

Article

Shedding Light on the Anthropogenic Europium Cycle in the EU–28. Marking Product Turnover and Energy Progress in the Lighting Sector

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Abstract: Phase-out strategies for incandescent bulbs in favor of advanced energy-efficiency lighting systems such as fluorescent lamps and solid-state technology have considerably reduced the energy use for lighting, but have also resulted in dependence on many critical materials like rare earth elements and shifted the attention to sustainable use and recovery of resources. In this work, a dynamic material flow model was developed to analyze the socio-economic metabolism of europium in the EU–28. The analysis shows that europium marked product turnover and progress in lighting efficiency, with this element being employed both in traditional and novel lighting technology to provide luminescence. The results also demonstrate that the current anthropogenic reserve could constitute an attractive source of secondary europium with substantial potentials for environmental benefits. However, nonexistent recycling and market forces hinder strategies for material circularity. In particular, the transition from fluorescent lamps to solid-state technology is quickly decreasing the demand for europium. This trend adds further constraints to the creation of a sustainable recycling industry for europium, with primary sources that might remain the preferable route to supply phosphors to future lighting systems.

Keywords: fluorescent lamps; light emitting diodes; energy efficiency; cathode ray tubes; in-use stock; REEs

1. Introduction

Lighting accounts for 19% of electricity consumption globally and 12% in the EU [1–3], in which the total domestic lighting consumption is expected to rise up to 100 TWh by 2020 due to growing wealth [4]. End-use energy efficiency lighting is a key component of the EU goals of contrasting climate change [1] and ambitious targets for reducing energy consumption (20% by 2020) improving energy efficiency (30% by 2030) which were adopted by the European Commission [5]. As a consequence, traditional incandescent light bulbs, which have long been produced but with very low efficiency, were subject to phase-out strategies in many countries (e.g., Japan, EU–28, Russia, Korea, China, USA, Australia) [6]. In contrast, the greatest energy efficiency investments in lighting were devoted to fluorescent lamps—from 2003 to 2007, the apparent consumption of compact fluorescent lamps (CFLs) increased by more than 400% as a consequence of incandescent bulb phase out [7]—and, more recently, to solid-state lighting (SSL) systems like light emitting diodes (LEDs) and organic light emitting diodes (OLEDs) [8]. Compared to incandescent lamps, in which visible light is given by a wire filament heated at a high temperature, the most energy-efficient lighting technologies are based on phosphors to exhibit luminescence. In fluorescent lamps, the phosphor powder coating the glass

provides bright color by converting the UV radiation, emitted from the interaction between mercury and electrons, to visible light [9]. In SSL technology, instead, light is emitted when electricity passes through a semiconductor chip containing phosphors [10]. The luminous efficacy (i.e., the ratio of the light output to the power consumed) of incandescent bulbs is in the range 16–25 Lm/W, with only 5% of the electrical power consumed being transformed into light, while the remaining 95% is dissipated as heat [7]. In contrast, the luminous efficacy fluorescent lamps and LEDs is in the order 50–80 Lm/W, with LEDs having a theoretical limit higher than 200 Lm/W [11].

Lighting standards are deemed to have considerable potentials for further energy savings [7,12], but are tied to a dependence on several critical resources [13] like rare earth elements (REEs) that are contained in the tricolor phosphors. The main set of REE phosphors is based on yttrium, lanthanum, cerium, europium, and terbium. Of most interest here, europium is utilized to provide both red color and blue color phosphors [14], and it is also employed in white LEDs [11]. However, despite the fact it was named after the continent of Europe [15], europium is entirely imported in the EU–28 [16], making the Member States highly dependent on foreign countries. This import reliance marks a potential vulnerability to supply restrictions [13]. Europium criticality is also increased by the absence of potential material substitutes [17], issues related to primary production including low crustal abundance—europium is one of the scarcest REEs in the Earth’s crust [18]—the “Balance Problem” [19], and the Chinese monopoly. Primary deposits of REEs in Europe were discovered in Greenland and Sweden, but the lack of beneficiation and early-stage processing capability hinders domestic extraction [20,21].

Resource efficiency practices are key levers for reducing elemental criticality [22,23]. Among others, enhancing end-of-life recycling would secure a sustainable supply of critical resources. In the case of europium, however, end-of-life recycling is almost nonexistent [13,17]. Market forces drive strategies for sustainable resource management like material recovery and recycling, but the generation of scrap at end-of-life is challenging to forecast. In this perspective, the characterization of anthropogenic material cycles by means of material flow analysis (MFA) techniques enables us to estimate material demand, scrap supply, and resource accumulation in modern societies [24–26]. The spectrum of materials analyzed by MFA practitioners is increasing. Metals used in large quantities were the most investigated elements [24,27], but REEs and specialty metals are expected to play a key role in promoting low-carbon technology so that developing MFA models will provide knowledge-based information to analysts and policy-makers. At the time of writing, literature works focusing on europium from a material flow accounting perspective are few. Du and Graedel [28,29] provided the first estimate of the global europium cycle, while BIO by Deloitte [16] and Guyonnet et al. [30] carried studies out for the EU–27. These studies discussed europium stock and flows in fluorescent lamps, which constituted the basis for future demand and supply projections [31].

