

Backlighting the European Indium Recycling Potentials

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Summary

With increased understanding of the effects of human activities on the environment and added awareness of the increasing societal value of natural resources, researchers have begun to focus on the characterization of elemental cycles. Indium has captured significant attention due to the potential for supply shortages and nonexistent recycling at end of life. Such a combination of potentially critical features is magnified for countries that depend on imports of indium, notably many European countries. With the aims of analyzing the dynamics of material flows and of estimating the magnitude of secondary indium sources available for recycling, the anthropogenic indium cycle in Europe has been investigated by material flow analysis. The results showed that the region is a major consumer of finished goods containing indium, and the cumulative addition of indium in urban mines was estimated at about 500 tonnes of indium. We discuss these results from the perspective of closing the metal cycle in the region. Securing access to critical raw materials is a priority for Europe, but the preference for recycling metal urban mines risks to remain only theoretical for indium unless innovations in waste collection and processing unlock the development of technologies that are economically feasible and environmentally sustainable.

Introduction

Growing interest in understanding the effects of human activities on the environment and added awareness of the societal value of natural resources in the form of materials, energy, space, and food (Drielsma et al. 2016; Dewulf et al. 2015) have brought the research to focus on the characterization of elemental cycles (Clift and Druckman 2016). These complementary perspectives require profound cognition of relationships between human and natural systems.

Historically, the most attention has been given to anthropogenic cycles of metals used in the largest amounts, such as iron, aluminum, and copper, to unveil patterns of material production and consumption and to settle strategies for securing long-term supply (Chen and Graedel 2012; Elshkaki et al. 2016; Ciacci et al. 2014; Pauliuk and Müller 2014; Allwood et al. 2010).

More recently, the potential risk of supply shortages and predictions of the increase of global population size and affluence

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have oriented governments, industries, and the research to expand the spectrum of their assessments to include metals with potential for supply concerns (U.S. National Research Council 2008; U.S. Department of Energy 2010; European Commission 2017; British Geological Survey 2015; Skirrow et al. 2013; Graedel et al. 2012).

For instance, indium is among the elements capturing a growing consideration due to its relatively high economic importance (Polinares Consortium 2012), lack of substitutes (Harper et al. 2015), extraction as a by-product from carrier metal ores (Schwarz-Schampera 2014), low recovery efficiency of processing and beneficiation (Lokanc et al. 2015), and non-existent recycling at end of life (EoL) (Graedel et al. 2011; Frenzel et al. 2017).

Indium demand is driven by high-tech applications that require its specialty characteristics. As conferred by its location in the periodic table near the borderline between metals and non-metals, indium shows ductility, malleability, conductivity, and transparency (Felix 2000; Chagnon 2000). In particular, the last two properties are the main reason for indium use in the semiconductor industry for flat panel displays and screens manufacturing, by far the most important applications of indium in technical uses.

Beyond its use as an electrode material in liquid crystal displays (LCDs), indium can be also found in the form of indium-gallium-nitride (InGaN) as a component of light-emitting diode (LED) semiconductor chips for screen back-lighting to enhance definition and brightness of LCD monitors (Buchert et al. 2012). The increasing use of coloured LED lights in everyday applications is spurring the manufacturing of semiconductor chips based on InGaN systems, which are generally utilized for emitting blue-green light. Semiconductor indium grades are mainly required for optoelectronic devices and fiber-optic communications.

In the thin-film photovoltaics (PV) industry, the manufacturing of copper-indium-gallium-selenide (CIGS) is still quite small compared to the crystalline silicon cell type, but the market for CIGS is growing (Fraunhofer Institute for Solar Energy 2016; Zimmermann 2015).

The heat-resistance property of indium is exploited in architectural glass coatings. In metallurgy, indium is utilized for the production of low-melting fuses, which are widely employed in fire-alarm systems, soldering alloys, thermal interface materials, dental fillings, and jewel manufacturing. Indium is also used in nuclear rods to measure the intensity and energy of streams of thermal neutrons in atomic reactors, and in aircraft motor bearings to both form stronger alloys and reduce the eroding action of the lubricating oils on the heated metal surface.

This set of highly specialized technical uses make indium very challenging to replace. A list of potential substitutes of indium in major applications has been identified (Harper et al. 2015), and, although the estimated substitute performance varies from adequate to good, available alternatives in current applications are often other scarce or critical metals themselves or indium competitors for specific end uses (e.g., gallium in metallurgy and the semiconductor industry).

The global demand for primary indium is in the order of 600 to 800 tonnes per year (tpy), but outlook for the indium market growth expects the global demand to increase at an annual rate of 5% to 10% (Schwarz-Schampera 2014). Indium mining is associated with base metals containing ores enriched in zinc, copper, lead, and tin. Low concentrations of indium, typically between 20 and 350 parts per million (Frenzel et al. 2017; Werner et al. 2017), in these minerals prevent the economic recovery of indium as a primary commodity, so this metal is obtained only as a by-product. The most common indium extraction technique processes ores enriched in zinc- and copper-bearing zinc minerals from underground mines. Most indium-bearing massive sulphide deposits are located in East and South-East Asia along the western Pacific plate boundary and in South America (Schwarz-Schampera 2014).

