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Regionalized life cycle inventories of global sulfidic copper tailings

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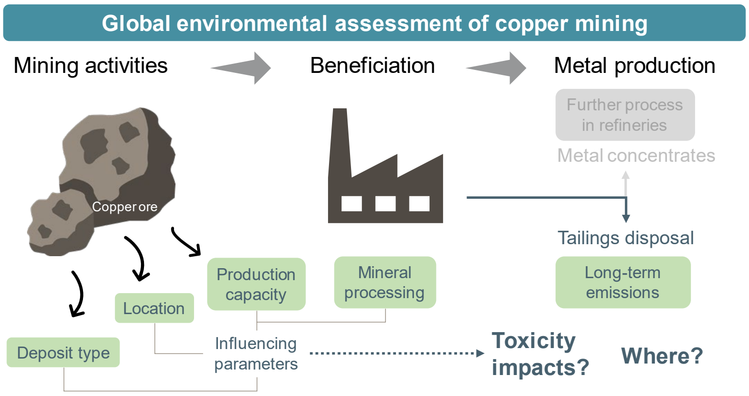
Abstract

Worldwide, an issue of copper production is the generation of mine waste with varying characteristics. This waste can pollute natural environments, and in particular the heavy metal emissions of the tailings may pose long-term consequences. Currently, life cycle assessments of mine tailings are hampered by both limited data availability in the metal production value chain and lack of appropriate methodologies. We collect data from 431 active copper mine sites using a combination of information available from the market research and technical handbooks to develop site-specific life cycle inventories for tailings disposal. The approach considers the influences of copper ore composition and local hydrology for dynamically estimating leached metals of tailings at each site. The analysis reveals that together, copper tailings from the large (i.e., porphyry) and medium-size copper deposits (i.e., volcanogenic massive sulfide and sediment-hosted) contribute to more than three-quarters of the total global freshwater ecotoxicity impacts of copper tailings. This strongly correlates with hydrological conditions leading to high infiltration rates. The generated inventories vary locally, even within single countries, showcasing the importance of site-specific models. Our study provides site-specific, dynamic emission models and thus improves the accuracy of tailing's inventories and toxicity-related impacts.

Keywords: site-specific inventory models, ore mining, mine tailings, ecotoxicity impacts, tailings geochemistry, metal production, mineral processing, life cycle assessment

Synopsis: This research combines state-of-the-art environmental and mineral processing frameworks, indicating highly variable impacts caused by sulfidic copper tailings deposited worldwide.

Graphical Abstract



Introduction

According to the UNEP-IRP Global Resources Outlook 2019, the use of natural resources has tripled in the last four decades[1], and if business as usual is maintained[2] in the production processes, the expected future environmental impacts will be exacerbated. Thus, it is imperative to more sustainably produce materials that support our modern lives. One key metal to respond to these challenges is copper. Notable examples are the use of copper as essential components in renewable energy systems, i.e., solar panels and wind turbines. With various possible use cases and incentives to transition to a low carbon economy, it is estimated that copper demand will grow up to four-fold in less than half of a century[3]. However, the environmental implications of this transition depend on the technological routes to satisfy future copper demands[4–6].

Various types of copper sulfide ore deposits are the primary source of metallic copper, accounting for 80% of copper resource[7]. The production of copper from ore deposits requires the separation of unwanted impurities such as silicates, carbonates, and sulfides. This comprises several activities that generate considerable wastes, such as waste rocks from mining and residues/slags from metallurgical processing and refining[8–10] (Figure S29). In between these processes, there is beneficiation: a technology prominently used to extract metals from ores. This requires the usage of chemicals and also produces mineral processing waste. These waste slurries, otherwise called tailings, are then discharged to legally operated storage facilities. Due to an inherently low concentration of copper in ores, tailings are generated at an enormous amount, accounting for more than 90% of the input ore[10,11]. Declining copper grades in deposits might worsen this situation, as it implies more tailings will have to be managed per kg of copper produced[12].

Environmental disruption related to the tailings generation and deposition is inevitable. Over time, poorly managed tailings can interact with the surroundings such as rainwater and oxygen, and subsequently initiate acid mine drainage[8], which leads to elevated heavy metals concentration in the environment. The composition of tailings can vary among copper mines due to different geological properties and processing schemes[13,14], the differences of which are important when considering leaching behaviors due to presence of acid-/ base producing minerals[15,16].

Several tools and databases are available to assess environmental performances of metal production value chains. In a broader context, the criticality assessment concepts by Graedel[17,18], Cimprich et al.[19], and Bach et al.[20] translate qualitative criteria into criticality scores by assessing environmental implications, supply risk, vulnerability to supply restriction, and socio-economic dimensions. Official public databases, such as pollutant release and transfer registers (PRTR)[21], record pollutants released to the environment, but with varying level of detail depending on the specific requirements of local environmental authorities[22–25]. Life cycle assessment (LCA) is a standardized approach to assess the processes' impacts throughout the entire metal value chain[26,27]. However, with respect to tailing emissions, many LCA studies fail to report the inventories due to lack of methods, data limitations, and unrealistic data collection efforts[28,29]. This persisting issue has been discussed[30] and worked around by other researchers in the LCA field by applying a waste-specific transfer coefficients model[31] that was initially developed for landfill emissions[32]. When specific mine data are available, one could also build the inventories for the current conditions as demonstrated by others[33–35]. While this might provide tailings details for sites operating under similar conditions, the major shortcoming is that the inventories are based on averaged data in multiple locations to represent specific mining production. Other drawbacks are the neglect of influential site-specific input parameters and, more importantly, the dynamics of leaching in the long term. Therefore, the results of LCA studies that include toxicity impacts from tailings may differ significantly or be completely underreported/ overreported[5,6,29,36,37].

The goal of this study is 1) to provide a global assessment of sulfidic copper tailings using state-of-the-art frameworks in minerals processing, hydrological modelling, and environmental assessment; and 2) to identify the environmental hotspots by a dynamic assessment at mine-site level, which provides a better understanding of the overall impacts of current and future copper production.

Materials and Methods

Methodology overview

Our main methodology built upon previous work in the advancement of mineral processing[26,38], subsurface environmental simulation[39], and the environmental impacts of mine tailings in the LCA of metal production[33,34,40]. The methodology integrated several key frameworks that link the workflows for estimating the environmental impacts of mine tailings (Figure 1 top). Because copper has the most comprehensive mineral and production database available[41] and is of high production volume with an increasing trend, we have chosen to apply these methods to sulfidic copper tailings.

