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Discrete-Point Analysis of the Energy Demand of Primary versus Secondary Metal Production

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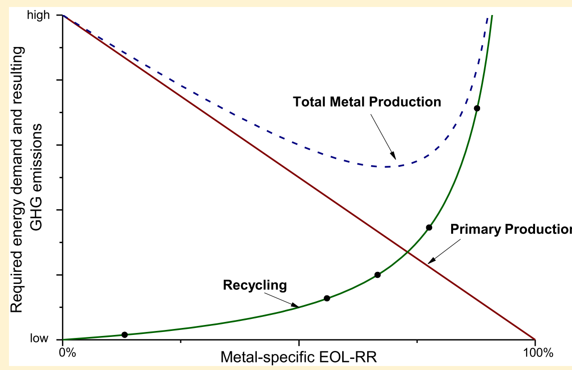
Supporting Information

ABSTRACT: The metal industry consumes large amounts of energy and contributes significantly, up to 10%, to global greenhouse gas (GHG) emissions. Recycling is commonly included among the most viable options for mitigating the climate forcing of metal production by replacing primary production. However, the recycling rates of metals are still incomplete and, in particular, do not exist for most specialty metals. Our empirical analysis of 48 metals shows that their recycling is mainly impeded by their low concentrations. In many cases, the metal concentration in end-of-life products is lower than that in natural ores. This phenomenon inevitably raises the question of the extent to which recycling can be conducted without losing its mitigating effects on climate change. We answer this question for two example metals, tantalum and copper, within the scope of Germany, a leader in recycling. For tantalum, the results show that a further increase in the end-of-life recycling rate (EOL-RR) could contribute to minimizing the overall energy consumption and GHG emissions, despite its low concentrations in end-of-life products. The energy requirements for recycling copper from end-of-life products already reach the magnitude of those for primary production. A further increase in EOL-RR must be examined in detail to ensure mitigating effects on climate change.



1. INTRODUCTION

There are two ways of metal production: primary production, i.e., extracting metals from ores (primary sources), and secondary production from discarded products, so-called end-of-life products (referred to as secondary sources hereafter), generally known as recycling. In general, recycling is proposed to fulfill several goals: to diminish the depletion of natural resources and to contribute to environmental and climate protection. The former goal is part of a controversial discussion.[1−6] This article is focused on the latter goal.

Primary metal production accounts for 6−10% of the global anthropogenic greenhouse gas (GHG) emissions[7,8] and has an enormous energy demand of approximately 8% of the global energy demand.[9] To mitigate environmental pollution and intensification of climate change by primary metal production, recycling is commonly included among the most viable options. Recycling reduces the need for primary production with all its related environmental and societal impacts and additionally minimizes the need for landfilling of discarded products and waste materials.[10] It is often stated in the literature that metal recycling is much more energy efficient and more environmentally friendly than metal production from ores.[7,11−16] However, these studies apply only to relatively pure metal scraps, such as new scraps. As reported by Rankin,[7] the energy requirements of recycling depend on the quality, especially the grade, of the scraps. Secondary metal sources are usually not available in their pure form but rather at low concentrations in numerous complex products with extensive material mixtures, which usually limits the recyclability of secondary metal sources, as has been shown in several studies via examples.[15,17−19]

A survey of the current end-of-life recycling rates (EOL-RRs) is published by the United Nations Environment Programme (UNEP).[20] Only 18 metals have EOL-RRs above 50%. Thirty-two metals are basically not recycled. High recycling rates are typically achieved for base metals, which are used in large quantities and relatively pure forms in most of their applications, such as iron, copper, or aluminum. Those metals that are produced in small quantities are, except for precious metals, not recycled or only recycled at rather low rates.[21]

Improving these recycling rates toward the vision of a circular economy is already a consensus in policies[22] and demanded by society and science.[23−26] Moreover, numerous studies have been performed to meet the increased demand to develop recycling technologies and increase EOL-RRs, in particular for specialty metals.[27−34] However, how do further increases in metal EOL-RRs, especially considering the rather low metal concentrations, compare to their primary production

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in terms of the energy demand and GHG emissions? Also, can this approach eventually lead to negative trade-offs in regard to climate protection? Ciacci et al. estimate that increasing EOL-RRs to their theoretical maximum will lead to significant energy savings and GHG emission reduction.[35] The main uncertainty in this study is that the recycling efforts are largely neglected, even though recycling, just like primary production, also requires energy and results in GHG emissions. These recycling efforts are crucial in the discussion on increasing EOL-RRs and minimizing the overall energy demand and GHG emissions of metal production.[36−38] In principle, the advantages of recycling compared to primary extraction need to be supported by evidence. Otherwise, it cannot be ensured that the steps toward a circular economy will promote sustainability and will lower environmental impacts.[39]

To show how end-of-life metal recycling performs in comparison to primary production, we present two approaches in a logical order. First, through comprehensive analysis of 48 metals, we show how these metals are diluted in the technosphere and how this dilution affects their EOL-RRs. In general, metal production can be viewed as an increase in concentration in the respective primary or secondary sources. This concentration increase is possible only through the use of energy,[40] which in turn results in GHG emissions. Second, using copper (Cu) and tantalum (Ta) as examples of industrial and specialty metals, respectively, with a high importance in modern technologies, we show how the energy demand of recycling evolves with increasing EOL-RR and how these results can be used to optimize recycling activities with the objective of mitigating the GHG emissions of the overall metal production system. In contrast to the existing investigations, which are merely of a theoretical nature,[7,36,37,41] we rely on empirical models and discrete data. Furthermore, we illustrate that such a single-metal approach in life-cycle assessment is limited to a certain extent due to the complexity of recycling systems and the interaction of metals.