The majority of primary indium metal is recovered from the residues of zinc ores (mainly sphalerite) concentration and smelting and from the recycling of zinc smelting dusts, slimes, drosses, and gases. After froth flotation steps, further beneficiation and conversion to indium metal can follow different techniques depending on the mineral feed and the nature of the treatment; hydrometallurgical stages and electrolytic refining are usually preferred routes for indium metal production. The largest country producers of indium metal are China, the Republic of Korea, Japan, and Canada, which together represent more than 90% of global primary indium production (U.S. Geological Survey 2017).

Secondary sources usually supplement global indium supply by about 1,000 tpy. New scrap, mainly spent indium-tin-oxide (ITO) sputtering targets and In-bearing electronic scrap, are generally recycled. However, similar to primary production, processing new ITO scrap for indium recovery has grown in parallel with ITO-producing countries, with ITO recycling occurring mainly in the same plants where the sputtering processing is carried out so that indium undergoes, in most cases, close-loop recycling (i.e., indium is utilized and recycled in the same product type) (Tolcin 2017; Zhang et al. 2015). Japan, China, and the Republic of Korea represent together about 90% of the global ITO production capacity, and the same countries are dominant in the recovery of indium from spent ITO as well. Indium recycling at EoL is nonexistent (Graedel et al. 2011), making primary production the main supply mean of indium worldwide.

The known natural deposits of indium seem to be large enough to meet the mid-term demand, but issues of long-term supply sustainability could occur due to the fast growth of (apparent) indium consumption rates (Frenzel et al. 2017). The U.S. Geological Survey (USGS) reported the world indium reserves (11 kilotonnes [kt]) and reserve base (16 kt) until 2007 (U.S. Geological Survey 2008). The Indium Corporation reports an estimate of about 50 kt of world indium reserves (Mikolajczak and Jackson 2012). Based on USGS estimates of global zinc and copper resources and reserves, the indium reserve was quantified at 125 kt (Schwarz-Schampera 2014). Werner and colleagues revised that estimate at more than 350 kt of indium content in known mineral deposits (Werner

et al. 2017). Additional indium stock (~15 to 25 kt) has been individuated in above-ground deposits such as tailings and ore residues, but selective indium extraction is challenging due to iron contamination (European Commission 2015b; Werner et al. 2017). The quantified indium deposits likely represents an underestimate of indium resource, and measurements have been revised frequently, pinpointing how challenging the quantification of available world resources of scarce metals can be. Mineral exploration and mining activity is ongoing, and the technological development can convert yesterday's uneconomic deposits in tomorrow's profitably extractable resources.

Such a combination of potentially critical features is magnified for countries that depend on imports of indium in its various forms (Ciacci et al. 2016b). This is the case for many European countries (Alfontzi 2003), for which the supply risk can be also enhanced by indium export restrictions such as those set up recently by China (Polinares Consortium 2012). Thus, the European Commission (EC) has included indium in its list of most critical raw materials, suggesting that securing indium supplies are a priority (European Commission 2017).

Curiously, Europe's concerns about the supply risk of indium are not entirely new, as the attention toward the discovery and exploitation of domestic indium dates back to the use of indigo blue dye that was responsible for giving indium its name. "Indium" comes from the latin term *indicum*, used for "Indian," being the bright blue pigment imported in Europe from India at a very high price. As told in Venetskii (1970): "... When, towards the end of the 18th century, the British navy cut off France from India and the other countries across the ocean, Napoleon, who looked forward to seeing his soldiers in their dark-blue uniforms in the subjugated capitals of the world, promised a colossal prize of a million francs to anyone who would find a method of obtaining the valuable indigo from European raw materials, and thus would help preserve the traditional color of the uniform of the triumphant army."

As mining explorations and geological surveys' assessments lay the base for quantifying primary deposits and virgin material supply, the characterization of anthropogenic material cycles can constitute a fundamental basis for assessing the size of in-use stocks (IUS), also known as "urban mines." Material flow analysis (MFA) is a primary methodology in industrial ecology (IE) (Clift and Druckman 2016) and can assist in the sustainable management of critical raw materials (Kim et al. 2015).

With this intention, a dynamic MFA perspective was applied to analyze the indium metabolism in Europe and estimate the magnitude of secondary indium sources available for recycling. The implications of improving indium recycling for reducing primary metal inputs and the associated environmental burdens were discussed to support Europe's transition toward the efficient closure of material life cycles. We expect that the results will provide novel insights into the regional indium patterns and constitute a fundamental basis for future assessments and decision making.

Material and Methods

A material flow model was developed for accounting the indium flows and IUS in Europe (EU-28) from 2002 to 2015. MFA requires extensive data gathering to provide inputs for the model: however, in contrast to metals used in large amounts such as iron and aluminium, primary data are rarely available for scarce metals like indium and scattered estimates are often the only information available. For this reason, we elicited the opinion of experts to settle exogenous variables and we applied the law of conservation of matter, which constitutes the basis of any MFA study, to balance for inflows and outflows along the metal's life cycle.

For each main phase of the anthropogenic indium cycle (i.e., mining, smelting and refining, fabrication and manufacturing, use, and EoL management), the model implemented in Microsoft Excel[®] accounted for annual inputs, process losses, process outputs, imports, and exports. A detailed description of the material flow accounting equations was reported elsewhere (Ciacci et al. 2017).