Our approach was divided into four main parts: 1) compilation of copper-active production and ore mineralogy for each production site, 2) process-based approximation of tailings composition, 3) site- and time-dependent life cycle inventory modeling of mine tailings emission, and 4) global impact assessment of sulfidic copper tailings over different time horizons.

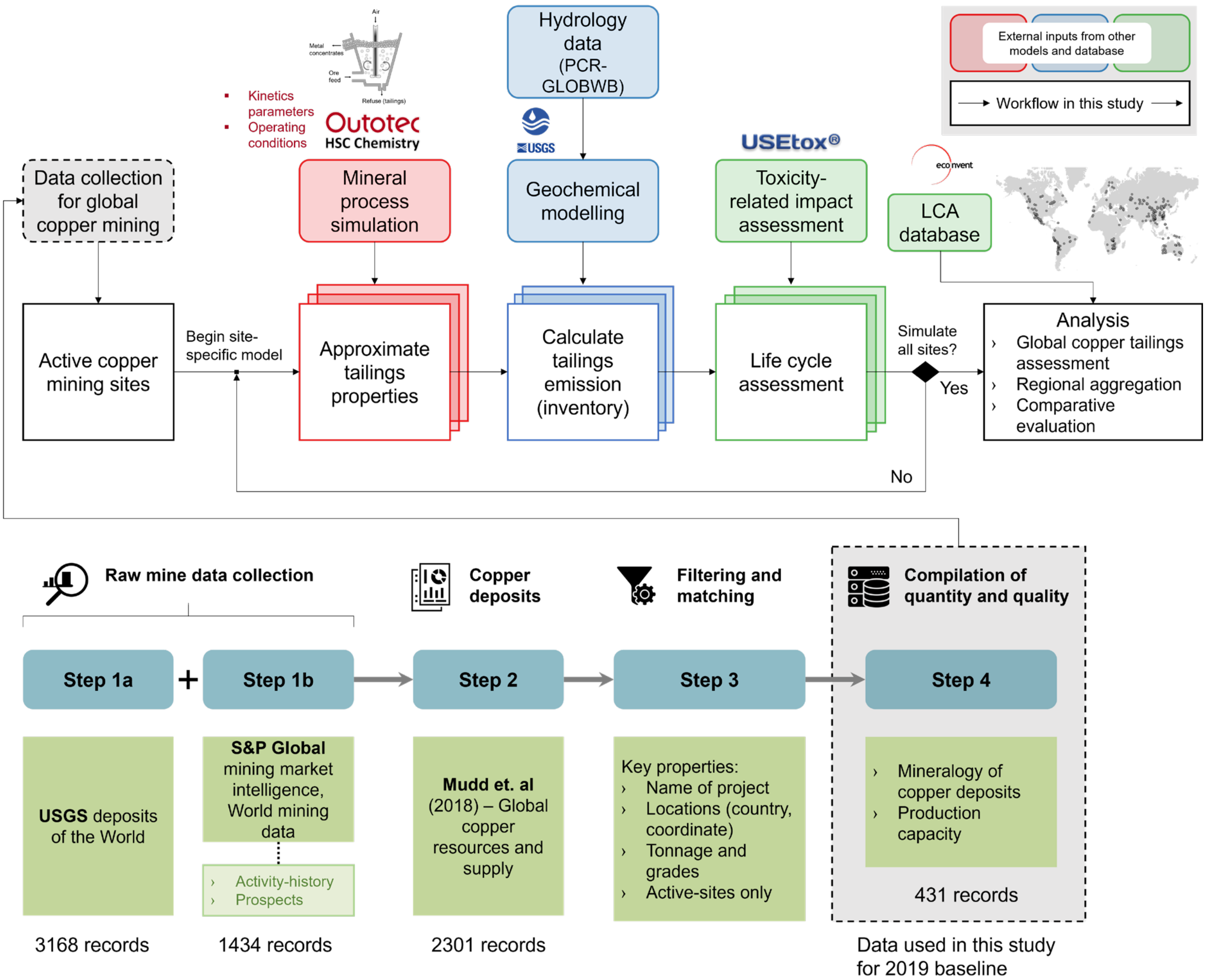


Figure 1. Schematic illustration of the methodology employed (top) and sources of copper production data (bottom)

Copper production data

The database compiled in this study combined extensive resources from 1) *U.S. Geological Survey Mineral Database*[42], 2) *S&P Market Intelligence Metals and Mining Report*[43], 3) *World Mining data 2020*[44], and 4) a rigorous copper deposit study[41]. Together they represented more than 75% of annual global sulfidic copper production in 2019, along with ore deposit characteristics, which indicate the mineralogy of each mine deposit for specific production sites. Of total production data, we specifically focus on the copper production process via pyrometallurgical pathway since it represents the dominant technology to produce copper (see SI-1 Section S2). If the data for ore deposits were unavailable in the previously mentioned databases, we linked them with the closest deposit sources based on its geographical coordinates. The workflow to obtain the baseline data for the global assessment is presented in Figure 1 bottom.

Beneficiation process modelling and simulation

Beneficiation of sulfidic deposits comprises a combination of physical and chemical processes to transform raw copper ore into metal concentrates and tailings as waste. In this study, we developed a systematic method to build a tailings composition database based on processing steps and as a function of ore characteristics. This is critical, as the mineralogy of each deposit defines the necessary separation process of valuable metals from non-valuable gangue materials, which can act both as buffering minerals and/or acid-accelerating agents in the tailings. The chosen separation process ultimately dictates the tailings properties of the tailings for every site. To complete this task, we classified the copper ore deposits based on the formation grouping of Heinrich and Candela[45], with additional sub-classification based on copper grades, buffering minerals, and other impurities[41,46]. Their approaches provide necessary classification guidelines, and all compiled active copper-sites are presented in Table S1.

To link the data from the previous step with a beneficiation process, we constructed simplified flowsheets (illustrated in Figure S1) with industrial process parameters in the software Outotec HSC Chemistry 10 “Flowsheet Simulation” feature[47]. This approach is similar to what others have done[48,49].