In this work, we developed a dynamic MFA model of europium in the EU–28 from 1990 to 2016 and provided the first comprehensive analysis on historical flows and in-use stock of this element. The main end-uses of europium were covered in the analysis, including cathode-ray tube (CRT) TVs and monitors, flat panel displays, small information technology (IT), fluorescent lamps and LED technology, which are increasingly applied in automotive and general lighting. The potentials for end-of-life recovery and for closing the europium cycle in the EU–28 are commented. In this view, the importance of life cycle assessment (LCA) for addressing issues associated with the environmental sustainability of REE recycling was previously asserted [32]. Aligned with this recommendation, a follow-up assessment was aimed at determining first-order estimates of annual potentials for energy savings and greenhouse gas (GHG) emissions reduction associated with sources of secondary europium in the EU–28. Lastly, a retrospective on europium demand and stock accumulation and their implications on material and energy efficiency in lighting is discussed in the results. This work is part of a paper series focusing on selected metals in the EU–28 deemed relevant for interlinkages between the metal supply-demand dynamic and energy systems [33,34]. We expect that the outcomes will be

informative for readers from the climate change and critical materials policy communities as well as to the lighting and electronics industries.

2. Materials and Methods

The anthropogenic cycle of europium was divided into its main life cycle stages including ore mining, production (consisting of beneficiation processes, metal refining and creation of semi-finished products), manufacture of finished products, use, end-of-life collection and sorting. The MFA model was built to account for all flows in europium metallic equivalents and to extend the analysis from 1990 to the most recent year possible (i.e., 2016). A detailed description of the accounting methods was previously reported [33]. Roskill Information Service [35] provided the historical europium demand for domestic production of europium-based phosphors and pigments, which are the precursors of almost all europium applications. Phosphor precursors have the same composition of lamp phosphors, but a different mineral structure. Further processing is required to achieve the desired form suitable for lamps. The average market composition of the tricolor phosphors is 55% red, 35% green and 15% blue phosphors [9], although individual mixtures are often manufacturer-specific.

Based on EU statistics of production and trade records (i.e., Prodcum, Comext) [36,37], we compiled a list of products containing europium (see Table S1 in the Supplementary Material). In the first approximation, we have assumed no losses of europium from the manufacture of semi-finished and finished products. Also, stockpiling at production facilities was assumed to be negligible [30]. We reviewed the existing literature to set mass percentages and express the results in europium content. The selected products were then aggregated into seven end-use application segments including automotive, CRTs (TVs and monitors), flat panel displays, small IT (e.g., smartphones, laptops), luminaires, fluorescent lamps, and LED lamps. The spectrum of miscellaneous applications of europium is quite wide and includes other uses such as anti-forgery marks in Euro bank notes, neutron absorber and control rods in nuclear reactors, and use in the optic industry [13,35,38,39]. However, due to a lack of information, these end-uses were not considered in this study.

Penetration rates of europium-containing products were considered and modeled to account for the use of europium-based phosphors and pigments in end-use markets (see Table S1 in the Supplementary Material). For instance, laptops and notebooks used to incorporate fluorescent lamps for backlighting illumination, but shifted towards LED systems more recently. We assumed a market penetration rate of 100% for europium-based lamps for retrofitting laptops and notebooks, but europium contents were set assuming that LED backlighting increased from 1% in 2005 to 92% and 99% in 2010 and 2016, respectively [11,40]. In many cases, records are reported in official statistics from 1995 to today. This time span exceeds the average lifetime of europium's end-uses considered in this analysis so that, with the exception of CRT TVs and fluorescent lamps, the initial stock of europium contained in products in use was assumed to be negligible in 1995. In general, new products on market that replace obsolete technology show an early growth period after which the demand may evolve according to different patterns [41]. For CRT TVs, because phosphors were being used for backlighting long before 1990, the initial stock was determined assuming that europium put on market in 1980 equaled 75% of CRT TVs put on demand in 1995 and a constant growth rate for 1980–1995. Similarly, for fluorescent lamps, the initial stock was set equal to zero in 1985, while a constant growth rate was applied for 1985–1995.

Annual net-inflows of europium to use (or apparent consumption, namely manufacture production—export and import) constitute the input to the dynamic stocks and flows accounting model. More precisely, given the amount of europium entering the use phase by application and the residence of products in use, the generation of europium flows at end-of-life and the annual accumulation in the anthropogenic reserve, or in-use stock, were determined. Finished products containing europium can be considered as protected environments so that dissipation during use of europium to the environment was considered to be nonexistent [42]. Table S2 in the Supplementary Material lists the parameters

utilized for the stocks and flows dynamic modeling. End-of-life collection rates were applied to annual europium outflows from use (see Table S3 in the Supplementary Material) to distinguish whether europium loss occurs during waste collection or subsequent processing.

Mapping anthropogenic material cycles often requires tackling issues related to data availability. The MFA model for europium in the EU–28 was filled with information from reliable sources in the literature while missing flows were estimated to satisfy the mass balance. However, some degree of uncertainty affects all MFA models and propagates to the results. For the model created, lower and upper bound values for the market penetration rate of europium-based goods and the europium contents in finished products (see Table S1 in the Supplementary Material) were used to run the uncertainty analysis. Sensitivity analysis and a comparison with historical data series on waste electrical and electronic equipment were also carried out to evaluate the reliability of the main outcomes.

The in-use stock constitutes a potential source of secondary europium. To what extent increasing europium recovery and recycling could result in first-order energy savings and GHG emission reduction was explored by combining LCA with elemental information provided by the MFA model developed. To this aim, two life cycle inventories were compiled to model the recycling of europium from spent fluorescent lamps on the basis of literature estimates and expert survey. More in detail, theecoinvent process “disposal, fluorescent lamps” [43] was referred to for the treatment and disposal of fluorescent lamp modeling (dry) dismantling, cutting, blowing and air exhaust cleaning. This is a multi-output process and delivers co-products, namely “disposal, fluorescent lamps”, “mercury, from fluorescent lamp treatment, at plant”, “glass cullet, from fluorescent lamp treatment, at plant”, “rare-earth activated phosphors, from fluorescent lamp treatment, at plant” and “secondary metals, from fluorescent lamp treatment, at plant”. By default, 100% of environmental impacts are allocated to the disposal process.