The historical indium refinery production in Europe is reported by several geological surveys (U.S. Geological Survey 2017; Brown et al. 2016) (see table S1 in the supporting information available on the Journal's website). Also, the European indium market by product and country were gathered from a market research and consulting company (Grand View Research 2017). Indium recovery during smelting operations depends on its concentration in the raw materials (Frenzel et al. 2017); refining efficiency rates from (Lokanc et al. 2015) were applied to convert the amount of raw indium statistics to the annual metal content processed.

Most indium is traded as unwrought metal and metal powder forms at the standard indium purity of 99.99% (4N). Eurostat (Eurostat 2017c) reports extra-EU (European Union) trade records for "8112.29.81 – unwrought indium; indium powders" according to the combined nomenclature (CN) (see tables S2 and S3 in the supporting information on the Web). Additional information on the trade of unwrought indium and indium powders from the major EU country exporters to the United States were found in mineral yearbooks (U.S. Geological Survey 2017).

The apparent consumption of refined indium (computed as domestic refinery production + imports – exports) constitutes the amount of indium demanded by fabricators to produce semifinished goods such as ITO powder, InGaN, CIGS, and indium-containing alloys. First-use market shares were estimated through surveys with industry and producer associations (Mikolajczak 2017; Kammer 2017; Omodeo 2017; Hagelüken 2017), (see table S4 in the supporting information on the Web).

Transboundary trades of indium-containing semifinished goods are difficult to identify and quantify because these goods are generally a subset of wider product categories recorded in trade statistic databases. For instance, the CN product code "8112.99 – Articles of rare earths/metals nes" very likely includes indium articles, but the classification is too aggregated to enable an individual estimate of contained indium. A further

limitation is also due to the limited number of years for which those records were reported.

Moreover, a considerable fraction of indium-containing alloys eludes the conventional classification schemes because of the lack of unequivocal definition. As an example, any alloys containing indium at >50% (w/w) are registered as unwrought indium, which can lead to double counting of the amount of indium imported into Europe. On the other hand, a very common alloy used in the optical industry has ~20% indium and more than 51% bismuth. This alloy is typically traded as unwrought bismuth with no information about indium content (Mikolajczak 2017).

Thus, to enable a representative description of the European indium supply chain, we combined the top-down approach and the bottom-up approach (Müller et al. 2014). First, we estimated the amount of indium utilized to fabricate domestic semifinished goods; second, we identified the list of finished goods that contain indium and gathered European production statistics (i.e., Prodcom) (Eurostat 2017b); then, indium contents were applied (see table S5 in the supporting information on the Web) to calculate the amount of indium demanded by European manufacturers to create finished goods. Last, we computed the annual quantities of net-traded indium as the difference of indium input to manufacturing from the output from fabrication. The list of finished goods containing indium and the relative metal content considered in the model are reported in the supporting information on the Web (see tables S6 and S7). Correspondence tables were then applied to match EU production records with the harmonized system and the standard international trade classifications for accounting indium flows in extra-EU trades.

The annual apparent indium consumption or the indium flow into use by major end-use segment entered the calculator model for the quantification of in-use stocks. To this end, average lifetimes and statistical distributions were set as described in (Harper et al. 2015) (see table S8 in the supporting information on the Web). Annual indium net-accumulation in (or net-depleted from) the European IUS was computed as the difference between the flow into use and the amount of indium contained in obsolete products generated at EoL. Dissipation of indium during use has been assumed negligible (Ciacci et al. 2015).

Existing data on the fate of indium at EoL are very limited and often contradictory. Metadata for EoL collection and pre-processing rates of principal waste categories that can contain indium were gathered from a previous study (Ciacci et al. 2017).

Limited quality of data influences the inherent uncertainty of MFA studies (Schwab and Rechberger 2017). Uncertainty analysis was run to test the robustness of the material flow accounting model and reconcile the estimates. Specifically, lower- and upper-bound data were used to compute uncertainty ranges for the magnitude of indium accumulated in the IUS and generated at EoL. Ranges of indium content in finished goods can be very broad so it was assumed that the distribution of indium contents follows a normal curve and that 99.7% (i.e., 3σ) of indium concentrations is included between the lower

and upper values listed in table S5 in the supporting information on the Web. Averages were computed as arithmetic means and standard deviation (σ) was deduced accordingly (see table S7 in the supporting information on the Web). For more details, we refer the reader to the Supporting Information on the Web.

Results and Discussion

The European Indium Cycle

Figure 1 displays the anthropogenic indium cycle in Europe in 2014. This year has been selected as a contemporary “snapshot” of indium flows and stock in the region. No major changes have occurred in general trends of indium flows in the time span considered.

According to the extensive review and the most accurate up-to-date data collection provided in Werner and colleagues (2017), the quantity of indium contained in the known European indium-bearing deposits amounts to about 20.6 kt, but little information was found in literature to address the actual mining and processing of indium in the EU. However, most indium processed in the region is likely imported in the form of zinc concentrates, fumes, and drosses.

Virgin indium member state producers are France (Nyrstar) and Belgium (Umicore). Umicore has a reported indium refinery capacity of 50 tpy (Caffarey 2012), while Nyrstar commissioned an indium metal production plant in 2012 that brought the smelter to produce about 40 tonnes (t) of indium in 2015 (Nyrstar 2016). A fire at the Nyrstar smelting facility in Aubay caused no indium production in 2016; however, indium metal production was expected to resume in 2017 (U.S. Geological Survey 2017).