Our approach simulated the behavior of flotation schemes in the beneficiation process through a ‘three-component’ minerals floatability process. It parameterizes the minerals flotation as a first-order kinetic equation[50,51]. This model yields the recovery of a mineral R_m to the flotation time t as follows:

$$\boldsymbol{R_m = m_f\left(1 - e^{-k_f t}\right) + m_s\left(1 - e^{-k_s t}\right) + 0 \cdot m_n} \quad (1)$$

Where

m_f and m_s represent the proportion of fast and slow particles, respectively;

k_f and k_s represent the flotation rate constant of fast and slow floating particles, respectively;

m_n represents the proportion of non-floating particles such that $m_f + m_s + m_n = 1$

Using equation (1), we approximated the characteristics of the tailings of each mine production site as a function of its ore deposits inputs and flotation process parameters. The flotation parameter details (kinetics data, recovery, minerals, and reagents) for each beneficiation process were primarily obtained from the *Handbook of Flotation Reagents*[52] and *Will’s Mineral Processing Technology*[53]. Other sources, such as a collection of flotation studies and patent literatures[54,55], were also used, specifically for the flotation of chalcopyrite-containing ore deposits. We then used the HSC 10’s “Geo Module” feature to extract mineral characteristics from the database. Since we aimed to approximate tailings composition based on publicly available data, the mineralogy input for different deposits were assumed to contain generic chemical compositions. The list of parameters used in building up the mineral processing simulations are exemplified in Figure S2 and tabulated in Table S2-S3.

Life cycle assessment of copper tailings disposal

Our study focuses on the end-of-life phase of waste, in this case, tailings from the production of copper. In accordance with the ecoinvent database[56], we first relate all emissions and impact to the waste-treatment service "disposal of 1 tonne of tailings from copper ore concentration at a specific mine site". We then extend the functional unit to "kg copper produced" by analyzing the entire production value chain, as this is the final purpose of copper mining (i.e., providing copper to the society). Finally, we also quantified total emissions and impacts of tailings for the entire mining sites, referring to one year of total copper production and the resulting tailings treatment.

To model tailing emissions at site-specific locations, we considered tailing characteristics, as a function of mine composition and processing technology (previous sections) and hydrological conditions. All heavy metal emissions in the study were allocated to copper production, representing the worst-case situation without allocation of part of the emissions and impacts to by-products. This represents a conservative approach, as the impacts allocated to copper will be overestimated. As all the inventory data is transparently presented in this paper, future research may apply other allocation techniques, such as by mass or economic value[57]. The remaining copper processing inventories were taken directly from the Life Cycle Inventory (LCI) database ecoinvent 3.6[56]. The illustration of the studied system is depicted in Figure 2.

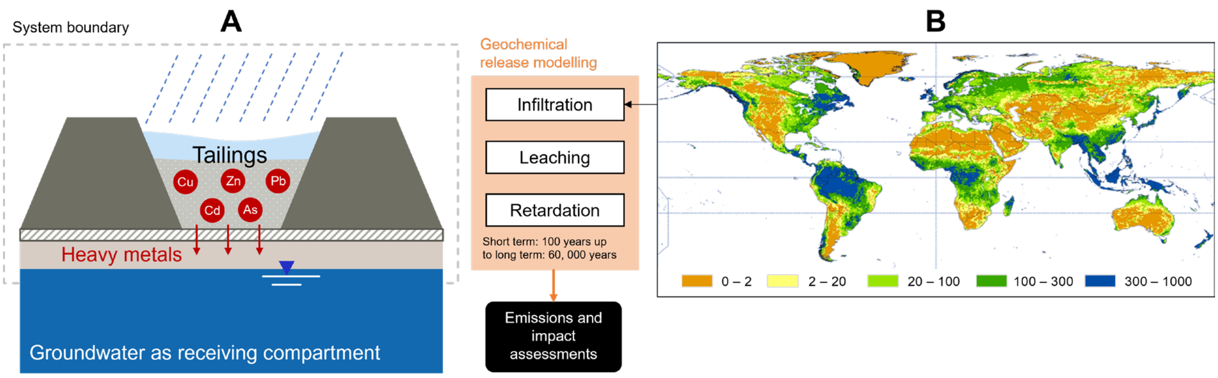


Figure 2. The schematic illustration of tailings emission model. Part A describes the tailings characteristics and metal species considered in this study. Part B shows the annual groundwater recharge, taken from the results of PCR-GLOBWB[58], in mm per year.

This study focuses on toxicity-related environmental impacts by using the latest midpoint impact categories recommended for life cycle impact assessment[59,60]. This includes freshwater ecotoxicity, for which impacts were quantified by applying global characterization factors (CFs) (defined in USEtox 2.12[61]) to leaching emissions. We assumed that all leached heavy metals would be transported to freshwater. The main reason for this simplifying assumption was the lack of groundwater CFs in USEtox 2.12. In the impact assessment, no spatial differentiation was considered. Furthermore, no emissions to air via dust were assessed in this study, assuming their contribution is small in the overall system[33] (see SI-1 section S16). This may be different at very arid sites.

We calculated the heavy metal emissions for short-term (100 years) up to a long-term period (60,000 years) for comparative purposes with the ecoinvent database. While this is an explicit (somewhat arbitrary) value-choice, our continuous long-term model allows future researchers to also define different time frames[62].

To predict emissions for a long time horizon, we applied geochemical modeling using the PHREEQC simulation[63]. This model allows the prediction of heavy metals concentration in the tailings, which are controlled by mineralogy and pH development[64,65]. We assumed that the technical lifetime of the storage basin integrity is limited, so that the technical barriers were neglected for the long-term assessment. All minerals in the tailings were assumed to come in contact with the leaching water (no enclosures) and could eventually seep into groundwater (controlled by solubility). We quantified the dissolved concentrations of Cu, Cd, Pb, Zn, and As in the leachate at equilibrium with a set of solid phases. These substances represent the most toxic and mobile heavy metals present in the leachate of copper tailings[66]. For As, the surface complexation reactions were obtained from the Dzombak model[67], which assumes that arsenic attaches on hydrous ferric oxide (HFO) surfaces. Parameters and thermodynamic reactions, which we included in PHREEQC speciation-solubility modeling, are provided in Table S4 and S13, following the approach of Hansen et al.[16] and Dijkstra et al.[68], based on the PHREEQC and WATEQ4F databases. This approach allows for consistent geochemical modelling for generation and mobility of leachate from mine waste. Another input parameter was a matrix infiltration rate for each site, which was taken from the global hydrology model PCR-GLOBWB 2[58]. We used the net groundwater recharge as the site-specific infiltration parameter in our calculation (see part B of Figure 2). This rate represents infiltration due to climate into the natural soil. We disregarded any alteration (typically reduction) of the infiltration rate by rehabilitation measures, as our assumption is that active rehabilitation will not be continued in the long term[69].

