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Regionalized life cycle inventories of global sulfidic copper tailings

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6 Abstract

7 Worldwide, an issue of copper production is the generation of mine waste with 8 varying characteristics. This waste can pollute natural environments, and in particular the 9 heavy metal emissions of the tailings may pose long-term consequences. Currently, life cycle 10 assessments of mine tailings are hampered by both limited data availability in the metal 11 production value chain and lack of appropriate methodologies. We collect data from 431 12 active copper mine sites using a combination of information available from the market 13 research and technical handbooks to develop site-specific life cycle inventories for tailings 14 disposal. The approach considers the influences of copper ore composition and local 15 hydrology for dynamically estimating leached metals of tailings at each site. The analysis 16 reveals that together, copper tailings from the large (i.e., porphyry) and medium-size copper 17 deposits (i.e., volcanogenic massive sulfide and sediment-hosted) contribute to more than 18 three-quarters of the total global freshwater ecotoxicity impacts of copper tailings. This 19 strongly correlates with hydrological conditions leading to high infiltration rates. The 20 generated inventories vary locally, even within single countries, showcasing the importance of 21 site-specific models. Our study provides site-specific, dynamic emission models and thus 22 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.

28 Graphical Abstract



30 Introduction

According to the UNEP-IRP Global Resources Outlook 2019, the use of natural 31 resources has tripled in the last four decades¹, and if business as usual is maintained² in the 32 33 production processes, the expected future environmental impacts will be exacerbated. Thus, it 34 is imperative to more sustainably produce materials that support our modern lives. One key 35 metal to respond to these challenges is copper. Notable examples are the use of copper as 36 essential components in renewable energy systems, i.e., solar panels and wind turbines. With 37 various possible use cases and incentives to transition to a low carbon economy, it is 38 estimated that copper demand will grow up to four-fold in less than half of a century³. 39 However, the environmental implications of this transition depend on the technological routes to satisfy future copper demands $^{4-6}$. 40

41 Various types of copper sulfide ore deposits are the primary source of metallic copper, accounting for 80% of copper resource⁷. The production of copper from ore deposits requires 42 43 the separation of unwanted impurities such as silicates, carbonates, and sulfides. This 44 comprises several activities that generate considerable wastes, such as waste rocks from mining and residues/slags from metallurgical processing and refining⁸⁻¹⁰ (Figure S29). In 45 46 between these processes, there is beneficiation: a technology prominently used to extract 47 metals from ores. This requires the usage of chemicals and also produces mineral processing 48 waste. These waste slurries, otherwise called tailings, are then discharged to legally operated 49 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}. 50 51 Declining copper grades in deposits might worsen this situation, as it implies more tailings will have to be managed per kg of copper produced¹². 52

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 drainage⁸, 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}.

60 Several tools and databases are available to assess environmental performances of 61 metal production value chains. In a broader context, the criticality assessment concepts by 62 Graedel^{17,18}, Cimprich et al.¹⁹, and Bach et al.²⁰ translate qualitative criteria into criticality 63 scores by assessing environmental implications, supply risk, vulnerability to supply 64 restriction, and socio-economic dimensions. Official public databases, such as pollutant 65 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 66 authorities^{22–25}. Life cycle assessment (LCA) is a standardized approach to assess the 67 processes' impacts throughout the entire metal value chain^{26,27}. However, with respect to 68 69 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 70 discussed³⁰ and worked around by other researchers in the LCA field by applying a waste-71 specific transfer coefficients model³¹ that was initially developed for landfill emissions³². 72 73 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 74 operating under similar conditions, the major shortcoming is that the inventories are based on 75 76 averaged data in multiple locations to represent specific mining production. Other drawbacks 77 are the neglect of influential site-specific input parameters and, more importantly, the 78 dynamics of leaching in the long term. Therefore, the results of LCA studies that include 79 toxicity impacts from tailings may differ significantly or be completely underreported/ overreported^{5,6,29,36,37}. 80

81 The goal of this study is 1) to provide a global assessment of sulfidic copper tailings 82 using state-of-the-art frameworks in minerals processing, hydrological modelling, and 83 environmental assessment; and 2) to identify the environmental hotspots by a dynamic 84 assessment at mine-site level, which provides a better understanding of the overall impacts of 85 current and future copper production.