Additional refined indium production (for about ~20 to 25 tpy) was reported from other EU countries, including Germany, Italy, the Netherlands, and the United Kingdom (U.S. Geological Survey 2015; Brown et al. 2016). These four countries have imported unwrought indium, indium powders, and indium new scrap and their activity includes indium alloys and compounds fabrication and ultra-refining of indium (i.e., up to 5N to 7N grades), mainly demanded by the semiconductor industry for PV and solar cells manufacturing (Schwarz-Schampera 2014).

No indium scrap from fabrication and manufacturing is left untreated. New scrap is collected sent out for refining, recycled, and returned to them as indium ingots, compounds, or any other form they needed to start the production. Negligible metal stockpiling was also considered to occur in the European indium industry (Mikolajczak 2017).

Europe plays a noteworthy role in the global processing of zinc ores and concentrates to refined and ultra-refined indium metal, but these indium forms are not necessarily used by domestic fabricators. In fact, the region is a net exporter of unwrought indium and the domestic net demand of metal indium to produce semifinished goods is in the order of 20 to 30 tpy (Grand View Research 2017; Mikolajczak 2017; Kammer

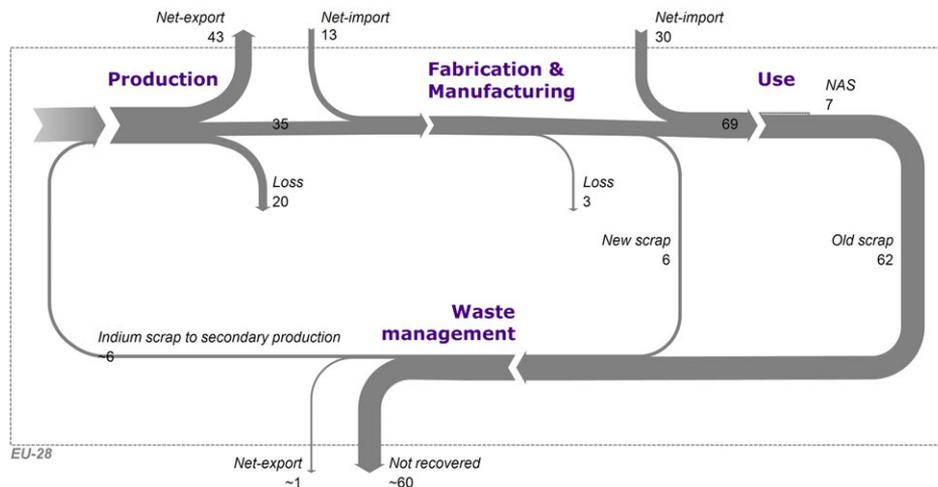


Figure 1 The 2014 anthropogenic indium cycle in the EU-28. NAS: net-addition to in-use stock. Values are in tonnes of indium.

2017; Omodeo 2017; Hagelüken 2017). It is perhaps surprising to note that this amount is much smaller than the global primary input to fabrication (i.e., ~600 t; Licht et al. [2015]), but the reason is that ITO production, the main end-use segment of indium, takes place outside the region, mainly in East Asian countries (i.e., Japan, Korea, and China) (U.S. Geological Survey 2017). There is very little manufacture of flat panel displays remaining in Europe. Glass coating occurring in Europe is mostly for heat-resistance glass used in architectural applications and other niche uses, accounting for 2 to 3 tpy of indium in total.

Alloys, PV, semiconductors, LEDs, solders, batteries, and other uses exist in Europe. The 20 to 30 tpy of indium demanded by first users was disaggregated to metallurgy (30%), PV (40%), glass coatings (10%), research, and other uses (20%). The market shares have remained relatively constant over the last decade, with PV that increased, dropped, and is now increasing again. Thermal interface materials are not manufactured in Europe (Mikolajczak 2017).

The amount of indium embedded into finished goods resulted at about 40 t in 2014, disaggregated among glass coating (66%), metallurgy (13%), electrical and semiconductors (19%), and research and other uses (3%). The net import of indium contained in semifinished goods was quantified at ~13 t. Most indium imports are in the form of ready-made ITO-coated products to be incorporated into finished goods.

The apparent demand (i.e., flow into use) of indium to provide goods and services to European consumers was almost doubled by net imports from countries outside the EU that raised the total flow into use at about 70 t of indium in 2014. The greatest part of these imports consisted of flat panel displays, vehicles, and electrical equipment and electronics (EEE). Glass coating remains the primary application segment (77%), followed by electrical and semiconductors (14%), metallurgy (8%), and research and other uses (1%).

The simulation of indium generated at EoL as a function of annual flows into use and product lifetime distribution parameters resulted in about 62 t indium embedded into obsolete

goods and EoL products in 2014, bordering literature estimates reported for 2012 (Deloitte 2015). The difference between the indium flow into use and that out of use in each year represents the amount of indium net accumulated into the IUS, which amounted at 7 t in 2014.

