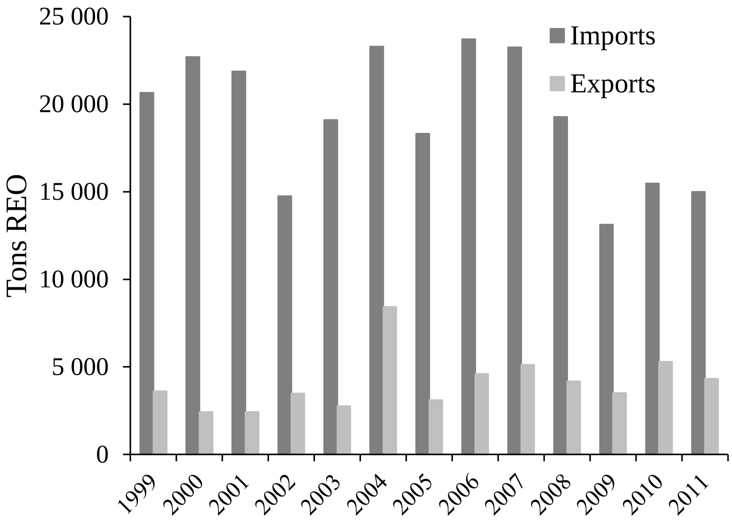

Figure 2. Estimated imports and exports of REOs in the EU-27, from 1999 to 2011 (based on EUROSTAT).

Figure 3 shows the proportion of imports into the EU-27 coming from China, as an illustration of the dependence on this producer.

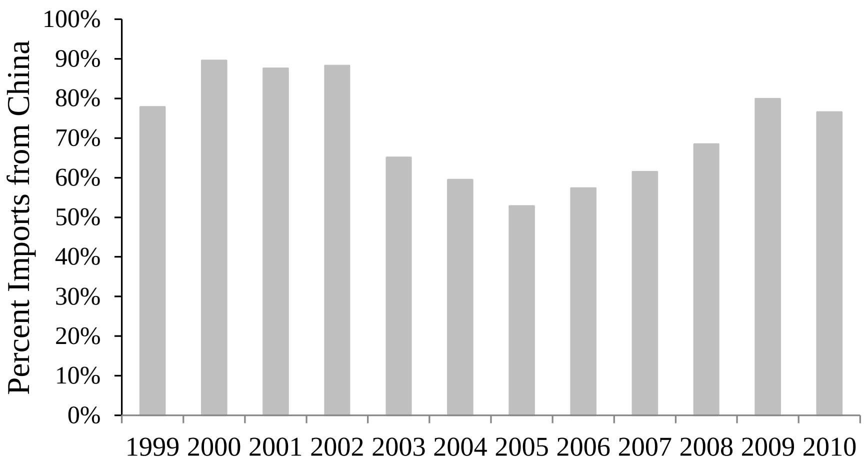

Figure 3. Illustration of the dependence of EU on imports from China

The distribution of REOs among the various market sectors for EU-27, was estimated based on Table 2.

Table 2. Estimate of relative REO consumption by market sector for Europe

Application	Magnets	Battery alloys	Metal alloys (excl. batteries)	Auto catalysts	Petroleum refining
% REO consumption	3.7%	7.6%	4.8%	27.3%	16.1%
Application	Polishing compounds	Glass additives	Phosphors	Ceramics	Other
% REO consumption	3.1%	18.6%	6.2%	6.2%	6.2%

REE requirements per application were based on Lynas (2010; see Table 1 of Jordens et al., 2013), with a correction for lamp phosphors according to Table 3.

Table 3. Typical composition for lamp phosphors

Oxide	La_2O_3	CeO_2	Tb_4O_7	Y_2O_3	Eu_2O_3
% REO	15	10	5	65	5

The disaggregation process yields results for the period 2006-2011 presented in Figure 4.