86 Materials and Methods

87 Methodology overview

88 Our main methodology built upon previous work in the advancement of mineral 89 processing^{26,38}, subsurface environmental simulation³⁹, and the environmental impacts of 90 mine tailings in the LCA of metal production^{33,34,40}. The methodology integrated several key 91 frameworks that link the workflows for estimating the environmental impacts of mine tailings 92 (Figure 1 top). Because copper has the most comprehensive mineral and production database 93 available⁴¹ and is of high production volume with an increasing trend, we have chosen to 94 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.





102 Copper production data

103 The database compiled in this study combined extensive resources from 1) U.S. 104 Geological Survey Mineral Database⁴², 2) S&P Market Intelligence Metals and Mining *Report*⁴³, 3) *World Mining data 2020*⁴⁴, and 4) a rigorous copper deposit study⁴¹. Together 105 106 they represented more than 75% of annual global sulfidic copper production in 2019, along 107 with ore deposit characteristics, which indicate the mineralogy of each mine deposit for 108 specific production sites. Of total production data, we specifically focus on the copper 109 production process via pyrometallurgical pathway since it represents the dominant technology 110 to produce copper (see SI-1 Section S2). If the data for ore deposits were unavailable in the 111 previously mentioned databases, we linked them with the closest deposit sources based on its 112 geographical coordinates. The workflow to obtain the baseline data for the global assessment 113 is presented in Figure 1 bottom.

114 Beneficiation process modelling and simulation

115 Beneficiation of sulfidic deposits comprises a combination of physical and chemical 116 processes to transform raw copper ore into metal concentrates and tailings as waste. In this 117 study, we developed a systematic method to build a tailings composition database based on 118 processing steps and as a function of ore characteristics. This is critical, as the mineralogy of 119 each deposit defines the necessary separation process of valuable metals from non-valuable 120 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 121 122 tailings for every site. To complete this task, we classified the copper ore deposits based on the formation grouping of Heinrich and Candela⁴⁵, with additional sub-classification based on 123 copper grades, buffering minerals, and other impurities^{41,46}. Their approaches provide 124 125 necessary classification guidelines, and all compiled active copper-sites are presented in Table 126 S1.

131 Our approach simulated the behavior of flotation schemes in the beneficiation process 132 through a 'three-component' minerals floatability process. It parameterizes the minerals 133 flotation as a first-order kinetic equation^{50,51}. This model yields the recovery of a mineral R_m 134 to the flotation time *t* as follows:

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

135 Where

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

137 k_f and k_s represent the flotation rate constant of fast and slow floating particles, 138 respectively;

139 m_n represents the proportion of non-floating particles such that $m_f + m_s + 140$ $m_n = 1$

141 Using equation (1), we approximated the characteristics of the tailings of each mine 142 production site as a function of its ore deposits inputs and flotation process parameters. The 143 flotation parameter details (kinetics data, recovery, minerals, and reagents) for each beneficiation process were primarily obtained from the Handbook of Flotation Reagents⁵² and 144 145 Will's Mineral Processing Technology⁵³. Other sources, such as a collection of flotation studies and patent literatures^{54,55}, were also used, specifically for the flotation of chalcopyrite-146 147 containing ore deposits. We then used the HSC 10's "Geo Module" feature to extract mineral 148 characteristics from the database. Since we aimed to approximate tailings composition based 149 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.

152 Life cycle assessment of copper tailings disposal

153 Our study focuses on the end-of-life phase of waste, in this case, tailings from the 154 production of copper. In accordance with the ecoinvent database⁵⁶, we first relate all 155 emissions and impact to the waste-treatment service "disposal of 1 tonne of tailings from 156 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 157 158 of copper mining (i.e., providing copper to the society). Finally, we also quantified total 159 emissions and impacts of tailings for the entire mining sites, referring to one year of total 160 copper production and the resulting tailings treatment.

161 To model tailing emissions at site-specific locations, we considered tailing 162 characteristics, as a function of mine composition and processing technology (previous 163 sections) and hydrological conditions. All heavy metal emissions in the study were allocated 164 to copper production, representing the worst-case situation without allocation of part of the 165 emissions and impacts to by-products. This represents a conservative approach, as the impacts 166 allocated to copper will be overestimated. As all the inventory data is transparently presented 167 in this paper, future research may apply other allocation techniques, such as by mass or 168 economic value⁵⁷. The remaining copper processing inventories were taken directly from the Life Cycle Inventory (LCI) database ecoinvent 3.6⁵⁶. The illustration of the studied system is 169 170 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-GLOBWB⁵⁸, in mm
 per year.