As shown in figure 2, from 2002 to 2015 the cumulative European IUS of indium amounts to nearly 500 t ($\pm 32\%$; figure S1 in the supporting information on the Web). This estimate is aligned with the results for Japan (Yoshimura et al. 2013; Nakajima et al. 2008), Taiwan (Chang et al. 2015), the United States (Goonan 2012), Australia (Werner et al. 2018), and the world (Licht et al. 2015). Scaling the global amount of indium that entered the use phase in 2011 (i.e., ~300; Licht et al. [2015]) to the EU levels by population and gross domestic product at purchasing power parity (GDP-PPP), that is, two major drivers for the apparent consumption of finished goods and services by consumers (Elshkaki et al. 2016), it can be estimated that about 60 t indium were demanded by the EU-28 in 2011. This value is comparable to our estimate of ~77 t for the same year. Carrying the same calculation out for the more developed world economies, it can be roughly inferred that the United States, Europe, China, and Japan all together demand about 60% of total indium into use.

Very often, the turnover of electrical goods and electronics occurs prior to these products reaching the end of their useful lives. A direct consequence is that a considerable fraction of secondhand EEE is traded transboundary. An StEP report (Baldè et al. 2016) quantified at about 60 kt the net export of laptops and desktops, flat panel screens, and mobile phones from the EU mainly to African and Asian developing countries. Applying the indium content ranges used in the model results in at least 1 t of indium being net exported annually by means of EEE transboundary flows. As stated by the authors, the amount of used EEE exported is likely an underestimate of the “real” flow and efforts are put forward to understand extra-EU trade of electronics, especially to contrast the illegal management of waste electrical and electronic equipment (WEEE) (Baldè et al. 2016).

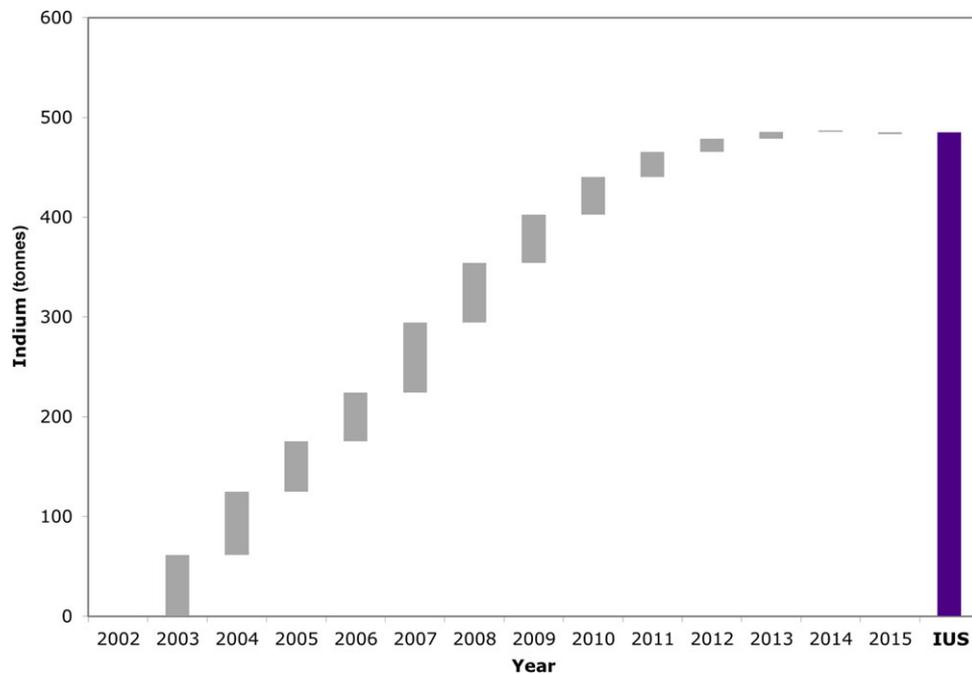


Figure 2 Annual and cumulative in-use stock (IUS) of indium in the EU-28.

However, the greatest fraction of indium at EoL remains unrecovered and lost. The European indium cycle is highly inefficient so that margins for improving indium recovery and recycling rates at EoL are worthy of attention. The implications of turning that potential into real capacity is discussed in the next sections.

Implications of Improving Indium Recycling for the Metal Supply

In recent years, the EC has placed considerable attention on tackling the challenges of securing access to raw materials and reaching sustainable production-consumption patterns. To this end, several initiatives have been adopted including the development of a methodology for assessing the criticality of raw materials (European Commission 2017), the launch of packages to support the transition toward greener energy systems (European Commission 2016), and the implementation of the circular economy (CE) model (European Commission 2015a). The latter approach puts recycling at the core of elemental cycles, and the recycling of urban mines is promoted as a main action to reduce primary material inputs and the associated environmental burdens.

As depicted in figure 1, the indium cycle is quite far from efficient recycling, and the closure of its cycle is hampered by nonexistent recovery at EoL. However, two important reflections can be drawn out.

First, the magnitude of indium generated at EoL and potentially available for recycling is greater than the amount of indium demanded by domestic fabricators and manufacturers, and is near to the estimated annual flow into use. Thus, a virtual basis for closing the indium cycle in the region does exist.

Second, the size of the cumulative IUS estimated by the model (i.e., ~500 t of indium) represents nearly the amount of refined indium globally demanded and it is approximately 15 times greater than the current amount of indium demanded by European fabricators. Therefore, not only a virtual closure of the indium cycle is theoretically possible, but also the potential for material circularity seems to be also relatively sustainable in the medium term.