However, only energy requirements and GHG emissions attributable to europium are of interest in the analysis. The International Organization for Standardization guideline 14,044 defines specific procedures for multi-output processes, with avoiding allocation as the preferable option [44]. Because allocation cannot be avoided here, and mass allocation is not the best procedure since the REE phosphor powder is contained in small quantities, an economic allocation was chosen to disaggregate the environmental burdens among the valuable outputs of the process. Energy required for the manufacture of electronic equipment is often more closely related to cost than to weight [45]. The market price of individual material streams was used as a value parameter for the allocation. Glass, aluminum, and the REE phosphors were considered the only valuable co-products of the process. No existing databases provide the life cycle inventory for supplemental energy requirements and GHG emissions released for extracting and purifying europium from the rare earth-activated phosphors concentrate. Together with yttrium, europium recovery from spent fluorescent lamps is reported to require weaker leaching solvents and temperatures than cerium-, lanthanum-, gadolinium- and terbium-based phosphors [46]. The only study found in the literature that applied LCA to europium recovery was based on lab scale processes [47], which are usually more intensive in both material and energy requirements than optimized scaled-up plants.

To our knowledge, the Solvay Loop Life Project [48] is the only process tested at full operating scale for the recovery of REEs from spent fluorescent lamps. It applied chemical attack and liquid–solid extraction steps to separate and concentrate the REE phosphor powder. Then, thermal treatment, filtration, washing, nitric acid attack, and REE separation stages were executed. Precipitation, filtration and calcination steps finalized the recycling of REEs. Based on expert survey [49], we set the plausible range of electricity consumption for the full-scale recovery and recycling of europium between 50–100 kWh/kg Eu recycled. This is a simplification, as it assumes that REEs are recovered at similar rates and completely from the phosphor powder. Additional energy inputs may also be needed to supply solvents and the technology infrastructure. The energy-related GHG emissions were computed using the European electricity production mix. The “Eu LCA Supplementary Material”

spreadsheet and Table S4 in the Supplementary Material summarize the mass quantities, market prices and the resulting cumulative energy demand (CED) and global warming potential (GWP) utilized in the analysis.

3. Results and Discussion

3.1. The Contemporary Europium Cycle in the EU–28

As REE mine extraction is nonexistent in the region, total europium supply in the EU–28 comes from import. However, some early stage processing for the separation of mineral mixtures into individual rare earth oxides and the production of phosphors and pigments occur in this region [31]. Overall, the EU–28 market penetration of europium in phosphor and pigment production was estimated to cover less than 5% of the global demand [35]. In recent years, the penetration of this sector in the total europium demand has reduced in favor of other specialty applications (see Figure S1 in the Supplementary Material). This trend may be a consequence of commodities that use less amounts of europium per unit of products, notably LED lamps, with respect to fluorescent lamps. The total domestic demand for europium in phosphors, pigments, and other specialty applications summed up to about eight metric tons of europium metallic equivalent (t Eu) in 2016, while the estimated input of europium to the European manufacture of finished goods resulted in 23 t Eu in the same year. Comparing the two flows, around 15 t Eu in the form of europium oxide, phosphor powder, semiconductor chips, and other semi-finished products were net-imported by domestic manufacturers. About 80% of the europium inflow was employed in fluorescent lamps, followed by luminaires. CRT TVs and monitors, automotive, flat panel displays, small IT and LED lamps made up the remaining 2%.

A cross section from manufacture to end-of-life management of the contemporary europium cycle in the EU–28 is displayed in Figure 1. All stocks and flows are representative of the year 2016 and are expressed in metric tons of europium metallic equivalent (t Eu). Europe used to be a producer of fluorescent lamps, but in the late 1990s, the production shifted to Asian countries (mainly China), increasing competition in the lamp market [50]. After years in which fluorescent lamps were net-imported in Europe, in 2016 the EU Member States net-exported CFLs for the equivalent amount of 1.4 t Eu. Automotive was the other end-use segment in which europium export was greater than import. Net-import of europium-containing products in the region characterized luminaires, monitors, displays, small IT, and LED lamps instead. The balance between imports and exports was approximately 1 t Eu in 2016, with the total flow into use resulting in 24 t Eu.

The dynamic MFA model simulated the accumulation to the in-use stock and the generation of europium at end-of-life over the time span considered. For 2016, the model estimated the net-depletion of the in-use stock at about 25 t Eu, while total europium at end-of-life resulted in 47 t Eu. Applying product-specific end-of-life collection rates, about 20 t Eu could have reasonably been recovered from spent fluorescent lamps in the EU–28 in 2016. Supplemented with an additional 5 t Eu deriving from obsolete CRT TVs and monitors, flat panel displays, small IT, luminaires, and LED lamps, this amount would sum up to 25 t Eu. However, no recycling procedures for europium were operating in 2016, with this element being lost during the disposal of waste material streams or dispersed as a tramp element in other material cycles.