175

176 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 177 178 freshwater ecotoxicity, for which impacts were quantified by applying global characterization factors (CFs) (defined in USEtox 2.12⁶¹) to leaching emissions. We assumed that all leached 179 180 heavy metals would be transported to freshwater. The main reason for this simplifying 181 assumption was the lack of groundwater CFs in USEtox 2.12. In the impact assessment, no 182 spatial differentiation was considered. Furthermore, no emissions to air via dust were assessed in this study, assuming their contribution is small in the overall system³³ (see SI-1 section 183 184 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 frames⁶².

189 To predict emissions for a long time horizon, we applied geochemical modeling using 190 the PHREEQC simulation⁶³. This model allows the prediction of heavy metals concentration 191 in the tailings, which are controlled by mineralogy and pH development^{64,65}. We assumed that

192 the technical lifetime of the storage basin integrity is limited, so that the technical barriers 193 were neglected for the long-term assessment. All minerals in the tailings were assumed to 194 come in contact with the leaching water (no enclosures) and could eventually seep into 195 groundwater (controlled by solubility). We quantified the dissolved concentrations of Cu, Cd, 196 Pb, Zn, and As in the leachate at equilibrium with a set of solid phases. These substances 197 represent the most toxic and mobile heavy metals present in the leachate of copper tailings⁶⁶. 198 For As, the surface complexation reactions were obtained from the Dzombak model⁶⁷, which 199 assumes that arsenic attaches on hydrous ferric oxide (HFO) surfaces. Parameters and 200 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 201 202 al.⁶⁸, based on the PHREEQC and WATEQ4F databases. This approach allows for consistent 203 geochemical modelling for generation and mobility of leachate from mine waste. Another 204 input parameter was a matrix infiltration rate for each site, which was taken from the global 205 hydrology model PCR-GLOBWB 2⁵⁸. We used the net groundwater recharge as the site-206 specific infiltration parameter in our calculation (see part B of Figure 2). This rate represents 207 infiltration due to climate into the natural soil. We disregarded any alteration (typically 208 reduction) of the infiltration rate by rehabilitation measures, as our assumption is that active rehabilitation will not be continued in the long term⁶⁹. 209

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.

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

219 Where, M_x (mg) is the product of total emissions for metal x in the defined t_n time 220 horizons, PR (1/m².a) represents the net matrix infiltration rate from the global hydrology 221 model, $t_{timestep}$ the time step (a) for every simulation within geochemical modeling, and 222 $C_x(t)$ the concentration of metal x in the leachate (mg/ l) at time t as the results from the 223 geochemical simulation. A (m²) is the surface area related to 1 tonne of tailings material and 224 was calculated from the following equation (3).

$$A = \frac{1 t \ tailings \ material}{\rho_{tailings} \cdot d} \tag{3}$$

225 Where $\rho_{tailings}$ (kg tailings/m³) and *d* (m) are the density and the thickness of the 226 tailings, respectively (see parameters used in Table S4).

227 Baseline scenario: Analyzing the environmental hotspots

228 The life cycle impact assessment (LCIA) results under different time horizons were 229 analyzed and mapped for each site to identify global hotspots. The environmental impacts 230 were quantified per kg of copper production and as total impacts per mine for one year of 231 mine operation. The latter were calculated by multiplying the mass of copper produced for 232 each site in 2019 with the results per tonne of copper. As copper mining activity only 233 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 234 the LCA report of copper supply chain analysis⁷²). The overall procedure is illustrated in 235 Figure S15. 236

Afterwards, we analyzed these spatially- and time-resolved mine tailings inventory 237 data in three ways. First, we analyzed the influences of ore deposits and metallurgical 238 239 processing configurations on the overall tailings' emissions. To further study the interaction 240 between ore deposit and infiltration rates, a sub-analysis was performed for a specific ore 241 deposit type with broad ranges of composition (Table S5-12). Second, the LCIA results of one 242 year of operation of all mines within any country were aggregated. Last, we compared the 243 results of this study with the eleven country specific sulfidic tailings inventory datasets in 244 ecoinvent 3.6^{56} . We matched our modeled inventory in this study following the country 245 classifications that ecoinvent implements (Figure S16). In all these steps, we chose a long-246 term time horizon (i.e., 60,000 years) to conservatively account for heavy metal emissions 247 being leached out from the system and to be consistent with the assumptions taken in 248 ecoinvent. For the evaluation of the effect of various choices concerning time horizons, we 249 also present results for a 100 year time horizon.