However, establishing and maintaining a virtuous recycling industry requires certain amounts of secondary indium sources are valuably processed in the long term. In the last 15 years, the annual amount of indium entering the use phase has remained below 100 tpy and increased from 60 tpy (2002) up to about 90 tpy (2009) and then has decreased to 70 tpy more recently. This apparent reduction in the demand for indium is also reflected in annual additions to IUS (figure 2). Thus, only if the demand for indium remains at the current levels or increases that latent circularity has the potential for closing the metal cycle and reducing significantly the reliance of Europe from net import of indium forms.

Figure 3 compares the historical accumulation of IUS per capita, obtained by dividing the anthropogenic indium reserve in the region by the European population, versus GDP-PPP per capita at constant 2011 international dollars. For 2002–2011, the data set shows rapid increases, even during the recession in 2009, but the trendline has remained relatively constant at about 1 gram of indium per capita in the last 5 years.

Although a complete saturation of the indium IUS per capita is unlikely in the near future considering projections for indium demand (Jackson 2012), a possible explanation for such a flattening can be found in that cathode ray tube style screens have been almost completely replaced by with LCD/LED screens.

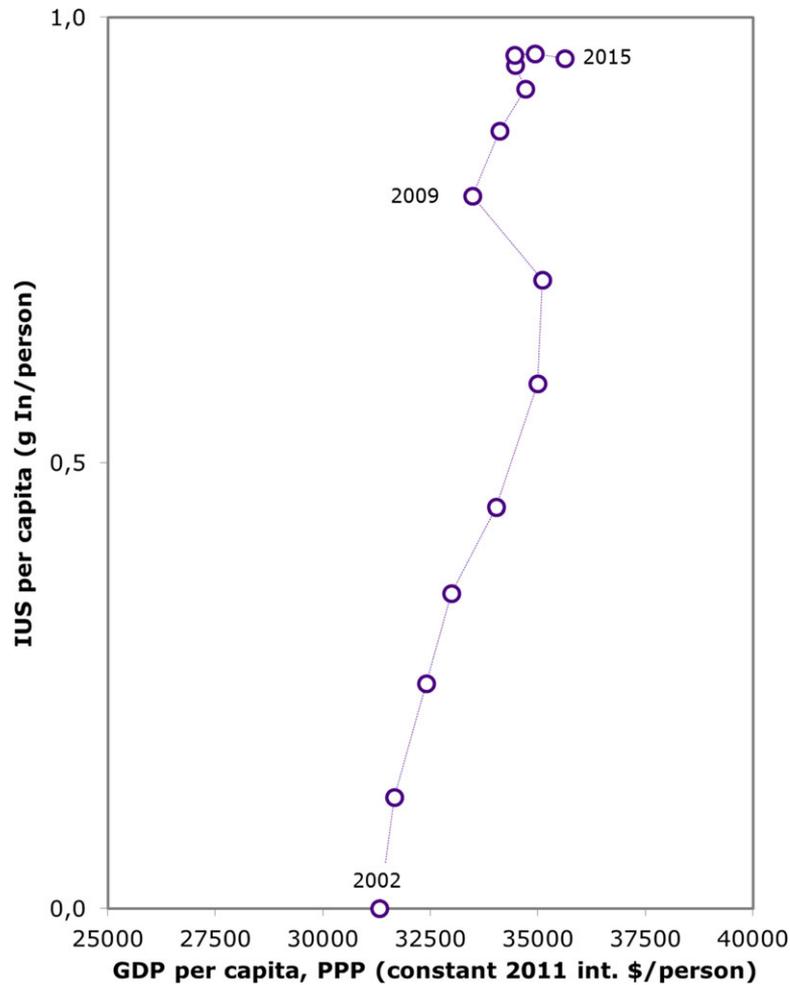


Figure 3 Intensity of the indium in-use stock (IUS) per economic activity in the EU-28. Per capita indium IUS versus per capita gross domestic product at purchasing power parity (GDP-PPP) at constant 2011 international dollars (\$).

However, according to a recent survey, the EU consumers are shifting toward larger display purchases, which have higher indium contents on a mass basis, when they enter the market to replace first generations of LCD screens (Schlösser et al. 2014).

Additional boost to the regional indium demand could derive from low-carbon energy systems such as wind turbines (Kim et al. 2015) and PV (Ylä-Mella and Pongrácz 2016), particularly in case of high market penetration of thin film solar cells (Stamp et al. 2014; Zimmermann 2017). In the PV sector, silicon (Si)-based cells accounted for more than 90% of the global production in 2015 (Fraunhofer Institute for Solar Energy 2016); thin-film technology has been on the market for 30 years, but CIGS and Cu-In selenide (CIS) are more recent, being in use for nearly a decade. At current levels, CIGS and CIS constitute together about one third of all thin-film PV technologies, but they are likely to become a primary technology for clean-energy production (European Commission 2016; Bleiwas 2010) and to compete with other electricity sources (Mercer 2015) in virtue of their better performance, less material intensity, and potentially lower manufacturing costs

than Si-based cells. Efficiency of CIS and CIGS solar cells is still less than traditional Si-modules, but the research suggests that the gap is narrowing (Solar Frontier K. K. 2016; Bleiwas 2010).

New application segments could also emerge in the coming years: For example, the replacement of lead and tin with indium in cryogenic metallurgy, automotive alloys, and low-melting alloys for security purposes is still limited today because of the high manufacturing costs but is believed to increase in the future (Omodeo 2017).