We ran the geochemical models for tailing of each sulfidic deposit type (i.e., porphyry, volcanogenic massive sulfide, skarn, sediment-hosted, magmatic sulfide, iron oxide, intrusion-related, and epithermal copper deposit in Table S5-S12 and Figure S6-S13) to obtain the concentration of heavy metal emissions over time for each site. Other minerals were normalized following the composition of copper in the deposit (Figure S5 and SI-1 section S4). The accumulated leached metals over the pre-defined time frames were then calculated following equation 2. This is similar to what was done in other contaminant release studies[70,71], but is adapted to the specific mine tailings composition, deposit dimensions and site-specific climatic conditions.

$$M_{x,\ total} = \sum_{t=t_0}^{t_n} PR \cdot A \cdot t_{timestep} \cdot (C_x(t)) \quad (2)$$

Where, M_x (mg) is the product of total emissions for metal x in the defined t_n time horizons, PR (l/m^2.a) represents the net matrix infiltration rate from the global hydrology model, $t_{timestep}$ the time step (a) for every simulation within geochemical modeling, and $C_x(t)$ the concentration of metal x in the leachate (mg/ l) at time t as the results from the geochemical simulation. A (m^2) is the surface area related to 1 tonne of tailings material and was calculated from the following equation (3).

$$A = \frac{1\ t\ tailings\ material}{\rho_{tailings} \cdot d} \quad (3)$$

Where $\rho_{tailings}$ (kg tailings/m^3) and d (m) are the density and the thickness of the tailings, respectively (see parameters used in Table S4).

Baseline scenario: Analyzing the environmental hotspots

The life cycle impact assessment (LCIA) results under different time horizons were analyzed and mapped for each site to identify global hotspots. The environmental impacts were quantified per kg of copper production and as total impacts per mine for one year of mine operation. The latter were calculated by multiplying the mass of copper produced for each site in 2019 with the results per tonne of copper. As copper mining activity only represents a part of the life cycle of copper production, we embedded our generated tailings inventory into the available primary copper production inventory of ecoinvent 3.6[56] (based on the LCA report of copper supply chain analysis[72]). The overall procedure is illustrated in Figure S15.

Afterwards, we analyzed these spatially- and time-resolved mine tailings inventory data in three ways. First, we analyzed the influences of ore deposits and metallurgical processing configurations on the overall tailings' emissions. To further study the interaction between ore deposit and infiltration rates, a sub-analysis was performed for a specific ore deposit type with broad ranges of composition (Table S5-12). Second, the LCIA results of one year of operation of all mines within any country were aggregated. Last, we compared the results of this study with the eleven country specific sulfidic tailings inventory datasets in ecoinvent 3.6[56]. We matched our modeled inventory in this study following the country classifications that ecoinvent implements (Figure S16). In all these steps, we chose a long-term time horizon (i.e., 60,000 years) to conservatively account for heavy metal emissions being leached out from the system and to be consistent with the assumptions taken in ecoinvent. For the evaluation of the effect of various choices concerning time horizons, we also present results for a 100 year time horizon.

Modeling future global copper tailings emissions

The primary supply of copper until 2050 provided by Elshkaki et al.[4] and Northey et al.[73] have been used to derive future projections of copper provision. In this study, we prepared three scenarios, namely copper supply in year 2030, 2040, and 2050 from the above-mentioned data sources. A data reconciliation, however, was necessary as the forecasted data from previous studies do not contain the location of expanded or newly established sites. Therefore, the data gaps for new mine and expansion projects were based on an undiscovered copper resources study[46], S&P feasibility study[43], and other reports[74,75] (see Figure S17).

Two approaches were taken for the development of site-specific copper supply:

1. Case of sites expansion (sites in the base scenario)

We estimated the copper grade decline until the year 2050 following Crowson[12], as described in equation (4) and applied the global decline rate to the specific grade of each mine, assuming continuous production with the same ore extraction rates over time.

$$\boldsymbol{G = 4 \cdot 10^{82} \cdot y^{-25.05}} \quad (4)$$

Where G is the copper ore grade (%) in year y.

The production of the mine sites in the current scenario is the starting point for this base case. We assumed the production remains constant until all the resources are depleted and replaced by the second case (see below). Thus, if time until depletion (resources/annual production capacity) is < 30 years, the mine is considered no longer operational by 2050.

2. Case of new sites (sites under pre-production in the base scenario)

According to ICMM[76], the life cycle of a mine prior to operation can be distinguished into two stages: exploration and construction (Figure S17). The discovery phase of copper production was excluded in this analysis, as it takes an average 20 years before a mine can finally operate[77].

Using these approaches (see SI-1, section S8), the newly opened sites will have initial copper grades according to the latest exploration data whenever information is available. Otherwise, we defined the grade as the highest achievable according to the USGS[42] deposit characteristics database. Finally, we combined the merged forecasted data with our method to estimate the environmental impacts of tailings until 2050.

Results and discussion

Global assessment of copper mine tailings

The toxicity impacts of all 431 assessed active copper sites in 2019 are shown in Figure 3A. The detailed results are available in the digital SI. These sites capture >75%[41–43] of global sulfidic copper tailings disposal. Most copper is mined from porphyry copper deposit type, which accounts for almost half of the total number of sites and is distributed across continents. The toxicity impacts per tonne of tailings due to porphyry copper tailings disposal are generally lower than from other deposit types. However, each mine has its own ore composition that directly influences the beneficiation scheme and respective tailings compositions. Our results (Figure 3 and SI-2 Table S2.3) show that the highest total annual toxicities per mine site are found in:

- Large-size, porphyry copper mining sites in the Americas (i.e., Chile, Peru, USA) and Asia (Indonesia, Papua New Guinea)
- Medium-size, sediment-hosted and volcanogenic massive sulfide-deposit sites in Canada, Africa (i.e., DR Congo, Zambia) and a few in Europe (Russia, Poland)