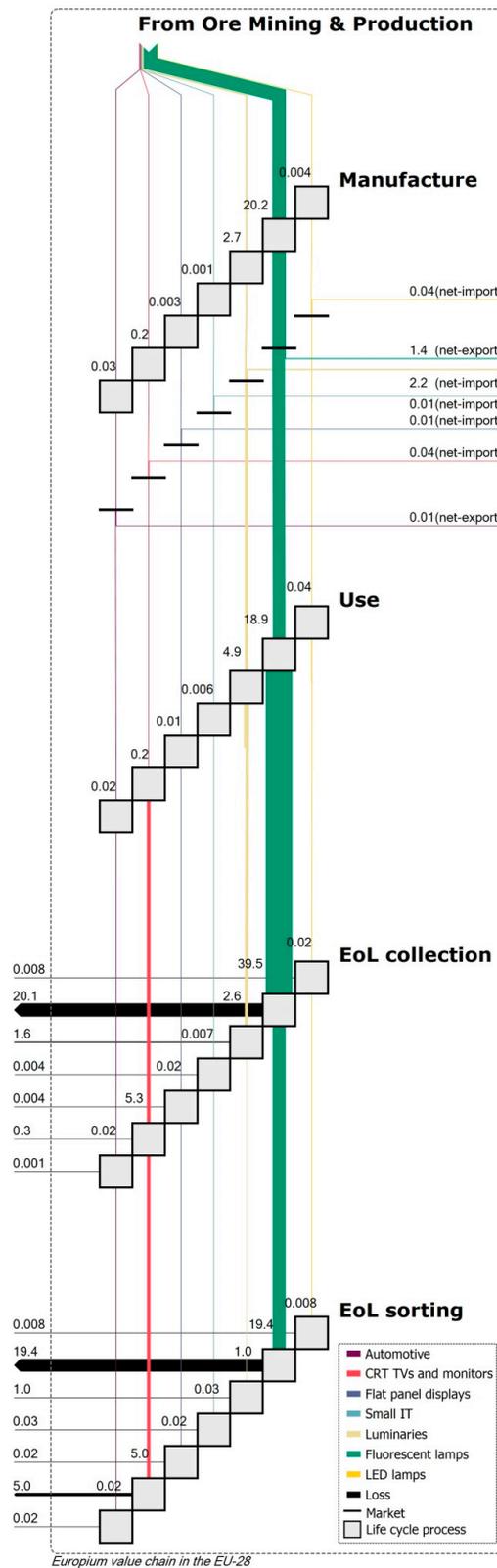


Figure 1. Cross section from manufacture to end-of-life (EoL) management of the 2016 europium cycle in the EU–28. Values are in metric tons of europium metallic equivalent. CRT = cathode-ray tube. IT = information technology. LED = light-emitting diode.

3.2. Potentials for End-of-Life Recovery and for Closing the Europium Cycle in the EU-28

The implementation of the circular economy model adopted by the European Commission [51] includes driving material circularity into anthropogenic cycles and setting essential conditions for end-of-life recovery and recycling. In this view, obsolete CRT TVs and monitors and spent fluorescent lamps are the two main end-uses by overall europium generated at end-of-life, while luminaires follow and the remaining applications are negligible (Figure 2 and Table S5 in the Supplementary Material). However, while CRT TVs and monitors are going to be less relevant in the future, fluorescent lamps will still constitute the main potential source of secondary europium in the coming years. Fluorescent lamps have short lifetimes [10,30,52,53] so that product turnover occurs quickly and the delay between inflow and outflow is relatively short, giving this application segment the greatest potential for europium recovery. The accuracy of estimation of material outflows from use depends on the reliability of the model developed. Uncertainty caused by different lifetime models may affect the outcomes of dynamic models [54–56]. In addition, product lifetimes might change over time. However, these changes are negligible for fluorescent lamps [52]. A sensitivity analysis (see Table S6 and Figure S2 in the Supplementary Material) showed that the estimates of this study are relatively robust and the main findings are not particularly impacted by different lifetime distribution parameters. Also, the estimation of europium-containing products out of use scaled by population and gross domestic product were compared with similar data from the Netherlands [52] and Italy [57] and attested good consistency between these results.

The common management route for spent fluorescent lamps collected at end-of-life begins with the removal of electrical contacts and metal ends from the lamps. Then, a series of physical processes (i.e., crushing, sweeping and sorting) is carried out to liberate individual material streams. The fluorescent powder containing phosphors is blown out from the glass surface by high-pressure air and heated at about 450 °C to remove mercury through desorption and distillation [9,58]. The only valuable material flows recovered are metals (e.g., aluminum), the glass and the electronic components. The resulting mercury-free fluorescent powder is a non-hazardous waste product and, as such, it can be disposed at a lower cost [59]. Policy regulations on mercury removal are driving efforts to improve the end-of-life management of fluorescent lamps. Additional momentum to collection and process efficiency could also derive from a growing interest in securing the supply of critical materials like europium. Spent fluorescent lamps are predicted to contain up to 25,000 t of REEs by 2020 [58], so the existence of management schemes for fluorescent lamps at end-of-life could supply substantial amounts of europium scrap and foster the implementation of dedicated recovery and recycling practices for europium.