Returning to the present situation, Europe has the features of a major consumer of indium-containing goods, and recovery and recycling the domestic IUS could constitute relevant means to secure the region with a secondary supply of indium. Ultimately, however, the whole matter boils down to the technical and economical feasibility of indium recovery at EoL. Today, indium is rarely recovered from any of its applications at EoL because secondary indium supply is more costly than primary production, and this condition is likely to continue unless more economically viable indium recovery processes are developed (Frenzel et al. 2017).

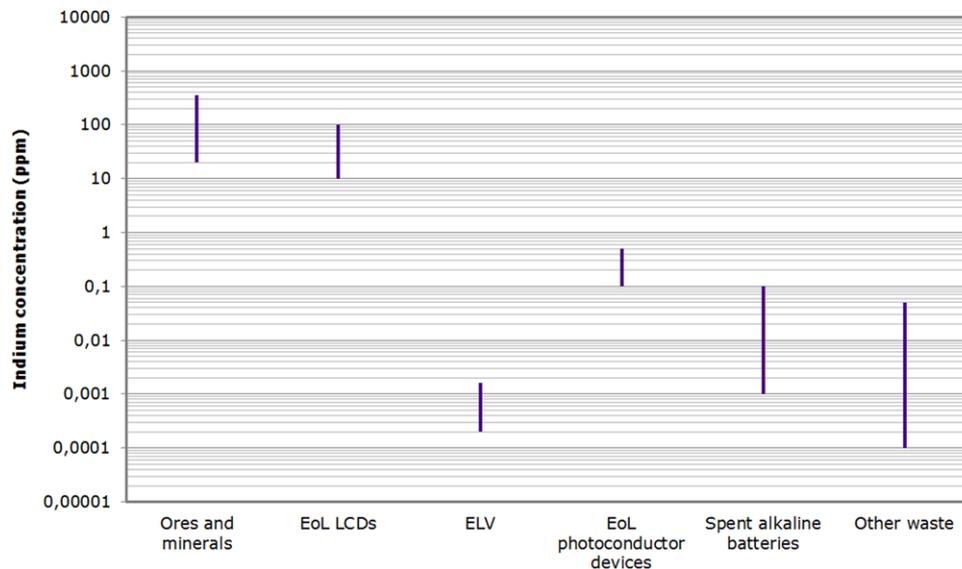


Figure 4 Indium concentration in primary and secondary sources. EoL LCDs = end-of-life liquid crystal displays; indium concentration based on (Buchert et al. 2012; Cucchiella et al. 2015; Nakajima et al. 2008; Takahashi et al. 2009; Chancerel et al. 2013; Rocchetti et al. 2016; Chang et al. 2015). ELV = end-of-life vehicle: indium concentration based on (Cullbrand and Magnusson 2011; Widmer et al. 2015). End-of-life (EoL) photoconductor devices: indium concentration based on (Andersson 2000; Solar Frontier K. K. 2016). Spent alkaline batteries: indium concentration (own assumption) based on (Werner et al. 2018). Other waste: indium concentration (own assumption) based on (Werner et al. 2018).

To reverse the status quo and to give the recycling of indium a primary role for meeting future demand, the issues of waste collection and treatment need to be virtuously addressed. Ineffective collection and separation of indium from waste and obsolete products at EoL are among the main hindrances to the closure of the indium cycle (Licht et al. 2015). Indium concentrations in some finished goods are comparable with those in minerals, and in some cases they are even higher (figure 4), but the huge variety and number of single products to be collected and processed have to face the issue of dispersion (Ueberschaar et al. 2017).

Potentially, the indium contained in solders and alloys could be functionally recycled (i.e., preserving indium's function in the new product) (Ciacci et al. 2015), if selective separation and recycling of alloys of known composition were carried out by alloy type (Hatayama et al. 2012). Yet, the management of EoL alloys is still driven by the recovery of major carrier metals, such as iron, aluminum, zinc, and copper, and the associated recycling routes do not enable to recover indium. In fact, indium tends to distribute into the metal phase or the slag phase where it is essentially lost as a tramp element bringing no functionality to the new (recycled) product (Hiraki et al. 2011; Reck and Graedel 2012).

Similarly, indium is currently unrecovered from WEEE notwithstanding that this waste category has respectable collection rates in general (Eurostat 2017a). Since the greatest indium recycling potential is connected with its application in glass coating due to large market penetration, amount of indium accumulated in use (see figure S2 in the supporting information on the Web), and high indium concentration (figure 4), several

processes have analyzed the recovery of indium from discarded LCDs. The general process framework consists of (1) collection of LCDs, (2) dismantling (either manual or mechanical disassembly), (3) treatment of displays (e.g., crushing, pyrolysis), and (4) recovery of indium. Processes for indium separation and purification include solvent extraction, liquid membrane separation, vacuum chlorinated separation, and vacuum carbon reduction (Zhang et al. 2015). Novel approaches have investigated the recovery of indium through ion exchange resins, biological metallurgy, and cross-current leaching with zinc cementation (Rocchetti et al. 2016). A combination of these techniques could eventually enhance process yields and reduce the operation costs as discussed in Rocchetti and colleagues (2015). Although many of those processes are promising, no recycling system for EoL indium has been fully established in Europe yet (Zhang et al. 2015).