2. MATERIALS AND METHODS

2.1. Concentration Ratio of Primary and Secondary Sources. Based on the studies by Graedel et al.[42] and Ciacci et al.,[35] the main end-use applications of 48 metals were identified, as these end-use applications accrue after their use phase as secondary metal sources. The metal concentrations of these secondary sources as well as their specific EOL-RRs were determined by a broad literature research. For each of the 48 metals examined, a current average concentration of the corresponding primary sources was also obtained from the literature. The collected and referenced data are provided in the Supporting Information in Chapter S1.1. The metal concentrations in the secondary sources are then compared with the average concentration in the primary sources. These ratios are in turn compared with the current EOL-RRs of the metals in the respective secondary sources.

2.2. Energy Demand and End-of-Life Recycling Rate. *2.2.1. General Information.* To optimize metal production with the objective of climate protection, the overall GHG emissions must be mitigated, which are directly related to energy consumption.[10,35] Therefore, cradle-to-gate (i.e., from ore mining or the collection of secondary sources to the pure metal) cumulative energy demand (CED) data for primary and secondary metal production are utilized to enable a comparison of these two methods. The CED accounts for both direct and indirect primary energy inputs of all relevant metal production stages and their relevant background processes.[43] Direct energy inputs are, for instance, fossil fuel combustion for crushing virgin ores and melting metal concentrates or electricity for shredding and sorting scraps. Examples of indirect energy inputs include primary energy for the production of nonenergetic inputs such as floating agents or chemicals for hydrometallurgical processes and electricity generation. Additionally, the CED is a good approximation to assess other environmental impacts.[44]

There are several theoretical considerations on how the CED of metal production develops with increasing EOL-RR.[7,36,37,41,45] These considerations all basically refer to the method depicted in Figure 1. The general consideration is that

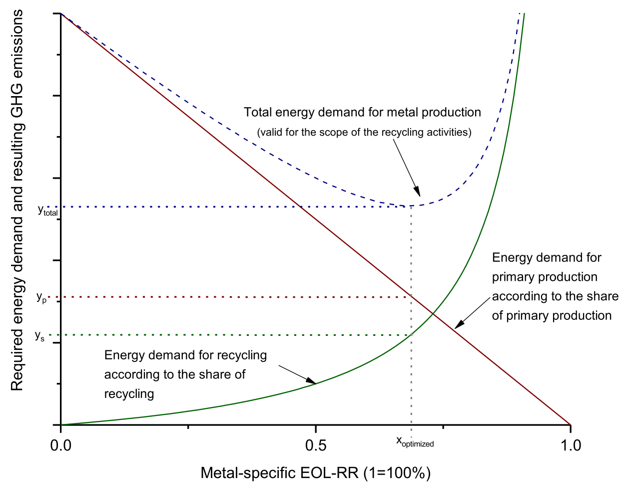

Figure 1. Qualitative description of the CED-optimized metal production system for the amount of metal masses available for recycling (modified from Stumm and Davis[36]). The metal production system is optimized when the total energy demand for metal production (combination of primary and secondary production) is minimal.

with increasing EOL-RR of a metal, the related energy requirement for recycling disproportionately increases because high-grade scraps will be recycled first, and for further EOL-RR increases, the more diluted sources need to be exploited. If a metal is partly dissipated, as is the case for many applications,[46] the CED will approach infinity. In proportion to the recycling rate increase, the CED of primary production is reduced by substitution with metal recycling. Therefore, the CED of primary production linearly decreases. The addition of both functions results in the total CED of metal production, which is valid within the scope of the recycling activities. The minimum of this line indicates the optimal combination of primary and secondary metal production for a specific metal regarding the CED (i.e., the CED-optimized EOL-RR). Recycling beyond this point would lead to a further increase in the overall CED and therefore to negative trade-offs. From the qualitative description of Figure 1, the CED-optimized EOL-RR is achieved at $x_{optimized}$. In this case, approximately 70% of the theoretically available metal mass in the secondary sources is recycled; the remaining 30% is produced from primary sources. The total CED to recycle 70% occurs at y_s, and the total CED of the remaining 30% of primary production is at y_p. The resulting $y_{total} = y_s + y_p$ is thus the average CED of the total mass of metal production (equivalent to an EOL-RR of 100%).

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Figure 1 could imply that secondary production could completely substitute primary production: this is definitely not the case due to permanently increasing metal demand.[47,48] Even if the EOL-RR would be 100%, which is only theoretically possible, there is still a need for primary metals. We would therefore like to stress that the analysis based on Figure 1 refers only to the amount of masses available for recycling or EOL-RR and not to the total metal demand.

The method is based on three assumptions. One assumption is that the recycled metal can be used as a full substitute for the metal from primary production (1:1 substitution). This condition is not always guaranteed. In the case of aluminum, for instance, the impurities caused by undesirable alloying elements cannot be removed, which reduces the quality.[49] To ensure that primary and secondary produced metals are full substitutes, we analyze metals that can be recycled to their pure form.