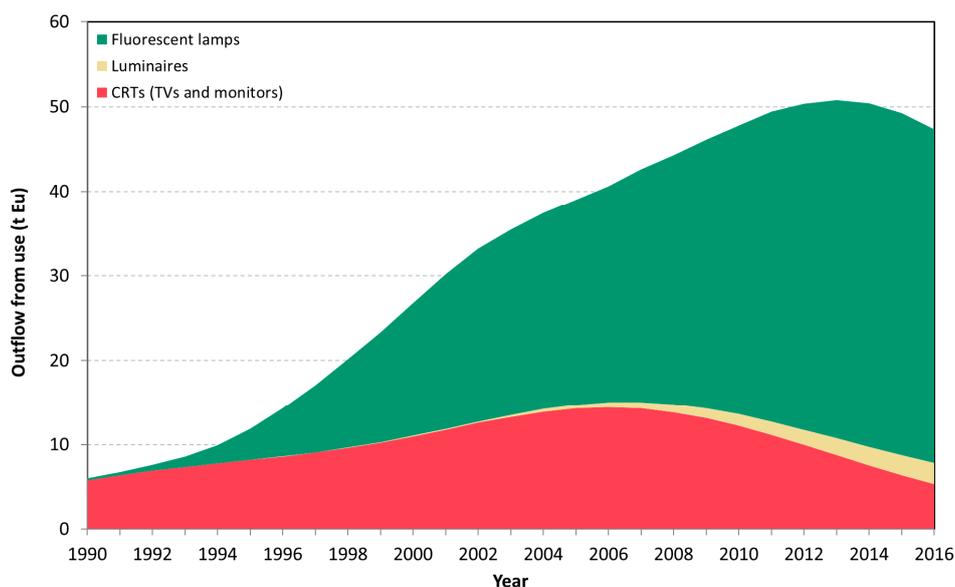


Figure 2. Historical europium outflow from use by selected end-use application segments.

Industry and academia are exploring processes for the recycling of REEs from spent phosphors, which might impact future supply-demand dynamics of europium. Binnemans and Jones [60] discussed three possible recycling routes for the phosphor powder from spent fluorescent lamps including (i) direct re-use of the recycled lamp phosphors, (ii) recycling of the various phosphors by physiochemical separation and (iii) chemical extraction of the phosphors. Quality issues and dedicated manufacture-based take-back systems would need to be considered for the first two options [10], making their direct use in the manufacture of new lamps only theoretical. Thus, despite being the most energy intensive process out of the three possible recycling approaches, the extraction of individual REEs by hydrometallurgy is commonly adduced as the preferable route to recover REEs [46,60]. The sequence of operations to extract europium from the mercury-free phosphor powder generally follows a series of chemical attacks such as acid or alkali digestion methods to dissolve REEs. These procedures, however, often undergo multi-stages and produce large volumes of waste streams that require further treatment. Once in solution, selective REE recovery from the concentrate can be carried out through, for instance, extraction (e.g., solvent extraction, ionic liquids extraction, supercritical fluid extraction), precipitation, electro-winning, or chromatography techniques [58].

REE recovery rates reported in the literature are generally >80%, but most research is carried out at a lab scale and the phosphor powders analyzed are often artificially prepared. The presence of impurities can affect the “real” spent fluorescent powder and would likely decrease the efficiency of the recycling procedure [9]. Based on the recovery performance of a full plant for REEs from spent fluorescent lamps, Machacek and colleagues estimated the REE separation and recovery efficiency rate at about 60% [10]. Multiplying this yield with end-of-life collection and sorting rates of spent fluorescent lamps in Europe (i.e., approximately 50%, based on [61]), the overall end-of-life recycling rate (EoL-RR) would be 30%. In the Circular Economy perspective, with an EoL-RR of 30%, the amount of europium potentially recyclable from domestic spent fluorescent lamps would approximate the European demand for europium in phosphor and pigment production at 2016 levels. Extending this calculation under the hypothesis that the end-of-life collection rate of spent fluorescent lamps increased near to 100%, the amount of europium potentially recoverable could equal the entire europium inflow to use in this region at 2016 levels. Thus, even modest improvements in the EoL-RR of europium would enable us to cover substantial fractions of the annual demand in the EU-28 by domestic secondary sources.

From the view of material circularity, a full sequence of manufacturing capacity would ensure the outlet to domestic producers and reduce the metal criticality of a country or a region [17,62].

However, while margins for increasing the annual domestic production of individual REEs has been reported [31], the results of this study demonstrated that the current manufacture of semi-finished products based on europium is considerably lower than the demand from domestic users so that more than 60% of europium inflow to use was imported in the form of semiconductor chips and other desired products in 2016. The preference for LED and OLED products, the production of which occurs mainly in Asian countries [35], further challenges the enhancement of the domestic manufacture industry. Thus, achieving efficient recovery and recycling of europium at end-of-life without enhancing the domestic manufacturing capability might only result in increasing exports of valuable forms of secondary europium from Europe, but no reduction in the regional import reliance nor in its criticality rate might occur.

Beyond preserving the natural capital and laying the base for the closure of the europium cycle, recycling would also offset the environmental impacts associated with primary europium supply. Globally, about half of current GHG emissions embodied in metal losses from phosphors at end-of-life were attributed to europium [63]. Applying the GWP and CED values defined in Table S4 in the Supplementary Material to the 2012–2016 average europium outflow from use in the EU–28, the annual budget of potential energy savings and GHG emissions reduction would amount to about 275 (92–554) TJ and 14 (5–27) ktCO₂eq, respectively. The resulting percent energy savings and carbon reduction are >85% compared to primary europium, aligning these results with literature estimates for other metals [64].