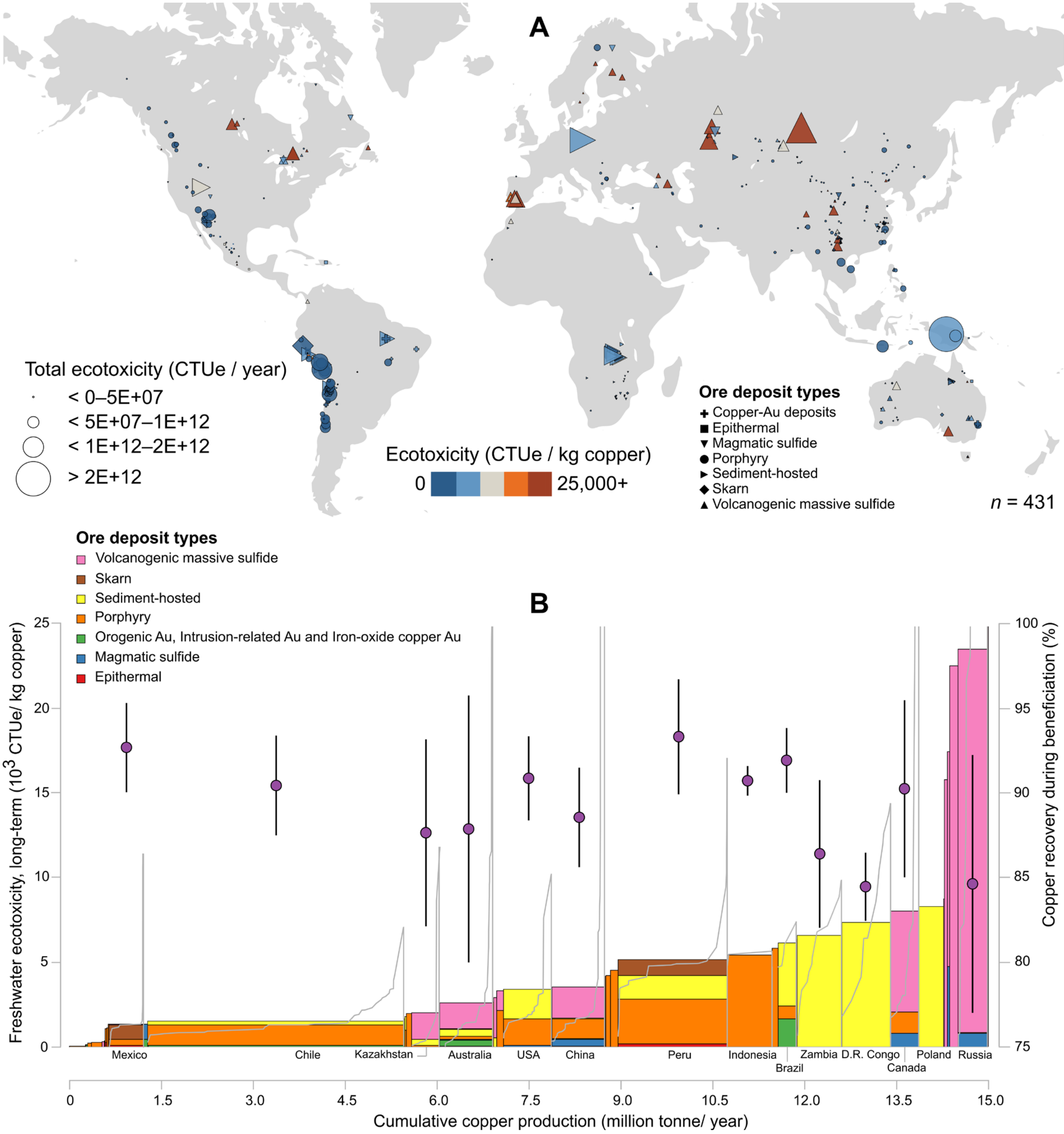


Figure 3. Part A shows the freshwater ecotoxicity (long-term) of copper production for each mine site (LCIA method: USETox). Three features are displayed: 1) total ecotoxicity (indicated by bubble size), 2) ecotoxicity per copper mass produced (indicated by color), and 3) the type of ore deposits (indicated by shape). Part B displays copper mine tailings freshwater ecotoxicity for each country and the distributions. 1) Stacked bars represent ore deposit types, 2) width is equivalent to annual production capacity, 3) left y-axis represents the toxicity impacts, both weighted average per country and spread per country shown in gray line, 4) right y-axis shows the ranges of copper beneficiation recovery with production-amount weighted average (purple circles) and error bars as weighted standard deviation.

Region-specific and country-aggregated assessment

For background LCI databases, country-level data is required and Figure 3B shows the variabilities that can exist in particular countries and how deposit types and beneficiation contribute to the results. The weighted global average for long-term freshwater ecotoxicity is 4.6×10^3 Comparative Toxic Units for Ecotoxicity (CTUe)/kg copper produced, while the median value is 2.0×10^3 CTUe/ kg copper produced. While Chile mainly sources from porphyry, with rather low impacts, countries like Australia, China, Peru, and Canada have more varying deposit types and therefore higher impacts. Since various deposit types require different beneficiation processes, the level of heavy metals in the tailings can change. In particular, for volcanogenic massive sulfide and sediment-hosted deposits found in Russia and DR Congo, the beneficiation performs particularly subpar[52,53].

Results show that nearly 70% of our worldwide ecotoxicity impacts are occurring in seven countries: Russia (17%), Peru (14%), Chile (10%), DR Congo (8%), Zambia (7%), Indonesia (6%), and Canada (5%). Details for all countries are shown in Figure S21 and Table S16.

Influences of climate conditions and ore deposit types

Higher net positive infiltration generally leads to larger amounts of heavy metals being carried to the soil and groundwater compartment (Figure S22). Ecotoxicity per kg of copper decreases with an increasing copper grade, but correlations are very weak (Figure S24). Volcanogenic massive sulfides and sediment-hosted deposits have relatively higher emissions in the same climatic conditions (i.e., infiltration rates between 40 – 140 mm/ year) due to higher amounts of pyrite but smaller buffering capacities (i.e., calcite and dolomite). However, several high-grade copper sites are situated in regions with low infiltration and thus relatively have low emissions (Figure S25).

Comparison of leaching and toxicity results to other studies

The comparison of our toxicity results with country-specific datasets in the ecoinvent 3.6[56] per tonne of tailings is presented in the top part of Figure 4A for short-term (100 years) and long-term (60,000 years) time horizons. For a short-term horizon, our study generally depicts lower toxicity impacts compared to ecoinvent. There is, however, high variability within countries and short-term emissions is of low importance for ecotoxicity in copper production (Figure 4B).