250 Modeling future global copper tailings emissions

The primary supply of copper until 2050 provided by Elshkaki et al.⁴ and Northey et 251 al.⁷³ have been used to derive future projections of copper provision. In this study, we 252 253 prepared three scenarios, namely copper supply in year 2030, 2040, and 2050 from the above-254 mentioned data sources. A data reconciliation, however, was necessary as the forecasted data 255 from previous studies do not contain the location of expanded or newly established sites. 256 Therefore, the data gaps for new mine and expansion projects were based on an undiscovered copper resources study⁴⁶, S&P feasibility study⁴³, and other reports^{74,75} (see Figure S17). 257 258 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 Crowson¹², 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.

$$G = 4 \cdot 10^{82} \cdot v^{-25.05} \tag{4}$$

263

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.

Case of new sites (sites under pre-production in the base scenario)
 According to ICMM⁷⁶, 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 operate⁷⁷.

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 USGS⁴² deposit characteristics database. Finally, we combined the merged forecasted data with our method to estimate the environmental impacts of tailings until 2050.

278 **Results and discussion**

279 Global assessment of copper mine tailings

280 The toxicity impacts of all 431 assessed active copper sites in 2019 are shown in 281 Figure 3A. The detailed results are available in the digital SI. These sites capture >75%⁴¹⁻⁴³ of 282 global sulfidic copper tailings disposal. Most copper is mined from porphyry copper deposit

283	type, which accounts for almost half of the total number of sites and is distributed across
284	continents. The toxicity impacts per tonne of tailings due to porphyry copper tailings disposal
285	are generally lower than from other deposit types. However, each mine has its own ore
286	composition that directly influences the beneficiation scheme and respective tailings
287	compositions. Our results (Figure 3 and SI-2 Table S2.3) show that the highest total annual
288	toxicities per mine site are found in:
289	- Large-size, porphyry copper mining sites in the Americas (i.e., Chile, Peru, USA) and
290	Asia (Indonesia, Papua New Guinea)
291	- Medium-size, sediment-hosted and volcanogenic massive sulfide-deposit sites in
292	Canada, Africa (i.e., DR Congo, Zambia) and a few in Europe (Russia, Poland)
293	



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 productionamount weighted average (purple circles) and error bars as weighted standard deviation.

302

303 Region-specific and country-aggregated assessment

304 For background LCI databases, country-level data is required and Figure 3B shows the

305 variabilities that can exist in particular countries and how deposit types and beneficiation

306 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 307 median value is 2.0 \times 10³ CTUe/ kg copper produced. While Chile mainly sources from 308 309 porphyry, with rather low impacts, countries like Australia, China, Peru, and Canada have 310 more varying deposit types and therefore higher impacts. Since various deposit types require 311 different beneficiation processes, the level of heavy metals in the tailings can change. In 312 particular, for volcanogenic massive sulfide and sediment-hosted deposits found in Russia and DR Congo, the beneficiation performs particularly subpar^{52,53}. 313

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.

318 Influences of climate conditions and ore deposit types

319 Higher net positive infiltration generally leads to larger amounts of heavy metals being 320 carried to the soil and groundwater compartment (Figure S22). Ecotoxicity per kg of copper 321 decreases with an increasing copper grade, but correlations are very weak (Figure S24). 322 Volcanogenic massive sulfides and sediment-hosted deposits have relatively higher emissions 323 in the same climatic conditions (i.e., infiltration rates between 40 - 140 mm/ year) due to 324 higher amounts of pyrite but smaller buffering capacities (i.e., calcite and dolomite). 325 However, several high-grade copper sites are situated in regions with low infiltration and thus 326 relatively have low emissions (Figure S25).

327 Comparison of leaching and toxicity results to other studies

The comparison of our toxicity results with country-specific datasets in the ecoinvent 329 3.6⁵⁶ per tonne of tailings is presented in the top part of Figure 4A for short-term (100 years) 330 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).

334 Ecoinvent's representation of the Rest of World (RoW) category may be especially 335 sensitive to regional details, as it aggregates data from several large copper producing 336 countries with varying deposit types and climate conditions (e.g., DR Congo, Poland, and 337 Brazil). Our analysis found a very wide range of ecotoxicity impacts for countries considered 338 in this category (from as low as 0.03 up to 440 CTUe/ tonne of tailings). We therefore suggest 339 that future RoW data includes the variabilities in uncertainties to indicate where large 340 differences in toxicities exist, and detailed assessments should be used to improve the data. In 341 addition to tailings composition, tailings management has a significant impact on toxicity. For 342 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 343 immediately, highlighting the need for alternative disposal methods⁸⁰. 344

345



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.