An additional assumption is that the average value of the current CED of primary production can be used. Based on this assumption, the CED for primary production decreases linearly with increasing EOL-RR due to the substitution approach. However, this is a simplification of the real situation of primary metal production as the CED of primary production varies according to ore content, mine type, and ore type.[50,51] The reason for this simplifying assumption is that it is currently not known in which order the primary sources will be substituted. In principle, it can be assumed that substitution is based on economic criteria, which are generally rather nontransparent. In addition, recycling is substituting future mines rather than existing ones due to the further increasing metal demand. The current average CED of primary metal production is therefore a first but quite legitimate approximation for our analysis, which is particularly conceptual in nature. If the CEDs of the actual substituted primary sources, which are currently not known, differ from the assumed average CED, the calculated CED-optimized EOL-RR may also be different. In addition, the average CED of primary production may vary over time as the ore grade and production technology change. We have analyzed both future developments and current values in our investigation.

Furthermore, it is assumed that the CED of recycling can be described in terms of the EOL-RR through a distribution function. In the best case, however, this function can only be an approximation since metals are incorporated in very inhomogeneous secondary sources. Therefore, in reality, the energy requirement function of recycling, qualitatively illustrated in Figure 1, exists as discrete points representing the secondary sources. In this article, analysis of the optimum is carried out for the first time with empirical discrete data points.

The EOL-RR accounts for all available secondary sources of a metal and is always time- and geographic-location-specific. The geographic location is also relevant in determining the collection and transport logistics as well as the efficiency and global warming potential (GWP) of electricity generation. In our study, the area is set to Germany, and the time period is fixed to 1 year. Germany has been selected as a technologically highly developed country with well-advanced recycling systems and technologies that achieve high recycling rates and can therefore be seen as a best-estimate scenario for recycling activities. Cu and Ta were selected due to several factors. These metals are typical examples of industrial and specialty metals, respectively, with a high significance for modern technologies and will therefore have a substantially increasing demand in the future.[47,52] In addition, these metals are representatives of recycling systems with high (Cu) and low (Ta) EOL-RRs.[21]

2.2.2. Mathematical Description and Data Generation. The connection between the CED and EOL-RR, qualitatively shown in Figure 1, is complex because several different secondary sources need to be considered. Each secondary source will be hereafter defined as x_i ($i = 1, 2, ..., n$), where n is the total number of sources. The secondary sources x_i are elements of set T and are regarded as objects with different characteristics. One characteristic is the metal mass $m(x_i)$ for each source x_i in the defined area and time period. Since the theoretically available mass $m(x_i)$ is not completely fed into recycling processes due to incomplete collection, collection rates $\mathrm{CR}(x_i)$, following the definition by the UNEP,[20] for each secondary source must be taken into account.

As recycling processes always experience process inefficiencies[53] and thermodynamic limitations,[54] the metal losses for each process ($j = 1, 2, ..., m$) also have to be considered by including recycling efficiencies $RE(x_i)_j$. Thus, the contribution of a secondary source (as a percentage) to the EOL-RR can be determined by the following equation

$$\frac{m(x_i)\cdot \mathrm{CR}(x_i)\cdot \prod_{j=1}^{m} \mathrm{RE}(x_i)_j}{\sum_{i=1}^{n} m(x_i)} \tag{1}$$

The CED necessary to recycle a metal from a secondary source x_i is hereafter defined as $\mathrm{CED_S}(x_i)$. System models have been developed to calculate the $\mathrm{CED_S}(x_i)$. These models cover all processes of the recycling chain, starting with the collection of end-of-life products through all manual and mechanical preprocessing steps to metallurgical recovery. Only the best available technologies with all relevant (energetic and non-energetic) inputs are considered. To calculate the CED of the collection and transport processes, we determined average distances based on real georeferenced data of the collection points and recycling facilities. For the data and model descriptions, see the Supporting Information (Chapters S1.2, S1.3, and S1.4).

Since recycling activities usually involve the recovery of several metals, the CED values of the different processes in the recycling models have to be allocated. In life-cycle assessment, there are two allocation methods proposed, namely, mass allocation and economic allocation. Bigum et al. state that from a waste management perspective, mass allocation should be preferred as the objective is to treat the received waste. Regarding recycling processes as metal production activities, allocation should be performed according to the (economic) value of the target metals, as the value of the metal is the key incentive for the recycling taking place.[12] Of course, we are aware that recycling fulfills both benefits at the same time, but, in general, recycling takes place only when the processes are economically feasible.[15,19] Thus, we apply mass allocation for the processes that provide no economic benefits. The latter is only the case for collection processes, which are in fact disposal measures of end-of-life products from an end-consumer perspective. For all other processes and transport chains, we choose economic allocation. In addition, we follow the recommendation by Ekvall and Finnveden[55] and reduce the allocation problem through subdivision as much as possible.

To identify the CED-optimized EOL-RR, the different secondary sources x_i must first be arranged according to their

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Table 1. Detailed Information about the Secondary Sources of Copper[a,b]

secondary source	details	concentration (g/g)	total mass $m(x_i)$ (t)	$\text{CED}_\text{S}(x_i)$ (MJ/kg)
cables	electrical cables (surface and underground cables, electronic appliances, etc.)	0.36	54 000	14
WEEE	consumer electronics, ICT equipment, and large and small household appliances	0.1059	154 000	21
ELVs	cars, trucks, and buses	0.0155	11 000	26
MSW	incineration ash from municipal solid waste (copper from discarded electronic products or other metallic applications)	0.00355	13 000	42
C&D	copper applications in building and construction (plumbing, heating, wiring, etc.)	0.00245	84 000	45

[a]The masses are based on data from the year 2014; for references and more information, see Chapter S1.3 in the Supporting Information [b]Electric and electronic equipment waste (WEEE), end-of-life vehicles (ELVs), municipal solid waste (MSW), construction and demolition waste (C&D).