Ecoinvent's representation of the Rest of World (RoW) category may be especially sensitive to regional details, as it aggregates data from several large copper producing countries with varying deposit types and climate conditions (e.g., DR Congo, Poland, and Brazil). Our analysis found a very wide range of ecotoxicity impacts for countries considered in this category (from as low as 0.03 up to 440 CTUe/ tonne of tailings). We therefore suggest that future RoW data includes the variabilities in uncertainties to indicate where large differences in toxicities exist, and detailed assessments should be used to improve the data. In addition to tailings composition, tailings management has a significant impact on toxicity. For instance, direct tailings discharge into the environment, such as practiced in notorious mine sites in Indonesia and Papua New Guinea, i.e., OK Tedi and Grasberg[78,79] pollute fresh water immediately, highlighting the need for alternative disposal methods[80].

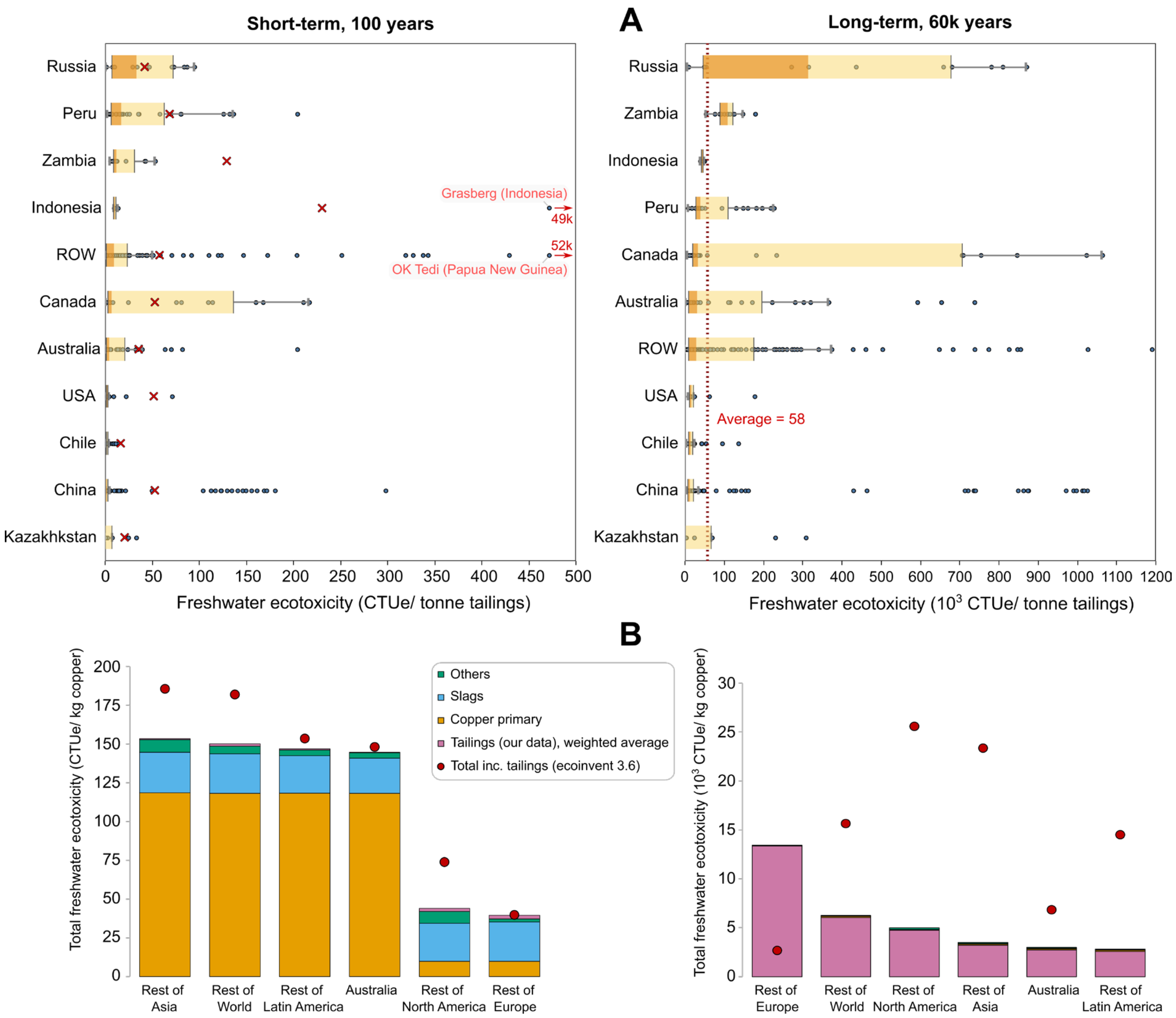


Figure 4. Part A: Freshwater ecotoxicity impacts quantified per tonne of tailings deposited for countries covered in ecoinvent. The red cross symbols indicate values from ecoinvent for the short-term horizon. Long-term variability from ecoinvent is not shown due to negligible differences between countries (hence, only a single average value as red dashed line). The width of each box represents 25th percentile Q1 (dark orange) and 75th percentile Q3 (yellow), while the whiskers represent 1.5*interquartile range from the Q1 and Q3. Any points outside the whiskers are outliers. The log-scale chart is presented in Figure S27. Part B: Freshwater ecotoxicity impacts per kg of copper for short-term (left) and long-term (right) perspectives in different world regions. Data to generate this chart is available in SI-2, Table S2.10-S2.11.

In the long-term time horizon, our analysis shows that toxicity results (median) are mostly lower than in ecoinvent due to differing tailings property modelling approaches. Ecoinvent's approaches, first, lacked differentiation between copper deposit composition on the individual mine level and, second, assumed almost complete leaching of all tailing's components in the long-term contemplations. Instead, in our study, we applied a set of systematic procedures and models to quantify tailings compositions and leaching at mine-level resolution.

To evaluate the ecotoxicity impacts of tailings in the life-cycle perspective of copper production, we performed an LCA study at a continent-level of ecoinvent (Figure 4B). In the short-term time horizon, the primary copper production process including smelting, refining, and slag deposition contribute more than 90% of the total ecotoxicity impacts for all continents. The findings are also supported by analyses done at higher granularity (Table S18), where there are generally negligible differences in ecotoxicity values between continents. In the long-term perspective, tailings dominate (>95%) the freshwater ecotoxicity impacts of copper production for all regions. It is therefore of utmost importance to properly assess toxicity impacts of tailings (see SI-1 section S14) and one should avoid ignoring differences between sites when performing comparative LCA studies.

