Environmental Perspectives on Emerging Resource Recovery Systems of Mine Tailings: A Life Cycle Consideration

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“Keberhasilan bukanlah milik orang yang pintar.

Keberhasilan adalah kepunyaan mereka yang senantiasa berusaha.”

– B.J. Habibie

Success is not possessed by educated people.

It belongs to those who try everlastingly.

Acknowledgment

I would like to take this opportunity to express my heartfelt thanks to everyone who has made this PhD journey and scientific endeavors possible! Feel free to skip this section (as it tends to be somewhat lengthy and personal); I promise you I will not feel offended. Nevertheless, I firmly believe this thesis deserves a proper space for acknowledging key individuals in both professional and personal spheres.

A significant milestone was once started somewhere at the opportune time. It is dream-like in retrospect to connect past dots: how I hopped on a TGV train from Paris *Gare du Lyon* and presented my research work, as part of the PhD selection process, in front of ESD members in *Hönggerberg* once upon a sunny day in 2018. Fast-forward four years, here I am, drawing stories out of vivid memories and articulating them in written forms.

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Of course, I can reminisce infinitely about people who inspire and lend their helping hands in this journey. Doing so will take up more space in this section, and I am afraid we will deviate excessively from this thesis' original objective. What I can surely tell is that every single kind person has dramatic impacts on my personal and professional endeavors. You are all remembered. Surely you, readers, have made this report accessible by making it land in your hands and / or on your digital screens. With this, I truly hope this thesis lives up to its promise to satisfy your curious minds.

Thank you to all kind individuals! Now let's switch gears to research: Are you ready?

Abstract

The extraction of minerals and metals is a prerequisite for the production and utilization of technology in various sectors such as infrastructure, energy, transport, and many other industries. Compounded by the need to supply mineral and metal resources for a sustainable energy transition and for global urbanization, mineral and metal mining and mining waste volumes, especially tailings, are expected to grow globally. In addition to long-term emissions and their adverse environmental effects, poor tailings management might lead to the collapse of waste storage, causing accidental environmental disasters. This pushes the mining industry to commit to developing alternative solutions for tailings management. Reprocessing and valorizing tailings using innovative metallurgical techniques can help reduce environmental burdens and reduce demand for virgin resources. To make sure that new tailings valorization technologies are sustainable, it is important to develop a sound scientific assessment for quantifying the environmental implications of mine tailings management using a life cycle perspective. A better understanding of the environmental impacts of tailings management can help identify the long-term consequences of current disposal options and clarify the benefits of improved tailings management practices. Despite recent policy encouragement for minimizing harm and exploiting new resources from mine waste, these implementations are still unclear. In this context, the guiding research question of this thesis is: *How can quantitative environmental methods support decision-making in resource recovery systems of mine tailings?*

In order to enable informed environmental decisions, this thesis provides information on: (i) the short- and long-term emissions and resulting environmental impacts of tailings disposal under different technological and geographical conditions, (ii) which emerging reprocessing technologies are suitable for mitigating the environmental impacts through tailings valorization, along with (iii) the impacts of a modeled full-scale implementation of these technologies and ultimately, (iv) the assessment of future environmental impacts of widespread mine tailings reprocessing and valorization in Europe, when considering future scenarios.

A mix of scientific approaches in the field of geochemistry, metallurgical process modeling, and environmental assessment methods are applied to provide a site-specific tailings model. The spatial coverage in the first study is global, which can then be zoomed in to analyze facility-level environmental impacts. Subsequently, various technology upscaling frameworks and engineering-based upscaling approaches are utilized to estimate the environmental performance of numerous new (currently lab-scale) value recovery technologies from tailings. This step allows performing prospective life cycle assessment (LCA), with the ability to compare such valorization alternatives with conventional tailings depositions. The parametric LCA models account for technology

inputs from process designers, considering interoperability between any new processes and allowing optimization of reprocessing and valorization routes. The models are then combined with a scenario analysis for the EU copper tailings management, considering changing future energy systems and metal/material flow dynamics.

This thesis is composed of three individual articles, in addition to an overarching introduction and conclusion. Article 1 investigates the site-specific life cycle inventories of copper tailings, capturing 80% of the world's copper production. This work demonstrates the importance of mechanistic modeling and spatial resolution for modeling tailings emissions. It identifies environmental hotspots of tailings deposition for prioritizing mitigation agendas across the globe. Article 2 considers innovative repurposing technologies for tailings, resulting in multiple reprocessing routes with several secondary products as added-value resources. The detailed characterization of environmental impacts induced by such effort and environmental benefits associated with secondary resources are critically assessed. The results of this study, i.e., parameterized and upscaled LCA models, can be leveraged to detect techno-environmental performance bottlenecks and indicate improvement potentials in the value chain. In Article 3, particular focus is given to the considerations of the future-oriented environmental assessment of copper tailings management in the EU. Combining datasets and approaches of Articles 1 and 2, Article 3 presents scenario-based LCA to estimate and compare the environmental impacts of different tailings treatment scenarios under various future perspectives, such as metal demand and energy transition. This article also aims to quantify the environmental benefits and impacts of alternative tailings management options. Environmental benefits related to climate change and ecotoxicity are primarily achieved through (i) offsetting energy-intensive construction materials, (ii) reducing tailings discharge volumes in the waste storage, and (iii) substituting primary products and hence saving tailings reprocessing and valorization impacts.

This thesis makes the following three contributions to the scientific literature. First, it develops input-dependent and site-specific models for quantifying emission releases from tailings, which vary across geological settings, climates, and ore processing technologies. Second, by integrating prospective approaches and fit-for-purpose treatment pathways, this thesis provides process-based LCA that demonstrates the holistic technological configurations for tailings repurposing from an environmental life-cycle perspective. Third, the scenario analysis for mine tailings disposal strategies enhances the existing understanding of tailings management's role in the LCA of copper by considering technological development and material systems. The environmental improvement potential of alternative tailings management to achieve 2050 climate targets is investigated.

Finally, the applied research presents three valuable insights for mining practitioners and policy decision-makers toward sustainable mine waste management. First, the global environmental ecotoxicity hotspots induced by copper tailings landfilling are characterized by regions with highly sulfidic ore types and high infiltration rates. Second, the emergence of resource recovery technologies to solve mine waste management challenges emphasizes two key elements: (i) the co-production of building materials such as cement and ceramics for maximum environmental benefits and (ii) on a process level, the continuous development of innovative technologies can further benefit from such prospective LCA due to transparent and modular nature of technology modeling. Third, large-scale reprocessing and valorization of tailings offer the potential to generate useful products from tailings and to reduce future environmental impacts. Tradeoffs exist between climate change and ecotoxicity impacts for different alternative tailings management scenarios. In addition to cradle-to-gate assessment, the environmental impacts associated with the use of tailings-derived products must also be carefully considered in future research. Supporting regulatory policies and incentives are needed to promote the use of secondary materials from mine tailings. The outcomes of this thesis can provide guidance on environmentally sensitive mining operations and future opportunities of tailings processing technologies.

Zusammenfassung

Die Gewinnung von Mineralien und Metallen ist eine Voraussetzung für die Herstellung und Nutzung von Technologien in verschiedenen Sektoren wie Infrastruktur, Energie, Verkehr und vielen anderen Branchen. Angesichts der Notwendigkeit, Mineral- und Metallressourcen für eine nachhaltige Energiewende und die globale Urbanisierung bereitzustellen, wird erwartet, dass der Abbau von Mineralien und Metallen und die Menge der Bergbauabfälle, insbesondere der Tailings, weltweit zunehmen werden. Zusätzlich zu den langfristigen Emissionen und ihren negativen Auswirkungen auf die Umwelt kann ein schlechtes Tailings-Management zum Zusammenbruch der Abfalllager führen, was unfallbedingte Umweltkatastrophen zur Folge haben kann. Dies zwingt die Bergbauindustrie zur Entwicklung alternativer Lösungen für das Tailings-Management. Die Wiederaufbereitung und Valorisierung von Tailings mit Hilfe innovativer metallurgischer Techniken kann dazu beitragen, die Umweltbelastung zu verringern und den Bedarf an neuen Ressourcen zu reduzieren. Um sicherzustellen, dass neue Technologien zur Valorisierung von Tailings nachhaltig sind, ist es wichtig, eine fundierte wissenschaftliche Bewertung zur Quantifizierung der Umweltauswirkungen des Tailings-Managements unter Berücksichtigung des Lebenszyklus vorzunehmen. Ein besseres Verständnis der Umweltauswirkungen des Tailings-Managements kann dazu beitragen, die langfristigen Folgen der derzeitigen Entsorgungsoptionen zu ermitteln und die Vorteile verbesserter Verfahren des Tailings-Managements zu verdeutlichen. Obwohl die Politik in jüngster Zeit die Minimierung von Schäden und die Nutzung neuer Ressourcen aus Bergbauabfällen fördert, ist die Umsetzung noch unklar. In diesem Zusammenhang lautet die leitende Forschungsfrage dieser Dissertation: *Wie können quantitative Umweltmethoden die Entscheidungsfindung in Systemen zur Verwertung von Tailings aus Bergwerken unterstützen?*

Um fundierte Umweltentscheidungen zu ermöglichen, liefert diese Dissertation Informationen über: (i) die kurz- und langfristigen Emissionen und die sich daraus ergebenden Umweltauswirkungen der Tailings-Entsorgung unter verschiedenen technologischen und geografischen Bedingungen, (ii) welche neu entstehenden Wiederaufbereitungstechnologien geeignet sind, die Umweltauswirkungen durch die Tailings-Valorisierung zu mindern, sowie (iii) die Auswirkungen einer modellierten grosstechnischen Umsetzung dieser Technologien und schliesslich (iv) die Bewertung der künftigen Umweltauswirkungen einer weit verbreiteten Wiederaufbereitung und Valorisierung von Bergwerks-Tailings in Europa unter Berücksichtigung künftiger Szenarien.

Zur Erstellung eines standortspezifischen Tailings-Modells wird eine Mischung aus wissenschaftlichen Ansätzen aus den Bereichen Geochemie, metallurgische Prozessmodellierung und Umweltbewertungsmethoden angewandt. Der räumliche

Erfassungsbereich in der ersten Studie ist global und kann dann vergrössert werden, um die Umweltauswirkungen auf Anlagenebene zu analysieren. Anschliessend werden verschiedene Rahmenwerke für die Hochskalierung von Technologien und ingenieurbasierte Hochskalierungsansätze verwendet, um die Umweltleistung zahlreicher neuer (derzeit im Labormassstab) Technologien zur Wertstoffrückgewinnung aus Tailings abzuschätzen. Dieser Schritt ermöglicht die Durchführung einer prospektiven Ökobilanz mit der Möglichkeit, solche Aufwertungsalternativen mit konventionellen Tailings-Ablagerungen zu vergleichen. Die parametrischen LCA-Modelle berücksichtigen die technologischen Eingaben der Prozessentwickler, berücksichtigen die Interoperabilität zwischen allen neuen Prozessen und ermöglichen die Optimierung der Wiederaufbereitungs- und Aufwertungsrouten. Die Modelle werden dann mit einer Szenarioanalyse für das Tailings-Management in der EU kombiniert, die sich verändernde zukünftige Energiesysteme und Metall- / Materialflussdynamiken berücksichtigt.

Diese Dissertation besteht aus drei einzelnen Artikeln sowie einer übergreifenden Einleitung und Schlussfolgerung. Artikel 1 untersucht die standortspezifischen Lebenszyklusinventare von Kupfer-Tailings, die 80 % der weltweiten Kupferproduktion abdecken. Diese Dissertation zeigt, wie wichtig die mechanistische Modellierung und die räumliche Auflösung für die Modellierung von Tailings-Emissionen sind. Sie identifiziert Umwelt-Hotspots für die Ablagerung von Tailings, um die Prioritäten für die Abhilfemassnahmen auf der ganzen Welt festzulegen. Artikel 2 befasst sich mit innovativen Technologien zur Wiederverwendung von Tailings, die zu mehreren Wiederaufbereitungswegen mit verschiedenen Sekundärprodukten als Mehrwertressourcen führen. Eine detaillierte Charakterisierung der Umweltauswirkungen, die durch solche Anstrengungen verursacht werden, und der mit den Sekundärressourcen verbundenen Umweltvorteile wird kritisch bewertet. Die Ergebnisse dieser Studie, d.h. parametrisierte und hochskalierte LCA-Modelle, können genutzt werden, um Engpässe in der technisch-ökologischen Leistung zu erkennen und Verbesserungspotenziale in der Wertschöpfungskette aufzuzeigen. In Artikel 3 werden insbesondere Überlegungen zu einer zukunftsorientierten Umweltbewertung für das Tailings-Management von Kupfer in der EU angestellt. Durch die Kombination von Datensätzen und Ansätzen aus den Artikeln 1 und 2 werden in Artikel 3 szenariobasierte LCA vorgestellt, um die Umweltauswirkungen verschiedener Tailings-Behandlungsszenarien unter verschiedenen Zukunftsperspektiven, wie Metallnachfrage und Energiewende, abzuschätzen und zu vergleichen. Dieser Artikel zielt auch darauf ab, die Umweltvorteile und -auswirkungen des alternativen Tailings-Managements zu quantifizieren. Der Umweltnutzen in Bezug auf Klimawandel und Ökotoxizität wird in erster Linie durch (i) den Ausgleich energieintensiver Baumaterialien, (ii) die Verringerung des Tailings-Ableitungsvolumens in der Abfalldeponie und (iii) die Substitution von Primärprodukten und damit die Einsparung von Auswirkungen der Tailings-Aufbereitung und -Valorisierung erzielt.

Die Dissertation leistet die folgenden drei Beiträge zur wissenschaftlichen Literatur. Erstens werden inputabhängige und standortspezifische Modelle zur Quantifizierung der Emissionsfreisetzungen aus Tailings entwickelt, die je nach Geologie, Klima und Erzverarbeitungstechnologie variieren. Zweitens bietet die Dissertation durch die Integration prospektiver Ansätze und zweckmässiger Behandlungspfade eine prozessbasierte Ökobilanz, die die ganzheitlichen technologischen Konfigurationen für die Wiederverwendung von Tailings aus einer ökologischen Lebenszyklusperspektive aufzeigt. Drittens erweitert die Szenarioanalyse für Entsorgungsstrategien für Abraumhalden das bestehende Verständnis für die Rolle des Tailings-Managements in der Ökobilanz von Kupfer durch die Berücksichtigung von technologischen Entwicklungen und Materialsystemen. Das Umweltverbesserungspotenzial eines alternativen Tailings-Managements zur Erreichung der Klimaziele für 2050 wird untersucht.

Schliesslich liefert die angewandte Forschung drei wertvolle Erkenntnisse für Bergbaupraktiker und politische Entscheidungsträger im Hinblick auf ein nachhaltiges Abfallmanagement in Bergwerken. Erstens sind die globalen Ökotoxizitäts-Hotspots, die durch die Deponierung von Kupfer-Tailings entstehen, durch Regionen mit stark sulfidischen Erzen und hohen Infiltrationsraten gekennzeichnet. Zweitens werden bei der Entwicklung von Technologien zur Rückgewinnung von Ressourcen zur Lösung der Probleme bei der Bewirtschaftung von Bergbauabfällen zwei Schlüsselelemente hervorgehoben: (i) die Koproduktion von Baumaterialien wie Zement und Keramik zur Erzielung eines maximalen Umweltnutzens und (ii) auf Prozessebene kann die kontinuierliche Entwicklung innovativer Technologien aufgrund der transparenten und modularen Natur der Technologiemodellierung von einer solchen vorausschauenden Ökobilanz weiter profitieren. Drittens bieten die Wiederaufbereitung und Verwertung von Tailings in grossem Massstab das Potenzial, aus Tailings nützliche Produkte zu erzeugen und künftige Umweltauswirkungen zu verringern. Für verschiedene alternative Szenarien des Tailings-Managements bestehen Kompromisse zwischen den Auswirkungen des Klimawandels und der Ökotoxizität. Neben der Bewertung des gesamten Prozesses müssen auch die Umweltauswirkungen, die mit der Verwendung von aus Tailings gewonnenen Produkten verbunden sind, in der künftigen Forschung sorgfältig berücksichtigt werden. Um die Verwendung von Sekundärstoffen aus Tailings zu fördern, sind unterstützende Regulierungsmassnahmen und Anreize erforderlich. Die Ergebnisse dieser Dissertation können eine Orientierungshilfe für umweltbewusste Bergbaubetriebe und zukünftige Möglichkeiten von Technologien zur Verarbeitung von Tailings sein.

Résumé

L'extraction de minéraux et de métaux est une condition préalable à la production et à l'utilisation de technologies dans divers secteurs tels que les infrastructures, l'énergie, les transports et de nombreuses autres industries. Compte tenu de la nécessité de fournir des ressources minérales et métalliques pour une transition énergétique durable et l'urbanisation mondiale, on s'attend à ce que les quantités de minéraux et de métaux extraits et de déchets miniers, en particulier les tailings, augmentent dans le monde entier. Outre les émissions à long terme et leurs effets néfastes sur l'environnement, une mauvaise gestion des tailings pourrait entraîner l'effondrement du stockage des déchets, provoquant des catastrophes environnementales accidentelles. Ce constat incite l'industrie minière à s'engager dans le développement de solutions alternatives pour la gestion des tailings. Le retraitement et la valorisation des tailings à l'aide de techniques métallurgiques innovantes peuvent contribuer à réduire les charges environnementales et à diminuer la demande de ressources vierges. Pour s'assurer que les nouvelles technologies de valorisation des tailings sont durables, il est important de développer une évaluation scientifique solide pour quantifier les implications environnementales de la gestion des tailings miniers en utilisant une perspective de cycle de vie. Une meilleure compréhension des impacts environnementaux de la gestion des tailings peut aider à identifier les conséquences à long terme des options actuelles d'élimination et à clarifier les avantages de meilleures pratiques de gestion des tailings. Malgré les récents encouragements politiques pour minimiser les dommages et exploiter de nouvelles ressources à partir des déchets miniers, ces mises en œuvre ne sont toujours pas claires. Dans ce contexte, la question de recherche directrice de cette thèse est la suivante : *Comment les méthodes environnementales quantitatives peuvent-elles soutenir la prise de décision dans les systèmes de récupération des ressources des tailings miniers ?*

Afin de permettre des décisions environnementales éclairées, cette thèse fournit des informations sur : (i) les émissions à court et à long terme et les impacts environnementaux résultants de l'élimination des résidus miniers dans différentes conditions technologiques et géographiques, (ii) les technologies de retraitement émergentes qui conviennent pour atténuer les impacts environnementaux par la valorisation des résidus miniers, ainsi que (iii) les impacts d'une mise en œuvre modélisée à grande échelle de ces technologies et, finalement, (iv) l'évaluation des impacts environnementaux futurs du retraitement et de la valorisation généralisés des résidus miniers en Europe, en considérant des scénarios futurs.

Une combinaison d'approches scientifiques en géochimie, en modélisation de processus métallurgiques et en méthodes d'évaluation environnementale est appliquée pour fournir un modèle de tailings spécifique au site. La couverture spatiale de la première étude est globale, et peut ensuite être agrandie pour analyser les impacts environnementaux au

niveau des installations. Par la suite, divers cadres de mise à l'échelle des technologies et des approches de mise à l'échelle basées sur l'ingénierie sont utilisés pour estimer la performance environnementale de nombreuses nouvelles technologies de valorisation des tailings (actuellement à l'échelle du laboratoire). Cette étape permet de réaliser une analyse du cycle de vie (ACV) prospective, avec la possibilité de comparer ces alternatives de valorisation avec les dépôts de tailings conventionnels. Les modèles paramétriques d'ACV tiennent compte des apports technologiques des concepteurs de procédés, en considérant l'interopérabilité entre tous les nouveaux procédés et en permettant l'optimisation des voies de retraitement et de valorisation. Les modèles sont ensuite combinés avec une analyse de scénario pour la gestion des tailings de cuivre de l'UE, en tenant compte de l'évolution des systèmes énergétiques futurs et de la dynamique des flux de métaux / matériaux.

Cette thèse regroupe trois articles distincts, chacun répondant à des objectifs de recherche spécifiques. L'article 1 étudie les inventaires du cycle de vie spécifiques au site des tailings de cuivre, qui représentent 80% de la production mondiale de cuivre. Ce travail démontre l'importance de la modélisation mécaniste et de la résolution spatiale pour la modélisation des émissions de tailings. Il identifie les points chauds environnementaux des dépôts de tailings afin d'établir des priorités dans les programmes d'atténuation à travers le monde. L'article 2 examine les technologies innovantes de réutilisation des tailings, qui donnent lieu à de multiples voies de retraitement avec plusieurs produits secondaires comme ressources à valeur ajoutée. La caractérisation détaillée des impacts environnementaux induits par ces efforts et les bénéfices environnementaux associés aux ressources secondaires sont évalués de manière critique. Les résultats de cette étude, c'est-à-dire les modèles d'ACV paramétrés et mis à l'échelle, peuvent être exploités pour détecter les goulets d'étranglement des performances techno-environnementales et indiquer les potentiels d'amélioration dans la chaîne de valeur. L'article 3 met l'accent sur les considérations relatives à l'évaluation environnementale prospective de la gestion des tailings de cuivre dans l'UE. En combinant les ensembles de données et les approches des articles 1 et 2, l'article 3 présente une ACV basée sur des scénarios pour estimer et comparer les impacts environnementaux de différents scénarios de traitement des tailings dans diverses perspectives d'avenir, telles que la demande en métaux et la transition énergétique. Cet article vise également à quantifier les avantages et les impacts environnementaux de la gestion alternative des tailings. Les avantages environnementaux liés au changement climatique et à l'écotoxicité sont principalement obtenus par (i) la compensation des matériaux de construction à forte intensité énergétique, (ii) la réduction du volume des rejets de tailings dans le stockage des déchets, et (iii) la substitution de produits primaires et donc l'économie des impacts du retraitement et de la valorisation des tailings.

Cette thèse apporte les trois contributions suivantes à la littérature scientifique. Premièrement, elle développe des modèles dépendants des intrants et spécifiques aux sites pour quantifier les émissions des tailings, qui varient selon les géologies, les climats et les technologies de traitement du minerai. Deuxièmement, en intégrant des approches prospectives et des voies de traitement adaptées, la thèse fournit une ACV basée sur les processus qui démontre les configurations technologiques holistiques pour la réutilisation des tailings dans une perspective de cycle de vie environnemental. Troisièmement, l'analyse de scénario pour les stratégies d'élimination des résidus miniers améliore la compréhension existante du rôle de la gestion des résidus dans l'ACV du cuivre en considérant le développement technologique et les systèmes de matériaux. Le potentiel d'amélioration environnementale de la gestion alternative des tailings pour atteindre les objectifs climatiques de 2050 est explorée.

En conclusion, la recherche appliquée présente trois aspects fondamentaux pour les praticiens de l'industrie minière et les décideurs politiques en matière de gestion durable des déchets miniers. Premièrement, les points chauds de l'écotoxicité environnementale mondiale induite par la mise en décharge des tailings de cuivre sont caractérisés par des régions présentant des types de minerais hautement sulfurés et des taux d'infiltration élevés. Deuxièmement, l'émergence des technologies de récupération des ressources pour résoudre les problèmes de gestion des déchets miniers met l'accent sur deux éléments clés : (i) la coproduction de matériaux de construction tels que le ciment et la céramique pour un maximum d'avantages environnementaux et (ii) au niveau du processus, le développement continu de technologies innovantes peut bénéficier davantage de cette ACV prospective en raison de la nature transparente et modulaire de la modélisation technologique. Troisièmement, le retraitement et la valorisation à grande échelle des tailings offrent la possibilité de générer des produits utiles à partir des tailings et de réduire les impacts environnementaux futurs. Il existe des compromis entre les impacts du changement climatique et de l'écotoxicité pour différents scénarios de gestion des tailings. En plus de l'évaluation du berceau à la porte, les impacts environnementaux associés à l'utilisation des produits dérivés des tailings doivent également être soigneusement pris en compte dans les recherches futures. Des politiques réglementaires et des mesures incitatives sont nécessaires pour promouvoir l'utilisation de matériaux secondaires issus des tailings miniers. Les résultats de cette thèse peuvent fournir des conseils sur les opérations minières sensibles à l'environnement et les opportunités futures des technologies de traitement des tailings.