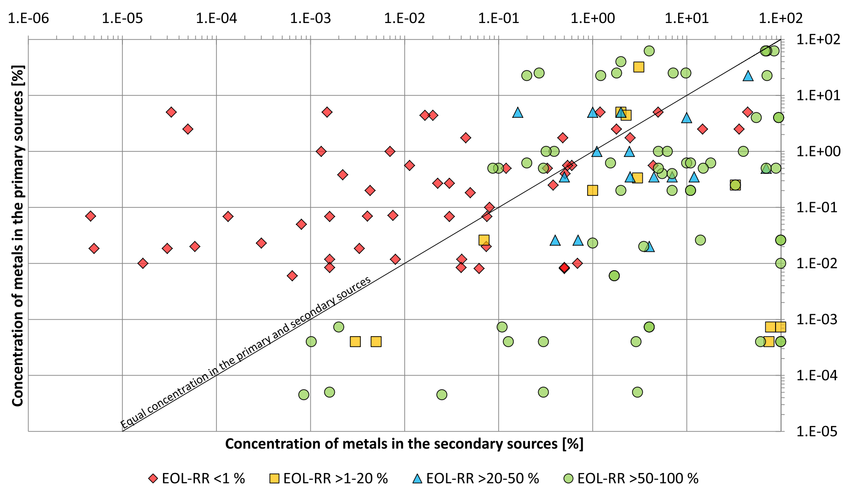

Figure 2. Comparison of the concentrations of 48 metals in their primary and secondary sources and the related EOL-RRs on a global basis. For each metal, the figure indicates one average concentration in its primary source but different concentrations in several secondary sources. Each data point therefore indicates the concentration of a secondary source in relation to the concentration of the primary source (for the data, see Chapter S1.1 in the Supporting Information).

$\text{CED}_\text{S}(x_i)$ value. The mathematical description is given in the following equation

$$\text{CED}_\text{S}(x_1) < \text{CED}_\text{S}(x_2) < \text{CED}_\text{S}(x_3)... < \text{CED}_\text{S}(x_n) \quad (2)$$

In regard to Figure 1, the total CED of metal production is minimal as long as recycling takes place only for secondary sources x_i with a lower $\text{CED}_\text{S}(x_i)$ than the CED of primary production CED_P. The latter values are gathered from the literature and life-cycle assessment databases. Mathematically, all secondary sources x_i, which should be recycled to minimize the total CED, are elements of set R as a part of the overall set T

$$R = \{x_i \in T/\text{CED}_\text{S}(x_i) < \text{CED}_\text{P}\} \quad (3)$$

The last secondary source x_i to be recycled in the defined order is the source with the highest $\text{CED}_\text{S}(x_i)$ in set R, which is defined in the following as x_k

$$\text{CED}_\text{S}(x_k) \equiv \max\{\text{CED}_\text{S}(R)\} \quad (4)$$

The EOL-RR where the total CED is minimal can therefore be expressed by

$$\frac{\sum_{i=1}^{k}\left(m(x_i)\cdot\text{CR}(x_i)\cdot\prod_{j=1}^{m}\text{RE}(x_i)_j\right)}{\sum_{i=1}^{n} m(x_i)} \quad (5)$$

The CED savings that can be realized by the optimized combination of primary and secondary production compared to single primary production can be calculated as follows

$$\text{CED}_\text{savings} = \sum_{i=1}^{k}\left(m(x_i)\cdot\text{CR}(x_i)\cdot\prod_{j=1}^{m}\text{RE}(x_i)_j \cdot(\text{CED}_\text{P} - \text{CED}_\text{S}(x_i))\right) \quad (6)$$

With the definitions from eqs 3 and 4, it is shown that $\text{CED}_\text{savings} > 0$.

3. RESULTS AND DISCUSSION

3.1. Concentration Ratio of Primary and Secondary Sources.

Each data point in Figure 2 indicates the metal concentration of a secondary source in relation to the concentration of the primary source. This analysis illustrates that the metal concentrations of the secondary sources can vary greatly and are in most cases quite low. In addition, in many secondary sources, metals are diluted to such an extent that their ore concentrations are even higher (valid for all metals located above the diagonal line in Figure 2). The latter is especially true for specialty metals, whose elementary properties are needed to improve modern technologies.[56] There are several extreme examples, such as cerium, in flat screen monitors with a concentration that is approximately 150 000 times lower than that in natural ores. However, most of the specialty metal applications depicted in Figure 2 have moderate factors below 100.