355

356 In the long-term time horizon, our analysis shows that toxicity results (median) are

- 357 mostly lower than in ecoinvent due to differing tailings property modelling approaches.
- 358 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
- 360 components in the long-term contemplations. Instead, in our study, we applied a set of

361 systematic procedures and models to quantify tailings compositions and leaching at mine-362 level resolution.

363 To evaluate the ecotoxicity impacts of tailings in the life-cycle perspective of copper 364 production, we performed an LCA study at a continent-level of ecoinvent (Figure 4B). In the 365 short-term time horizon, the primary copper production process including smelting, refining, 366 and slag deposition contribute more than 90% of the total ecotoxicity impacts for all 367 continents. The findings are also supported by analyses done at higher granularity (Table 368 S18), where there are generally negligible differences in ecotoxicity values between 369 continents. In the long-term perspective, tailings dominate (>95%) the freshwater ecotoxicity 370 impacts of copper production for all regions. It is therefore of utmost importance to properly 371 assess toxicity impacts of tailings (see SI-1 section S14) and one should avoid ignoring 372 differences between sites when performing comparative LCA studies.

373 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}

383

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.

400 In addition to the freshwater ecotoxicity-related impacts completed in this study, we 401 also conducted environmental impacts for human-toxicity and other LCIA impact categories using the ReCiPe 2016, endpoint, Hierarchist version⁸¹ and the Environmental footprint (EF) 402 403 method⁸², which also provide an aggregated single-score impact result. Other processes (i.e., 404 copper refinery) in the value chain show a higher contribution in the total results, namely due 405 to particulate matter and gaseous emissions from smelters. Results are sensitive to the 406 methods chosen, but metal emissions from tailings are still responsible for ecotoxicity and 407 human toxicity-related to tailings impacts (contributing to around 27–45% of overall 408 processes, see SI-1 section S15 and SI-2 Table S2.14).

409 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:

413 Flotation and tailings approximation approach. In our analysis, the copper extraction 414 efficiency spans from 75 - 90%, which is high considering today's industry standards⁷. Main 415 parameters that were used in the mineral process simulations were taken from aggregated 416 plant data in technical handbooks and based on approximations from computer simulations 417 and steady-state plant operations. In reality, copper grades in the feed stream might fluctuate 418 and plant variability (e.g., shutdown, market demand, etc.) should be dynamically captured in 419 future research. Dynamic simulation models for 431 beneficiation processes were not possible, as the accurate operating conditions and detailed flowsheets for each facility are 420

421 generally confidential. In the future, this might become feasible, since mining companies are 422 increasingly encouraged to open their asset's performance through several global 423 standards/frameworks as obligatory key indicators⁸³. Additionally, we only modeled the 424 beneficiation process as a single-stage circuit (Figure S1), while advanced grinding and 425 flotation techniques⁸⁴ could optimize particle liberation from the ore and thus reduce toxicity 426 of tailings (see SI-1 section S12). It might become economically attractive to do so and should 427 then be investigated in the future assessment.

428 Modeling of the infiltration rate. In this study, a simplified hydrologic model from the output of PCR-GLOBWB2⁵⁸ was used as the core approach. The annual net infiltration data 429 430 as output of the model provides key inputs for the geochemical modelling, assuming that the 431 values remain constant for the duration of the simulation. Since tailings add an additional soil 432 layer, infiltration rates may be limited due to low hydrological conductivity as a result of 433 small grain size. Additionally, covers might limit short term infiltration. However, previous 434 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 435 436 involves centuries to thousands of years as time-steps, climate change effects should be 437 considered in future research.

438 Further research could consider the role of tailings rehabilitation for quantifying 439 leachate emissions based on infiltration rates. This could examine the actual field operation in 440 different regions of the world, like collection and treatment of leachate. To estimate the 441 effects on long-term leaching, scenarios of rehabilitation efforts would need to be set up. 442 While in our analysis we assumed that such activities would not be continued during the 443 leaching period of many thousands of years, other assumptions could lead to diminished 444 leaching. However, in such scenarios the ongoing effects of the rehabilitation efforts would 445 also need to be considered, including additional energy consumption and resources.