Table of contents

Glossary

Terms	Description
AMD	Acid mine drainage is the formation of highly acidic water rich in heavy metals. This acidic water forms through the chemical reaction of surface water and shallow subsurface water with rocks that contain sulfur-bearing minerals, resulting in sulfuric acid
Circular economy	An economic system in which products and materials are designed in such a way that they can be reused, reprocessed, recycled, or recovered and thus maintained in the economy for as long as possible, along with the resources of which they are made, and the generation of waste is avoided or minimized
CSA	Calcium sulfoaluminate cement, one of the low-carbon alternatives to ordinary Portland cement (OPC); Unlike OPC, the use limestone materials in CSA are partly substituted by incorporation of industrial waste and secondary materials in the formulation
Geopolymer / inorganic polymer	Inorganic aluminosilicate polymer that forms solid ceramic-like materials at near ambient temperatures. It is a subset of alkali-activated materials and produced through the reaction of aluminosilicate materials with an alkaline activator, e.g., alkali hydroxide, silicate, carbonate, or sulfate
GHG	Greenhouse gas, any gas that absorbs net heat energy emitted from Earth's surface and reradiating it back to Earth's surface, hence causing greenhouse effects and thus contributing to climate change
IAM	Integrated assessment modeling aims to provide policy-relevant insights into environmental change and sustainable development issues by considering broad socio-economic and technological developments and their consequences over time
Industrial ecology / IE	The study of systemic relationships between society, the economy, and the natural environment, which focuses on the use of technology to reduce environmental impacts; Examples of methods in IE include LCA, MFA, and others
LCA / Ökobilanz (German) / ACV (French)	Life cycle assessment, a standardized tool for evaluating the environmental performance of products and processes comprising four main phases: goal definition and scope, inventory analysis, impact assessment, and interpretation

LCI	Life cycle inventory, the methodology step that involves creating an inventory of input and output flows, that is, energy, resources, and emissions, for a product / service system
FU	Functional unit, the function of a studied system that serves as the reference basis for calculations in LCA
LCIA	Life cycle impact assessment, refers to the methodology steps for evaluating the potential environmental impacts by converting the LCI results into specific impact indicators
MFA	Material flow analysis, an analytical method to quantify flows and stocks of materials in a defined system
Prospective / ex ante LCA	LCA concepts that promote the integration of environmental criteria in early stages of technology development toward responsible research and innovation
SSP	Shared socio-economic pathways are scenarios of projected socio-economic global changes up to 2100. They are used to derive forecasts of environmental impacts (e.g., greenhouse gas emissions) with different climate policies
Tailings	Unwanted materials / waste streams left over after separation of the valuable fraction from the uneconomic fraction (gangue) of ores

Chapter 1

Introduction

This chapter introduces the context and the research questions motivating the work. An overview of the thesis is presented, which also serves as the background for the work elaborated in **Chapters 2 – 4**.

Chapter 1: Introduction

1.1 Background

Metal and mineral mining is a critical pillar of economic growth for energy, technology, and infrastructure. Globally, the industry provides staple materials to sustain modern societies across sectors for centuries (Rankin, 2012). Rapid globalization worldwide in parallel stimulates the increased use of minerals and metals. According to the *Global Resources Outlook* report by OECD (2019), the world's material consumption is expected to double from 2017 levels. Recognizing the industry's vast size is one difficulty; achieving expansion for a good quality of human life while delivering vital resources without destroying local ecosystems is another. The latter objective is also popularly known as resource decoupling (IRP, 2020). On top of inevitable globalization demand, issues revolving around low-carbon transition might add another layer of complexity to material supply. It is crucial to acknowledge that such a transition is material intensive. Many studies suggest that the future green revolution will strongly drive the metal demand in the decades ahead (Elshkaki et al., 2018; Giurco et al., 2019; Gloaguen et al., 2022; Herrington, 2021; Lee et al., 2020). Mining demand for technology commodities such as lithium, graphite, and cobalt might increase by more than 20 times (IEA, 2021). With respect to all these motives and concerns, mining activities will remain a vital part of the economy in the near future.

A sharp concern in the mineral industry is that the exploitation and use of mineral resources generate considerable quantities of waste. Among the largest waste streams associated with mining are tailings (Mudd, 2007; C. Wang et al., 2014). According to the latest estimates, approximately 8 billion tons of tailings are deposited annually (Oberle et al., 2020), commonly stored in around 3400 active storage facilities worldwide (Franks et al., 2021). Future trends in mining, such as rising metal demand and declining ore grades, may compound the challenges of tailings management, as the rate of waste produced can be dramatically amplified (Calvo et al., 2016; Franks et al., 2011; Norgate et al., 2007). During the stockpiling period, tailings deposition may leave long-term environmental legacies in the form of acid mine drainage potential (Lottermoser, 2010). This is characterized by low pH and elevated concentrations of heavy metals and metalloids (Kossoff et al., 2014), which could seep into the surrounding areas and pollute natural environments such as groundwater, especially if there is a lack of control or complete control loss over long time horizons. In the worst-case scenario, insufficient and poor management of tailings facilities might result in dam failures, causing devastating social, economic, and environmental devastation to the local community (Owen et al., 2020; Quadra et al., 2019; Queiroz et al., 2018). In any case, wastes from operational and inactive mines represent a major environmental challenge for mine operators, local communities, and governing regulators. This generates urgent calls for appropriate management of mine tailings to minimize disaster and pollution risk.

Many international stakeholders increasingly share efforts to join forces and share commitments to adopt best practices on tailings management. At the international level, initiatives such as Global Tailings Review (globaltailingsreview.org) have released "Global Industry Standard on Tailings" (2020), led by the International Council on Mining and Metals (ICMM), United Nations Environment Programme (UNEP), and Principles for Responsible Investment (PRI). The standard aims to prevent catastrophic failures and improve the safety of mine tailings facilities at all stages of their lives, including closure and post-closure. At the regional level, such as in Europe, the European Directive 2006 / 21 / EC (2006) lays down measures, procedures, and guidance to prevent and reduce, as far as possible, any adverse effects on the environment and human health resulting from the management of extractive waste. All member states are required by this directive to create an inventory of closed and abandoned mining waste sites that cause or have the potential to cause significant adverse environmental consequences.

Today, many guidance and consensus on waste management have attempted to encourage the integration of sustainable development principles into mine planning and operations (Bellenfant et al., 2013; Franks et al., 2011; Xu et al., 2019). This aligns with the sector's industrial ecology thinking, which promotes effective recycling, reprocessing, and rehabilitation of mine wastes (Lèbre & Corder, 2015; Lottermoser, 2011). Such a recovery-oriented approach to waste management could be appealing from a financial and environmental standpoint. In addition to limiting the environmental implications of mining waste disposal, there is the possibility of cost savings in remediation and income increases from recovered and waste-derived products. While sustainable mine waste management for tailings sounds compelling, the situation in most countries, including Europe, tells otherwise. A comparative mine waste assessment in several European countries found that there is still room for improvement in the valorization and recycling potential of mine waste (Žibret et al., 2020). This implies that crude collections of mine waste inventories might miss the opportunities to recognize mine tailings repositories as resources. Such findings deserve attention, as estimates suggest that mine waste represents 27% of the EU's current waste output (Eurostat, 2020).

In response to the above needs, an increasing number of projects have been established to address mining waste issues through technological innovations in the EU (Blengini et al., 2019). Such collaboration frequently involves mining operators / asset owners, research institutes, and policy stakeholders: critical components in scaling up solutions due to the engagement of actors from many sectors (Kinnunen & Kaksonen, 2019). After all, transforming mine residues into resources helps secure supply to minimize future risks related to poor governance, limited material substitutability, and low recycling rates (Løvik et al., 2018; Santillán-Saldivar et al., 2021). To develop sustainable reprocessing solutions for mine tailings, a European Training Network (ETN) for the "Remediation and Reprocessing of Sulfidic Mining Waste Sites" (etn-sultan.eu) was launched with the support of the EU Framework Programme for Research and Innovation Horizon 2020. From 2018 to 2022, the interdisciplinary and intersectoral consortium developed innovative methodologies to i) assess the resource potential of Europe's tailings and ii) to explore techniques for comprehensive metal extraction / recovery and valorization of tailings resources, covering all the links in the reprocessing value chain. The work reported in this thesis was completed as part of the aforementioned ETN project, with an emphasis on novel environmental assessment techniques for mine tailings.

This chapter covers the following topics. First, the environmental implications of standard tailings management and alternative / emerging resource recovery technologies are highlighted. Second, the scientific approaches for assessing waste management systems from an environmental standpoint are pointed out. The thesis goals and pertinent research questions are developed using current knowledge and identified research gaps. Finally, a guiding structure on how this thesis addresses each research question is presented.

1.2 State of research

1.2.1 Tailings storage and acid mine drainage pollution

Tailings are bulk materials generated from the beneficiation step of the metal ore. The fraction of tailings to ore can range from 90–98% for some porphyry copper ores to 20–50% for other minerals (Nagaraj, 2005). There are several options to manage these tailings streams: riverine disposal, submarine disposal, backfilling, and storage behind dammed impoundments (Lottermoser, 2010). Direct discharge of tailings materials into freshwater bodies such as river and seas raise controversy and are mostly prohibited in many areas due to considerable adverse impacts on the local environment, disrupting river sedimentation, water quality, and biodiversity (IIED & WBCSD, 2002; Vogt, 2012). The standard disposal method is to discharge tailings streams into engineered storages, with the facilities often termed tailings ponds or dams.

Mine tailings storage facilities are designed to endure several decades. They are subject to close monitoring and maintenance not only throughout the entire active life of the mine but far beyond their operational years during the closure and abandonment phase (Komljenovic et al., 2020). Despite engineering designs, however, reactive components such as acid-generating minerals (e.g., pyrite, pyrrhotite, galena, sphalerite, etc.) may still get exposed to water and atmospheric oxygen. Such oxidative weathering of reactive sulfidic minerals through a series of reactions is the precursor of acid mine drainage or commonly called AMD (Kossoff et al., 2014; Park et al., 2019). It is well known that AMD is a serious environmental concern of mine tailings, especially when large quantities of sulfidic minerals are present and promote high solubility of metallic species, e.g., lead, copper, cadmium, arsenic, and other metals, causing major pollution events (Dold, 2014; Nieva et al., 2018; P. Wang et al., 2019). Depending on the characteristics of the tailings and local conditions, this process can take many years, if not centuries, and cause long-lasting negative impacts on the environment.

1.2.2 Toward resource recovery from mine tailings

Global concerns about the availability of sustainable and adequate supplies of the minerals and metals needed by society are rising (Rankin, 2012). Metal consumption has consistently grown over the previous century and is predicted to accelerate in response to global expansion, economic growth, particularly in emerging nations, and the need for a low-carbon future (IEA, 2021; World Bank, 2020). Against the background of increasing needs for sustainable minerals and metals, this situation has pushed many countries across the globe to reassess resource recovery from inactive stocks; this includes repurposing mine tailings (Shaw et al., 2013; Tayebi-Khorami et al., 2019).

According to Edraki et al. (2014) and Lottermosser et al. (2011), resource recovery may represent a potential strategy to mitigate the environmental impacts of mine tailings via reprocessing, recycling, valorization, and other rehabilitation efforts. The benefits of this approach are twofold: (i) reduce the volume of mine tailings to be discharged into tailings dams / impoundments, thereby saving space for landfilling and limiting AMD impacts, and (ii) displace traditional, but resource- and energy-intensive materials through adding value to untapped resources. Many of these strategies are based on well-established metallurgical techniques during ore beneficiation, mineral processing, and refining, including physical-based gravity separation and surface-based froth flotation (Whitworth, Forbes, et al., 2022). In recent years, there have been a number of novel applications regarding reprocessing and valorization of mine tailings that can be found in the literature:

- Construction materials: As raw materials for alkali-activated materials / geopolymers (Kiventerä et al., 2020; Mabroum et al., 2020), cementitious binders (Maruthupandian et al., 2021; Vargas & Lopez, 2018), calcium sulfoaluminate cement / CSA (Martins et al., 2021), ceramic (Veiga Simão et al., 2022).
- Secondary metals: Base metals recovery and critical minerals extraction via chemical leaching, bioleaching, or other novel metallurgical processes (Gaustad et al., 2021; Spooren et al., 2020; Whitworth, Vaughan, et al., 2022).

However, variability in the mineralogical, chemical, and physical characteristics of mine tailings implies that tailor-made metallurgical methods are usually required for tailings reprocessing and valorization. In response, the EU recognized research and development interest in this area (Blengini et al., 2019; Løvik et al., 2018), which funded several projects dealing with material recovery from mining / metallurgical residues, such as the Horizon 2020 programs. Furthermore, proper characterization and mapping of materials at the mine waste source, including tailings, is necessary to fully account for potential resource assessment concerning the quantity, quality, and location (Heuss-Aßbichler et al., 2020). With this information at disposal, the share of secondary materials flowing in the EU economy may be boosted from the current suboptimal level of around 10% (A. Mayer et al., 2019). Turning the conceptual promise of tailings reprocessing and valorization into a wider sustainability contribution at larger scales needs a scientific basis from an environmental perspective.

1.2.3 Quantifying environmental impact of mine tailings management

Assessing the potential environmental impact caused by tailings deposition provides a first step for identifying contamination risks and future rehabilitation planning. Similarly, reprocessing and valorizing tailings materials necessitate critical evaluation such that environmentally sustainable and technologically feasible innovations can contribute to the cleaner production of mineral resources.

Many studies have attempted to characterize the environmental impacts of mine tailings management [e.g., (Adiansyah et al., 2017; Brooks et al., 2019; Doka, 2008, 2017; Islam & Murakami, 2021; Reid et al., 2009; X. Song et al., 2017)]. Depending on the purpose of the characterization and time horizon considered, there are different approaches available to estimate tailings emissions: conceptual framework (Adiansyah et al., 2015), dam disaster / hazard risk assessment (Islam & Murakami, 2021; Nungesser & Pauliuk, 2022; Owen et al., 2020), leaching tests and / or environmental and human health risk assessment (Cappuyns et al., 2014; Helser et al., 2022), empirical calculations (Doka & Hischier, 2005; Rader et al., 2019), and geochemistry / reactive transport modeling (Muller et al., 2022; Muniruzzaman et al., 2020). The numerous studies conducted on mine tailings reflect many problems associated with short-term, harmful consequences such as dam failures and long-term, persistent consequences such as metal leakages to soil and groundwater.

Life cycle assessment (LCA) is a decision-making tool to measure the environmental impacts of products / services from a systems perspective (Hellweg & Milà i Canals, 2014). In the wake of increased sustainability awareness across industries, LCA has also been widely employed among practitioners and researchers in the mining and mineral sector (Farjana et al., 2019; Nuss & Eckelman, 2014; Pimentel et al., 2016). Beylot and Villeneuve (2017) relied on companies' environmental reports to source life cycle inventory (LCI) data in the LCA of mine tailings management. Yet, the usefulness of LCA in the case of mine tailings management, however, is frequently hampered, despite being commonly observed as one of the environmental hotspots in the i) supply chain of primary metals (Peña & Huijbregts, 2014; Tao et al., 2019) and ii) metal-based products and systems like battery electric vehicles (Arvidsson et al., 2022; Nordelöf et al., 2014). One reason is the absence of site-specific LCI data regarding tailings management and the associated emissions, which may lead to the inaccurate total neglect of waste management systems in impact assessment (Edraki et al., 2014; Santero & Hendry, 2016). Even if tailings assessment is considered in the system, the current widely used Ecoinvent database is derived from a generic tailings composition with high uncertainties and clearly lacks input-specific assumptions as well as spatial representativeness (Reinhard et al., 2019; Yellishetty et al., 2009).

In response to these shortcomings, there has been a rise in how to complement LCA with other scientific approaches to enhance data quality and scientific rigor. Building upon the findings of Pell et al. (2021), Reuter et al. (2019), and Segura-Salazar et al. (2019), the following notable examples can be found in the literature:

- *Geometallurgy* involves understanding mineralogical and metallurgical ore characteristics to generate mineral databases that can be integrated into a predictive model for mineral processing design. These mineral databases contain chemical properties and elemental composition that can be accessed to create specific orebody knowledge / feedstock and capture details required for the process modeling purposes, as represented by the software packages, e.g., HSC Geo (Metso: Outotec, 2022) and FactSage (GTT Technologies, 2022).
- *Process simulation and flow sheeting* tools can be employed for the respective orebody knowledge and the associated mineral processing with the LCA method. The design of various process configurations aims to translate simulation outputs (i.e., energy and mass flows) to obtain holistic environmental impact indicators. In the last decade, the process simulators are not only able to help generate mass-based process models but also incorporate robust algorithms that allow reconciliation of process chemistry and its governing thermodynamics, making the LCIs and LCAs results reliable (Abadías Llamas et al., 2019; Pell et al., 2019; van Schalkwyk et al., 2018). Examples of well-established mineral processing simulators are HSC Chemistry (Metso: Outotec, 2022), USIM PAC (Caspeo, 2022), METSIM (METSIM International, 2022), and JKSimMet (JKTech, 2020).
- *Geochemical and reactive transport modeling* has the capability to capture the evolution of tailings chemistry, such as complex chemical and dissolution processes, potentially leading to the formation of mine drainage (Hansen et al., 2008). According to Steefel et al. (2015), geochemical modeling tools contain a wide range of equilibrium and kinetically controlled geochemical reaction processes, which include, among others: i) mineral dissolution and precipitation reactions, ii) homogeneous (aqueous phase) reactions, and iii) surface complexation diffuse layer reactions. Simulating dynamics of mine tailings with geochemistry approaches can enable the creation of LCI and LCA under different parameters (e.g., tailings composition and rainwater infiltration rates), offering the opportunities to extrapolate such site-specific models to other sites. Some works have demonstrated this concept for

residual waste deposition / landfilling and used the predicted data as emission estimates (Kalonji-Kabambi et al., 2020; Pabst et al., 2017; Salmon & Malmström, 2004). For these purposes, prominent geochemical models are available: PHREEQC (Parkhurst & Appelo, 2013), MIN3P (K. Mayer et al., 2012), and PFFLOTRAN (Lichtner et al., 2015).

In the last decade, LCA applications for emerging processes have been extensively reviewed by many authors [e.g., (Cucurachi et al., 2018; Hetherington et al., 2014; Moni et al., 2020; Thonemann et al., 2020; van der Giesen et al., 2020)]. Prospective or ex-ante LCAs are applied to support process designs and spot environmental improvement potentials at the early-stage development. A comparative assessment of emerging processes with existing mature processes requires to prospectively model the full-scale implementation of the emerging processes and consider potential scaling / learning effects. This is necessary to enable a fair comparison, as emerging processes are likely to undergo process improvements through scaling and learning, while mature technologies have already been optimized. This could be solved systematically by following recommendations for prospective LCA studies (Arvidsson et al., 2018; Bergerson et al., 2019; Buyle et al., 2019). This involves systems analysis by modeling the foreground and background systems of interest at mature technological levels and full scale.

Similar to standard / retrospective LCAs, appropriate implementation of prospective LCAs needs high-quality and thorough technical data. However, inventory data for such emergent processes is scarce. Therefore, given the low maturity of processes under development and hence limited meaningful data at the early stage, upscaling approaches are needed. According to Tsoy et al. (2020) and Parvatker & Eckelman (2019), the following methods are available from the literature to address data gaps and to generate LCIs associated with the process modeling of novel technologies:

- Estimations of environmental impacts of similar technologies using scaling relationships (Caduff et al., 2011, 2014; Dick et al., 2004).
- Engineering-based calculations (Piccinno et al., 2016), chemical process simulations, usually supported by specific tools / software (Capello et al., 2007; Segura-Salazar et al., 2019), and proxy for chemical reactions, e.g., via stoichiometric calculations (Hischier et al., 2005).
- Predictions of environmental impacts for chemicals using molecular structure models that are based on neural networks (Karka et al., 2022; R. Song et al., 2017; Wernet et al., 2012)
- Technological learning curves and effects (Bergesen et al., 2016; Caduff et al., 2012; Thomassen et al., 2020).

Besides LCIs in the foreground system of prospective LCAs, the development of technologies can significantly benefit from an integration of future scenarios built upon well-established narratives, e.g., UNEP-IRP geo outlook (UNEP, 2019) and long-term energy perspectives (IEA, 2022). After all, the construction of prospective LCA requires harmonized narratives and approaches both in the foreground and background systems (Arvidsson et al., 2018), which have been increasingly used as the methodological basis according to the review of Bisinella et al. (2021). In practice, this involves using additional models such as prospective material flow analysis (MFA), energy system models, external databases of climate scenarios, and other foresight-specific tools. Mine tailings generation is tied to the production of metals / minerals and therefore is also affected by the trends in mining. Numerous scenario-based studies have conducted the environmental impacts of future metal production involving a combination of one or more trends in technological alternatives, efficiency improvements, background energy mix changes due to transition, and material supply / demand [e.g.,(Elshkaki et al., 2018; Giurco & Petrie, 2007; Kuipers et al., 2018; Kulczycka et al., 2016; Rötzer & Schmidt, 2020; Van der Voet et al., 2019)]. A forward-looking analysis led by Ciacci et al. (2020) explored the evolving copper supply, demand, and associated GHG emissions in the EU through scenario modeling. Their study demonstrated the successful combined use of scenario analysis, dynamic MFA, and LCA to assess impacts at the EU level. In general, it is discovered that while environmental impacts per functional unit (i.e., kg of metal) may decrease with time, total environmental impacts may grow in the coming decades mainly due to population and economic growth, as also reported by Yokoi et al. (2022).

Scenario modeling is critical to obtain prospective background LCA databases when doing prospective LCA. Integrated assessment models (IAMs) are among the robust, widely used frameworks for modeling energy systems and industrial processes in future scenarios. Mendoza Beltran et al. (2020) applied IAM models from IMAGE to update background systems, given economic and technical constraints / objectives in the defined future scenario pathways. The future scenario pathways are borrowed from the shared socio-economic pathways (SSPs). The SSPs are part of a scenario framework established by the climate change research community to facilitate the integrated analysis of future environmental implications, adaptation, and mitigation (Riahi et al., 2017). Under the SSPs, there is a wide range of scenarios, covering multiple narratives and assumptions describing alternative socio-economic developments, from business as usual to sustainable development (Fricko et al., 2017). As of now, two LCA-related tools have been developed to ease the linking of IAMs with prospective inventory databases: i) via the superstructure approach (Steubing & de Koning, 2021) and ii) the streamlined coupling approach (Sacchi et al., 2022). Such integration tools allow systematic database creations and incorporation of scenarios in prospective LCAs, similar to studies conducted for future metals supply (Harpprecht et al., 2021; Meide et al., 2022).

Detailed environmental impact quantification can help to prioritize actions toward reprocessing and valorizing particular materials from extractive residues (Joyce & Björklund, 2020). While LCA—one of the prominent industrial ecology tools—has performed forward-looking assessments of scenarios, the roles and impacts of secondary resources mining (i.e., tailings reprocessing and valorization) in the sustainable future supply of future materials have rarely been investigated. This calls for follow-up investigations, knowing that multiple GHG reduction measures are needed to reach decarbonization goals (Watari et al., 2022; Yokoi et al., 2022). This literature review shows the applicability of LCA and its advancement to directly support science-based recommendations and policymaking, essentially to understand the environmental benefits / tradeoffs of mine tailings repurposing strategies.

1.2.4 Research gaps

The literature review of sections 1.2.1 – 1.2.3 reveals several research gaps concerning detailed mine tailings assessment (local and global) and evidence on the actual sustainability performance of reprocessing through emerging technologies. Specifically, the following research gaps were identified:

- Spatial and temporal inventory assessment needs to be performed in the life cycle of tailings management. While scientific approaches and process / geochemical models for estimating characteristics and environmental consequences of tailings are readily accessible, they were historically used independently in silos. Combining relevant scientific approaches and integrating them into LCA can i) allow accurate, site-dependent environmental impact quantification of the tailings disposal practice, ii) provide a full picture of where the environmental hotspots are located, primarily when global scale assessment is pursued, and iii) enable time-specific emissions to be assessed for processes that occur for hundreds or thousands of years, such as metal leaching behaviors.
- Various resource recovery technologies may emerge and help mitigate the environmental liabilities of mine tailings. Although such strategies show early promise, deploying new technologies necessitates additional resource consumption and process adaptation, which might counterbalance the expected environmental benefits. Compared with other waste-to-product concepts in the mineral industry, like bauxite residues or base metal scraps, LCAs on mine tailings use / exploitation for secondary resources are rare. Thus, a systematic approach is needed to assess these emerging metallurgical technologies' environmental performance.
- Modeling systems with changing parameters such as energy transition and metal supply / demand dynamics is necessary for future-oriented environmental assessment of tailings reprocessing and valorization technologies. Combining scenario development from IAMs and systems thinking in LCA is needed to characterize the potential environmental benefits and impacts of alternative tailings management technologies.
- There has been growing interest in repurposing mine tailings for valuable products in Europe. Nevertheless, the implementations of technologies are only done at lab or pilot scale in some sites. The environmental assessment of large-scale deployment of

tailings management is thus lacking. To fully understand the contribution of tailings reprocessing to material valorization and climate mitigation, it is necessary to develop a bottom-up environmental assessment that can be combined with future scenarios regarding metal demand trajectory, energy transition, and resource recovery pathways.