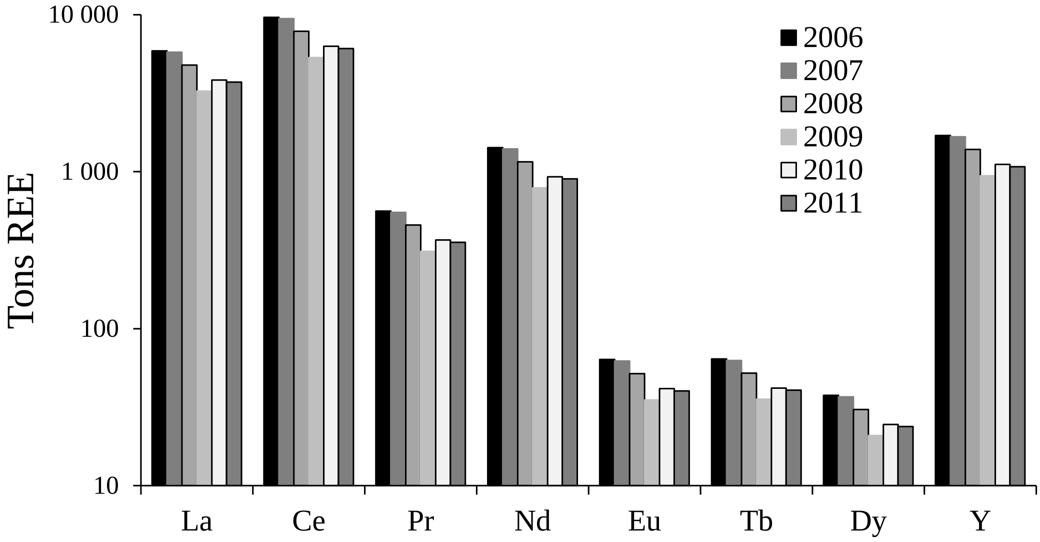

Figure 4. Estimated imports of REEs in the EU-27, from 2006 to 2011.

Data mining is ongoing with respect to the estimation of flows of REEs contained in manufactured products within the EU-27. Concerning in-use stocks of REEs, a first estimate can be obtained from the values proposed by Du & Graedel (2011a, b, c), considering the share of the EU-27 in the Gross World Product (21.4% in 2011). An advantage of this approach is that the data of Du & Graedel are constrained by world production data for permanent magnets etc.. Whereas estimating in-use stocks in the EU-27 by integrating imports and generation of products containing REEs over a period of time, is faced with the significant uncertainties relative to (i) the proportions of REEs in functional units (e.g. magnets) used in the products (e.g. audio equipment) and (ii) the proportion (weight%) of functional unit within these products. Using the GDP/GWP ratio, the in-use stocks of Table 3 are obtained.

Table 4. Estimates of in-use stocks of REEs in the EU-27, based on Du & Graedel (2011a)

REE	La	Ce	Pr	Nd	Eu	Tb	Dy	Y
In-use stocks (tons)	18 447	30 730	10 721	29 275	86	150	1 840	1 477

3.2 Data reconciliation in MFAs in presence of epistemic uncertainties

A methodology for data reconciliation under fuzzy constraints was developed and is described in Dubois et al. (2012a). In order to enhance the method's diffusion, a MATLAB code was programmed that implements the methodology. It has been applied to test cases and to a "real" MFA, described in van Beers et al. (2005), of flows and stocks of copper in the Australian economy. Figure 4 is an example comparing results obtained for the flow of waste to the environment and using two alternative methods. The first method relies on a classical representation of uncertain parameters using single probability (normal) distributions, defined by their average and standard deviation. Data reconciliation is obtained by minimizing an objective function expressed as the sum of squares of the differences between estimated and reconciled flows, weighted by their respective standard deviations. The second method relies on a possibilistic representation of uncertain parameters, using intervals with preferred values (resulting in triangular possibility distributions, or fuzzy numbers). For the comparison, the range around the preferred value was taken equal to the standard deviation used for the probability distributions.