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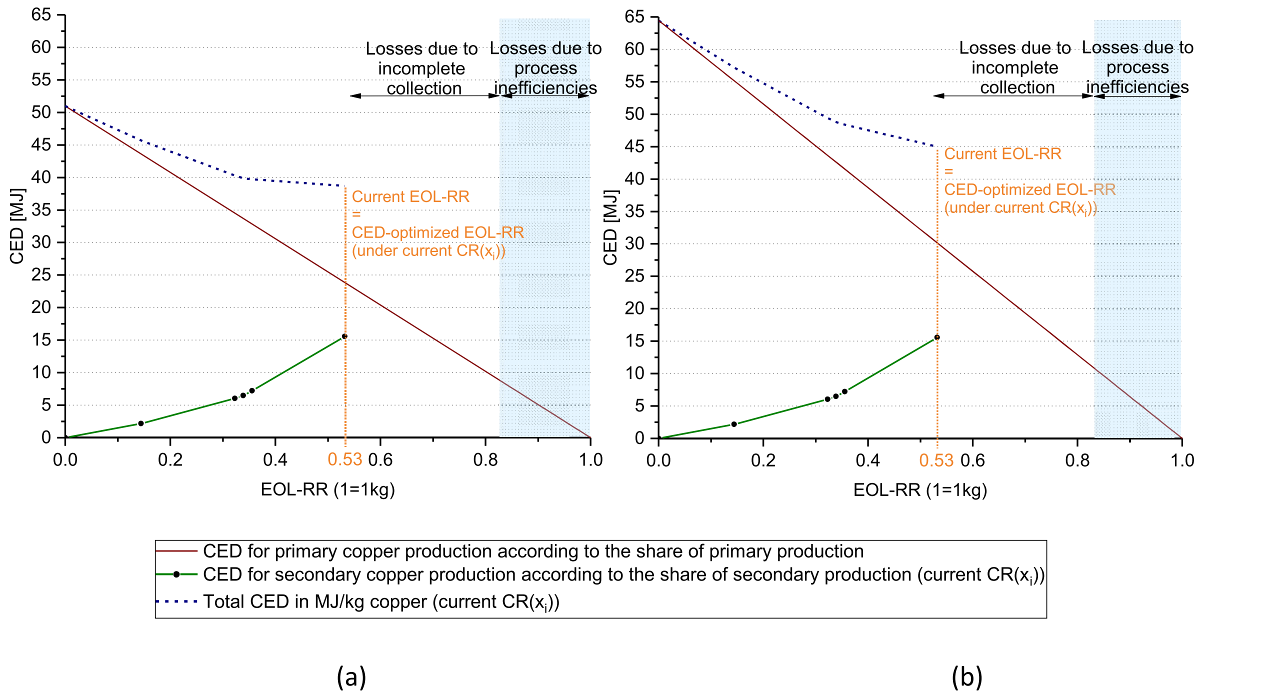

Figure 3. Application of the optimization method for the CED-optimized EOL-RR in Figure 1 to copper production. The EOL-RR is scaled to 100% equaling 1 kg of copper. (a) Current situation of primary copper production; (b) future situation of primary copper production. The values are valid for Germany.

This analysis provides an empirical explanation for the various and mostly quite low EOL-RRs of metals as published by the UNEP.[20] If the concentration of a metal in the secondary source is lower than that in the primary source, recycling tends not to occur and vice versa. Especially for higher concentrations, however, there are several exceptions. These exceptions are essentially due to mass effects, i.e., metals from secondary sources with lower concentrations than those in primary sources are also recycled since these metals occur in large quantities and suitable economies of scale can be achieved. A typical example is the copper in buildings. The same effect occurs in the reverse sense. Thus, for example, the concentrations of rare-earth metals such as cerium in glass-polishing agents are considerably higher than those in ores, but the available quantities are very small (see Chapter S1.1 in the Supporting Information).

3.2. Energy Demand and End-of-Life Recycling Rate.

3.2.1. Copper. As shown in Figure 3, the CED of Cu recycling increases disproportionately with increasing EOL-RR. This phenomenon can essentially be ascribed to the different concentrations of the various secondary sources listed in Table 1. Currently, the EOL-RR of Cu in Germany is approximately 53%. The remaining 47% is the losses due to process inefficiencies and incomplete collection. Thus, all collected secondary sources of Cu are exploited as far as the current recycling system allows due to technical and economic limitations.

The current global production of primary Cu is mainly based on sulfuric ores, processed via the pyrometallurgical route, and oxidic ores, processed via hydrometallurgical processes. The International Copper Association reports a CED of 51 MJ/kg Cu for the average global primary Cu production.[57] This value can be validated by other studies.[58,59] For future development, it might be that the world's primary Cu production will increasingly shift toward the hydrometallurgical processing route,[51] which requires 65 MJ/kg Cu.[7] This higher CED value compared to the current global average is due to the use of chemicals in hydrometallurgy and to the fact that the pyrometallurgical processing route of sulfuric ores is an exothermic process.[60] However, this forecast is of course only a rough estimation as possible efficiency gains in the production technology are not considered.

The results show that Cu recycling of the analyzed secondary sources under current (Figure 3a) and future conditions (Figure 3b) contributes to the overall CED savings. Nevertheless, for some secondary sources the CED(x_i) are definitely on the same order of magnitude as the CED of primary production (see Table 1). This finding proves that Cu recycling is by no means always much more energy efficient than primary production, as is often claimed in the literature.[10,61,62] With slightly lower qualities of the secondary sources, it is quite possible that recycling offers no more energetic and environmental advantages but disadvantages.