Although the technological feasibility and environmental sustainability of europium recovery seems plausible, the economical preference of recycling is still unsure due to several reasons. First, the decrease of europium prices experienced since 2011 has affected the financial feasibility of the recycling industry. The total cost for europium recycling from spent lamps includes costs for collection at end-of-life and further processing for elemental recovery. In addition, the grade of europium recovered, reagent costs, energy consumption, inefficiency during end-of-life collection and sorting, mandatory recycling quotas based on the mass of product, and decreasing concentrations in products are limiting factors in the recovery efficiency process [10,47,60,65]. Second, the transition from fluorescent lamps to SSL technology is expected to make the demand for europium peak and then decline [66] because the quantities of REEs required per unit of SSL are up to some orders of magnitude lower than those needed to produce fluorescent lamps. This trend could lead to an oversupply of primary europium compared to its future demand, which in turn would add further constraints to secondary material recovery. In the next section, historical trends of europium inflow and stock changes are analyzed and discussed to provide evidence about the future demand and supply for europium in the EU–28.

3.3. Europium as a Marker of Product Turnover and Progress in Lighting Efficiency

The specialty properties of europium have been crucial in technological advancements in lighting. For instance, the breakthrough of color televisions and screens was possible thanks to the use of europium as no bright phosphors were available for the red color until that time [39]. Then, CFLs made a revolution in efficient residential lighting. However, although CFLs were invented in the 1970s, a wider scale adoption started only after the 1990s, when the performance and cost of CFLs improved remarkably. CFLs needed to undergo several technological advancements and market changes before they were largely accepted by consumers [50]. Over the time span analyzed, the apparent consumption of europium increased from 10 t Eu in the 1990s up to a peak of 60 t Eu in the late 2000s, mainly driven by fluorescent lamps (Figure 3, see also Table S5 in the Supplementary Material). The penetration of fluorescent lamps in the lighting sector has been supported by government actions to mitigate climate change and increase energy efficiency [6]. Today, the progress towards more efficient lighting standards is fostering a worldwide market transition from fluorescent lamps to SSL [67]. Furthermore, national legislations aiming to eliminating hazardous substances add primary motivations to shift from fluorescent lamps to mercury-free lighting technologies such as LEDs and OLEDs.

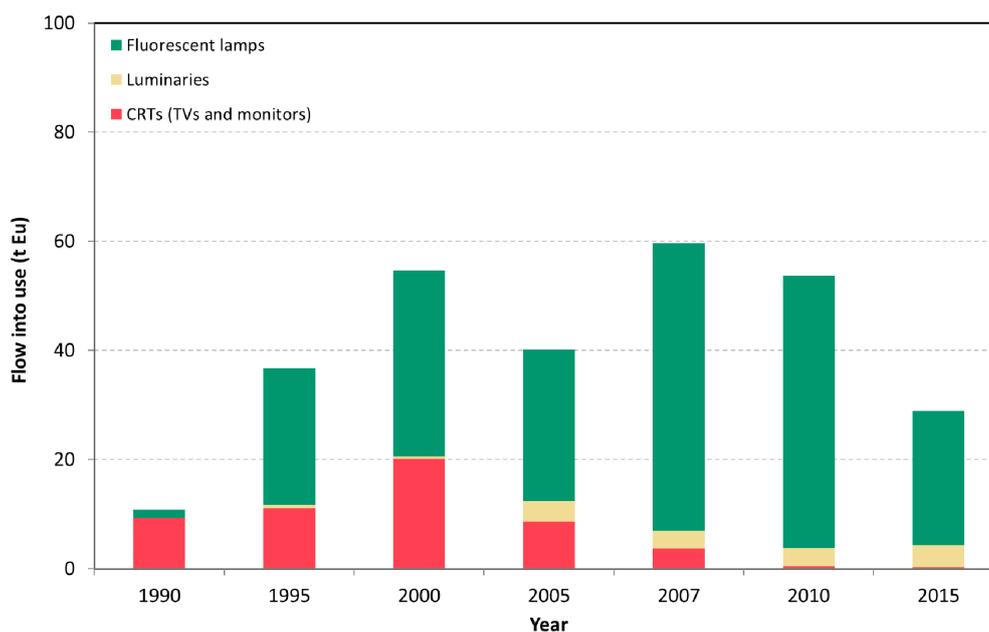


Figure 3. Europium flow into use by main end-use application segments for selected years.

SSL systems are attractive because of their longer lifetime and versatility of uses. LEDs and OLEDs can produce white light in different color tones (from warm to cold) and offer many different solutions for lighting, enabling, for instance, full integration into furniture elements [1]. This versatility has driven LED technology to spread out from traditional end-use segments (i.e., automotive and road lighting) to displays and TVs, and very recently to general lighting, which is the largest lighting market by revenue. Automotive represents about 20% of the LED market, while backlighting is shrinking compared to previous years due to an already very high market penetration of LED retrofitting, lower sales for liquid crystal displays and monitors, and forecast for OLED-based products [67]. Some challenges remain to be tackled before full market saturation by SSL technology. For instance, previous analyses suggested that, similarly to the historical uptake of CFLs, SSL systems would need a relevant cost reduction before becoming competitive on the market [50]. In addition, although some LED products are manufactured with good quality, many LEDs on market are still unable to provide satisfying variations of, for instance, correlated color temperature and color-rendering indexes, so improving their lighting properties is a priority. Anyway, as shown in Figure 3, the demand for europium in fluorescent lamps is decreasing considerably. The results provide evidence that the europium inflow to use has almost halved over the last decade, confirming some previous expectations on the decline of the demand for europium in Europe [31,35].