Figure 4 compares flows before and after reconciliation, obtained using the two methods. In the probabilistic case, the reconciled average value is seen to be shifted with respect to the initial estimate. In the possibilistic case, the reconciled flow lies within the initial estimate, but has a lower maximum level of possibility (likelihood). This is an indicator of the level of consistency of the reconciliation. The advantage of the least-squares method is that it is very robust: a solution will be found. On the other hand, this solution may not be at all realistic. This is because normal probability distributions are defined between $\pm \infty$; hence a solution can be found in areas of very low probability (see Dubois et al., 2012b). With the second method on the other hand, if flows are incompatible, the method cannot find a solution, as there will be no interval of values for which all constraints of mass balance and parameter membership are satisfied.

Another drawback of the probabilistic method is the justification of single normal probability distributions, when faced with incomplete/imprecise information. As argued by Dubois and Guyonnet (2011) and others, expert judgment is often better suited to possibilistic representation (nested intervals) than an arbitrary selection of single probability distributions.

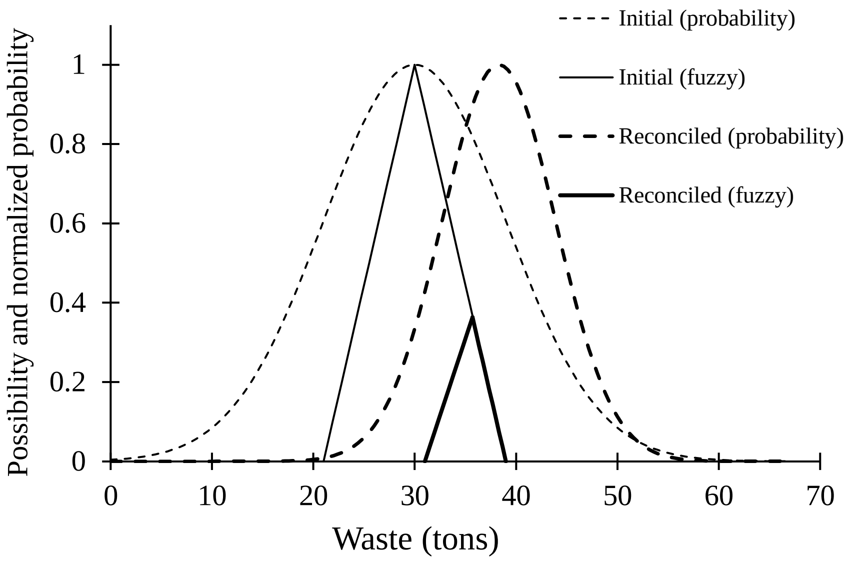

Figure 5. Comparison between initial and reconciled data, for an exemple flow (waste), using two alternative modes of uncertainty treatment and data reconciliation: probability and least-squares minimization versus possibility and fuzzy reconciliation

5. CONCLUSIONS AND FURTHER PROSPECTS

The primary aim of the ASTER project is to perform a material flow analysis (MFA) for rare earth elements in the EU-27. While the project is at its first year, some interesting results have been obtained. Building upon previous work, such as the Öko-Institut report (2011), results presented in this paper show a history of EU-27 imports and exports since 1999. A methodology for disaggregating this data into individual REEs, based on previous work, provides estimates of specific REE imports and exports.

Also, the issue of data reconciliation in material flow analysis has been examined, with a critical evaluation of the classical approach of probabilistic data representation and least-squares reconciliation. An alternative method, based on possibility theory, has been proposed and implemented in an ergonomic MATLAB environment. It is projected to apply the proposed approach to the material flows of rare earth elements in the EU-27.

Results highlight the enormous potential of lithospheric stocks in continental Europe, compared to in-use stocks. However, this should not be seen as justification to reduce efforts with respect to secondary (recycled) resources, as recycling is essential in order to (i) avoid emissions related to primary resource development and (ii) reduce waste streams and elimination. Considering current REE consumption growth rates (on the order of 5% annually; Alonso et al., 2012), reliance on primary sources is unavoidable, while secondary sources are an essential complement in order to increase resource eco-efficiency (see e.g. Grosse, 2010; Guyonnet al., 2011; Reuter et al. 2012).

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