To address these identified research gaps, I developed objectives, goals, and pertinent research questions described in the next section.

1.3 Goals and research questions

This thesis aims to investigate the environmental impacts of both existing tailings disposal and emerging mine tailings management alternatives from a life cycle perspective. To approach the overarching objective, four goals are defined: 1) improve the environmental assessment for tailings by considering site-dependent factors influencing leaching behaviors and identifying the global hotspot mining sites, 2) characterize the environmental performance of emerging tailings reprocessing technologies, 3) integrate scenario modeling into the environmental assessment of large-scale deployment of tailings management, and 4) quantify the circularity and environmental impacts associated with conventional and alternative mine tailings management, accounting for future trends such as metal demand trajectory, energy transition, and resource recovery pathways.

To reach these goals, the following research questions (RQ) are formulated:

RQ 1: Regionalization of tailings life cycle inventory

How can mine tailings disposal emissions be modeled and improved by considering site-specific factors and characteristics of tailings? Where do global environmental hotspots occur?

RQ 2: Prospective LCA approaches for the case of tailings reprocessing and valorization

How can small-scale laboratory and pilot experiments to recover materials from mine tailings be used to assess the environmental performance of a future full-scale implementation of reprocessing and valorization technologies?

RQ 3: Large-scale potential for the use of tailings-based products as industrial resources

What is the potential of recovering minerals and metals from EU mine tailings to produce industrial products? Can alternative tailings management contribute to reducing environmental impacts (and if so, how much)?

These research questions shall be addressed for the case studies in Europe and worldwide. The developed methodologies, however, are adaptable to cases in other locations and types of extractive residues.

1.4 Structure of the thesis

This PhD thesis is a cumulative dissertation encompassing three scientific articles. Each research question (section 1.3) was analyzed in a separate publication and is presented individually in **Chapters 2-4**, followed by the concluding remarks and ways forward (**Chapter 5**). In appendices A-C, each chapter is supplemented with additional method descriptions and results. The research articles forming the structure of the thesis are depicted in Figure 1.1.

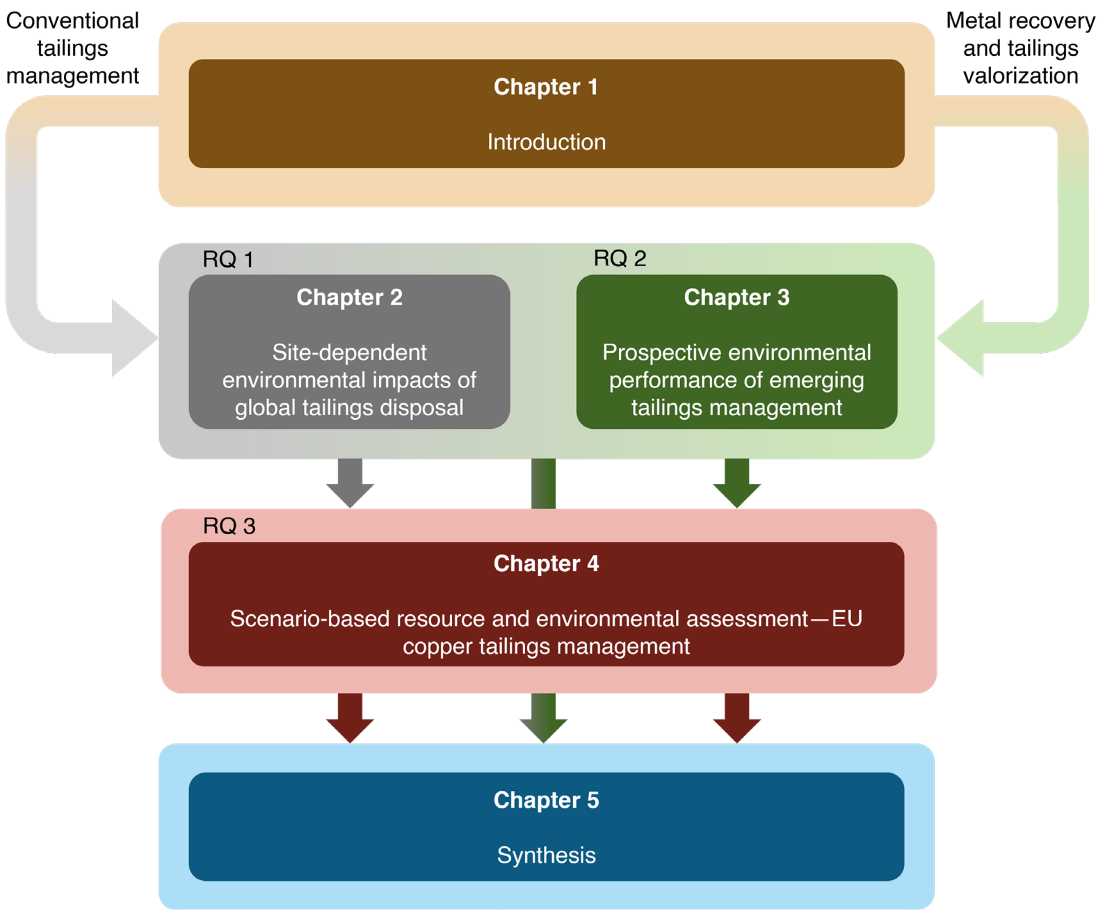

Figure 1.1: Simplified scheme providing the thesis structure.

Chapter 1 introduces the background, the main objectives, and the research questions of the thesis.

Chapter 2 uses geochemistry and metallurgical process modeling to quantify the long-term emissions from tailings deposition, considering local conditions (i.e., hydrology) and compositions of the tailing materials. These emissions can then be assessed with life cycle assessment methodology to analyze the environmental impacts of worldwide sulfidic copper tailings.

Chapter 3 assesses the environmental performance of copper tailings reprocessing and valorization technologies developed in the frame of the project. Primary data from experiments and technology designers' experiences in metallurgical engineering and material sciences are collected as inputs in the process modeling. Technology upscaling methods related to prospective LCA are applied. Environmental hotspots in the resource recovery system are identified and may guide process optimization toward more sustainable reprocessing of metal tailings.

Chapter 4 leverages the methodologies from earlier chapters to develop a scenario-based environmental assessment for tailings management in the EU. The EU serves as a regional case study, considering the increasing number of extractive residue innovation projects in the value chain of mine tailings reprocessing. I seek to derive scientific evidence on whether and which environmental gains are possible. In this study, I incorporate scenario analysis, technological development, and life cycle assessment—together representing elements for forward-looking analysis—to prospectively quantify the recycling and environmental potential of emerging resource recovery technologies.

Chapter 5 synthesizes all the research performed throughout this thesis (**Chapters 2-4**). Key conclusions of this thesis are drawn. Scientific and practical contributions are discussed for LCA / research community, industry practitioners, and policymakers. The critical appraisal elaborates on the challenges managed in conducting the study and the recognized limitations. In the outlook, I propose recommendations for potential future research and supporting informed decision-making processes.

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Chapter 2

Regional life cycle inventories for global sulfidic copper tailings

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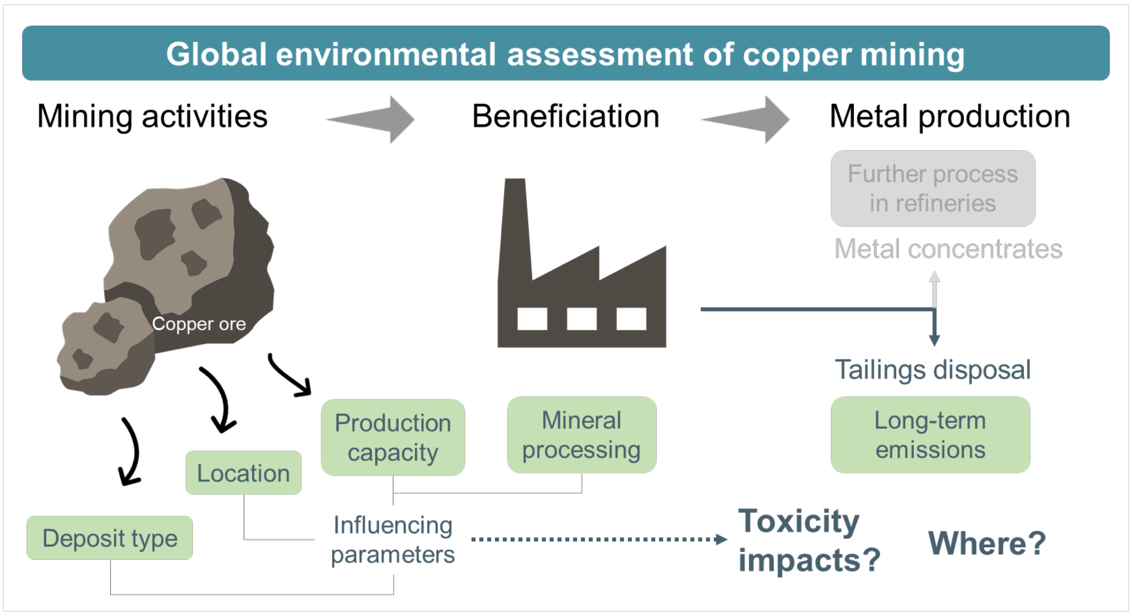
Chapter 2: Regionalized life cycle inventories of global sulfidic copper tailings

Graphical abstract



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 the hydrological condition that enhances the metal leaching in long-term horizons. 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, tailings geochemistry, metal production, mineral processing, life cycle assessment

2.1 Introduction

According to the UNEP-IRP Global Resources Outlook 2019, the use of natural resources has tripled in the last four decades (IRP, 2019), and if business as usual is maintained (UNEP, 2019) 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 (World Bank, 2020). However, the environmental implications of this transition depend on the technological routes to satisfy future copper demands (Elshkaki et al., 2018; Kuipers et al., 2018; Van der Voet et al., 2019).

Various types of copper sulfide ore deposits are the primary source of metallic copper, accounting for 80% of copper resource (Schlesinger et al., 2011). 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 (Gordon, 2002; Lottermoser, 2010; Mudd, 2010) (Figure A29). 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 (Calvo et al., 2016; Gordon, 2002). Declining copper grades in deposits might worsen this situation, as it implies more tailings will have to be managed per kg of copper produced (Crowson, 2012).

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 (Lottermoser, 2010), 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 (Kesler & Simon, 2015; Revuelta, 2018), the differences of which are important when considering leaching behaviors due to presence of acid- / base producing minerals (Dold, 2014; Hansen et al., 2008).

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 et. al (2012), Nassar et. al (2012), Cimprich et al. (2017), and Bach et al. (2016)

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) (UNECE, 2021), record pollutants released to the environment, but with varying level of detail depending on the specific requirements of local environmental authorities (Cooper et al., 2017; Johnston Edwards & Walker, 2020; Kolominskas & Sullivan, 2004; Mudd, 2007). Life cycle assessment (LCA) is a standardized approach to assess the processes' impacts throughout the entire metal value chain (Nuss & Eckelman, 2014; Segura-Salazar et al., 2019). 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 (Pell et al., 2019; Stewart et al., 2012). This persisting issue has been discussed (Althaus & Classen, 2005) and worked around by other researchers in the LCA field by applying a waste-specific transfer coefficients model (Doka, 2017) that was initially developed for landfill emissions (Doka & Hischier, 2005). When specific mine data are available, one could also build the inventories for the current conditions as demonstrated by others (Beylot & Villeneuve, 2017; Peña & Huijbregts, 2014; Song et al., 2017). 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 (Kuipers et al., 2018; Memary et al., 2012; Norgate & Haque, 2012; Pell et al., 2019; Van der Voet et al., 2019).

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.

2.2 Materials and Methods

2.2.1 Methodology overview

Our main methodology built upon previous work in the advancement of mineral processing (Reuter et al., 2019; Segura-Salazar et al., 2019), subsurface environmental simulation (Steefel et al., 2015), and the environmental impacts of mine tailings in the LCA of metal production (Beylot & Villeneuve, 2017; Muller et al., 2019; Song et al., 2017). The methodology integrated several key frameworks that link the workflows for estimating the environmental impacts of mine tailings (Figure 2.1 top). Because copper has the most comprehensive mineral and production database available (Mudd & Jowitt, 2018) 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.

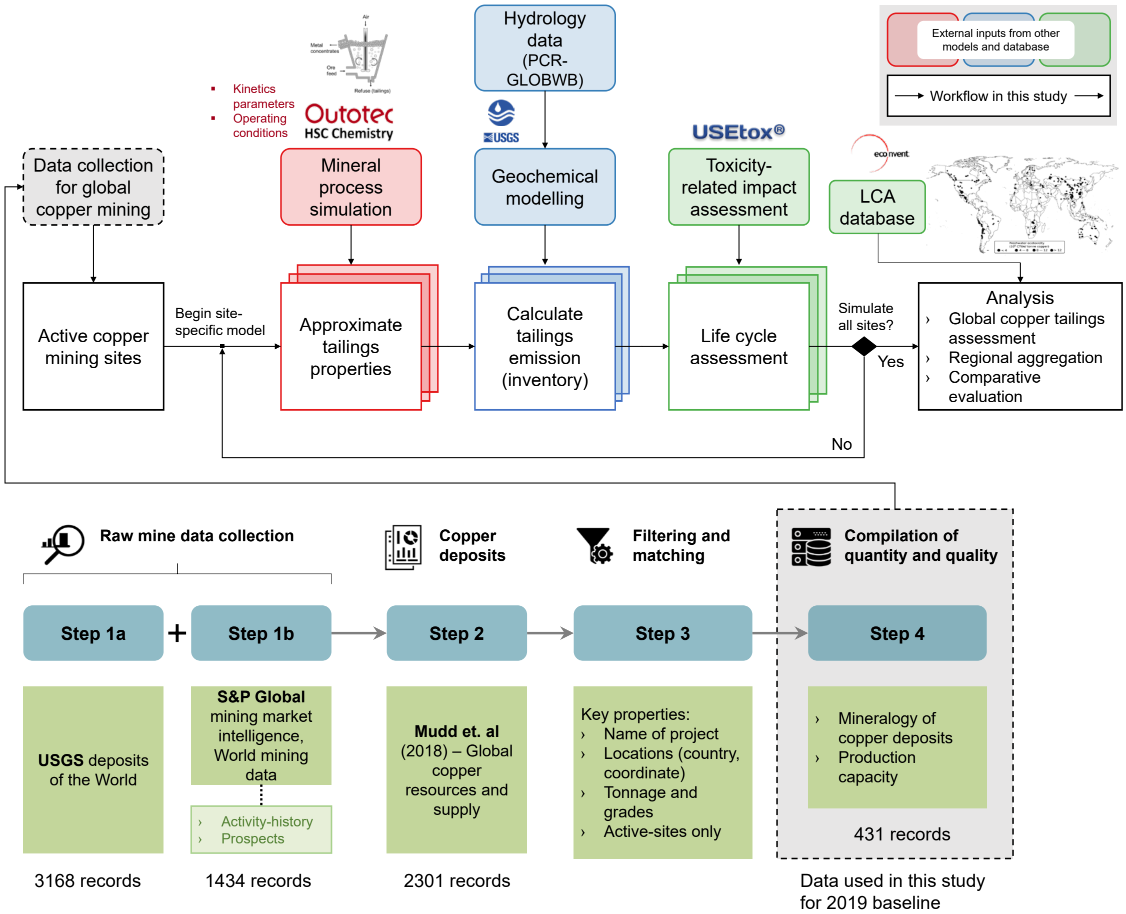


Figure 2.1: Schematic illustration of the methodology employed (top) and sources of copper production data (bottom).

2.2.2 Copper production data

The database compiled in this study combined extensive resources from 1) *U.S. Geological Survey Mineral Database* (USGS, 2019), 2) *S&P Market Intelligence Metals and Mining Report* (S&P, 2020), 3) *World Mining data 2020* (Austrian Federal Ministry of Agriculture Regions and Tourism, 2020), and 4) a rigorous copper deposit study (Mudd & Jowitt, 2018). 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 Appendix A2). 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 2.1 bottom.

2.2.3 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 (2013), with additional sub-classification based on copper grades, buffering minerals, and other impurities (Hammarstrom et al., 2019; Mudd & Jowitt, 2018). Their approaches provide necessary classification guidelines, and all compiled active copper-sites are presented in Table A1.

To link the data from the previous step with a beneficiation process, we constructed simplified flowsheets (illustrated in Figure A1) with industrial process parameters in the software Outotec HSC Chemistry 10 “Flowsheet Simulation” feature (Metso Outotec, 2020). This approach is similar to what others have done (Jose-Luis et al., 2019; Michaux et al., 2020).

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(Bu et al., 2017; Ruuska et al., 2012). 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* (Bulatovic, 2007) and *Will's Mineral Processing Technology* (Wills & Finch, 2016). Other sources, such as a collection of flotation studies and patent literatures (Duan et al., 2003; Fuerstenau et al., 2007), 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 A2 and tabulated in Table A2-A3.

2.2.4 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 (Ecoinvent, 2021), 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 (Santero & Hendry, 2016). The remaining copper processing inventories were taken directly from the Life Cycle Inventory (LCI) database ecoinvent 3.6 (Ecoinvent, 2021). The illustration of the studied system is depicted in Figure 2.2.

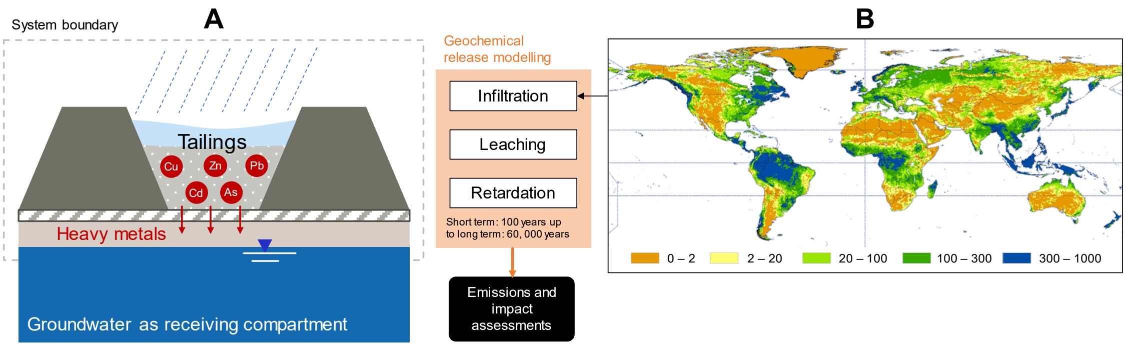


Figure 2.2: The schematic illustration of tailings emission model. Part A describes the tailings characteristics and metal species considered in this study. Part B shows the annual groundwater recharge, taken from the results of PCR-GLOBWB (Sutanudjaja et al., 2018), in mm per year.

This study focuses on toxicity-related environmental impacts by using the latest midpoint impact categories recommended for life cycle impact assessment (Bulle et al., 2019; Rosenbaum et al., 2008). This includes freshwater ecotoxicity, for which impacts were quantified by applying global characterization factors (CFs) (defined in USEtox 2.12 (USEtox, 2019)) 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 (Beylot & Villeneuve, 2017) (see Appendix A16). 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 (Bakas et al., 2015).

To predict emissions for a long time horizon, we applied geochemical modeling using the PHREEQC simulation (Parkhurst et al., 2013). This model allows the prediction of metalloid releases from the tailings, which are controlled by mineralogy and pH development (Cappuyns et al., 2014; Jarošíková et al., 2017). 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 (Nordstrom, 2019). For As, the surface complexation reactions were obtained from the Dzombak model (Dzombak & Morel, 1990), 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 A4 and A13, following the approach of Hansen et al. (2008) and Dijkstra et al. (2008), 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 (Sutanudjaja et al., 2018). We used the net groundwater recharge as the site-specific infiltration parameter in our calculation (see part B of Figure 2.2). 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 (Rowe & Islam, 2009).

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 A5-A12 and Figure A6-A13) 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 A5 and Appendix A4). 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 (Kosson et al., 1996; Schwab et al., 2014), but is adapted to the specific mine tailings composition, deposit dimensions and site-specific climatic conditions.

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

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).

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

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 A4).

2.2.5 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 (Ecoinvent, 2021) (based on the LCA report of copper supply chain analysis (Streicher-Porte et al., 2010)). The overall procedure is illustrated in Figure A15.

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 A5-12). 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 (Ecoinvent, 2021). We matched our modeled inventory in this study following the country classifications that ecoinvent implements (Figure A16). 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.

2.2.6 Modeling future global copper tailings emissions

The primary supply of copper until 2050 provided by Elshkaki et al. (2018) and Northey et al. (2014) 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 (Hammarstrom et al., 2019), S&P feasibility study (S&P, 2020), and other reports (Amos et al., 2018; SRK Consulting, 2016) (see Figure A17).

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 (2012), 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.

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

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 (2015), the life cycle of a mine prior to operation can be distinguished into two stages: exploration and construction (Figure A17). The discovery phase of copper production was excluded in this analysis, as it takes an average 20 years before a mine can finally operate (Ali et al., 2017).

Using these approaches (see Appendix A8), 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 (2019) deposit characteristics database. Finally, we combined the merged forecasted data with our method to estimate the environmental impacts of tailings until 2050.

2.3 Results and discussion

2.3.1 Global assessment of copper mine tailings

The toxicity impacts of all 431 assessed active copper sites in 2019 are shown in Figure 2.3A. The detailed results are available in the digital SI. These sites capture >75% (Mudd & Jowitt, 2018; S&P, 2020; USGS, 2019) 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 2.3 and Appendix-2 Table A2.3) 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)

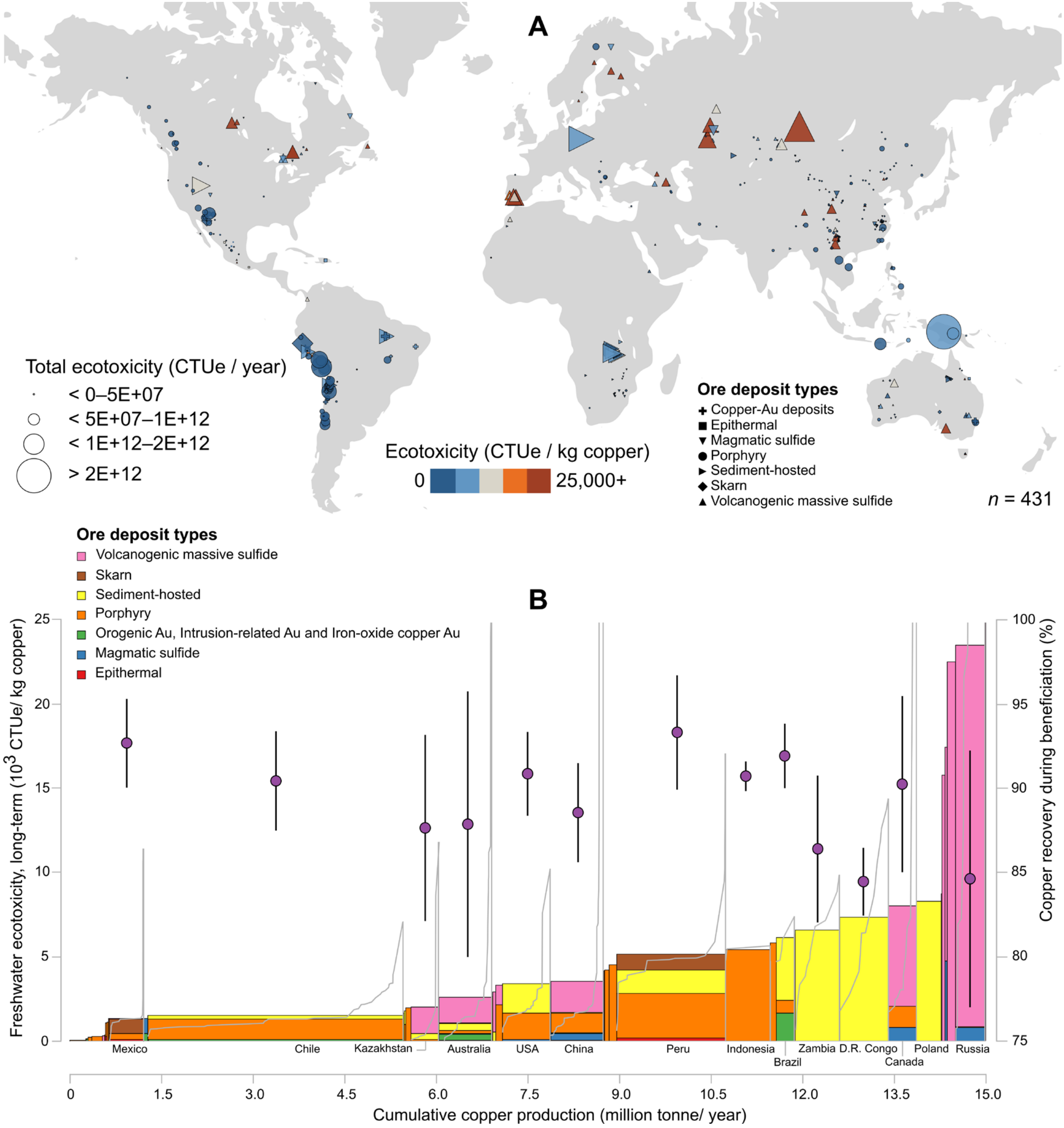


Figure 2.3: Part A shows the freshwater ecotoxicity (long-term) of copper production for each mine site (LCIA method: USETox). Three features are displayed: 1) total ecotoxicity (indicated by bubble size), 2) ecotoxicity per copper mass produced (indicated by color), and 3) the type of ore deposits (indicated 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.