Impacts of future primary copper production

Freshwater toxicity impacts in the upcoming decades based on projected future production are shown in Figure 5. Globally, copper tailings were responsible for 6.8E+13 CTUe/year in 2019, which represents the baseline for the following analysis. According to the projection of primary copper mining from other studies[4,73,77], the production will reach a peak level at 2030 and flatten after 2050 thanks to direct reuse from stocks and availability from recycling streams.

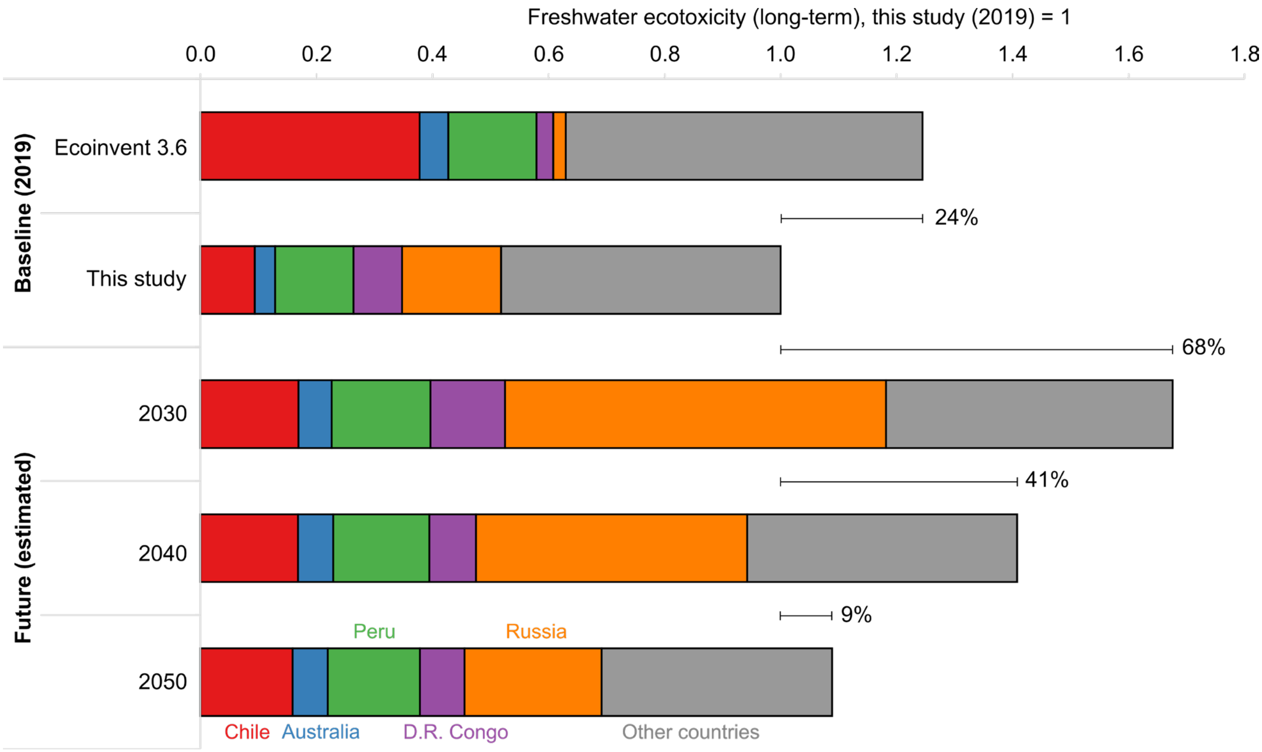


Figure 5. Freshwater ecotoxicity impacts from one year of operation of all global sulfidic tailings (long-term time horizon of 60'000 years), including future projection of copper-extraction amounts from other studies[4,73]

The increase in copper production for both site expansions and new discoveries will influence the environmental implications caused by tailings deposition. It is anticipated that Chile will continue to be the top copper producer for the next three decades. However, strong production increases are predicted for Russia, Australia, and DR Congo, with the discovery of high-rank copper deposits of volcanogenic massive sulfide and sediment-hosted deposit types[11,12,41] in high infiltrating regions, which have tendencies towards higher toxicity levels in the tailings.

Once society shifts steadily towards secondary copper resources (after 2030), a decrease in toxicity impacts is anticipated, from a ratio of 1.68 in 2030 and 1.41 in 2040 to 1.09 in 2050 (compared to the baseline year 2019, Figure 5 and Table S14). The primary copper demand in 2030 largely affects the increase of ecotoxicity impacts more than degrading ore grades quality. The impacts caused by ore grade decline start to appear after

2030, where lower quality ore grades in 2050 show 32% contribution at the highest (Table S14). Although much of the copper is provided through recycling in the scenario for 2050, primary copper extraction and impacts from sulfidic copper tailings are still expected to increase.

In addition to the freshwater ecotoxicity-related impacts completed in this study, we also conducted environmental impacts for human-toxicity and other LCIA impact categories using the ReCiPe 2016, endpoint, Hierarchist version[81] and the Environmental footprint (EF) method[82], which also provide an aggregated single-score impact result. Other processes (i.e., copper refinery) in the value chain show a higher contribution in the total results, namely due to particulate matter and gaseous emissions from smelters. Results are sensitive to the methods chosen, but metal emissions from tailings are still responsible for ecotoxicity and human toxicity-related to tailings impacts (contributing to around 27–45% of overall processes, see SI-1 section S15 and SI-2 Table S2.14).

Discussions of modelling approaches and data

While the results allow for a more detailed assessment of copper tailings' impacts and thus also better representation of averaged impacts, the following key sources of uncertainties and limitations in this study need to be noted:

*Flotation and tailings approximation approach***.** In our analysis, the copper extraction efficiency spans from 75 – 90%, which is high considering today's industry standards[7]. Main parameters that were used in the mineral process simulations were taken from aggregated plant data in technical handbooks and based on approximations from computer simulations and steady-state plant operations. In reality, copper grades in the feed stream might fluctuate and plant variability (e.g., shutdown, market demand, etc.) should be dynamically captured in future research. Dynamic simulation models for 431 beneficiation processes were not possible, as the accurate operating conditions and detailed flowsheets for each facility are generally confidential. In the future, this might become feasible, since mining companies are increasingly encouraged to open their asset's performance through several global standards/frameworks as obligatory key indicators[83]. Additionally, we only modeled the beneficiation process as a single-stage circuit (Figure S1), while advanced grinding and flotation techniques[84] could optimize particle liberation from the ore and thus reduce toxicity of tailings (see SI-1 section S12). It might become economically attractive to do so and should then be investigated in the future assessment.