446 Geochemical modelling in PHREEQC. We applied the 1D geochemical reactivetransport model using PHREEQC⁶³ and took default equilibrium reactions available from 447 448 PHREEQC and WATEQ4F databases (Table S13). We also added arsenic speciation into these databases, which leaches depending on ferrihydrite concentration^{87,88}. More complicated 449 450 models would require a concerted data-intensive computational effort due to a high level of 451 parameterizations. Additionally, microbial activities might also contribute to changing 452 conditions, but due to the long-term duration of the models, a quasi-equilibrium state is 453 assumed to dominate instead of kinetically-controlled mechanisms^{15,89}. Both Cu and Zn in this 454 study have been generally leached out (~60%) after a period of 60,000 years. Besides that, we 455 neglected emissions from other trace metals such as silver, gold, molybdenum, and others in 456 the tailings due to lack of established geochemical reactions in the database.

457 *Choice of time horizon.* Tailings or landfill impacts in LCA generally apply an 458 arbitrary time-horizon choice, which is a subjective decision. LCA practitioners should clearly 459 communicate the time-horizon choice in their study. We followed what has been used in the 460 ecoinvent database, differentiating short-term and long-term time horizons. However, one of 461 the advantages of this study is the ability to model emission inventory for any time frame, 462 using the temporally differentiated concentration curves displayed in Figures S6 - S13463 together with location specific infiltration rates and the tailings' composition. This also allows 464 for comparing leachate concentrations to toxicological thresholds and, hence, for an 465 assessment of risks.

466 *LCA and uncertainties.* Since ore deposits geochemistry varies across sites and within 467 sites, the generated inventories (i.e., emissions) can vary even within a single ore deposit at 468 the same location. Although we did not consider for within-site variation in this paper, we 469 validated some results of our model with the currently operating copper site from our project 470 partner⁹⁰ and Chilean field sampling data⁹¹, including sensitivity cases for the modeled sites

471 (Figure S19 and S20). Deviations of model results against sampled data were within a472 reasonable range.

473 The model developed in this study can be used to generate dynamic LCIs, and 474 therefore allows dynamic LCIAs of metal emissions from tailings. This could become 475 relevant if, in the future, groundwater emissions are explicitly modeled, as environmental 476 processes in the soil and groundwater can be slow. In the absence of characterization factors 477 for groundwater, we used here characterization factors for freshwater as surrogates, where the 478 temporal dimension is less of an issue. Once characterization factors for the groundwater 479 become available, a dynamic LCIA could be performed, complementing the dynamic 480 inventory analysis presented in this paper.

481 *Future primary copper mining*. We combined primary copper production data from other studies^{12,73,92} with forecasted mining projects from the mining database⁴³. The studies 482 483 have different underlying assumptions than the database, and it is possible that technology 484 might change in future. Thus, our study is only applicable for the business-as-usual scenario 485 of mining technology. In the context of resource discovery and availability, above studies 486 assumed generally declining ore quality. However, future technologies may allow production 487 with better efficiency, and hence there is a chance to improve overall copper extraction rate. 488 Additionally, the appearance of low-cost and advanced mine exploration technologies might 489 enable access to currently undiscovered copper deposits, as estimated from several 490 studies^{46,77,93}. Moreover, different ore deposit types may have different rates of decline. These 491 factors, however, are beyond the scope of this study.

492 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-ARENDAL⁹⁴ 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 waste⁹⁵. 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.

509 The generated inventory datasets can be applied for future studies whenever the need 510 arises to compare the LCA studies that involve tailings (i.e., in the background data). Together 511 with an allocation approach, they can also be used to quantify impacts for by-products of 512 copper production. The results presented here contribute to the set of publicly available LCI 513 datasets for mine tailings and can supplement or get integrated into existing databases (i.e., 514 ecoinvent) that currently have limited area-/technology-coverage and are based on simpler 515 modeling techniques. The data can also help to complement the information provided by 516 official pollutant databases like PRTR, which can be applied both at mine-site and regional, 517 specifically when long-term assessments are needed.

518 **Outlook**

519 Our results can be connected to the LCA of copper production value chains and 520 provide additional insight on upstream environmental impacts, and thus contribute to

521	understand the importance of improving resource efficiency in metal supply chains ⁹⁶ . It can
522	also serve as a screening tool to help decision-makers to prioritize tailings/mine sites
523	remediation. This might include reprocessing for manufacturing other products ⁹⁷ as a basis for
524	the long-term environmental remediation and valorization98-100 of mine tailings. The
525	appearances of novel technologies under active development such as solvo-metallurgy ¹⁰¹ or
526	bio-hydrometallurgy ¹⁰² are promising options for tailings reprocessing schemes, which might
527	be implemented in tailing remediation models in future research.

528 Associated Content

529 (S) Supporting Information

530 Details and further results. These materials are available free of charge via the internet at:531 http://pubs.acs.org.

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