As all Cu-containing secondary sources are currently being fed into appropriate recycling systems, there are still two options for improving the EOL-RR of Cu: reducing the losses due to process inefficiencies or improving the collection rate. Reducing the process inefficiencies will probably result in higher energy demands. This phenomenon needs to be examined in detail to ensure that negative trade-offs are avoided. The assumptions made by Ciacci et al.[35] whereby further improvements in the EOL-RR will immediately lead to a lowering of the overall GWP are quite uncertain. Enhancing the collection rates of end-of-life products is presumably associated with no or only marginal increases in effort. The CED per unit mass Cu, as listed in Table 1, will therefore likely remain unchanged. This estimation is based on the fact that the average distance for collection does not change with increasing collection rate in the defined area. Additionally, an increase in the collection rate is not only an energy concern

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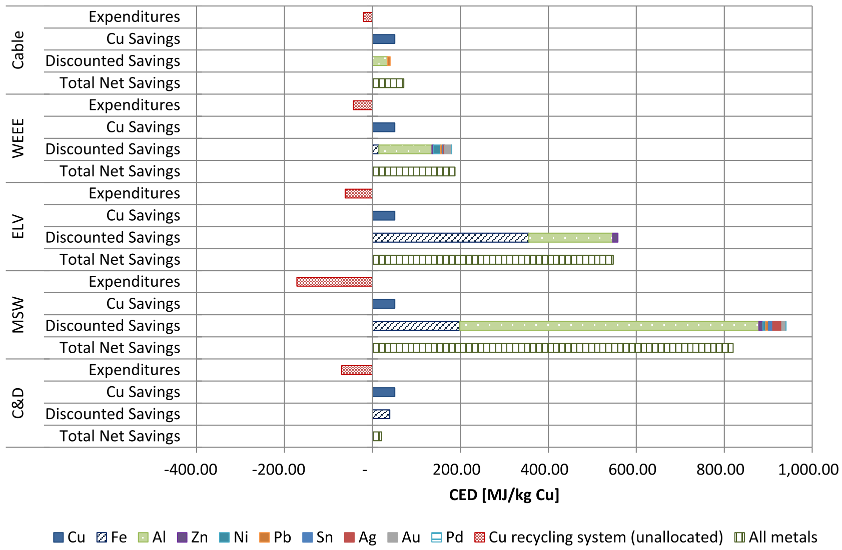

Figure 4. CED savings and expenditures of the expanded recycling system of copper for all analyzed secondary sources (for the data, see Chapter S1.3 in the Supporting Information).

but rather a societal or political challenge, as seen from the example of smartphones.[63,64] Hence, it can be assumed that there is still a great potential for increasing the EOL-RR of Cu in Germany with additional savings in the overall CED and GWP. However, since there are no data available for increasing the collection rates, this assumption is subject to certain uncertainties.

According to the results of the recycling model presented in Table 1, the average energy demand of end-of-life Cu recycling in Germany is 28 MJ/kg, which results in 1.7 kg CO_2 equiv/kg Cu. Based on the current EOL-RR of Cu in Germany, its recycling saves a total of 3900 TJ CED and 403 000 t CO_2 equiv per year. The mentioned increases in the collection rates of the analyzed secondary sources, without increases in the collection effort per mass unit of the secondary source, could lead to further CED (up to +63%) and GWP (up to +58%) savings.

However, these results are subject to several uncertainties. Secondary sources usually consist of numerous materials incorporated in complex matrix systems. The recycling of one specific metal from such complex systems fulfills not only the purpose of secondary production of this specific metal, but, usually, the process also supports the recycling of all other metals incorporated in this product. For Cu, this is true in two respects. Cu is a collector metal for several other metals, especially precious metals, and enables their recycling.[60] Furthermore, if Cu is not separated, the recycling of other metals can be hindered.[65] High impurity levels of Cu, for instance, in iron, will lower its quality and require the addition of pure primary metal.[66] Hence, the analysis of the recycling of one specific metal in a multimetal recycling system by allocation, as is usual in the sense of life-cycle assessment, is not able to cover these dimensions.

For Cu recycling, we propose an additional perspective that covers the total recycling system of the respective secondary sources to support the interpretation of the above-mentioned results. Figure 4 depicts the total CED expenditures and savings (achieved by substituting the primary material) of the extended Cu recycling system. The savings of all byproduct metals present during all recycling steps are discounted with their additional expenditures, e.g., the expenditure for the further processing steps of precious metal recycling out of the anode slime of Cu electrolysis or aluminum sorting and remelting after mechanical separation from the secondary source. It is clear in Figure 4 that all expanded Cu recycling systems are profitable regarding the total CED net savings. If the recycling of Cu from these secondary sources is reduced or even completely stopped, it can be assumed that the overall savings would decrease, according to the role of Cu in such recycling systems as a carrier or an impurity for other metals. Moreover, many of the CED expenditures of the recycling system accrue with or without Cu recycling, e.g., collection or shredding processes. Such challenges cannot be solved by improving the allocation rules, as proposed by Stamp et al.[67] A product-centric approach as proposed by Reuter et al.[68] is definitely one step in the right direction, but the approach does not cover the more complex reality involved. Rather, a kind of secondary-source-centered approach is required, covering all metals and materials contained in all of the products being processed together in the mechanical and metallurgical steps.

3.2.2. Tantalum. The calculated CEDs of end-of-life Ta recycling as well as detailed information about its secondary sources are presented in Table 2. The high CEDs are mainly due to the quite low concentrations of Ta in its secondary sources. In electronic products, the concentrations of Ta range from 0.004 to 0.119%.[25] Thus, large amounts of electronic scraps need to be processed to recover a small amount of Ta, thus resulting in a high CED per mass unit.