2.3.2 Region-specific and country-aggregated assessment

For background LCI databases, country-level data is required and Figure 2.3B 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 (Bulatovic, 2007; Wills & Finch, 2016).

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 A21 and Table A16.

2.3.3 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 A22). Ecotoxicity per kg of copper decreases with an increasing copper grade, but correlations are very weak (Figure A24). 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 A25).

2.3.4 Comparison of leaching and toxicity results to other studies

The comparison of our toxicity results with country-specific datasets in the ecoinvent 3.6 (Ecoinvent, 2021) per tonne of tailings is presented in the top part of Figure 2.4A 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 2.4B).

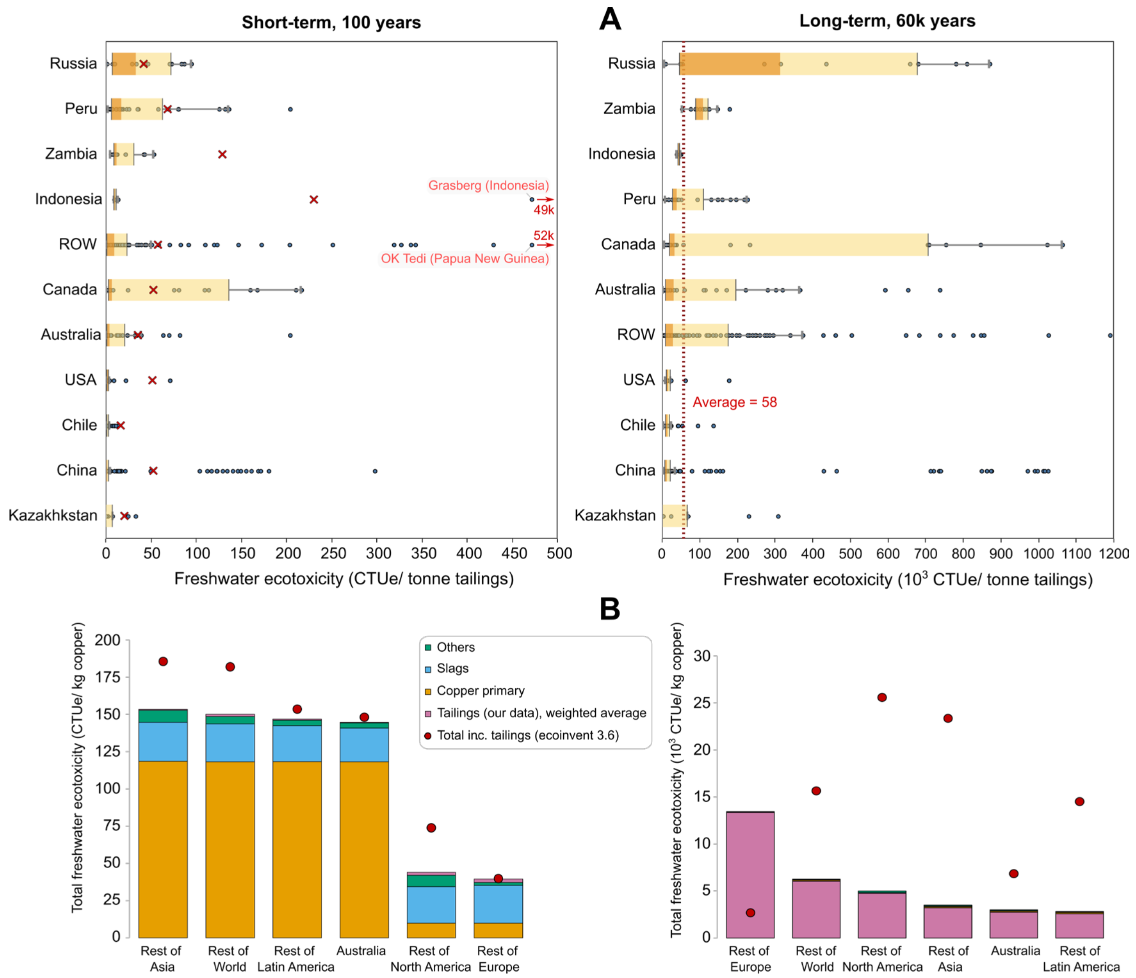


Figure 2.4: Part A: Freshwater ecotoxicity impacts quantified per tonne of tailings deposited for countries covered in ecoinvent. The red cross symbols indicate values from ecoinvent for the short-term horizon. Long-term variability from ecoinvent is not shown due to negligible differences between countries (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 A27. 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 Appendix-2, Table A2.10-A2.11.

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 (Earthworks, 2012; Vogt, 2013) pollute fresh water immediately, highlighting the need for alternative disposal methods (Mudd et al., 2020).

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 2.4B). 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 A18), 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 Appendix A14) and one should avoid ignoring differences between sites when performing comparative LCA studies.

2.3.5 Impacts of future primary copper production

Freshwater toxicity impacts in the upcoming decades based on projected future production are shown in Figure 2.5. 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 (Ali et al., 2017; Elshkaki et al., 2018; Northey et al., 2014), the production will reach a peak level at 2030 and flatten after 2050 thanks to direct reuse from stocks and availability from recycling streams.

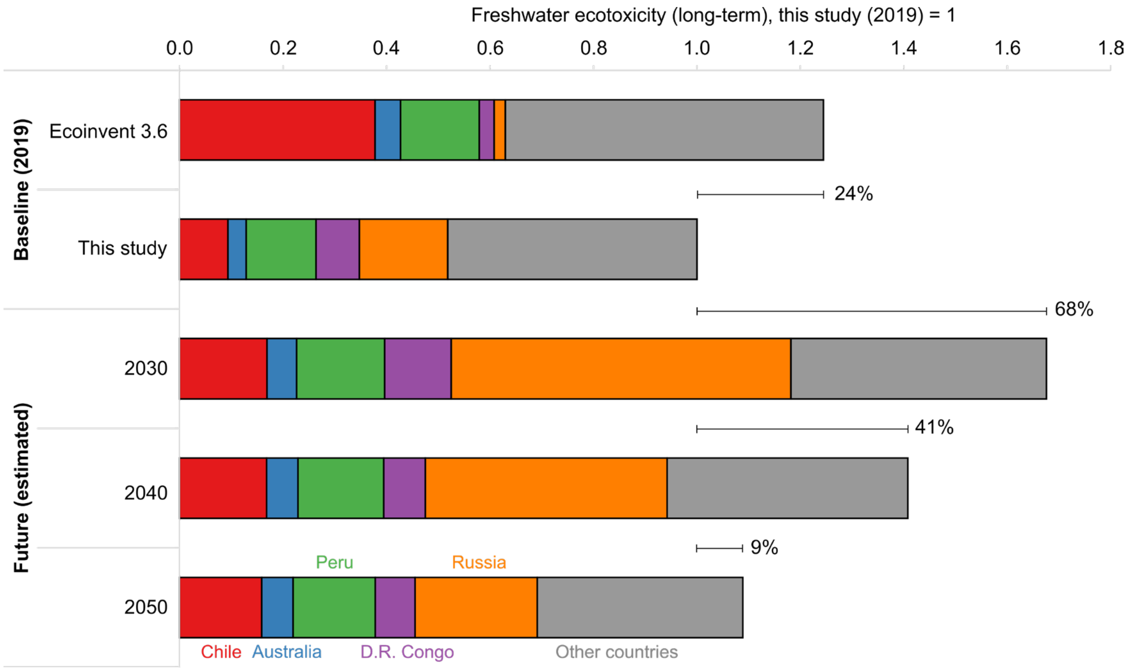


Figure 2.5: Freshwater ecotoxicity impacts from one year of operation of all global sulfidic tailings (long-term time horizon of 60'000 years), including future projection of copper-extraction amounts from other studies (Elshkaki et al., 2018; Northey et al., 2014).

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 (Calvo et al., 2016; Crowson, 2012; Mudd & Jowitt, 2018) 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 2.5 and Table A14). 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 A14). 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 (Huijbregts et al., 2017) and the Environmental footprint (EF) method (Fazio et al., 2018), 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 Appendix A15 and Appendix-2 Table A2.14).

2.3.6 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 (Schlesinger et al., 2011). 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 (Valenta et al., 2019). Additionally, we only modeled the beneficiation process as a single-stage circuit (Figure A1), while advanced grinding and flotation techniques (Kohmuench et al., 2018) could optimize particle liberation from the ore and thus reduce toxicity of tailings (see Appendix A12). 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 (Sutanudjaja et al., 2018) 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 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 (Döll et al., 2014; Hanasaki et al., 2018; Sutanudjaja et al., 2018)), 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 (Parkhurst et al., 2013) and took default equilibrium reactions available from PHREEQC and WATEQ4F databases (Table A13). We also added arsenic speciation into these databases, which leaches depending on ferrihydrite concentration (Appelo et al., 2002; Lefticariu et al., 2019). 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 (Chen et al., 2014; Dold, 2014). 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 A6–A13 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 (SULTAN, 2019) and Chilean field sampling data (SERNAGEOMIN, 2020), including sensitivity cases for the modeled sites (Figure A19 and A20). 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 (Crowson, 2012; Elshkaki et al., 2016; Northey et al., 2014) with forecasted mining projects from the mining database (S&P, 2020). 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 (Ali et al., 2017; Hammarstrom et al., 2019; Levin et al., 2020). Moreover, different ore deposit types may have different rates of decline. These factors, however, are beyond the scope of this study.

2.3.7 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 (GRID-Arendal, 2020) 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 (Hudson-Edwards, 2016). 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 2.3 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.

2.4 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 (Lèbre et al., 2017). 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 (Blengini et al., 2019) as a basis for the long-term environmental remediation and valorization (Edraki et al., 2014; Kinnunen & Kaksonen, 2019; Park et al., 2019) of mine tailings. The appearances of novel technologies under active development such as solvo-metallurgy (Li et al., 2020) or bio-hydrometallurgy (Falagán et al., 2017) are promising options for tailings reprocessing schemes, which might be implemented in tailing remediation models in future research.

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Chapter 3

Prospective environmental assessment of reprocessing and valorization alternatives for sulfidic copper tailings

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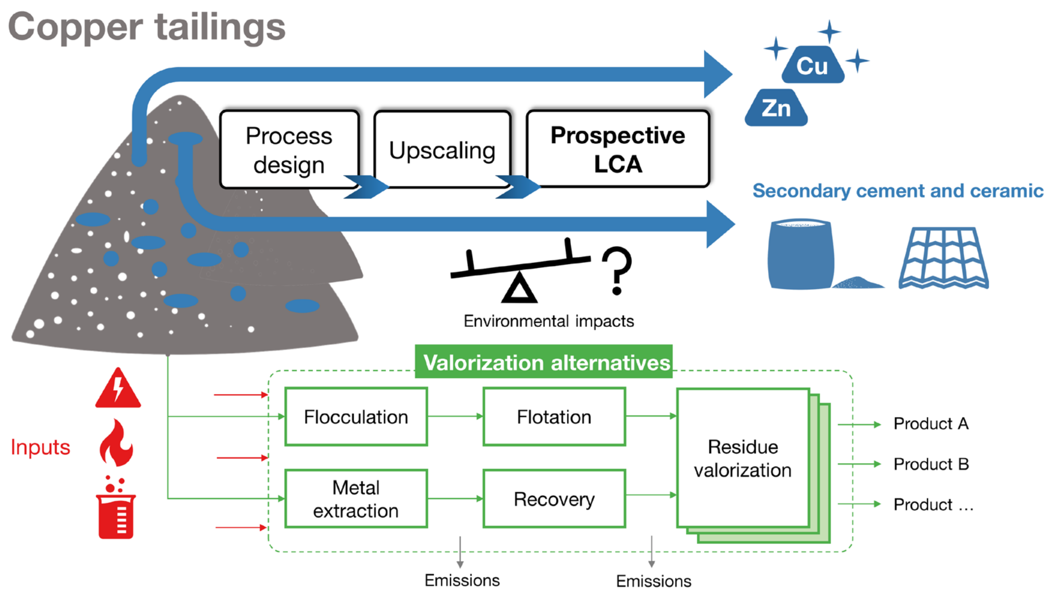
Graphical abstract



Highlights:

- The environmental impacts of reprocessing copper mine tailings were studied.
- Process modeling and technology upscaling were performed for inventory analysis.
- Tailings reprocessing resulted in climate-positive impacts from a life cycle perspective.
- Mineral recovery is essential to ensure significant benefits from reprocessing systems.
- Implications for the sustainable mining industry and policymakers were discussed.

Abstract

Waste from primary mining operations, especially mine tailings, receive much attention as potential secondary resources that can transform liabilities into resources. The primary intention is to minimize mine tailings disposal problems through volume reduction while recovering secondary resources for industrial materials. However, the environmental benefits and tradeoffs behind these approaches remain unclear. This study conducts a process-based life cycle assessment (LCA) study to investigate multiple reprocessing pathways of copper tailings to co-produce secondary metals and building materials. Four design options representing different value-recovery routes are constructed to assess the environmental burdens of reprocessing chains and the associated benefits from displaced virgin products. This study assesses emerging technologies in a prospective LCA, with projections of bottom-up foreground process modelling and background data like energy supply scenarios. Our analysis reveals that the assessed technologies can only be beneficial when all co-products are utilized. Results indicate that reprocessing may save from 25 up to 930 kg CO_2-eq per tonne of treated copper tailings. Potential environmental benefits depend on the reprocessing routes, technology upscaling parameters, and quality of secondary products. We propose recommendations to enhance the environmental performances of mine tailings reprocessing strategies, such as switching to low-impact chemical alternatives and optimizing energy use. Our findings can support the sustainable development of the metal industry in terms of waste management and secondary resource use.

Keywords: life cycle assessment, prospective study, environmental impacts, mine waste, circular economy, waste valorization

3.1 Introduction

Worldwide, industrial-scale mining generates a large volume of waste in the form of tailings residues and waste rock. According to the UNEP 2019 report (UNEP, 2019), copper production alone generates around 4 billion tonnes of tailings annually that should be disposed of. This volume is predicted to increase as our metal demands grow over time (Elshkaki et al., 2018; Watari et al., 2021).

Tailings disposal causes long-term environmental pollution through metal leaching or so-called acid mine drainage (Lottermoser, 2010). Furthermore, poor facility management carries dam structural risks, which inadvertently could lead to huge-scale collapses (Franks et al., 2021). The recent 2019 *Brumadinho* mine accident, among other man-made disasters, was a wake-up call for the entire sector, pinpointing that the long-term storage for such a voluminous waste is not a future-proof solution (Roche et al., 2017). Therefore, pursuing other mitigation efforts is vital to manage mine tailings better, preventing long-term environmental issues and associated hazards.

Better designs for mine waste handling and alternative approaches are already proposed to support cleaner production and environmentally responsible processes. As extensively reviewed by other researchers (Adiansyah et al., 2015; Edraki et al., 2014), examples of feasible technologies are tailings thickening to decrease water infiltration that leads to leaching, desulphurization to limit acid mine drainage, and co-disposal for mine backfill materials. Meanwhile, there are reprocessing strategies for converting tailings to secondary products. The idea has recently become a public interest, in conjunction with the circular economy concept, as it holds the possibility to solve waste problems by turning them into resources (Lèbre et al., 2017; Tayebi-Khorami et al., 2019).

In the context of reprocessing and recycling (Park et al., 2019), tailings are treated to extract metals and mineral residues. Many researchers have already suggested and developed these technologies with a high degree of success in their experiments. For example, one study uses desulfurization flotation techniques to separate sulfur-rich streams (Broadhurst et al., 2015) and generate by-products while simultaneously decreasing acid mine drainage potential. Another study relies on a combined magnetic – flotation, which can further improve technical performances and extract metals (Huang et al., 2020), and reduce the need for primary mining. If metal contents are high (< 0.5%-wt in copper tailings), other researchers encourage leaching or other separation techniques to maximize metal recoveries (X. Li et al., 2020; Mäkinen et al., 2020; Passos et al., 2021). Some other studies focus on using the mineral fractions, primarily consisting of SiO_2, Fe_2O_3, Al_2O_3, and CaO, as the main ingredients for building materials (Ahmari & Zhang, 2012; Gou et al., 2019; Kim & Park, 2020; Leite Lima et al., 2019). While these experiments show promising results, the research was performed with basic or no environmental assessments. Reprocessing of mine tailings also does not automatically make it ecologically benign, for example, due to higher energy demand and unintended contaminant releases. Given these concerns, a thorough environmental assessment is needed to examine whether these new processes bring the expected environmental benefits, especially when dealing with system-wide perspectives and large-scale applications.

For evaluating the environmental performances of mine tailings reprocessing schemes, one can conduct a life cycle assessment (LCA). This method is standardized and aims to holistically quantify the environmental impacts of resource inputs and emissions over the relevant life cycle phases (ISO, 2006). As such, LCA can fill the knowledge gap in the reprocessing of tailings, made possible by systematically calculating environmental impacts. When multiple secondary materials are generated from different treatment routes, it is also crucial to account for all alternatives. Conventional LCAs – due to their generally retrospective nature – are not ideal for modeling the environmental impacts of future systems and possible scenarios, which may involve up-scaling of lab or pilot-scale data to large-scale production in the foreground system, as well as projected background data. Prospective / ex-ante LCAs attempt to resolve these issues by adapting early-stage processes in the environmental assessment of modeled future systems (Arvidsson et al., 2018). Thus, with the help of ex-ante LCA frameworks (Tsoy et al., 2020) and upscaling methodologies (Parvatker & Eckelman, 2019) for complementary data provisions, prospective LCA offers a chance to assess the environmental profiles of emerging technologies that are still at a lab scale.

The main objective of this study is to construct LCA models applied to copper tailings reprocessing and valorization. We develop life cycle models for an operational mine site with specific waste characteristics, constituting various processing routes based upon continuously refined experiments. These were conducted by research partners in the

H2020 ETN SULTAN project (European Training Network for the Remediation and Reprocessing of Sulfidic Mining Waste Sites, www.etn-sultan.eu). This LCA study provided feedback loops for prioritizing technological improvements in processing chains. We also explore the implications of varying key parameters in the processes and evaluate performances with respect to technological changes. Ultimately, the presented study contributes to addressing transparently the environmental performances of mine waste reprocessing for decision-making purposes.

3.2 Methods

3.2.1 Goal and scope

This process-based LCA aims to quantify the life cycle environmental impacts of the different reprocessing chains at a tailings site located in Portugal. Currently, tailings are generated from the beneficiation process of metal ores, after which these slurries are sent to the tailings management facility for deposition (Escobar et al., 2021). In this work, the developed LCA model represents various conceptual reprocessing routes, which comprise novel technologies to recover metals from the waste streams and valorize cleaned residues (Figure 3.1) and are compared to the reference case without tailings treatment (direct landfilling). The functional units (FU) are then: the disposal of one tonne of sulfidic mine tailings (specifications in Appendix Table B1) and the production of materials and other by-products according to reprocessing routes (details in Appendix B2). In summary, the FUs include:

- The disposal of 1 tonne sulfidic tailings,
- The production of 1.56 tonne CSA cement,
- The production of 4.1 tonne ceramics,
- The production of 0.69 tonne geopolymer (equiv. to ordinary Portland cement),
- The production of 2.9 kg copper and 7.5 kg zinc,
- The production of 110 kg sulfuric acid,
- The production of 182 MJ heat energy

We assume zero-burden waste, which excludes the environmental loads caused before the waste generation in the previous life cycles (Ekvall et al., 2007). The system expansion approach is applied, accounting for the credits for the avoided productions (Schrijvers et al., 2020). In the base case, both secondary metals and building materials are assumed to behave comparably similar to the substituted primary products, translating to standard 1:1 substitution ratio (Laurent et al., 2014; Viau et al., 2020). Hence, the impacts of the use and end-of-life phase are excluded in this study. Moreover, relevant information about the use and end-of-life considerations of the resulting secondary products is yet unexplored, preventing any reasonable analysis. The influences of substitution ratios are explored in the sensitivity analysis.

Net environmental impacts of the alternative routes are then calculated as the difference between new reprocessing routes and the reference, comprising: (i) credits for substitution of metals, building materials, and byproducts made from tailings, that otherwise need to be supplied from primary production sources, (ii) credits for prevented long-term landfill emissions to water due to conventional tailings disposal. It has to be noted that the reprocessing system includes additional consumption of resources such as chemicals, aggregates, and other consumables for the production of secondary materials and thus, more than 1 tonne cement or ceramics are produced per tonne tailings in Routes A-1, A-2 and B-1 (Table 3.1). All these additional resource consumptions are included in the inventory models.

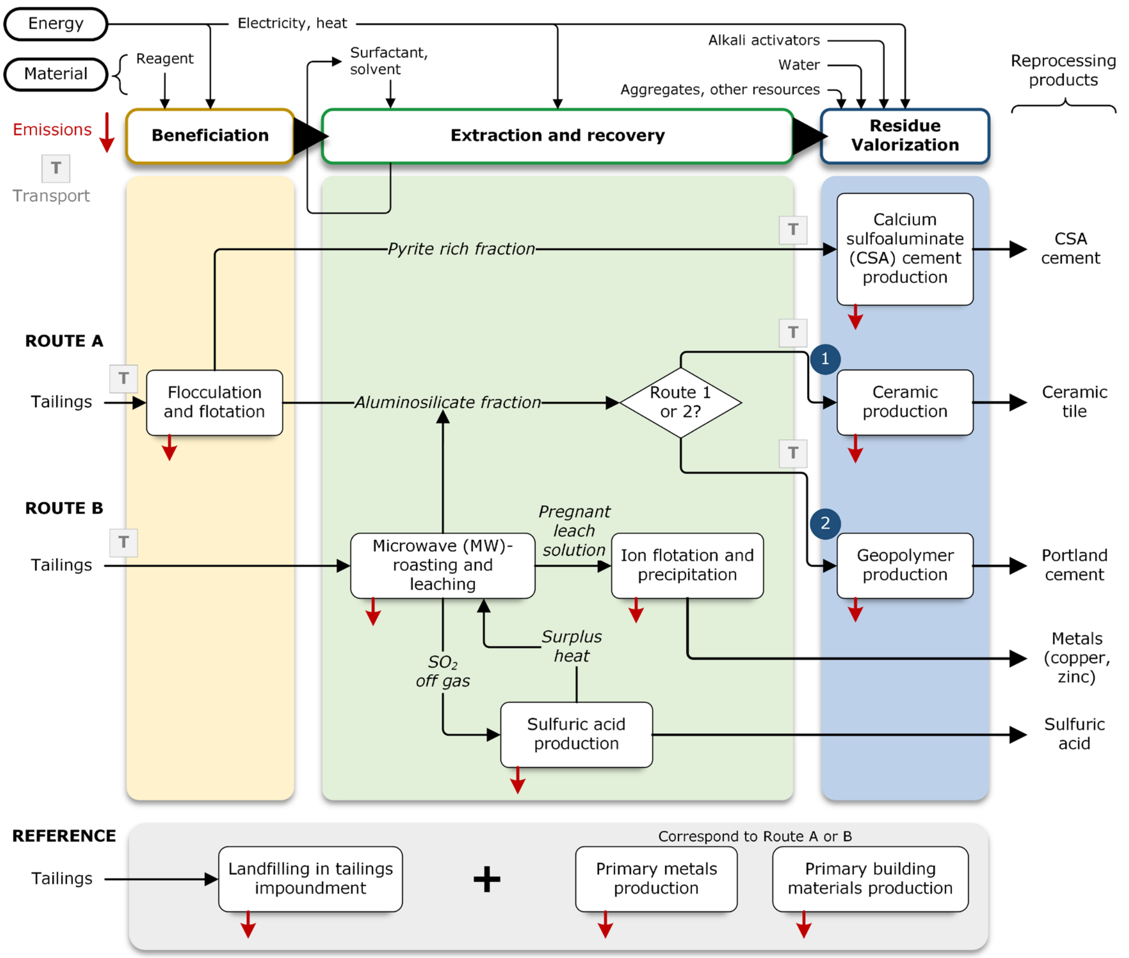


Figure 3.1: System boundaries of the analyzed systems, representing alternative tailings reprocessing route A (building materials production only), route B (secondary metals recovery and building materials production), and the reference 'direct landfilling' route.