Implications of Improving Indium Recycling for Environmental Sustainability

Beyond the reduction of primary material inputs, the CE approach also aims at decreasing the environmental impacts associated with the extraction and processing of natural resources by means of EoL recycling. Therefore, environmental sustainability can act as a strong driver for improving secondary indium recovery.

From this perspective, the results of this study can be used as a foundation to unveil the nexus between metals, energy, and climate change. The transition toward greener energy systems for a low-carbon society requires to implement the emerging

technologies for which indium is an essential component on a wide scale. In this sense, the potential contribution of indium recycling for future metal supply to provide greener energy systems is strongly interconnected with the potential impacts of energy savings and carbon emissions reduction associated with indium recycling.

In life cycle assessment (LCA) terms, the benefits of recycling can be computed as avoided impacts from primary production routes. As discussed above, the IUS constitute a significant source of secondary indium that, if not recovered and fed into the metal supply chain, has to be likely derived from virgin sources. Indium losses are probably irrelevant to mitigate climate change globally (as opposed to metals like iron and aluminium; Ciacci et al. [2016a]), but still the European indium IUS can contribute to making the European metal industry more sustainable. The supply of the amount of indium embodied into the IUS from primary sources would equal the energy requirement of 0.9 petajoules and the emission of nearly 80 MtCO₂-eq as estimated by using an existing LCA data set (Classen et al. 2009). These quantities represents the supply of about 50,000 t of cast iron at the current level.

Of course, these potential impacts are not entirely avoidable, as the energy required for separating and recycling indium at EoL will also result in the emission of greenhouse gases. However, very little information is available in literature on the environmental implications of secondary indium processing. Amato and colleagues provided the inventory of material and energy inputs for treating LCD waste through the cross-current leaching process (Amato et al. 2017) (see table S9 in the supporting information on the Web). Based on those data, however, the environmental impacts attributable to the sole indium recovery are much greater than those associated to primary indium production. Making indium recycling the better environmental option is also complicated by additional impacts, including those for collecting and transporting waste containing indium, for establishing the required recycling infrastructure, and for refining indium to commercial grades. On the other hand, scaling the pilot process up to the industrial scale can likely reduce the environmental impacts per unit of indium recycled. Other factors that can play in favor of indium recycling include the recovery of other metals from waste like discarded LCDs (Hagelüken 2008), which would enable to allocate the environmental impacts among the valuable recoverables, greater market penetration rates of renewable sources in the electricity production mix, and the adoption of strategies for resource efficiencies (e.g., design for recycling, ecodesign) (Ylä-Mella and Pongrácz 2016; Ardente et al. 2014).

Conclusion

The results of this work provided the first investigation of the indium cycle in Europe and discussed the inherent implications of improving indium recycling at EoL. MFA is confirmed to be a suitable and versatile methodology to analyze historical patterns of material demand and supply, laying the base for further assessments encompassing scenario analysis,

environmental impact evaluations, analysis of socioeconomic metabolisms, and similar.

The challenge with scarce metals like indium is that a lack of reliable and easily accessible data usually affects material flow accounting, from individual activity of mining operations onward. This work has also demonstrated that when investigating material cycles at the national or regional scale, the straightforward utilization of global averages needs to be carefully verified, as exemplified by market shares of indium in major application segments. Furthermore, there is a need for harmonizing the existing product classification systems of goods, as the current tariff codes are often too vague and macro-aggregated to enable the quantification of critical metals contained in small concentrations. The vulnerability of a country to possible supply restrictions of critical materials includes the country's reliance on imports, which, in turn, passes through the unequivocal characterization of traded goods.

In addition, the lack of studies and poor details on the environmental performance of available processes for indium recovery from obsolete flat panel displays, semiconductors, and similar products leaves uncertain if EoL recycling of indium would actually result in net environmental benefits. In IE, recycling is generally preferred to primary material production, but that preference needs to be supported by evidence. More research in this direction is hence desirable.

On a wider scale, the work showed that Europe is a major consumer of finished goods containing indium. This feature gives the EU a significant potential for secondary indium supply and for closing the metal loop in the region. However, several hindrances prevent to turn that potential into real capacity. Securing access to critical raw materials is a priority for the EC, but the preference for recycling the metal urban mines risks to remain only theoretical for indium unless innovations in waste collection and processing unlock the development of technologies that are economically feasible and environmentally sustainable. Such a change cannot occur without a merging and fruitful cooperation between governments, industry, and the research.

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Supporting Information

Supporting information is linked to this article on the *JIE* website:

Supporting Information S1: This supporting information includes information on indium refinery production capacity in Europe (Table S1), extra-EU import and export of unwrought indium and indium powders (Table S2 and Table S3, respectively), first-use and end-use market shares of indium in the EU-28 (Table S4), ranges of indium content in selected end-use products (Table S5), correspondence between finished goods and UNU-KEYS for conversion from product weight to indium content (Table S6), average indium contents and relative standard deviation applied in the study (Table S7), lifetime statistics by end-use application of indium (Table S8), life cycle inventory of indium recovery from end-of-life liquid crystal displays (LCD) (Table S9), uncertainty analysis results (Figure S1), and comparison between the indium cumulative in-use stock (IUS) and the output from use estimated by major application segment of indium (Figure S2).