As Ta recycling has so far taken place only for alloys and carbides,[69] its EOL-RR in Germany is only approximately 11% (see Figure 5 and Supporting Information S1.4). The largest share of tantalum is currently used in capacitors in electronic products.[52] The Ta contents in these potential secondary sources are currently lost during the conventional recycling processes of electronic scraps.[70] However, Ta recycling is technically feasible[25] and is already done for capacitor production scraps.[71] Since the current Ta primary production from the tantalite ore requires an enormous CED of approximately 4000 MJ/kg Ta[72,73] (see also Chapter S1.5 in the Supporting Information), the CED-optimized EOL-RR is at the technically feasible maximum (25%) of Ta recycling. Therefore, Ta recycling from end-of-life capacitors can

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Table 2. Detailed Information about the Secondary Sources of Tantalum[a,b]

secondary sources		concentration (g/g)	total mass $m(x_i)$ (t)	$CED_S(x_i)$ (MJ/kg)
capacitors	mobile phones	1.2×10^{-3}	7.2×10^{1}	6.0×10^{2}
	smartphones	8.3×10^{-5}	5.6×10^{-2}	3.4×10^{3}
	tablets	1.5×10^{-4}	2.2×10^{-3}	1.2×10^{3}
	notebooks	8.1×10^{-4}	3.7×10^{-1}	5.3×10^{2}
	desktops	3.9×10^{-4}	2.0×10^{1}	6.0×10^{2}
	servers	7.1×10^{-5}	1.9×10^{-1}	6.5×10^{2}
	HDDs	3.4×10^{-4}	5.6×10^{-1}	5.6×10^{2}
	FSMs	4.4×10^{-5}	9.3×10^{-2}	1.8×10^{3}
	ELVs	5.5×10^{-6}	9.5×10^{-1}	3.7×10^{3}
carbides (cutting and drilling tools)		4.0×10^{-2}	4.0×10^{-2}	2.2×10^{1}
superalloys		4.8×10^{-2}	4.8×10^{-2}	2.4×10^{-1}
oxides (in optical glasses)		1.1×10^{-3}	1.1×10^{-3}	3.0×10^{-1}

[a]The masses are based on the data from the year 2013; for references and more information, see Chapter S1.4 in the Supporting Information [b]Hard disk drives (HDDs), flat screen monitors (FSMs), end-of-life vehicles (ELVs).

contribute to a decrease in the overall CED under the current conditions, as depicted in Figure 5a. In the future, Ta will increasingly be mined as a byproduct of lithium,[74] and the related CED will decrease significantly to approximately 900 MJ/kg.[59] However, the CED-optimized EOL-RR only decreases marginally to 24% (see Figure 5b).

As already mentioned in the case of Cu, these results are valid only for the current collection rates, and the CED-optimized EOL-RR will probably shift toward the technically feasible limit (69%) if the collection rates increase.

Based on the calculated data, the average CED for end-of-life Ta recycling is 564 MJ/kg Ta, and the GWP reaches 34 t CO_2 equiv/kg Ta. The total net savings due to the current EOL-RR of tantalum in Germany are approximately 5.4 TJ and 362 t CO_2 equiv per year. The CED-optimized EOL-RR would improve these savings to 11.8 TJ and 781 t CO_2 equiv, respectively. Enhanced collection rates could further improve the savings by up to 160% (CED) and 161% (GWP).

In contrast to Cu recycling as an example of multimetal recycling, the Ta recycling system is a more isolated system. The recycling of capacitors, for instance, with the exception of dismantling the printed wiring boards on which the capacitors are located, is not interconnected with any byproducts. Unless Ta is separated specifically for recycling purposes, the metal ends up in the slag in most metallurgic processes and therefore does not contaminate other metal streams.[75] In such cases of isolated recycling systems, the proposed single-metal approach is expedient, and the optimized EOL-RR results are valid.

For Ta, as well as for many other specialty metals, the problem of the small amounts available for recycling exists. In 2013, only 12 t of Ta in end-of-life capacitors were theoretically available for recycling in Germany. Although quantities will increase in the coming years,[76] intelligent and probably transboundary recycling logistics are needed to cope with the small quantities widely distributed across the different secondary sources and to attain suitable economies of scale.

Of particular interest in Ta recycling from capacitors is that although the concentrations in these secondary sources can be lower than those in ores, recycling is more energy efficient than primary production. This phenomenon is due to the significant concentration increase by an average factor of 27 by simply dismantling the printed wiring board from the entire electronic product (see Chapter S1.4 in the Supporting Information). This effect is generally a major advantage of recycling as long as the target metal is located in one or a few specific and accessible product components, such as printed wiring boards. It can also be observed for neodymium magnets incorporated in electronic products.[27] However, this effect has its limits. Taking the example of indium in electronic products, which