*Modeling of the infiltration rate***.** In this study, a simplified hydrologic model from the output of PCR-GLOBWB2[58] was used as the core approach. The annual net infiltration data as output of the model provides key inputs for the geochemical modelling, assuming that the values remain constant for the duration of the simulation. Since tailings add an additional soil layer, infiltration rates may be limited due to low hydrological conductivity as a result of small grain size. Additionally, covers might limit short term infiltration. However, previous assumptions on relatively constant infiltration rates are justified, as precipitation and regional changes remain stable in the short term (in a span of 50 years[58,85,86]), but for a projection that involves centuries to thousands of years as time-steps, climate change effects should be considered in future research.

Further research could consider the role of tailings rehabilitation for quantifying leachate emissions based on infiltration rates. This could examine the actual field operation in different regions of the world, like collection and treatment of leachate. To estimate the effects on long-term leaching, scenarios of rehabilitation efforts would need to be set up. While in our analysis we assumed that such activities would not be continued during the leaching period of many thousands of years, other assumptions could lead to diminished leaching. However, in such scenarios the ongoing effects of the rehabilitation efforts would also need to be considered, including additional energy consumption and resources.

*Geochemical modelling in PHREEQC***.** We applied the 1D geochemical reactive-transport model using PHREEQC[63] and took default equilibrium reactions available from PHREEQC and WATEQ4F databases (Table S13). We also added arsenic speciation into these databases, which leaches depending on ferrihydrite concentration[87,88]. More complicated models would require a concerted data-intensive computational effort due to a high level of parameterizations. Additionally, microbial activities might also contribute to changing conditions, but due to the long-term duration of the models, a quasi-equilibrium state is assumed to dominate instead of kinetically-controlled mechanisms[15,89]. Both Cu and Zn in this study have been generally leached out (~60%) after a period of 60,000 years. Besides that, we neglected emissions from other trace metals such as silver, gold, molybdenum, and others in the tailings due to lack of established geochemical reactions in the database.

*Choice of time horizon***.** Tailings or landfill impacts in LCA generally apply an arbitrary time-horizon choice, which is a subjective decision. LCA practitioners should clearly communicate the time-horizon choice in their study. We followed what has been used in the ecoinvent database, differentiating short-term and long-term time horizons. However, one of the advantages of this study is the ability to model emission inventory for any time frame, using the temporally differentiated concentration curves displayed in Figures S6 – S13 together with location specific infiltration rates and the tailings' composition. This also allows for comparing leachate concentrations to toxicological thresholds and, hence, for an assessment of risks.

*LCA and uncertainties***.** Since ore deposits geochemistry varies across sites and within sites, the generated inventories (i.e., emissions) can vary even within a single ore deposit at the same location. Although we did not consider for within-site variation in this paper, we validated some results of our model with the currently operating copper site from our project partner[90] and Chilean field sampling data[91], including sensitivity cases for the modeled sites

(Figure S19 and S20). Deviations of model results against sampled data were within a reasonable range.

The model developed in this study can be used to generate dynamic LCIs, and therefore allows dynamic LCIAs of metal emissions from tailings. This could become relevant if, in the future, groundwater emissions are explicitly modeled, as environmental processes in the soil and groundwater can be slow. In the absence of characterization factors for groundwater, we used here characterization factors for freshwater as surrogates, where the temporal dimension is less of an issue. Once characterization factors for the groundwater become available, a dynamic LCIA could be performed, complementing the dynamic inventory analysis presented in this paper.

*Future primary copper mining***.** We combined primary copper production data from other studies[12,73,92] with forecasted mining projects from the mining database[43]. The studies have different underlying assumptions than the database, and it is possible that technology might change in future. Thus, our study is only applicable for the business-as-usual scenario of mining technology. In the context of resource discovery and availability, above studies assumed generally declining ore quality. However, future technologies may allow production with better efficiency, and hence there is a chance to improve overall copper extraction rate. Additionally, the appearance of low-cost and advanced mine exploration technologies might enable access to currently undiscovered copper deposits, as estimated from several studies[46,77,93]. Moreover, different ore deposit types may have different rates of decline. These factors, however, are beyond the scope of this study.

Application of results

We conclude that this study is representative of active copper sites (75 – 80% of total production). The results for copper tailings display how dramatically site-specific parameters can influence the LCA results of metal production. Our model can be modified and replicated for other metals and is directly usable for metals co-mined with copper such as lead, arsenic, or zinc. Additionally, the assessment for abandoned mine sites remains necessary but was not performed in this study due to a lack of structured data. The GRID-ARENDAL[94] UNEP Program recently developed a portal (Global Tailings Portal) to standardize tailings storage facility risk evaluations. Unfortunately, the portal does not document the data for closed mine sites that might cause long-term environmental burdens.

We also are able to identify regions with high environmental concerns due to tailings deposition. It answers previous calls on the concerted effort to predict impacts and thus, enable prioritization for mitigating impacts of uncontrolled disposal of mine waste[95]. Country and region level results can be used to improve a country's tailings management quality – thereby minimizing the risk of any dam's spillover or breakdowns. Results in Figure 3 also provide broad information for mine operators to continuously improve the recovery efficiency of their flotation plants if there is a huge loss of materials to the tailings.

The generated inventory datasets can be applied for future studies whenever the need arises to compare the LCA studies that involve tailings (i.e., in the background data). Together with an allocation approach, they can also be used to quantify impacts for by-products of copper production. The results presented here contribute to the set of publicly available LCI datasets for mine tailings and can supplement or get integrated into existing databases (i.e., ecoinvent) that currently have limited area-/technology-coverage and are based on simpler modeling techniques. The data can also help to complement the information provided by official pollutant databases like PRTR, which can be applied both at mine-site and regional, specifically when long-term assessments are needed.

Outlook

Our results can be connected to the LCA of copper production value chains and provide additional insight on upstream environmental impacts, and thus contribute to understand the importance of improving resource efficiency in metal supply chains[96]. It can also serve as a screening tool to help decision-makers to prioritize tailings/mine sites remediation. This might include reprocessing for manufacturing other products[97] as a basis for the long-term environmental remediation and valorization[98–100] of mine tailings. The appearances of novel technologies under active development such as solvo-metallurgy[101] or bio-hydrometallurgy[102] are promising options for tailings reprocessing schemes, which might be implemented in tailing remediation models in future research.

Associated Content

(S) Supporting Information

Details and further results. These materials are available free of charge via the internet at: http://pubs.acs.org.

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