Table 3.1 summarizes the processes and routes analyzed in this paper, which are mainly created by gathering direct feedback from researchers working on the reprocessing techniques of the four routes in the project (SULTAN, 2018), and supplemented by literature-based data and discussions with industry representatives.

Table 3.1: Overview of reprocessing steps generated secondary products, and displaced primary products in all routes for the treatment of 1 tonne tailings, compared to the reference route. Credits from secondary products are in brackets.

	Process steps	Route A-1	Route A-2	Route B-1	Route B-2
Environmental burdens	Beneficiation	Flocculation-flotation	Flocculation-flotation	-	-
	Extraction	-	-	MW-roasting and leaching	MW-roasting and leaching
	Recovery	-	-	Ion flotation and precipitation	Ion flotation and precipitation
	Residue valorization	Sulfur rich fraction: CSA cement production Aluminosilicate fraction: Ceramic production	Sulfur rich fraction: CSA cement production Aluminosilicate fraction: Geopolymer production	Aluminosilicate fraction: Ceramic production	Aluminosilicate fraction: Geopolymer production
Environmental credits	Avoided tailings landfill	1 tonne of sulfidic copper tailings			
	Displaced metals	-	-	Primary metals (copper 2.9 kg and zinc 7.5 kg)	Primary metals (copper 2.9 kg and zinc 7.5 kg)
	Displaced building materials	Primary CSA cement (1.56 tonne) Primary ceramic roof tile (4.1 tonne)	Primary CSA cement (1.56 tonne) Primary Portland cement (0.69 tonne)	Primary ceramic roof tile (3.4 tonne)	Primary Portland cement (0.58 tonne)
	Other by-products	-	-	Primary sulfuric acid (110 kg), heat (182 MJ)	Primary sulfuric acid (110 kg), heat (182 MJ)

Primary background data sources were taken directly from the latest Ecoinvent 3.7.1 (Ecoinvent, 2021) cut-off database, accessed using LCA software Activity Browser (Steubing et al., 2020). Overall, the model facilitates a grave-to-gate LCA model incorporating all relevant material and energy inputs to reprocess mine tailings. As for the substituted primary materials production, Ecoinvent 3.7.1 equivalent inventory and literature data were adapted to reflect only primary production routes. Table B26 lists all Ecoinvent 3.7.1 processes used or adapted for the substitution of the primary products. The life cycle impact assessment indicators are described in Appendix B3: We included mid- and endpoint indicators (Hierarchist version) from ReCiPe v1.13 (Goedkoop et al., 2013) for a complete set of impact categories, which also allows full aggregation into a single score for interpretation of overall impacts. Additionally we applied cumulative energy demand (CED) (Frischknecht et al., 2015), which is often used in material assessments, and USEtox toxicity-related indicators (Rosenbaum et al., 2008), which are important for assessing tailings impacts on the environments. The selection of indicators aims at covering the most relevant impact categories in the context of waste management and resource recovery from mine waste, besides allowing comparability with other studies in the field.

3.2.2 Life cycle inventory analysis: Modeling tools

Life cycle data are taken from in-house experiments (SULTAN, 2018), adapted to commercial scale using ex-ante LCA frameworks, and supplemented using secondary data. Technology-specific inventory calculations (Piccinno et al., 2016) are implemented in this study and fulfill the necessary information to complete missing data. Upon previous recommendations, procedures below summarize the inventory modeling hierarchy:

i) When accurate data of mature processes are readily available, this information is directly used
ii) Judgments from experts and process developers are considered for defining relevant equipment setups and process parameters
iii) Make use of engineering-based calculations (Piccinno et al., 2016) for estimating inventory inputs
iv) If data gaps are present after previous steps, similar large-scale applications are used as proxy data

A summary of the prospective modeling methods and the life cycle inventory calculations can be found in Appendix Table B2. The following subsections describe the associated technologies and modeling of the reprocessing routes.

A. Flocculation-flotation

To separate sulfur rich fraction from the original stream (route A), a combined flocculation-flotation process is employed based on the tailings desulfurization study of Broadhurst et al. (2015), complemented with data from project partners. Polyacrylamide and xanthate, which act as flocculants and collector agents, are used to improve pyrite separation. While polyacrylamide is present in the Ecoinvent database, we incorporate xanthate production from another study (Kunene, 2014). Overall, the modified beneficiation for tailings imitates industrial flotation techniques (Norgate & Haque, 2010), excluding the grinding process. Ultimately, these altogether constitute the beneficiation stages to separate pyrite residues fraction from the gangue materials rich in aluminosilicate fractions (Figure B1). Dewatering processes are installed for both outflows before each separate stream continues to valorization units. These processes are described in sections E – G.

B. Combined microwave-roasting and chemical leaching

The purpose of route B is to recover copper and zinc from the tailings by a combination of pyrometallurgical (microwave roasting) and hydrometallurgical techniques (ammoniacal leaching, ion flotation, and chemical precipitation), as illustrated in Figure B2. Since the tailings are in slurry form, they have first to be homogenized and dried. Contrary to route A, the sulfur content of tailings is removed through subsequent steps of drying and roasting. Xanthopoulos et al. (2021) and Ozer et al. (2017) reported that tailings release sulfur off-gas and undergo mineral changes according to the following reactions.

Pyrite oxidation causing mass loss occurs at 380 – 500 °C (eq. R1-R4)

$$2FeS_2 \rightarrow 2FeS + S_2 \tag{R1}$$

$$S_2 + 2O_2\,(g) \rightarrow 2SO_2\,(g) \tag{R2}$$

$$FeS_2 + O_2\,(g) \rightarrow FeS + SO_2\,(g) \tag{R3}$$

$$FeS + 2O_2\,(g) \rightarrow FeSO_4 \tag{R4}$$

Hematite formation causing another mass loss occurs at 500 – 730 °C (eq. R5)

$$2FeSO_4 \rightarrow Fe_2O_3 + SO_2\,(g) + SO_3\,(g) \tag{R5}$$

For the scaling up of microwave furnaces, we use a calculation procedure from the work of Bermúdez et al. (2015) to estimate the specific energy consumption for the large-scale implementations. After the microwave treatment method, the leaching process takes place with a mixture of ammonia and ammonium carbonate solutions as lixiviants. The optimum leaching efficiencies for copper (75%) and zinc (86%) are obtained from partner's results (Xanthopoulos et al., 2021). We design the leaching process as continuously stirred tank reactors as suggested by Piccinno et al. (2016), running parallel to accommodate the scale similar to what is practiced in the industry. The metal-containing leachate or pregnant leach solution then proceeds to a subsequent ion flotation and precipitation unit to recover copper and zinc, respectively. The stripped solid residues are processed in the hypothetical valorization units to produce ceramic roof tiles (route B-1) or geopolymer binder (route B-2).

C. Ion flotation and precipitation

The coupled process to recover metals from leachate is based on Xanthopoulos et al. (2021), a practical application from the theoretical ion flotation studies (Doyle, 2003). Copper is selectively separated over zinc from the pregnant leaching solution by ion flotation, while zinc in the aqueous phase is recovered by precipitation. Initially, the conditioning step—modeled as continuous stirred tank reactors (Piccinno et al., 2016)—

mixes leachate with sodium dodecyl surfactant. Another frother, such as ethanol, is also added to promote foam that physically collapses. This phenomenon helps separate the aqueous parts that contain soluble zinc from the original leachate. Copper and zinc recovery for the upscaled systems is assumed to be the same as its lab-scale results (Xanthopoulos et al., 2021): 85% and 95%, respectively. In scaling up this process, we employ a closed-loop solvent recycling system in ion flotation via continuous distillation (Xanthopoulos & Binnemans, 2021), as similarly done for waste solvent treatments (Amelio et al., 2014; Luis et al., 2013). As a result, 95% of the solvent chemicals are regenerable, with the required energy and chemical inputs, namely electricity, steam, nitrogen gas, and cooling water taken from the software Ecosolvent v.1.0.1 (Capello et al., 2007). However, 2.5% of makeup is still required to compensate for the loss of these chemicals. Figure B3 depicts the setup of the ion flotation and precipitation described above.

D. Sulfuric acid production

Sulfur off-gases are generated from the pyrite roasting process in route B. A safer alternative to directly emitting these gases into the environment is its use as feedstock to produce sulfuric acid. Owing to the dense concentration of SO_2 in the off-gas, pyrite roasting has been viewed as an alternative technique to the standard sulfur burning (Runkel & Sturm, 2009). Some adaptations are still needed, however. The entering off-gas must be cleaned through wet cleaning steps. The cleaning step functions to cool down and remove dust particles that are evaporated during the roasting process. 99.8% conversion of SO_2 into sulfuric acid can be achieved in a series of exothermic reactions, occurring in an absorption tower equipped with packed beds of vanadium oxide catalyst. Our model transfers this excess heat to the other processes that would otherwise require natural gas from external sources. The life cycle inventory data for sulfuric acid production is collected from typical best available technologies (European Commission, 2007) in Europe. The process flowchart is presented in Figure B4.

E. Calcium sulfoaluminate production

Calcium sulfoaluminate cement belongs to the alternative materials that have the potential to lower impacts using secondary, underutilized calcium and sulfur sources as the principal binding chemistry (Gartner, 2004). Akin to ordinary Portland cement, the CSA production involves raw material acquisition, calcination, and gypsum additions. The main changes in our CSA model relate to input materials and process emissions. The presence of ye'elimite ($Ca_4(AlO_2)_6SO_3$) as active clinker phase instead of alite (Ca_3SiO_5) brings advantages in decreasing firing temperature by 100°C to 250°C during clinkerization of CSA cement (Telesca et al., 2019). The inventory input for clinker materials is modeled based on Pires Martins et al. (2020, 2021), where highly sulfidic tailings constitute 14%-wt of raw meal mixture. To generate the entire life cycle model at a large scale, we combine our design mix with the industrial CSA plant data. We rely on the work of Ren et al. (2017), which documents the inventory data from operating CSA cement plants in China for both the conventional CSA clinker and waste-derived CSA clinker. The latter case fits our study regarding energy consumption but is adapted to tailings as the raw material instead of slag and bauxite residues (Figure B5).

F. Ceramic production

Aluminosilicate materials are well-known as suitable inputs for building ceramics, as demonstrated by Veiga Simão et al. (2021). Their work incorporated up to 10%-wt of tailings as alternative raw materials for substituting virgin material inputs in roof tiles. 0.5 wt% of barium carbonate was added to the roof tile mixture to fix the soluble sulfates, thus preventing drying efflorescence. Upon this basis, the amount of tailings in the ceramic mixtures of industrial production is linearly upscaled from the lab experiments. For other life cycle inventory data, mainly related to energy consumption at the plant, we use the European ceramic plant's performance available in Ecoinvent (Ibáñez-Forés et al., 2011). Aside from auxiliary data, adjustments to the proportion of tailings that replace virgin materials reflect changes in the upscaled ceramic production model (Figure B6).

G. Geopolymer production

Another use of aluminosilicate materials is the production of geopolymer (Hassan et al., 2019). This product can be a promising candidate to substitute Portland cement to make concrete or mortar, avoiding the need for high-temperature calcination. Based on the work of Niu et al. (2021), tailings-derived geopolymer can be made with suitable alkali activator formulations and reaction settings. We use sodium silicate and sodium hydroxide to mechanochemically activate tailings before the polymer-like structure can form. In our model, the consumption of alkali activators is linearly upscaled from the lab-scale experiments. A crucial step in the modeling is to simulate the manufacturing processes that reflect industrial-scale production with a relevant set of equipment. A recent study by Niu et al. (2021) provide an elaborate approach to upscale the production of geopolymer using a process simulator and generate life cycle inventory data. We specifically adopt an optimized geopolymer making simulation into the current LCA model. The described process is shown in Figure B7.

3.2.3 Sensitivity analysis

We perform two sensitivity analyses that are related to the process variables, explained in the following:

1) *Process parameters in the scale-up activity*. We create two spectrums representing best and worst cases to explore the influence of changing key parameters. In general, resource consumption for new processes is tested with (+50%, -25%) deviation from the base

values, while mature processes such as ceramic and CSA cement plants have (+25%, -10%) differences. The amounts of recovered metals are varied by defining the overall assumed maximum (95%) and worst (50%) recovery efficiencies. Solvent recycling in the worst cases is reduced to 50% capacity compared to the base case. Lastly, the particulate matter capture systems upgrade can decrease 90% of the emitted particles in the best case. Table B28 lists all altered variables, explanations of assumptions, and references for data in the sensitivity analysis.

2) *Transport distance*. No transport distance is considered in the base case, assuming all processes take place in the same location. We added lorry (i.e., truck) road transport as additional variables to investigate the effect of delivering raw waste to metal extraction and valorization plants. Short (50 km) and long (300 km) transportation distances are defined based on the manufacturing locations in Portugal (Figure B12).

Additionally, two other sensitivity analyses are conducted, mainly associated with the changes in the background electricity mix and secondary material substitution. We evaluated the implications of changing those factors both, one at a time and simultaneously.

1) *Energy transition*. According to the EU decarbonization plan (European Commission, 2020), renewables' shares will increase over time, with progress depending on the country's efforts. Herein, the Portugal 2030 national energy and climate plan (Environment Portugal, 2019) reflects the changes of the future energy mix. The country will shift from fossil-based power generation (coal 26%, natural gas 24%) to renewables (total contribution of 80%). Details of the energy mix are shown in Table B30.

2) *Substitution ratio for material credits*. To avoid overcounting benefits from the avoided virgin materials in the base case (1:1), we adjusted the substitution ratio of secondary materials. Following conservative approaches, secondary materials might have impurities or lower technical performances, which decrease the substitutability of the displaced products. Value-corrected substitution ratios for secondary metals were taken from aluminum (Koffler & Florin, 2013) as a proxy for copper and zinc, while the values for secondary building materials are taken from other waste-derived material studies (Hassan et al., 2019; Rigamonti et al., 2020) (ratios reported in Table B29).

3.3 Results and discussion

3.3.1 Environmental impact of mine tailings reprocessing

A comparison of life cycle environmental impacts for all reprocessing routes is presented in Figure 3.2. In most cases, reprocessing of tailings can result in net negative impacts, but there are also situations where the environmental burdens of reprocessing are more significant than material credits. Residue valorizations for all routes, particularly involving ceramic production, dominate the shares of environmental impacts in almost all categories. This is mainly because of the volumes: 3 – 4 tonne ceramics are produced for every tonne of tailings input, compared to 1.56 tonne of CSA cement and a 0.6 – 0.7 tonne of geopolymer. With relatively better environmental performances than their primary counterparts, the valorized building materials yield benefits that outweigh the resources added to reprocessing in most cases.

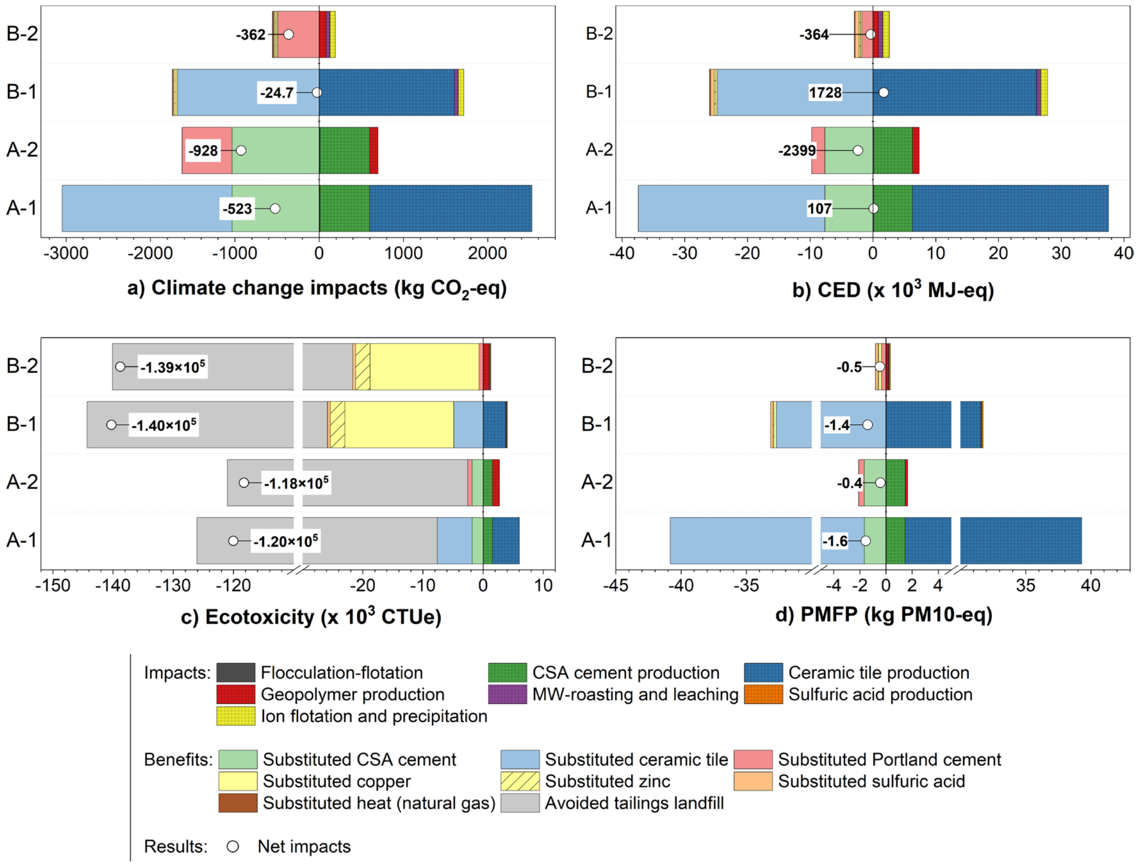


Figure 3.2: The environmental impacts for treating 1 tonne of sulfidic tailings in each evaluated route, including impacts from the reprocessing steps and the impact credits (negative impacts are equivalent to environmental benefits) from displaced primary materials and avoided landfilling. CED = cumulative energy demand, PMFP = particulate matter formation potential.

Also, the production of secondary metals through leaching and ion flotation (route B-1 and B-2) gives apparent benefits for the toxicity impact category, despite small production quantities for zinc and copper in the overall system. Approximately 3 kg of copper and 7.5 kg of zinc are recovered, avoiding primary copper and zinc ore processing and subsequent tailings disposal. With either scheme (route B), heavy metal emissions from tailings that often cause groundwater contamination can be prevented.

However, the magnitude of benefits should be evaluated correctly for both building materials and secondary metals. They are dependent on the quality of secondary displacing products and the choice of the product replaced in the market. Tailings-derived building products may contain impurities that can lower their ability to substitute primary products or even prevent their application, despite better mechanical performances (Kinnunen et al., 2018; Mabroum et al., 2020). Another aspect is related to base metal production. We use Ecoinvent global market data to represent copper and zinc production in this study. In reality, the emissions from regionalized upstream production vary substantially from one site to another (Adrianto et al., 2022).

To analyze hotspots in every reprocessing step, we present the percentages of contribution in Table 3.2. Electricity and fossil fuel consumption appear in most reprocessing chains, signifying its importance in the foreground system with varying degrees of concern (from below 10% to above 90%). Apart from energy-related contributors, the preparation of chemicals such as solvent mixtures for leaching, surfactants for ion flotation, and alkali activators for geopolymer manufacturing also gives a fair share of impacts due to their energy-intensive production.

Table 3.2: The hotspots in the alternative treatments of mine tailings (individual or combined share above 80%). CC = climate change impacts, CED = cumulative energy demand, FE = USETox freshwater ecotoxicity, PMFP = particulate matter formation potential, SS = Recipe (H, A) Aggregated Single Score. Yellow and red shaded cells indicate values between 40% - 70% and higher than 70%, respectively.

Route	Reprocessing steps	Hotspots	% contribution				
			CC	CED	FE	PMFP	SS
A	Flocculation-flotation	Electricity use for dewatering and flotation	**96%**	**95%**	**89%**	**92%**	**94%**
B	MW-roasting and leaching	Ammoniacal solutions as lixiviants	35%	35%	**53%**	25%	35%
		Electricity use for roasting and leaching	33%	28%	35%	**59%**	33%
		Thermal energy use for drying and heating	27%	32%	3%	7%	27%
B	Ion flotation	Energy use for solvent recovery	**81%**	**78%**	**40%**	**69%**	**76%**
		Electricity use for ion flotation unit	9%	8%	22%	17%	9%
		Consumption of surfactant, conditioning agents	7%	12%	27%	12%	13%
A	CSA cement	Direct emissions from calcination process	**73%**	0%	1%	**41%**	**40%**
		Consumption of clinker fuels in cement kiln	10%	**78%**	**65%**	10%	35%
		Cement ingredients (limestone, bauxite, gypsum)	4%	6%	9%	22%	9%
A2, B2	Geopolymer	Preparation of alkali activating solutions	**84%**	**80%**	**94%**	**86%**	**84%**
A1, B1	Ceramic roof tile	Thermal energy use at the ceramic plant	**60%**	**67%**	10%	1%	30%
		Electricity consumption at ceramic plant	33%	27%	**61%**	3%	17%
		Particulates from drying and firing process	0%	0%	0%	**95%**	**48%**

The hotspot analysis suggests that bringing together energy efficiency measures and more sustainable chemical consumption can be guiding principles for process improvement potentials. From the operator's viewpoint, one can focus on increasing the plant energy efficiency and particulate matter abatement controls at ceramic sites. Waste heat or biomass as a heat source can be considered if the local supply of such fuels is abundant and affordable. The importance of low-impact alkalis (Adesanya et al., 2021) may also overtake sodium hydroxide and silicate's role as promising constituents for geopolymer production. Ultimately, these all together can be stretched even further when decarbonization is in place, albeit externally reliant. The implications of the above strategies will be exercised in the sensitivity analysis.

3.3.2 Breakeven analysis: secondary metal production pathway

In situations when there is no valorization due to nonexistent markets for selling building materials, our analysis shows that the recovery of metals alone cannot offset the burdens of tailings reprocessing for most impact categories (Figure 3.3). Reprocessing and recovering metals from the current grade of tailings (0.46%-wt copper, 0.92%-wt zinc), according to climate change (CC) impacts, cumulative energy demand (CED), and fossil depletion potential (FDP), intensify overall impacts, with values 2 – 4 times larger than primary metal production. Despite these setbacks, other indicators like particulate matter formation potential (PMFP), metal depletion potential (MDP), and toxicity-related methods exhibit impacts > 90% lower than their primary routes. Notably, the sole metal recoveries are acceptable for all impact categories only when the tailings' metal concentration is high enough. We find that higher quality tailings with a copper grade of 1.2 – 1.8%-wt are essential to reach the breakeven point of this reprocessing scheme in route B without credits from valorized mineral fractions. Despite its high energy demand, route B through controlled roasting and leaching offers a side benefit of turning original tailings into a more stable residue (Kamariah et al., 2022): another opportunity for mitigating potential environmental impacts associated with sulfidic tailings.

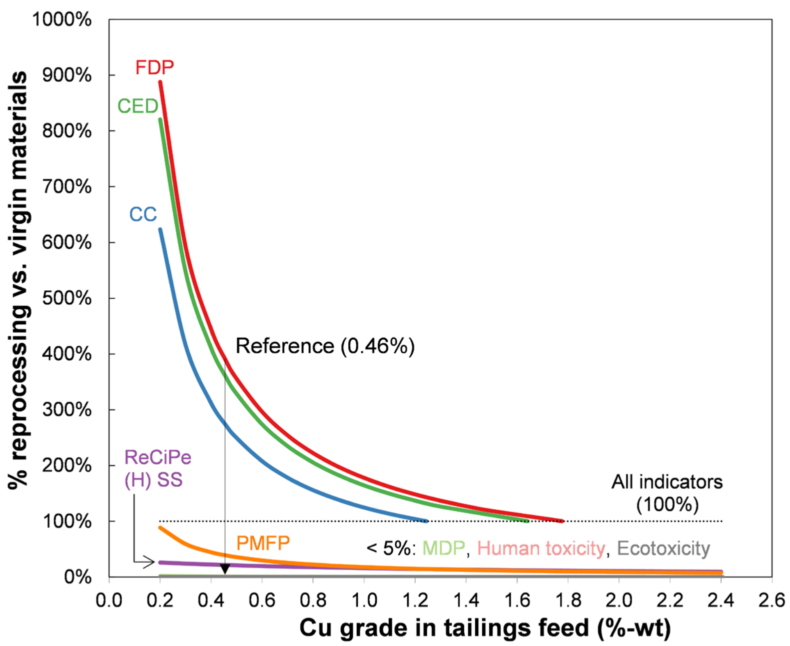


Figure 3.3: Cu in tailings vs. ratio of reprocessing to metal-only credits (route B without valorization). MDP, human toxicity, and freshwater ecotoxicity are hardly visible due to low values below 5%. FDP = fossil depletion potential, MDP = metal depletion potential, ReCiPe (H) SS = single score using ReCiPe method at endpoint level.