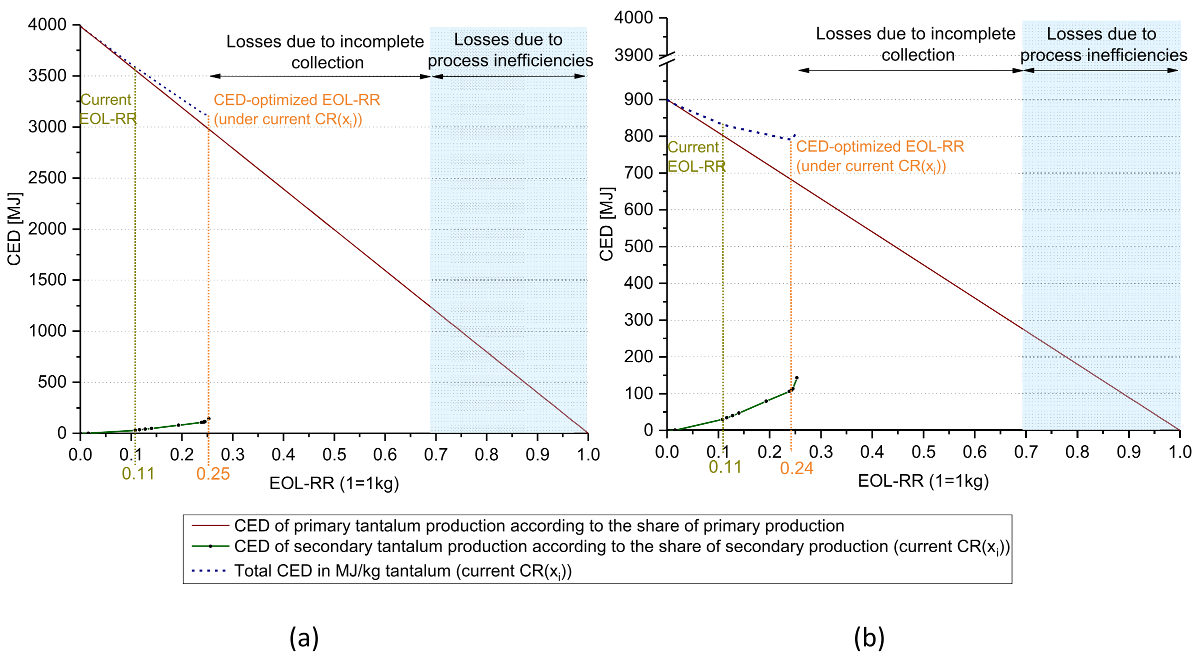

Figure 5. Application of the optimization method for the CED-optimized EOL-RR in Figure 1 to tantalum production. The EOL-RR is scaled to 100% equaling 1 kg tantalum. (a) Current situation of primary tantalum production; (b) future situation of primary tantalum production. The values are valid for Germany.

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occurs in LCDs, its concentration is 10−100 ppm.[77] The concentration in ores is 20−350 ppm,[78,79] which is not substantially higher; nevertheless, the CED of recycling is much higher than that of the production from ores.[31]

3.3. Overall Discussion. The optimization approach we have proposed is aimed at mitigating the impacts of metal production on climate change by treating recycling not as an end in itself, in terms of closing material cycles, but rather as an additional option of metal production that requires energy and leads to environmental pollution, just as is the case with primary production. Unlike the existing considerations, which are merely theoretical, we have determined realistic and discrete data to calculate the CED of end-of-life metal recycling based on comprehensive empirical system models, which proves that recycling can significantly contribute to reducing climate change but by no means without limits. Even Cu recycling, a prime example of a well-functioning recycling system, requires a similarly high CED for its low-concentration secondary sources as that of primary production. This finding, combined with the knowledge of the partly enormous dilution of metals in the technosphere, implies that completely closed cycles in terms of a circular economy would lead to negative trade-offs and intensify climate change. Nevertheless, in certain cases, the recycling of very low-concentration secondary sources can also be energetically advantageous, as the example of Ta recycling shows. Continuous improvements in recycling technologies can further enhance these advantages.

Finally, we would like to address two methodological limitations. First, we assume that primary production would be substituted by secondary production in such a way that primary production could be assessed with an average CED. However, CEDs for primary production also vary depending on mine type, ore type, and ore grade. Thus, the question is, what is actually being substituted? If the decisive factor for substitution were the CED, we could assume an order of substitution according to the merit-order principle, known from the energy sector. This means that the primary sources with the highest CED will be substituted first, followed by the more-energy-efficient ones. In reality, substitution is likely to be based on economic aspects. In this context, investments made for several decades and economies of scale should be taken into account. It can therefore be assumed that the primary production system tends to react inelastically to substitution and that the most expensive mines are not necessarily the first to be decommissioned. All of these considerations are based on the assumption that already existing mines will be substituted. However, as demand for metals is growing steadily, recycling is replacing mines that do not yet exist. The assumption of an average CED for substituted primary production is a first approximation, and there is still a lot of research to be done in this area to refine our initial results further.

The second limitation relates to our comparison of metal concentrations, where we compare the concentrations of metals in their initial sources. Here, we would like to point out the byproduct production, which is common in both cases of metal production, primary and secondary. Such byproduct productions enable the extraction of the low-concentrated metals, in particular, minor metals, only because they are favored by the extraction of the major metal. Tellurium, for example, is relatively low concentrated in primary ores at 0.008%;[80] its concentration after the extraction of copper, the related major metal, is about 2−10%.[81] Only at this point, the extraction is economically sensible. The same effect can also be observed in secondary production, e.g., in the recycling of WEEE. However, especially in secondary production, but also in primary production, concentrations of byproducts in the sources can be decisive for whether a source is mined. Therefore, it is not possible, on the basis of current knowledge, to clearly define where in the production processes the concentrations are most meaningful for such a comparison. Our analysis is consequently only a first indication of how the metal concentration ratio of primary and secondary sources influences recycling activities.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.9b05101.

Metal concentrations in secondary and primary sources; modeling and mathematical description; data for the copper and tantalum recycling system models, including input data, allocation factors, mass balances, and results in terms of CED and GWP; and CED and GWP for primary production of tantalum and copper (PDF)

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Notes

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