As material stocks are often a better proxy for services to people than material inflows [68], the cumulative reserve of secondary europium can provide a complementary insight into both europium demand dynamics and their effect on energy use for lighting in the EU-28. Figure 4 shows the evolution of the europium in-use stock in the EU-28 (see also Figure S3 in the Supplementary Material). The accumulation process underwent a rapid increase from 66 (55–75) t Eu in the 1990s to near 405 (188–641) t Eu in 2011, mainly driven by CRT TVs and monitors, and fluorescent lamps. This is a pattern common to other product stocks. As discussed by Chen and Graedel [41], in many cases, product stocks increase quickly for a given period of time after the product is introduced into the market. Then, different “wave” patterns may occur depending on the dynamics of market saturation. These authors cited CRT TVs and monitors as examples of products showing a definite pattern of their stock and undergoing a replacement to flat panel displays, which contain a lower amount of europium per unit of product. Europium is functional for both products, but it is also an essential component in the most advanced lighting systems so that this element can constitute an interesting marker of product turnover in lighting. The transition from fluorescent lamps to LED/OLED technology, which is

more efficient, more durable and devoid of mercury, is likely reflected in the net-depletion of the in-use stock in recent years, which has reduced the amount of europium stocked in the anthropogenic reserve by about 329 (137–545) t Eu in 2016. This estimate is aligned with the results presented by Guyonnet and colleagues for the EU–27 (i.e., 200 t Eu in 2010) [30] and Du and Graedel for the world (400 t Eu in 2007) [28,29]. Also, a detailed estimation of electrical and electronic equipment in stock in the Netherlands was provided by Wang and colleagues [52]. Applying the europium mass contents used in this work to the amount of fluorescent lamps accumulated in the Dutch stock, about 0.7 (0.3–1.0) g Eu/inhabitant would result. These values are close to our estimate of 0.5 (0.2–0.8) g Eu/inhabitant for fluorescent lamps in stock in the EU–28.

The retrospective on the in-use stock provided in this study enables complementary insights on the historical relation between europium inflows, stock changes, and the goal of increasing energy efficiency in lighting. More specifically, in Figure 5, the energy use for residential lighting in the EU–28 [69] is plotted against the in-use stock of europium-based lamps (i.e., fluorescent lamps and LED lamps). Overall, the energy used for residential lighting has likely increased until the point when fluorescent lamps have replaced incandescent lamps. As CFLs approached the saturation of the lighting market, the energy use for lighting decreased. In recent years, the uptake of SSL technology, mainly driven by LED systems, combined with the outflow of discarded fluorescent lamps from use is reducing the in-use stock, but no apparent changes in energy use for residential lighting are noticeable. Depending on how quickly SSL technology will be taken up by consumers and substantial progress in lighting performance will be achieved, LEDs and OLEDs have interesting potentials for maximizing the effect of energy efficiency progress by reducing the energy use for residential lighting, the demand for europium, and its net-accumulation in the in-use stock.

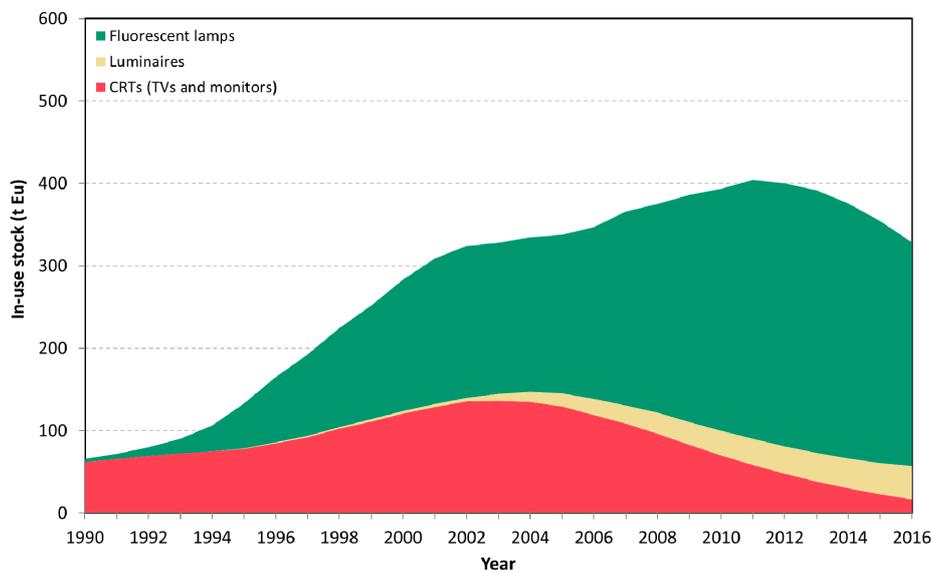


Figure 4. The anthropogenic reserve (or in-use stock) of secondary europium in the EU–28. Cumulative results.