Theoretically speaking, on the one side, this proposal might be preferable for ancient sites where copper concentration in tailings can reach as high as 1%-wt (Nash, 2003). On the other side, it may be challenging for some other tailings where the copper grades are already lower than 0.3%-wt due to technological upgrades or different ore characteristics. According to historical assessments of Chilean porphyry copper tailings (Alcalde et al., 2018), the current production facility has better beneficiation performances than decades ago, with the average tailings copper grades detected around 0.1%-wt. In such cases, mineral valorization pathways will be needed.

3.3.3 Model sensitivity

Scale-up and process parameters. The results of varying process parameter values in life cycle inventory modeling are presented in Figure 3.4. While most of the four reprocessing routes are below virgin production impacts (28 out of 32 indicators) according to base cases, they perform even better than the base cases for all indicators when we applied best case assumptions. Complementary to Figure 3.3, applying best case assumptions would reduce impacts notably for CC, CED, and FDP categories, but metal-only recovery still cannot help reach breakeven lines for low-grade tailings (Figure B10). Overall, the sensitivity during scale-up activity causes a range of implications to different routes, inducing changes from 1% to 80% impact additions or reductions.

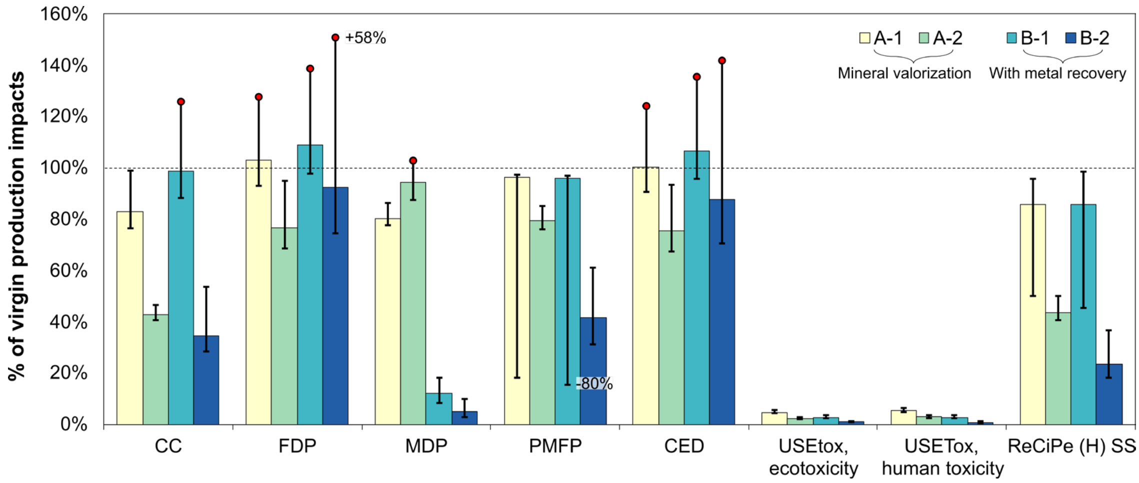


Figure 3.4: Comparison of reprocessing impacts of all routes to virgin material impacts for corresponding cases. The whiskers indicate best cases (lower range) and worst cases (higher range). For worst cases, red dots indicate values that exceed the 100% threshold.

Routes A-1 and B-1 with the goal to maximize ceramic products reveal high values, surpassing the impact of corresponding virgin for CC, FDP, and CED because of thermal energy and electricity consumption at the manufacturing plant. Next to that factor, particulate emissions are another major cause for PMFP and ultimately ReCiPe (H) endpoint indicators. With proper dust and particulate abatement devices, results indicate that reduction potentials are applicable for these specific routes, combined with other strategies described in the hotspot analysis.

Meanwhile, routes B-1 and B-2 with metals recovery show moderate gains to MDP, human toxicity, and freshwater ecotoxicity indicators—having less than 30% impacts of virgin production even in the worst-case perspective. For both routes, results of several impact categories are highly variable, indicating sensitivity of the result to many novel technologies used in the inventory models. Route B-2, for instance, has the entire portfolio of emerging processes and products, namely MW-roasting, leaching, ion flotation, and geopolymer processes. When applied at a large scale, processes with non-standard equipment such as microwave roasting furnaces and geopolymer plants would entail high uncertainties in the assumptions. Although it appears as the route with the lowest impacts, one should treat the findings with care due to the novelty of the process chain B-2.

Transport distance. When it comes to sensitivity with the transport distances, delivery by truck inflicts a low to moderate increase to overall impacts. Here, the causes are mainly the bulk transfer of raw tailings to a third party for reprocessing, namely the fresh feedstock and cleaned residues to its respective manufacturing plants. Adding 50 km transport by truck contributes to less than 2% of overall impacts. However, for longer distance travels (300 km), specific indicators such as CED, CC, and toxicity-related categories begin showing a 4 – 5 % rise, as shown in Figure B13.

Energy transition. Decarbonized electricity supply improves life cycle performances for all routes, as depicted in Figure 3.5. An 11% decrease from the base case is expected for the 2030 electricity mix (blue crosses), notably for impacts associated with energy-consuming processes in the foreground system. Nevertheless, a trade-off exists shall this transition occur. MDP and ecotoxicity indicators show opposite trends, mainly due to extra metal requirements for low-carbon power generation, i.e., solar PV and wind (Kleijn et al., 2011), which lead to background emissions from associated metal mining. This side effect is even more pronounced in a hypothetical 100% solar PV scenario for the electricity of the reprocessing facility (Figure B14), demonstrating the importance of responsible sourcing of electricity supply.

Substitution ratio. Having secondary products of inferior quality reduces the substitution ratios (Table B29) and makes overall environmental performances worse than base cases (Figure 3.5, red squares), with an average increase of 14% across all indicators and routes. Consequently, lower secondary material credits – particularly those gained from displaced ceramic and cement – would render some processing routes unsustainable.

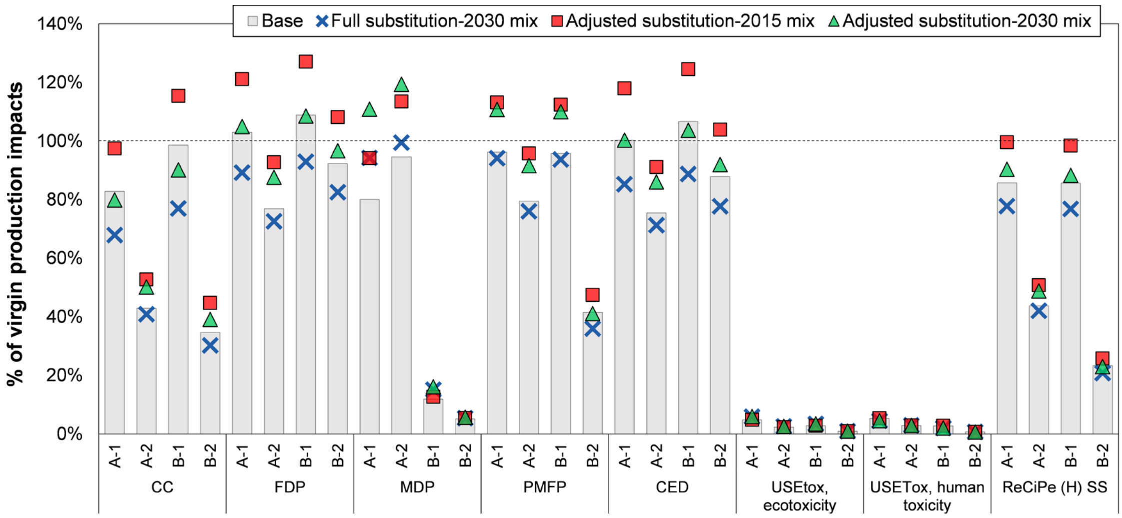


Figure 3.5: Relative changes of the environmental impacts if we modify the electricity mix (blue crosses), the substitution ratio (red squares), and both at the same time (green triangles).

When both factors are applied simultaneously, the results are generally situated between impacts induced by them, somewhat balancing the consequences (Figure 3.5, green triangles). Nevertheless, this combined effect can also lead to higher impacts than the individual effects, such as for A-1 and A-2 concerning MDP and toxicity indicators.

3.3.4 Implications for technology designers and policymakers

The results indicate that tailings reprocessing have varying potential to improve overall sustainability. The assessments are meant to provide first-hand calculations for the early-stage technological innovations and enlighten the contributions of these new processes toward sustainable mining. We could derive the following possible implications to technology direction and policy development:

Hotspot analysis highlights research and development needs. By knowing the important drivers of environmental impacts, we can identify the weak parts of the reprocessing chain and propose guiding principles for improving overall sustainability performances.

Build (pilot) facilities near or at the waste sites. Aside from exclusions of transport cost and impacts, this decision would foster synergies among entities, knowledge transfer, and better control of material exchanges at the future industrial symbiosis. Moreover, the established innovation cluster could accelerate scientific research in these areas, which may potentially generate economic revenues during operation or even after primary mining ceases production.

Signal for technological innovations and waste-derived product legislations. Small to medium demo / pilot enterprises can show evidence for real-life implementations early, where other jurisdictions may mirror such a success story. However, central to harnessing benefits are legal standards for products made from waste. As illustrations, introducing quality certifications and minimum shares of recycled materials in public procurement can ensure the viability of mass production and incentivize such plans in the EU and beyond.

3.3.5 Limitations and future research

This study has limitations that may provide starting points for future research. Some technologies are still at nascent stages, implying a probability of modifications in the long run. Due to a lack of detailed prognosis, these changes and scale differences were partially neglected in the early assessment. Additionally, more advanced technologies may materialize to extract different minerals and metals, such as combined bio-brine leaching to co-produce lead and zinc (Ye et al., 2017) or biosorption techniques to glean rare earth elements (Jin et al., 2017). To understand the true impacts of value recovery pathways from tailings, follow-up investigations of new technologies and constantly monitoring their evolutions are imperative.

Analogous to previous insights, opportunities exist to replace virgin chemicals consumed in the assessed process with bio-based material substitutes. For instance, xanthate could be replaced with cellulosic nanofiber materials, an emerging nanocomposite with similar performances made from renewable sources (Sharma et al., 2019). Similarly, other emerging surfactants may substitute sodium dodecyl sulfate, halving its emissions compared to the standard ethylene oxide productions (Nogueira et al., 2019). The compatibility of these bio-based chemicals with the current setups needs to be reevaluated in future research.

The four routes were constructed based on simplified secondary product classifications and mass flow analysis. Mathematical optimization techniques could be developed for this study, constraining the resource consumption and, in parallel, maximizing the environmental performances with defined objectives (Vadenbo et al., 2014). The optimal solutions are probably new pathways with branches of more diverse products not yet captured in the reference routes of this study. For future study, one may also perform an optimized collection route and then treat the mine waste at designated locations so that financial cost, travel efficiency, and environmental factors can be integrated into the assessment.

As exercised in the sensitivity, the product quality for secondary materials was calculated with value-corrected substitution ratios. To be more precise and guarantee safe application of recycled building materials, one could extend the life cycle phases included in this study by conducting detailed analyses for these specific waste-derived products. We would anticipate successful stabilization of harmful substances in the finished products, as confirmed by other tailings-derived materials studies (Kiventerä et al., 2018, 2019; B. Li et al., 2020). Future work could track potential leaching emissions during the use and disposal phase of the secondary products to explicitly model the downstream impacts for avoiding burden shifts.

3.4 Conclusions

This research presented the results of applying life cycle thinking tools in an early-stage assessment of mine tailings reprocessing and valorization. The developed assessments provided a solid understanding of the contributions to the environmental profiles of various emerging techniques. There are expected impact reductions among conceptual reprocessing routes, but none of them could avoid burden shifts from a life cycle perspective. The prospective nature of the LCA was able to highlight critical aspects of the process chains that should be enhanced for achieving a more sustainable manufacturing route.

The results suggested that the mineral valorization steps are significant contributors in many impact categories (CC, CED, PMFP), while metal recoveries are particularly beneficial for metal depletion and toxicity-related indicators. The concentration of metals in tailings determines the magnitude of benefits one can get from metal recovery-only routes. In terms of sensitivity, waste transport to the processing site showed a small to moderate contribution to overall impacts, indicating the importance of on-site treatments. On the one hand, decarbonized electricity—powering the whole process—could reduce the impacts of alternative tailings treatment. However, on the other hand, secondary materials with lower quality than substituted virgin products, primarily building materials, may limit the environmental benefits initially gained. Future research could extend the scope of LCA by including the use and end-of-life stages to evaluate impacts more accurately from any downstream processes that wish to incorporate such tailings-derived materials. The early-stage life cycle evaluation can accelerate technological innovations in the mining industry to improve the sector's sustainability.

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Chapter 4

Toward sustainable reprocessing and valorization of sulfidic copper tailings: scenarios and prospective LCA

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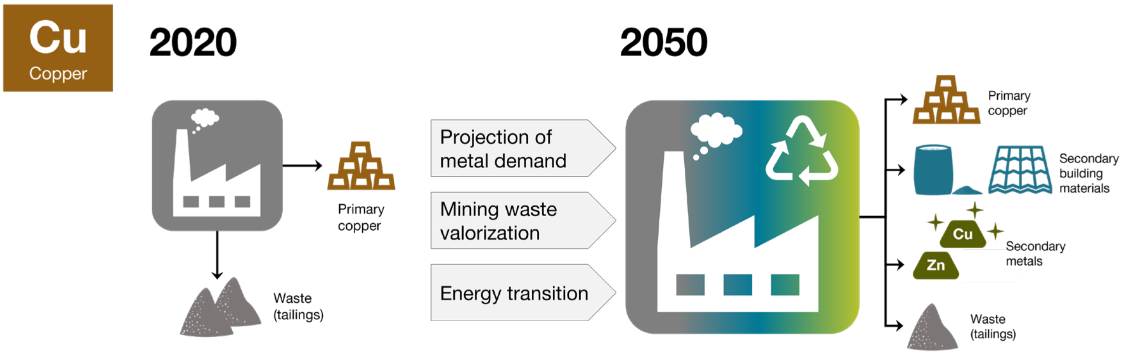
Chapter 4: Toward sustainable reprocessing and valorization of sulfidic copper tailings: scenarios and prospective LCA

Graphical abstract



Highlights:

- Environmental impacts of copper tailings reprocessing in the EU are quantified.
- Future scenario narratives are leveraged to create prospective life cycle assessment models.
- Copper tailings reprocessing can mitigate GHG emissions and toxicity impacts in 2050.
- Tailings reprocessing can supply up to 2% of future European copper demand.
- Tradeoffs exist between climate change and ecotoxicity impacts for different reprocessing.

Abstract

There has been increasing attention recently to reprocessing of mining waste, which aims to recover potentially valuable materials such as metals and other byproducts from untapped resources. Mining waste valorization may offer environmental advantages over traditional make-waste-dispose approaches. However, a quantitative environmental assessment for large-scale reprocessing, accounting for future trends and a broad set of environmental indicators, is still lacking. This article assesses the life cycle impacts and resource recovery potential associated with alternative waste management through mine tailings reprocessing at a regional scale. Sulfidic copper tailings in the EU were selected as a case study. We perform prospective life cycle assessments of future reprocessing scenarios by considering emerging resource recovery technologies, market supply & demand forecasts, and energy system changes. We find that some reprocessing and valorization technologies in future scenarios may have reduction potentials for multiple

impact indicators. However, results for indicators such as climate change and energy-related impacts suggest that specific scenarios perform sub-optimally due to energy / resource-intensive processes. The environmental performance of reprocessing of tailings is influenced by technology routes, secondary material market penetration, and choices of displaced products. The trade-off between climate change and energy related impacts, on the one hand, and toxicity impacts, on the other hand, requires critical appraisal by decision makers when promoting alternative tailings reprocessing. Implementing value recovery strategies for building material production, can save up to 3 Mt CO_2-eq in 2050 compared to business as usual, helping the copper sector mitigate climate impacts. Additional climate mitigation efforts in demand-side management are needed though to achieve the 1.5 °C climate target. This work provides a scientific basis for decision-making toward more sustainable reprocessing and valorization of sulfidic tailings.

Keywords: Mine waste, resource recovery, circular economy, life cycle assessment, scenario analysis

4.1 Introduction

The demand to solve waste accumulation problems and to supply resources sustainably have accelerated progress in emerging value recovery technologies (Rankin, 2017; Shaw et al., 2013). The mining sector is no exception. Among the most environmentally threatening waste problems is the disposal of mine tailings. When handled poorly, tailings can be the precursor of acid mine drainage, posing toxic contamination to the surroundings, even long after mines have ceased operations (Lottermoser, 2010). Currently, management options rely mostly on engineered storage through landfilling or backfilling (Kalisz et al., 2022). In the case of storage facilities, there are structural risks associated with long-term durability. Failures to manage such integrity-related risks may lead to dam collapses and environmental catastrophes (Schoenberger, 2016). Approximately 8 billion tonnes of tailings are generated annually, 46% of which comes from copper production, according to the latest estimates in the Global Tailings Review (Mudd & Boger, 2013; Oberle et al., 2020). These figures are supposed to grow as more minerals are consumed worldwide to support growth trends in emerging regions (Elshkaki et al., 2018; Herrington, 2021). Moreover, low-carbon power production such as solar, wind, and tidal, requires metals – a large fraction of which is fulfilled with primary mining (Lee et al., 2020; Valero et al., 2018; Vidal et al., 2013). Consequently, safe and sustainable solutions must be found for large quantities of mine tailings.

Many researchers and practitioners have been looking for improved management options with better environmental, social, and economic outcomes. With the advantages of gaining access to secondary materials and reducing waste volume, Edraki et al. (2014) and Whitworth et al. (2022) highlight value-adding opportunities in tailings reprocessing to recover metals and minerals. According to Spooren et al. (2020), extractive waste residues, such as tailings, may contain metal concentrations that can be higher than what can be found in the range of current economic ore grades of primary ores. Recent advancements in pyro-, hydro-, bio-, and solvo-metallurgical processing for metal extraction / recovery may capitalize on these undervalued stocks and make mine waste a resource. In addition to stranded valuable metals, the leftover residues can also be processed through valorization steps. Such steps add value by transforming residues into industrial materials, avoiding landfilling (Binnemans et al., 2015). In recent years, many studies have demonstrated viable production of alternative cement and ceramics derived from tailings (Ahmed et al., 2021; Martins et al., 2021; Niu et al., 2020; Pyo et al., 2018; Veiga Simão et al., 2021). Through valorization, tailings can also be used as raw materials in the secondary production of alkali-activated polymers: low-carbon substitutes for today's emission-intensive products such as ordinary Portland cement (Bernal et al., 2016; Mabroum et al., 2020). These opportunities generate growing interest among stakeholders and manufacturers to identify technically promising resource-recovery technologies with market and sustainability potential.

In the EU, recent years have witnessed a surge in innovations and research developments that aim to secure metals with high economic importance and avoid supply disruptions (Løvik et al., 2018). Policymakers have increasingly linked the contribution of emerging mine waste management technologies to overarching initiatives such as the European Green Deal (European Commission, 2019) and the Circular Economy Action Plan (European Commission, 2020). To translate plans into tangible findings for policy support, Blengini et al. (2019) provide various estimates of the potential recovery of several minerals compared to the current demand. Based on their simplified analysis, the authors concluded that the co-production of low-volume materials of high values and high-volume bulk minerals must be performed together to make the process environmentally viable and resource efficient. This is especially the case when specific metals are found at low concentrations in the mining waste heaps or landfills. In the EU, an innovative and integrated resource recovery research project SULTAN (https://etn-sultan.eu/) investigated the valorization of sulfidic mine waste from primary mining activities. SULTAN's core technologies include metal extraction / recovery via, e.g., microwave / chemical assisted leaching and mineral residue valorization, aiming to convert waste into various industrial materials and create environmental benefits. While the idea seems initially favorable, collecting waste materials and processing them to useful products require energy inputs and resources. This may lead to unintended consequences and failures to reduce the net environmental impacts. Therefore, the environmental benefits and impacts need to be assessed.

Life cycle assessment (LCA) is a standardized method to assess the environmental impact throughout the life cycle stages of a product / service, including raw material extraction to the disposal process (ISO, 2006). Known for its ability to identify environmental hotspots, LCA is also increasingly applied in the minerals industry (Segura-Salazar et al., 2019). LCA studies of mine tailings treatment generally find that waste reprocessing and valorization strategies tend to reduce environmental impacts in comparison to conventional tailings management, but not always (Adiansyah et al., 2017; Adrianto & Pfister, 2022; Grzesik et al., 2019; Song et al., 2017; Vargas et al., 2020). Variability in feedstock characteristics, treatment pathways, and potential secondary products will determine the net environmental performance as well as technical and economic applicability of these reprocessing and valorization options (Beylot et al., 2022). Some studies incorporate scenario modeling to build forward-looking analysis or prospective LCA. Those studies have analyzed that parameters like metal supply, technology efficiency, production routes, and background energy system may significantly influence the resulting environmental impacts (Ciacci et al., 2020; Elshkaki et al., 2018; Harpprecht et al., 2021; Kuipers et al., 2018; Rötzer & Schmidt, 2020; Van der Voet et al., 2019). No analysis has so far evaluated large-scale reprocessing of tailings through prospective LCA, accounting for the combined effects of various future scenarios.

This study aims to quantify the environmental benefits, impacts, and tradeoffs of large-scale deployments of copper tailings reprocessing and mineral valorization technologies in the EU. The prospective nature of this assessment requires scenario modeling. To assess secondary production potential in future scenarios, we estimate the available volume of secondary products and compare them with the primary demand in 2050 based on market forecasts. The anticipated environmental footprints are assessed for a multitude of indicators to detect potential environmental burden shifting. Environmental performances for different scenarios are explored by incorporating projections in the energy transition, technological improvements for the primary copper sector, and resource-recovery technologies for copper tailings.

4.2 Methodology

In this study, we develop a framework to quantify the environmental performance of tailings reprocessing and the potential replacement from the recovered products. Figure 4.1 gives an overview of framework elements. This covers several steps, which are explained in the following sections: (4.2.1) goal and scope, (4.2.2) scenario development, (4.2.3) modeling approach and data, (4.2.4) background inventories, (4.2.5) assessment of environmental benefits and impacts of the investigated scenarios, and (4.2.6) sensitivity analysis.

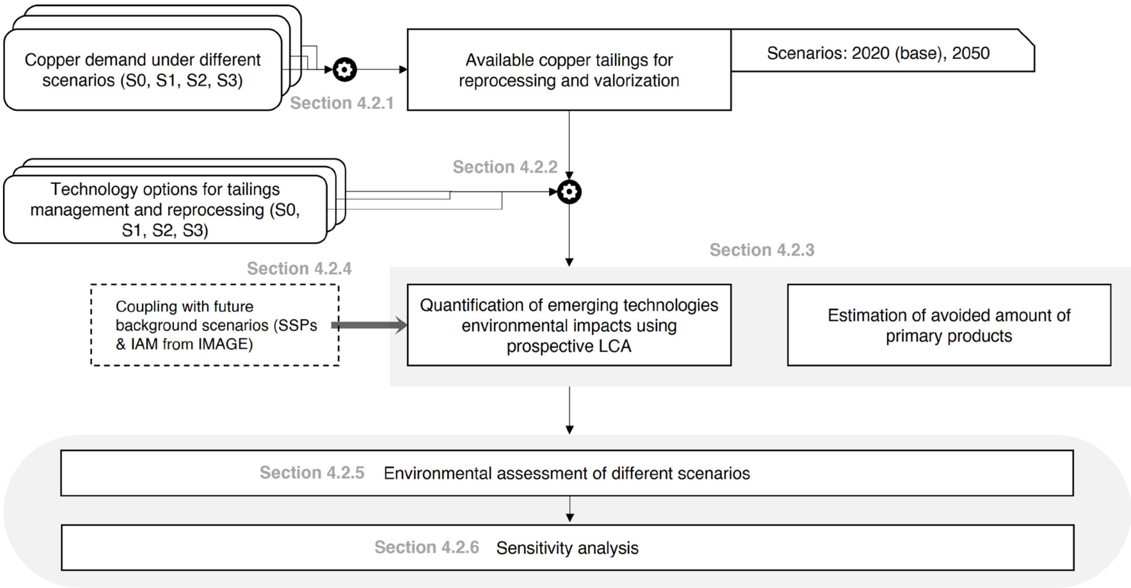


Figure 4.1: Workflow of the study. SSP: shared socioeconomic pathways, IAM: integrated assessment model.

4.2.1 Life cycle assessment: goal and scope

The goal of this study is (1) to evaluate the environmental benefits and tradeoffs between the secondary resources potential and energy / materials needed to perform the resource-recovery systems and (2) to estimate the large-scale impacts of copper tailings reprocessing in the EU. System-wide environmental analyses are performed to simulate the environmental implications of recycling / reprocessing sulfidic copper tailings. The zero-burden assumption is applied, i.e., the environmental burdens of copper tailings generation are excluded (Ekvall et al., 2007). The functional unit (FU) of this study is defined as "the treatment and management of sulfidic copper tailings arising in the EU in the year 2020 / 2050". The system expansion approach is applied to assign the credits for the avoided primary productions. The substitution effects of secondary products from these alternative processes are considered in the modeling, potentially substituting the primary production of materials (Ekvall, 2020; Schrijvers et al., 2020). Specifically for offsetting products / services, a systematic selection procedure is applied based on current and future production trends (section 4.2.3D). In addition, the nature of this study involves prospective elements such as emerging recovery technologies and future energy scenarios, which encompasses changes in foreground and background systems.

4.2.2 Scenario development

Initially, a baseline scenario in 2020 is developed based on historical production data of copper in the EU from a combination of sources: statistics from international copper study group and commodity market intelligence platform (ICSG, 2021; S&P, 2020). Whenever available, site-specific data (i.e., volume and feedstock characteristics) for each mine site and the country is retrieved from the global sulfidic copper tailings assessment of Adrianto et al. (2022).

Future copper needs and hence, mining activities will determine the future availability of copper tailings and reprocessing potential. Three scenarios for 2050 are explored based on projected, prospective dynamic material flow analysis linked with resource scenarios of the previous studies by Ciacci et al. (2020) and Elshkaki et al. (2018). These are then coupled with the climate scenarios and future projections taken from the shared socioeconomic pathways (SSPs) with varying climate protection measures (Riahi et al., 2017).

The SSP2 "middle of the road" scenarios are selected in this study, which forecast developments similar to current trends without considerable changes in the development trajectories (O'Neill et al., 2017; van Vuuren et al., 2017). In addition to the baseline SSP2 scenario, restrictive climate policy scenarios are combined with the representative concentration pathways (RCPs) to reach stringent radiative forcing targets (Fricko et al., 2017). Projection of energy use / supply inventories and socio-economic information in the SSP2 scenarios are derived from the widely used integrated assessment models (IAMs) IMAGE (Stehfest et al., 2014). All of the SSP2 scenarios in this study assume climate mitigation in the background energy systems leading to a radiative forcing of 1.9 W / m^2 in 2100, which corresponds to 1.5°C maximum global temperature increase in 2100 relative to pre-industrial levels. For scenario 1, only conventional tailings management is applied, in line with the business-as-usual scenario. Scenario 2 relies on resource-recovery technologies with higher maturity levels and less product novelty / complexity than scenario 3, i.e., the production of industrial waste-based ceramics in scenario 2 (see section 4.2.3C for detailed comparison). These two scenarios are specifically designed to model technological innovations already described in the previous study (Adrianto & Pfister, 2022). The linking of scenarios and reconciliations of narratives result in three future scenarios, as summarized in Table 4.1.

Table 4.1: Scenario definitions.

	Tailings management options – Metal demand scenarios	**Background energy systems and equivalent SSP-RCP narratives***
Baseline scenario	S0: Business as usual (BAU) route	Current energy systems
Future scenarios	S1: BAU route – Toward equitability 2050	Climate mitigation (1.5°C scenario), in line with SSP2-RCP 1.9 W / m^2
	S2: Mineral valorization route – Toward equitability 2050	
	S3: Metal and mineral recovery route – Toward equitability 2050	

Note: *Scenarios are chosen to be as consistent as possible among each other, following the IPCC special report guidelines (IPCC, 2018). Metal demand scenarios were taken from the study of Ciacci et al. (2020).

The storylines developed for each management scenario are explained as follows:

- Business-as-usual scenario in 2020 and scenario 1 in 2050

  Copper tailings are either stored in the dam and / or backfilled. The volume of backfilled materials depends on the mine site's configuration and site information (section 4.2.3B). Backfills also require additional materials and energy consumption, such as cement binder, slags, diesel, and electricity in the operational phase. In the year 2050, it is assumed that all land mining operations will install backfilling operations to manage their tailings as one of the current best practice approaches.

- Mineral valorization route, scenario 2 in 2050

  Technology improvement and successful commercialization allow building materials such as ceramics and alternative cement to be partly produced through tailings valorization. By 2050, there will be a trend toward cleaner energy mixes with less fossil resource dependence. Industry and consumers steadily accept tailings-based products in standard applications, which help substitute primary products.

- Metal and mineral recovery route, scenario 3 in 2050

  Further technology efficiency improvements and renewable energy systems are anticipated in this scenario. A notable advancement in the recycling technologies has enabled high purity metal recycling to be feasible. Emerging products such as alkali-activated binders (i.e., geopolymer as binder alternative to ordinary Portland cement) are assumed to enter the market. There is also a possibility to generate additional byproducts, such as sulfuric acid, thanks to the downstream processing of SO_2 gases.

4.2.3 Modeling approach and data

A. Demand projection and prospective tailings flows

Ciacci et al. (2020) estimated the potential demands for copper in the EU in 2050 using scenario analysis. These include demands for standard applications, i.e., construction, infrastructure, industry, transport & mobility, and consumer goods. To estimate total demands, copper demands for standard applications are added together with the transition demand of 1.5 Mt / year for clean energy technologies (section C.1.1 of the Appendix). Despite this additional increase, Europe’s copper mine production is expected to stay at the current level of 0.8 Mt / year, according to the metal outlook report (Gregoir & Van Acker, 2022). This domestic copper supply is used to estimate the potential volume of copper tailings. To account for copper grade declines, it is assumed that the degradation of copper ore grades follows the power regression relationship according to Crowson (2012). Copper tailings are produced from different mines, and thus it is important to fully characterize the quality and quantity of copper tailings at each site.

This was performed by considering site-specific data of the generated copper tailings in the baseline / future scenarios using market data from the S&P market intelligence platform (S&P, 2020) and regionalized environmental assessment of sulfidic copper tailings (Adrianto et al., 2022). Therefore, this study only focuses on tailings assessment for active copper sites, as the site-specific tailings data from abandoned mines or closed operations are not completely available.

B. Existing copper tailings management life cycle inventory

The following section concerns the BAU and future scenario 1, as defined in section 4.2.2. Tailings management in Europe mainly involves two options: 1) tailings disposal / landfilling in the storage facility and 2) backfill for underground operation support (JRC, 2018). The share of landfilling to backfilling is dependent upon site configuration. This ratio for landfilling and backfilling at each site is reported in the EU best available technologies document for tailings and waste rock management. The backfilling share is approximately 10% of total tailings in 2020 (European Commission, 2009). For the year 2050, it is assumed that a higher ratio of 30% for backfilling will be applied (Garbarino et al., 2020).

For the first method via landfilling, tailings may contain heavy metals and interact with the environment, which may generate long-term emissions to the freshwater bodies. Landfilling of copper tailings is modeled using the site-specific end-of-life inventories from the study of Adrianto et al. (2022). Meanwhile, the backfilling operation datasets are derived from the primary LCA data of the actual backfill plants (Reid et al., 2009). The latter is assumed to represent copper tailings' backfilling plant unit processes. However, the resource consumption (i.e., cement, diesel, quicklime, etc.) and emissions during operation from the original study are adjusted to the capacity of copper sites under the current research. Cement stabilization of the backfilled residues was assumed to prevent any leaching emissions.

C. Emerging copper tailings valorization life cycle inventory

For the two future scenarios (scenarios 2 and 3), it is assumed that tailings management options are a function of combined technologies in the reprocessing routes. Figure 4.2 shows the developed process flowsheet for large-scale resource recovery efforts for copper tailings.

We employ prospective LCA for foreground and background systems (Arvidsson et al., 2018). Adrianto et al. (2022) modeled large-scale production of emerging resource recovery systems for copper tailings in foreground systems. They provided life cycle inventories based on suitable technology upscaling methods for respective technologies (section C.1.2 of the Appendix). The background systems, such as future energy (i.e., power generation and heat) mixes, are based on the IAM IMAGE SSP2-RCP 1.9, which forecasts energy scenarios up to 2050, aligning with the SSP narratives (van Vuuren et al., 2012). The datasets for other materials and background datasets pertinent to the system in this analysis are explained in the following sections.

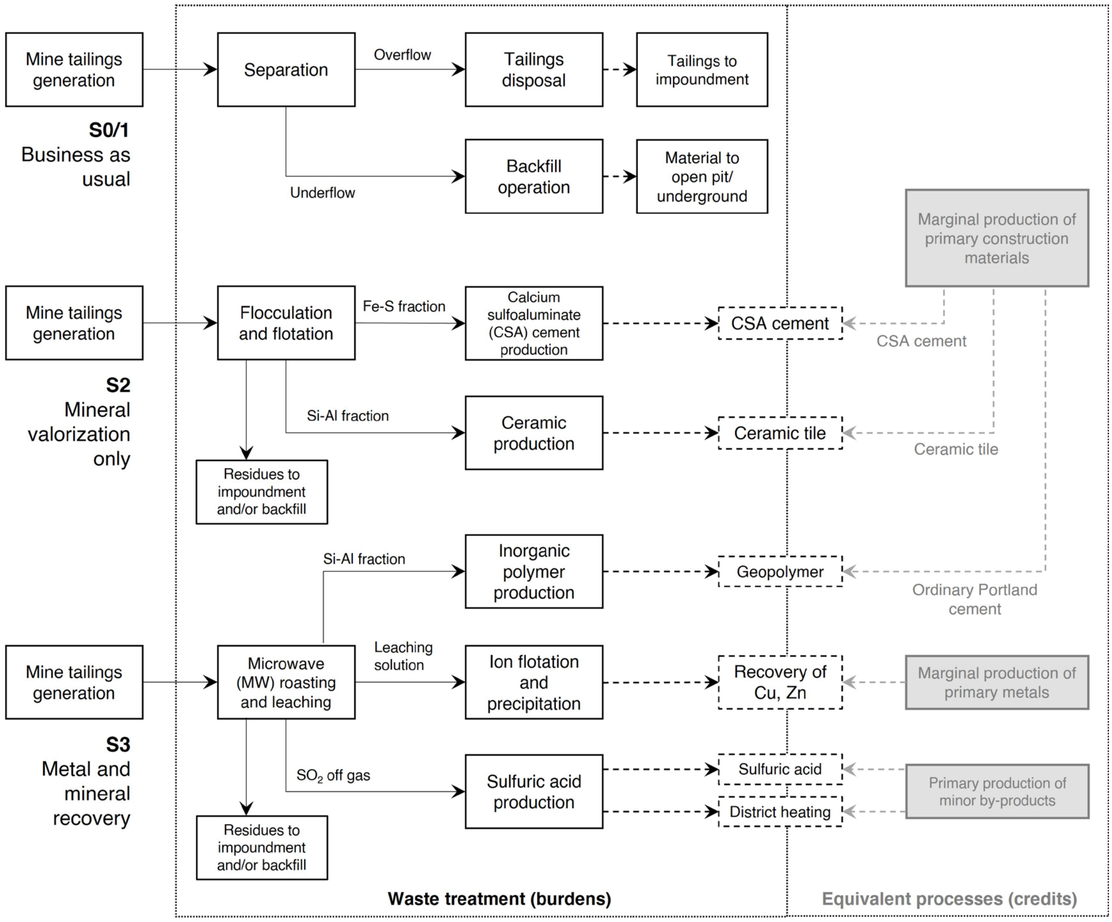


Figure 4.2: Management options for copper tailings applying standard disposal practices S0 / S1 (Years 2020 and 2050) and two alternative resource recovery scenarios, S2 and S3 (Year 2050).

D. Marginal technologies for substituted products

As mentioned previously, this work applies a system expansion or substitution approach. Consequently, selecting the appropriate displaced products / processes is a key part of LCA studies (Vadenbo et al., 2017). We follow the identification approach of marginal data developed by Weidema et. al (2004; 2009) for determining affected market processes. The approach has the advantage of determining possible marginal production without economic models and price information. Here, the long-term physical changes in supply, i.e., production quantities and growth trends of materials in different regions were taken into account (see section C.5.1 in Appendix). There are two sub-scenarios in the environmental assessment of this study. For S0 and S1, no substitution approach is applied since the systems do not produce substituting secondary products.

Meanwhile, for the year 2050 (S2 and S3), capital investment and technological breakthroughs may play roles and are considered to reflect progress for both existing and new technologies. We made performance estimations based on forecast and material outlook for specific products, considering future-oriented environmental assessments of the construction materials (Alig et al., 2021). In the base cases, it is assumed that all secondary production routes are based in Europe, i.e., secondary production replaces primary European production (Table C9). The assumptions made and details for the marginal production technologies (referring to sensitivity in section 4.2.6) for each relevant process are the following:

- **Calcium sulfoaluminate (CSA) cement**. CSA cement is commercially produced for many applications where high early strength and rapid setting developments are necessary, such as patching roadways, bridge decks, airport runways, tunneling, and others. EU cement research statistics reported that small quantities are made in Europe, which can be applied according to technical approvals (ECRA and CSI, 2017). It is assumed that in 2020, 0.1% of the traditional cement market will be taken by CSA cement, and this number will grow to 15% in 2050. These values follow market penetration rates for alternative cement from holistic cement review studies (Favier et al., 2018; Habert et al., 2020).
- **Ceramic**. Most European ceramics are produced domestically in Italy, Germany, and Spain (Cerame-Unie, 2021). These internal ceramic producers are identified as the marginal production process. It is assumed that theoretical efficiency upgrades will materialize in the future, as described in the best available technology document (European Comission, 2007; Ros-Dosdá et al., 2018). Besides that, aggressive emission reduction strategies for the year 2050 are also taken from the EU ceramic association roadmaps (Cerame-Unie, 2021).
- **Ordinary Portland cement**. We rely on IEA cement technology roadmaps to define future cement production's environmental performance (IEA, 2018). If not stated in the roadmaps, technological upgrades are taken from the best available technology document (JRC, 2013) and European efficient cement manufacturing (Croezen & Korteland, 2010). Monoethanolamine (MEA) based CO_2 capture technologies with 90% absorption efficiency are considered in future cement production routes. We assume this technology is the marginal production for the European cement market in 2050, while those imported from major players in India and China are defined as alternative marginal suppliers in the sensitivity analysis.
- **Copper and zinc**. According to the IEA critical minerals special report (2021), refined copper would be globally sourced from a mix of countries. As alternative sourcing strategies, the EU imports copper mainly from Latin America, i.e., Chile and Peru (Gregoir & Van Acker, 2022). Copper production via pyrometallurgical smelting technologies remains the major production pathway worldwide. Aside from domestic production, copper produced via pyrometallurgical smelters from Chile and Peru is assumed to be the next marginal technology. For future production, energy savings potential was taken into account, assuming a reduction in electricity and fuel demand by 20% and 55%, respectively (Kuipers et al., 2018; Kulczycka et al., 2016). Zinc would

be produced from mines and refineries using electrometallurgical smelting technologies (Van Genderen et al., 2016). From a recent zinc commodity report (USGS, 2022), China would remain the largest producer and is hypothetically assumed to be the marginal supplier. For future zinc production, energy demand (i.e., electricity and natural gas) are reduced by 12% according to the optimized energy consumption capacity (Qi et al., 2017).

- **Sulfuric acid and heat**. Over the last decades, a steady increase in sulfuric acid use for phosphate and sulfate fertilizers has driven its global demand (King, Davenport, et al., 2013). Since the market is distributed widely across regions, sulfuric acid production from elemental sulfur burning and heat generation (natural gas) is assumed to occur in Europe. The parameters for future sulfuric acid plants are taken from the best available technology document (European Commission, 2007).

4.2.4 Environmental background inventories

To facilitate the creation of prospective life cycle inventories covering future background systems, the software 'premise' is used to integrate future scenarios (Sacchi et al., 2022). This generates a systematic, complete set of prospective LCA databases containing results from the IAM IMAGE for SSP2 RCP 1.9 scenarios. The background data related to energy and material consumption in LCA are taken from Ecoinvent 3.8 database (Ecoinvent, 2022), which comply with the material types and grades applied for the study context whenever possible.

4.2.5 Environmental impact modeling

All scenarios are evaluated by LCA using various environmental indicators: climate change (IPCC, 2014), USEtox toxicity-related impacts (Rosenbaum et al., 2008), cumulative energy demand (Frischknecht et al., 2015), abiotic depletion potential (van Oers et al., 2002), and ReCiPe 2016 endpoint categories (Huijbregts et al., 2017). This selection of impact indicators aims to capture the most relevant impact categories when dealing with waste management and metal / mineral processing and supports comparability with other LCA studies. The environmental impact assessment is performed using the Activity Browser software (Steubing et al., 2020).

4.2.6 Sensitivity analysis

Sensitivity analyses are performed to test the robustness of the results and the influence of modeling choices. First, the market penetration rates of secondary products are varied from the default case, resulting in two cases: high market penetration (HM case) and worst-case assumptions (Table C7 in Appendix). Second, the substitution ratio of secondary materials made from tailings relative to primary materials is varied from 0.5 to the assumed default ratio 1. Ratios of substitutability might change due to differences in technical performance, perceived functionality, and market response factors, according to Vadenbo et al. (2017). This includes the effect of impurities in the products that may prevent product acceptance in the market. Third, the identified marginal productions may influence the substitution benefits for each secondary product and thus ultimately change the net environmental impacts of tailings management scenarios. In the coming decades, market shifts are expected. They might deviate from the current predicted industry trends, i.e., declining material production in the domestic market while increasing dependence on global imports of finished goods or vice versa. These would lead to changes in marginal technologies for such products and thereby define corresponding marginal suppliers outside the EU (Table C9).

4.3 Results and discussion

4.3.1 Secondary production from the reprocessing of copper tailings

Table 4.2 depicts how much secondary material can be produced from tailings in the EU and the volume of materials that can substitute their primary counterparts. For construction materials (i.e., ceramic and cement) across all scenarios, around 10-15% of market penetration was assumed due to market demand / supply constraints. This substantially limits the maximum scale-up potential of tailings valorization in industrial products. These effects are pronounced for ordinary Portland cement products. For illustration, less than 5% of OPC market share is assumed to be substituted by tailings-based geopolymer in 2050.

Table 4.2: Secondary production potential vs. material demand in EU. Volume unit in million tonnes.

Scenario	Secondary Material	Maximum possible secondary supply	Primary material substituted	Total demand forecast in 2050	Adjusted secondary demand	Fraction of secondary material uptaken in the market	Data source (for demand)
2	Ceramic tile	539	Ceramic tile	72[a]	61[i]	11%	(Cerame-Unie, 2021; Ceramic World Web, 2021)
	CSA cement	127	CSA cement	25[b]	19[ii]	15%	(Habert et al., 2020; Kelly et al., 2018)
3	Geopolymer	64	OPC cement	167[c]	6[ii]	10%	(Cembureau, 2022; IEA, 2018)
	Copper	0.1	Primary copper	4.6	0.1	Could be 100%	(Gregoir & Van Acker, 2022)
	Zinc	0.08	Primary zinc	2.9	0.08	Could be 100%	(Gregoir & Van Acker, 2022)
	Sulfuric acid	12	Sulfuric acid	25[d]	12	Could be 100%	(ChemIntel360, 2022; King, Davenport, et al., 2013)

Note: a = annual growth rate of 4.1% from 2020 to 2050; b = CSA cement takes 15% of OPC demand share due to alumina availability; c = assumed stable consumption in Europe throughout the century; d = future demand is forecast through the current Europe consumption trajectory.
i = assumed to be 85% of the primary demand according to the green procurement projection (European Commission, 2016; Sapir et al., 2022); ii = market penetration and raw ingredient availability are taken from the study of Habert et al. (2020)

Secondary cement products will likely face production constraints due to the scarcity of raw ingredients (Habert et al., 2020; Scrivener et al., 2018). The limited availability of raw materials is widely recognized as the main hindrance to the rapid scale-up potentials of CSA cement (Gartner & Sui, 2018) and geopolymer (Provis, 2018). CSA cement production chain requires alumina sources such as bauxite, which competes directly with aluminum metal production. To overcome this issue, high alumina or clay substitutes suitable for CSA cement manufacturing are under investigation (Galluccio et al., 2019; Negrão et al., 2022). For a similar reason, the scale-up rates of geopolymer are also limited by the conventional alkali activators like sodium silicate in the value chain. Untapped resources of raw materials such as glass waste and red mud (Joyce et al., 2018; Mendes et al., 2021) can be exploited to produce geopolymers with similar mechanical strength to conventional ones. Therefore, large-scale production of these two types of cement depends on the availability of abundant, technically feasible, and cost-competitive alternative raw materials.

In contrast, market demand can absorb the entire volume of recovered metals in scenario 3, except for geopolymer. Increased reprocessing and recycling rates of copper tailings in the EU can mitigate dependence on imported materials or domestic virgin production and help retain the value of recovered materials within the regional economy (Figure 4.3). Recovering base metal from copper tailings could satisfy 2% and 3% copper and zinc total demand, equivalent to a 12% and 11% increase in domestic European copper and zinc production, respectively. Note that our study only considers on the residual minerals present in tailings produced by operational mines. The actual recovery and economic potential might be larger than estimated in this study, if copper tailings storage facilities from closed operations are included (Araya et al., 2021). The advent of novel technologies and a rising appetite for metals sourced within the EU might become a driver to develop advanced reprocessing projects for mine waste repositories (Lèbre et al., 2017; Suppes & Heuss-Aßbichler, 2021; Tunsu et al., 2019).

In addition to secondary metals and construction materials, scenario 3 has the potential to produce other byproducts, such as sulfuric acid. While sulfuric acid is not a primary purpose of reprocessing, operating pyrite roasting plants might offer additional revenue streams in the future, especially when the petroleum and natural gas industry declines due to decarbonization efforts and thus, limit the supply of elemental sulfur from sour gas (King, Moats, et al., 2013). To this end, pyrite roasting could become a promising pathway for producing sulfuric acid (Ober, 2002; Runkel & Sturm, 2009).

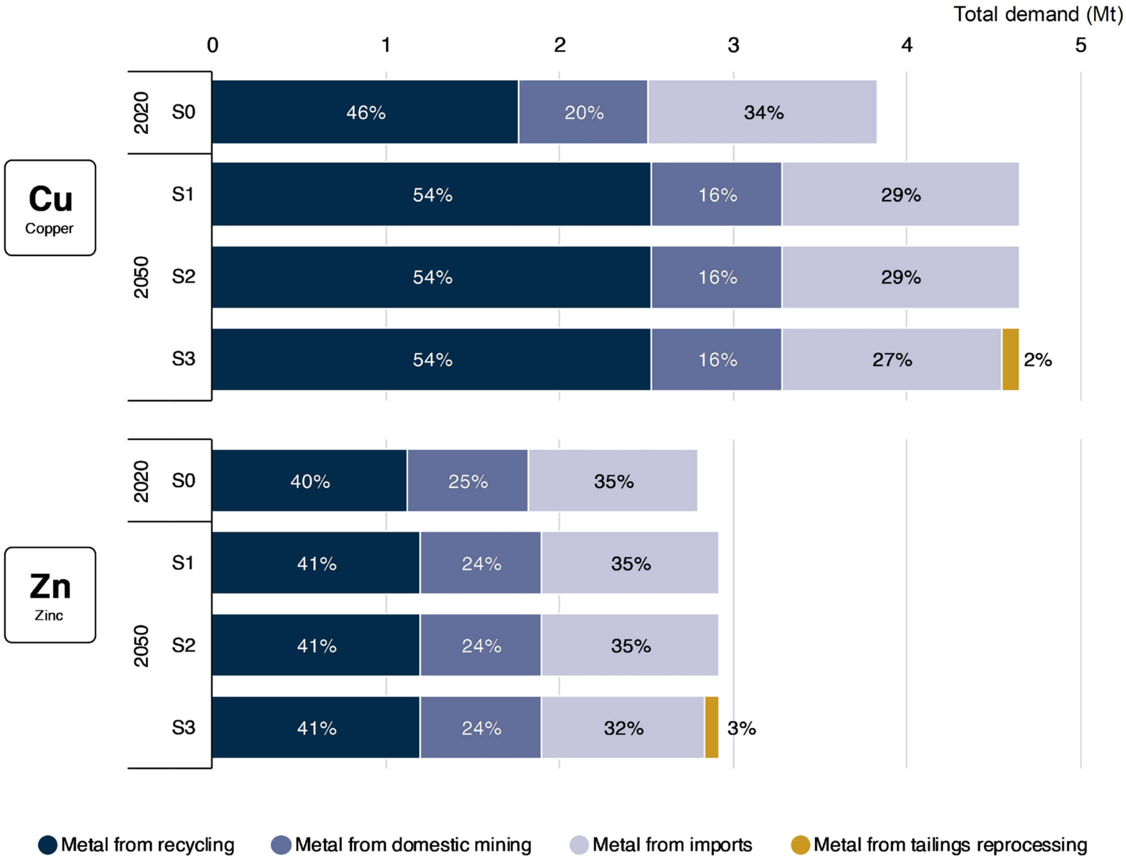


Figure 4.3: The share of metal supply (copper and zinc) from various sources, including domestic extraction, recycling, import, and copper tailings reprocessing. Bars' length denotes the total metal demand in current and future scenarios, adapted from other studies (Ciacci et al., 2020; Gregoir & Van Acker, 2022). Numerical details in Tables C1-C3 in the Appendix.