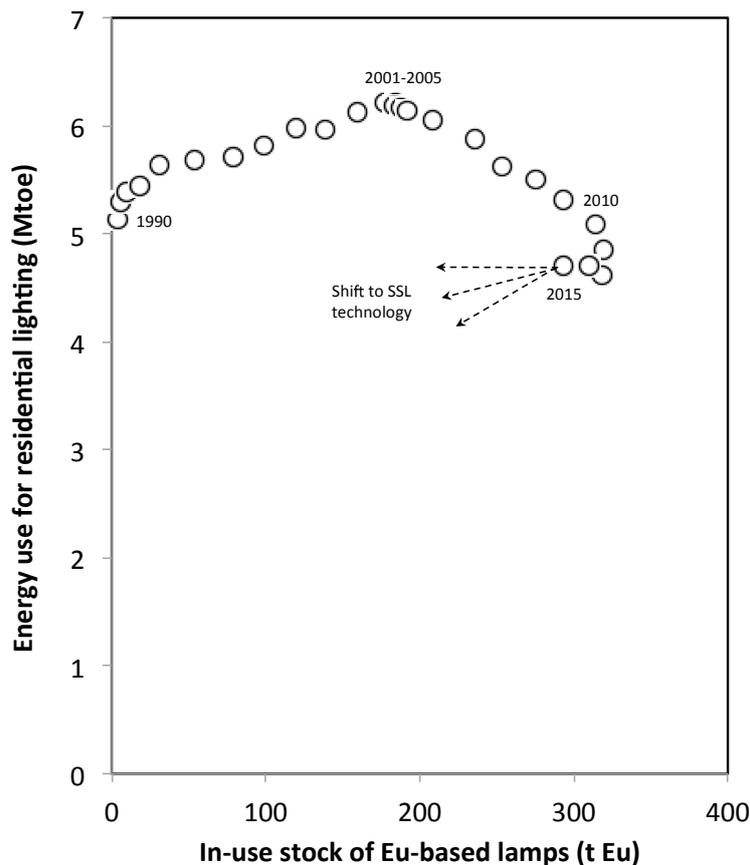


Figure 5. Energy use for residential lighting [69] versus in-use stock of europium-based lamps (including fluorescent lamps and LED lamps). SSL = solid-state lighting. Mtoe = million tons of oil equivalent.

Providing a better service to users with less resources is at the core of measures of environmental sustainability like dematerialization [70]. On the elemental level, the results show that a reduction of the europium in-use stock has occurred along with the adoption of more energy efficient lighting technology. It has been argued that dematerialization is an effective strategy of resource efficiency enhancement and environmental conservation only if qualitative inputs remain constant [22]. However, SSL technology demands more materials than traditional lighting systems due to heavier weights and the presence of electric components. Despite this, most of the environmental impacts in lighting are related to the use phase [53,71], so accounting for energy inputs to the production of lighting systems seems to have marginal effects on the main findings. Energy efficiency achievements and the shift towards low-carbon electricity systems will mitigate climate change [12], but unexpected rebound effects resulting from re-spending of cost savings on more energy intensive goods and services might erode the potential resource gains [72] and require further investigation.

4. Conclusions

This work provided a first detailed analysis of the anthropogenic cycle of europium in the EU-28, covering its main end-use application segments over multiple years, which increased the knowledge about the global socio-economic metabolism of this element. Uncertainty and sensitivity analysis showed a good reliability of the model developed, which might serve as a basis for further studies aiming at improving the overall accuracy and precision of the estimation of europium flows and stock, particularly in light of the takeover of fluorescent lamps by SSL systems. To the same goal, the inventory of products containing europium, their market penetration rates as well as plausible metal contents were made accessible to the readers in the Supplementary Material.

The results showed that the EU–28 is a main apparent consumer of europium-based products. Recovering and recycling europium from products at end-of-life, like spent fluorescent lamps (by far the most interesting source of secondary europium), could theoretically secure a sustainable supply to European manufacturers at a preferable environmental burden compared to primary sources. The existing end-of-life management of spent fluorescent lamps for mercury removal could speed up the establishment of a recovery route for europium and other REEs used in phosphors. However, if material circularity is to be implemented in the europium cycle, an expansion of the current European manufacture capacity seems to be needed to increase the outlet for domestic producers of europium-based commodities, or enhancing secondary production might only result in an increase of europium exports with no relief from the import reliance for this region.

The replacement of incandescent bulbs with fluorescent lamps and SSL technology has reduced the energy use for lighting [2] and, along with this market transition, the first retrospective on europium use in phosphors provided in this study has demonstrated that this element marked progress in material turnover and energy efficiency in the lighting sector. However, the transition to LEDs and OLEDs has determined a decrease in the demand for europium at about 50% over the last five years. While primary production of europium will likely continue in the coming years, as this element is co-produced with other REEs and adds value to mine extraction activities [66], this potential oversupply of primary europium might reduce the chances of exploiting secondary sources unless financial incentives to recycling are implemented [31] and new application segments make the demand for europium upswing.

New demand for europium could occur due to, for instance, increasing use of white LEDs and the expansion to new markets such as quantum hard drive manufacturing [73] and alternating current-sourced LEDs [74]. However, the declining trend is more pronounced in the demand for europium than for other REEs as lighting is the major application segment of this element. Creating a resilient European recycling industry for europium remains hindered by nonexistent recovery at end-of-life as well as by market forces. At the time of writing, a recovery in europium demand and its market prices seems unlikely, so primary sources might remain more technically and economically preferable than recycling to supply phosphors to future lighting systems.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2079-9276/7/3/59/s1>, Table S1: List of Prodcom commodities, end-use segment correspondence, associated market penetration rates of europium in each product, and europium contents with references applied in the model; Table S2: Parameters used in the dynamic MFA model; Table S3: Collection rates of end-of-life products containing europium; Table S4: Life cycle assessment factors employed in the calculation of potentials energy savings and greenhouse gas emissions reduction associated with europium recycling in the EU–28; Figure S1: Market shares of europium demand for semi-finished products fabrication; Table S5: Europium flow into use, net-addition to stock, and outflow from use by end-use segment for selected years; Table S6: Alternative lifespan distribution for fluorescent lamps (used for sensitivity analysis); Figure S2: Sensitivity analysis results for europium outflow from use in fluorescent lamps in the EU–28; Figure S3: Uncertainty analysis results for the cumulative in-use stock of europium in the EU–28.

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