4.3.2 Life cycle environmental impacts: baseline and future

Figure 4.4 shows the environmental performances of copper tailings management in the baseline year (Scenario 0) and the future scenarios with different treatment options (Scenario 1, 2, and 3). Positive values represent the environmental burden caused by managing tailings in the facility storage and performing backfill operations. The negative values represent the environmental credits of replacing and thus avoiding impacts of manufacturing primary metals and building products. Negative overall values (black crosses) mean that the management of copper tailings has net environmental benefits and is favorable for the selected indicators.

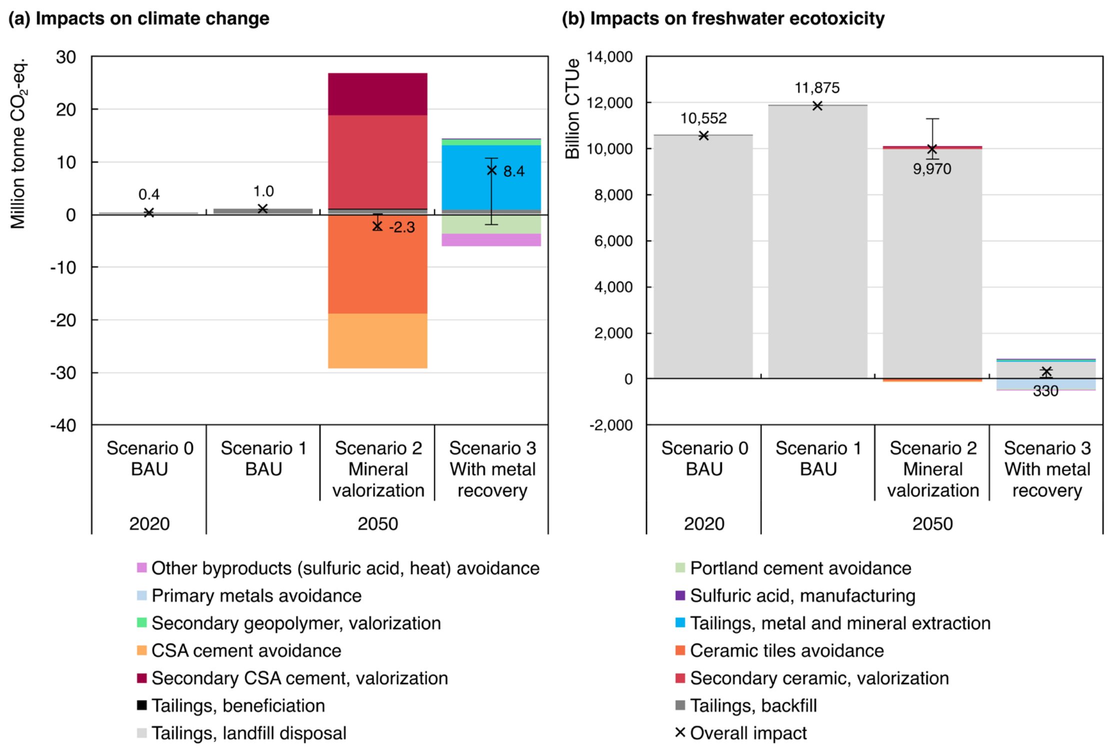


Figure 4.4: Prospective environmental impact from the management of copper tailings in EU under different treatment options. Two midpoint impact categories are shown: a) IPCC 2013 Climate change, b) USEtox freshwater ecotoxicity (see Appendix C for further indicators). The high and low whiskers indicate the possible variation in product market penetration (worst case and HM case, Table C7 in Appendix).

We found that implementing current tailings management options (scenarios 0 and 1) would always generate net impacts across indicators. Moreover, the total impacts of scenario 1 are always higher than scenario 0, as both scenarios implement the same combination of disposal and backfill operation, but scenario 1 has higher demand of copper. Declining ore grades would contribute to the growing volume of waste from metal processing in 2050 (Calvo et al., 2016), despite relatively stable domestic copper production in Europe throughout the mid-century (Gregoir & Van Acker, 2022). In scenarios 0 and 1, freshwater ecotoxicity impacts are higher than in the other scenarios due to long-term freshwater contamination by heavy metal leaching, potentially leading to acid mine drainage. Even if European countries were not found to be an individual global hotspot for toxicity impacts caused by tailings landfilling, the sum of all impacts in the region should not be underestimated in the aggregate (Adrianto et al., 2022).

Scenario 2 offers net benefits on climate change, cumulative energy demand, and resource depletion environmental indicators. Producing secondary ceramic tiles and CSA cement (with the amount specified in Table 4.2) can save up to around 2 Mt CO_2 eq. in 2050. If a lower quantity of secondary materials is available in 2050 (Table C7 in Appendix), the resulting net benefits for all three indicators would instead turn into net impacts. Furthermore, although a reduction of ecotoxicity impacts can be expected (16% decrease from scenario 1), there are still substantial tailings disposal environmental risks that must be managed safely in the future.

One way to minimize ecotoxicity impact potentials is by extracting the acid-generating compounds and metals from copper tailings, as applied in scenario 3. Converting pyritic compounds into other byproducts such as sulfuric acid and recovering companion metals, can significantly reduce ecotoxicity impacts. Besides the lower potential of leaching from the disposal of residues, supplemental material from tailings reprocessing may also substitute primary production, that otherwise would generate voluminous toxic waste such as tailings in the upstream metal ore processing. Gleaning metals from low-quality ores / deposits, as analyzed by Norgate and Jahanshahi (2010), comes at high resource expense, leading to burden shifts to energy-related impact indicators. In contrast to the previous study by Adrianto and Pfister (2022) that assumes unlimited demand for secondary products, this study shows that after credits from all secondary products are accounted for, a net environmental impact remains. Still, scenario 3 offers drastic reductions in ecotoxicity impacts compared to other scenarios. This advantage becomes crucial given the significant contribution of copper production to the global ecotoxicity impacts of metal resources (IRP, 2019).

The net impacts turn to net benefits under best-case assumptions for geopolymer market penetration (Figure 4.4, low whiskers). Therefore, GHG emissions of scenario 3 may be lowered by: 1) exploration of other metal / mineral extraction techniques to further reduce energy and resource (i.e., ceramic / cement ingredients and leaching agents) consumption during reprocessing, since the proposed processing methods in the future are close to the theoretical limits; and 2) the capability to substitute ordinary Portland cement at larger volumes domestically, or to partially sell in international markets beyond the EU boundaries.

4.3.3 Sensitivity analysis

The effects of modifying variables in LCA—such as the origin of substituted products and the definition of substitutability for product displacement—deserve further investigation. Our results were reproduced using different assumptions (section C.5.1 of the Appendix). Overall net GHG footprints for scenario 2 range from -2 to -21 MtCO_2-eq (Figure 4.5A). If ceramic tiles production in China were displaced instead of Europe (base case), the overall net environmental benefits of scenario 2 would increase by almost one order of magnitude. The reason is the energy-intensive process of primary ceramics production, which in China is supplied mainly by coal-based electricity (Wang et al., 2020), while in Europe, it is electricity- and natural gas-based. While there is also potential to lower GHG emissions when displacing OPC cement without CCS in scenario 3 (Figure 4.5B), these measures are insufficient to entirely compensate for the high GHG emissions caused by secondary metal recovery. Primary copper production via pyro- and hydro-metallurgical routes is projected to only make a slight difference in performance as the background energy system moves toward carbon neutrality and foreground technology efficiency improves (Kuipers et al., 2018). Furthermore, with the small volume of secondary metals recovered in scenario 3, changing marginal suppliers has negligible effects on overall GHG performance.

Regarding varying substitution rates for both scenarios, Figure C5 shows how sensitive the net GHG impacts are when the substitution factors for secondary products are changed simultaneously (section C.5.2 of the Appendix). For scenario 2, having secondary ceramic and CSA cement with substitution ratios above 0.8 is crucial to keep the net GHG balance negative. For SRs < 0.5, scenario 2 would perform even worse than scenario 3, which has no GHG mitigating effects in the default case.

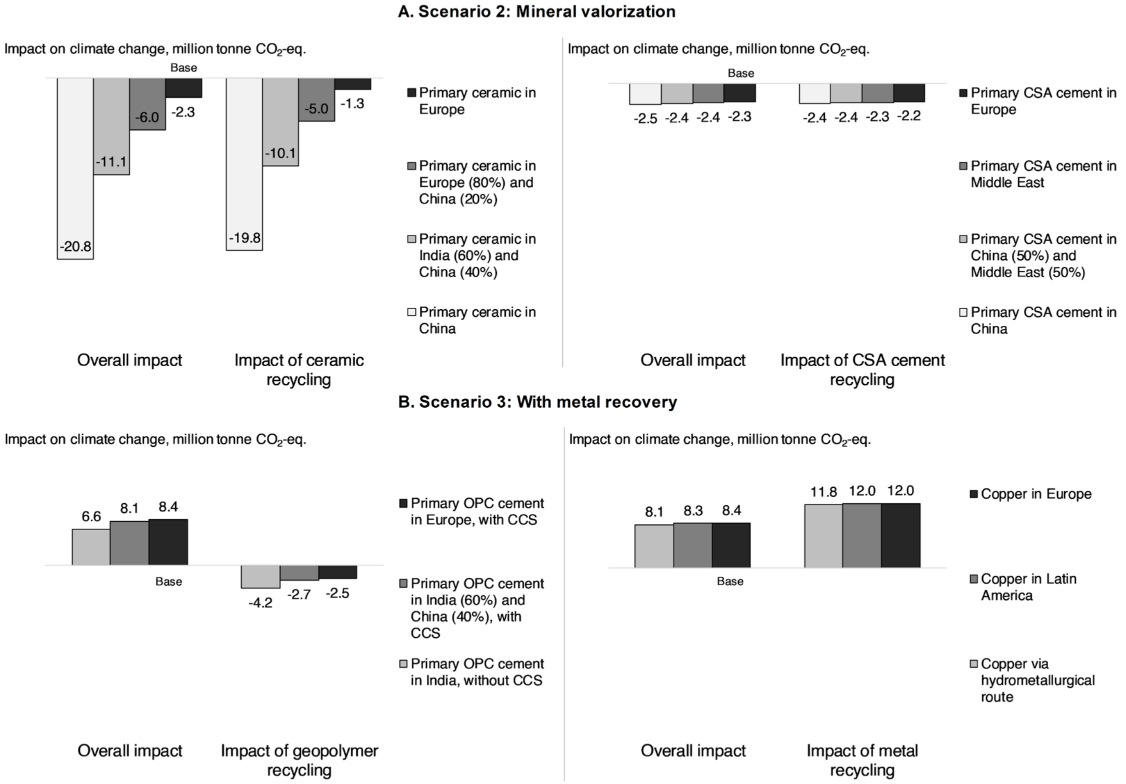


Figure 4.5: Change of marginal technologies for primary material production: effect on climate change impacts for scenarios 2 (top) and 3 (bottom). Overall impact refers to the total impact of all processes. In contrast, impact of recycling shows the net impact of recycling secondary products, i.e., reprocessing burdens minus credits from selected marginal production separately.

4.3.4 Contextualizing the impact of copper tailings management

One of the eminent challenges in the copper sector is to satisfy growing copper demand while meeting climate goals. As the energy system decarbonization progresses, copper production can also benefit from such a transition (Figure 4.6). Moreover, the trajectories of future demand under different scenarios dictate how much copper should be supplied (Ciacci et al., 2020). When alternative tailings reprocessing strategies are applied as in scenario 2, GHG emissions can be mitigated with the expected future secondary market demand. By contrast, scenario 3 does not lead to net GHG savings due to the high energy consumption for the metal extraction, as discussed in section 3.2. Yet, this is different for high market penetration rates of secondary cement (Table C7 in Appendix). However, even with energy efficiency improvements, decarbonization of the power sector, and improved tailings management, additional collective measures are needed to achieve the total GHG emission targets for the EU copper sector. To meet the 1.5°C decarbonization goals, additional reductions of approximately 36% (scenario 2) and 50% (scenario 3) are required to close these emissions gaps (Figure 4.6).

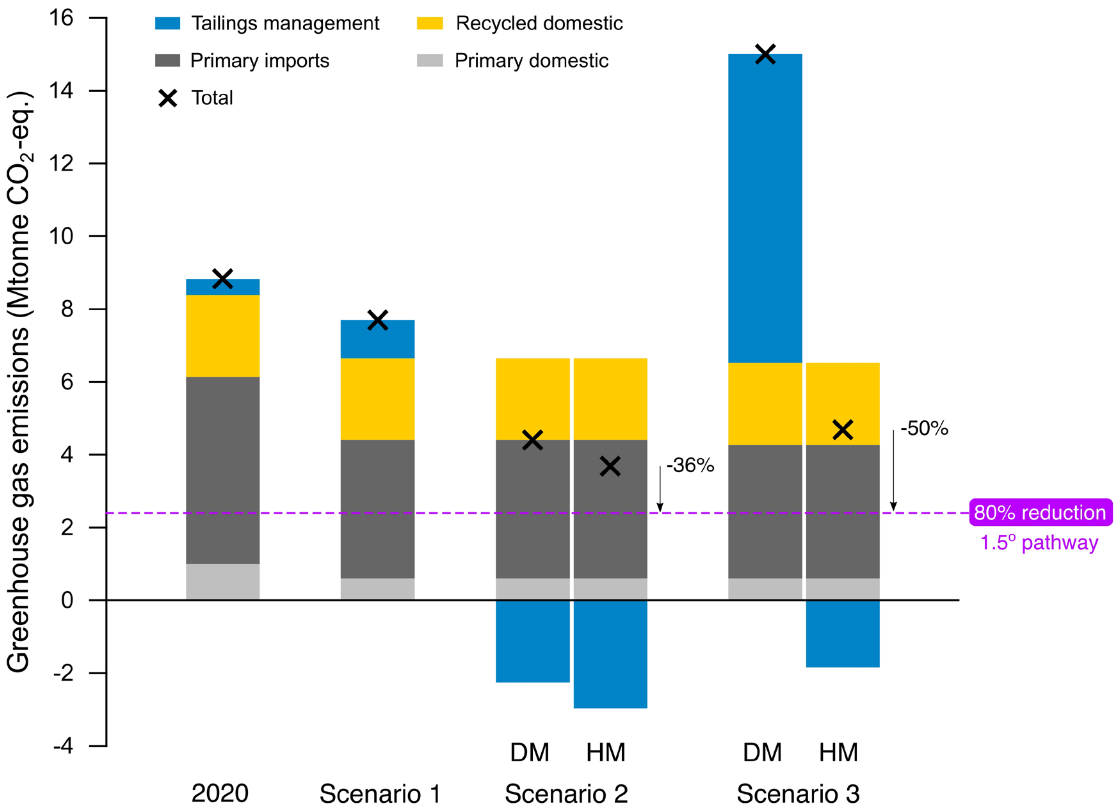


Figure 4.6: Estimation of greenhouse gas emissions embodied in copper demand in the EU according to different scenarios. GHG emissions with alternative tailings management and different secondary product market penetration are compared for each scenario. The dotted lines indicate the reduction of GHG emissions as required in the industry roadmaps (European Copper Institute, 2014). Consumption-based accounting is applied. Numerical details are presented in Appendix section C.8. DM: Default market penetration rates (base case), HM: High market penetration rates (HM case).

It is crucial to note that Europe's copper emission occurs mainly outside the territorial boundary according to the consumption based GHG accounting. Consumption-based accounting for the sector, which sums both emissions occurring in the domestic economy and embedded in imports from other countries, indicates that copper imported from abroad is responsible for more than 50% of the sectoral emissions induced by EU metal consumption (Table C13). A similar finding was discussed for countries with few or no mining activities in other European countries (Mayer et al., 2019; Muller et al., 2020), calling for the roles of additional climate change mitigation measures in reducing carbon footprints beyond territorial boundaries.

For deep decarbonization in the copper sector by 2050, Watari et al. (2022) discuss the importance of multiple measures on both, production side innovations and demand side management. Given that no silver bullet exists, a diffusion of different strategies is essential to meet the emissions reduction target. Central to today's context, this includes GHG-saving copper production, electrification, and aggressive recycling. While waiting for the core technological innovations to scale on time, other key levers, such as more efficient use of copper for the same services and product lifetime extension, could narrow or even bridge the emission gaps.

Based on the scenario modeling, reprocessing copper tailings in the EU could avoid approximately 2 – 3 Mt CO_2-eq. in 2050. The emission targets set by the European Commission (2018) imply a reduction of 128 Mt CO_2-eq. in 2050 for the "2.C metal industry" category (European Environment Agency, 2022). Thus, implementing system-wide reprocessing of tailings (HM case) in Europe would result in the avoidance of 1.5% for scenario 3 to 2.3% for scenario 2 of the total reduction measures in the category "2.C metal industry" (Table 4.3).

While these estimated GHG reduction values are uncertain, the magnitude indicates how many benefits or tradeoffs alternative waste management can generate. Most importantly, due to the transboundary nature of product displacement, the impact reduction for the two sub-scenarios in Table 4.3 that also account for GHG savings outside the EU, should be interpreted with caution. Although the global industry may benefit from implementing this approach regardless of location, the GHG reporting and inventory assessment for such cross-sectoral cooperation between entities must be carefully resolved to avoid double counting of GHG savings.

Table 4.3: Contribution of the copper tailings reprocessing to Europe's GHG reduction targets in 2050.

Years	1990	2020	2050	2050, only EU production		2050, displacement outside EU borders	
		S0	S1	S2	S3	S2	S3
Total GHG emissions, all categories[a], Mt CO_2-eq.	4,633	3,068	232	232	232	232	232
Net GHG emissions, metal industry[a], Mt CO_2-eq.	135	64	7	7	7	7	7
% reduction from 1990 levels, metal industry[b]	-	53%	95%	95%	95%	95%	95%
Reduction targets relative to 1990, all categories, Mt CO_2-eq.	-	1,565	4,401	4,401	4,401	4,401	4,401
Reduction targets relative to 1990, metal industry, Mt CO_2-eq.	-	71	128	128	128	128	128
Tailings management impacts[c], Mt CO_2-eq.	-	+0.4	+1.0	-2.3	+8.4	-21.1	+6.3
Tailings management impacts[c], Mt CO_2-eq. (HM case)	-	-	-	(-3.0)	(-2.0)	(-21.9)	(-11.2)
% tailings management impacts / reduction targets of all categories	-	+0.0%	+0.0%	-0.05%	+0.2%	-0.5%	+0.1%
% tailings management impacts / reduction targets of all categories (HM case)	-	-	-	(-0.1%)	(-0.04%)	(-0.5%)	(-0.3%)
% tailings management impacts / reduction targets of the metal industry	-	+0.6%	+0.8%	-1.8%	+6.5%	-16.4%	+4.9%
% tailings management impacts / reduction targets of the metal industry (HM case)	-	-	-	(-2.3%)	(-1.5%)	(-17.1%)	(-8.7%)

Note: For material displacement outside the EU in 2050, high-impact production from marginal sensitivity tests was chosen. HM case represents the scenario with high market penetration for secondary products. Positive (red) and negative (green) values are color-coded. More discussions can be found in the Appendix section C.9.

[a] GHG inventory data for 1990 and 2020 from EEA (2022), covering six source and sink categories: 1. energy, 2. industrial processes, and product use, 3. agriculture, 4. land use, land use change and forestry, 5. waste, and 6. Other;

[b] Defined GHG emission targets for both business as usual and decarbonization vision from EU commission (2018), assuming percentages apply equally across categories. The targets are used to estimate GHG inventory data in 2050.

[c] Own calculation (Figure 4.4 and Figure 4.5).

The evaluated case represents one example of climate mitigation solutions through waste management. Other breakthrough technologies beyond our analysis might penetrate the market and become commercialized, amplifying the GHG reduction potential through improved energy and resource intensity. For example, various types of tailings have been regarded as promising storage for the carbonation process, enabling CO_2 capture for emissions offset (Bullock et al., 2021; Wilson et al., 2014). Such solutions should also be assessed with LCA to complement the present study.

4.3.5 Implications for practice

This study has implications for business activities in the copper and materials industry. Today, business opportunities and sustainability standards in the copper sector have been focusing on technological upgrades and decarbonization of the production system. One area that lacks investigation is understanding the role of waste management through a life cycle assessment combined with a metal scenario outlook. Our research shows secondary production potential by reprocessing copper tailings in the EU.

Implementation barriers include heterogeneity of material properties, economic uncertainty, fragmented legislation, and conflicting corporate cultures / values (Almeida et al., 2020; Sibanda & Broadhurst, 2018). Additionally, in the context of EU mine tailings valorization, the lack of relevant regulatory standards for waste-based materials and financial incentives represent key bottlenecks in accelerating the use of industrial byproducts over virgin building materials (Kinnunen & Kaksonen, 2019). Beyond that, additional work is critical to demonstrate the applicability of new products at the desired scale. Tight regulations might sometimes prevent scalability even when the technologies are proven. The industry must be willing to go through national approval processes with often differing political and regulatory conditions before such products can successfully enter the market (Material Economics, 2022).

Our analysis reveals tradeoffs between climate change and ecotoxicity impacts for scenarios 2 and 3. Although small GHG reductions are possible by 2050, exploring additional strategies to meet future climate ambitions is imperative to meet the Paris climate agreement. Reijnders (2021) proposed the idea of near-zero waste production of copper, making use of the geochemically scarce elements and mineral matrix considerably lost in tailings, slags, and dust during the mining and refining stages. Assessing novel metallurgical processes and improving the recoverability of these elements / minerals may open doors for additional ecological benefits.

4.3.6 Opportunities for future work

The material demand projection and forecast based on established scenarios and integrated assessment models are uncertain. Our results should be understood as exploratory projections rather than the prognosis. Furthermore, the marginal processes in the substitution modeling were selected based on semi-quantitative methods using industry technological roadmaps. They did not consider dynamic market interaction, i.e., price elasticities, economic equilibrium, or trade barriers resulting from conflicts (e.g., sanctions). Lastly, while this study makes a compelling case for unveiling the impacts and benefits of reprocessing scenarios, subsequent stages in the LCA are missing, such as use- and end-of-life phases. Rigorous testing, such as leaching, aging, tearing, and recycling under different use and disposal conditions, is necessary for products using secondary materials. Providing these results can enable a comprehensive environmental comparison between secondary products from tailings and their primary equivalences. The ultimate goal is an extensive assessment that can strengthen decision-making and policy designs to support concrete system-wide solutions. Integrated analyses like Golev et al. (2022), combined with the presented framework, may enhance information on the sustainable management of mine tailings.

4.4 Conclusions

This study was set out to answer whether environmental benefits from secondary production through the reprocessing of tailings outweigh the associated environmental burdens. Built upon a previous site-specific assessment of mine tailings and future scenarios, a prospective LCA approach was employed here to assess the large-scale environmental impacts of reprocessing. Overall, the main conclusions of this analysis are as follows:

- Reprocessing copper tailings in the EU decreased freshwater ecotoxicity impacts compared to traditional waste management options. Other environmental benefits included GHG performance for scenario 2 with mineral valorization due to the large displacement of primary building materials such as cement and ceramic. However, scenario 3 with metal recovery showed an increase in climate change impacts compared to all other scenarios due to the energy-intensive metal recovery process for extracting metals at low concentrations.
- Secondary metal recovery from tailings, valorization of the mineral matrix as substitutes for building materials, and sulfuric acid production from pyrites could help meet the growing demand for these products in the EU. For building material production, the constrained availability of raw materials in the current supply of alumina sources and alkali activators could hamper efforts to scale production. This might limit the market penetration rates of these products to 10 – 15% of the total secondary supply.
- Regarding contribution to EU climate targets by 2050, around 2 – 3 Mt CO_2-eq. of savings can be generated by implementing alternative copper tailings management, equivalent to a 1.5 – 2.3% reduction in the metal industry category. Looking at the EU copper sector alone, this GHG performance is still insufficient to curb climate change compatible with the 1.5°C pathway. Additional strategies on top of what has been presented, such as demand-side management, material efficiencies, and breakthrough metallurgical innovations, must be explored altogether to close the emission mitigation gaps meaningfully.

In summary, our findings confirm the potential opportunities for tailings reprocessing and valorization at a large scale. There are still potential pitfalls, such as the net GHG impacts of reprocessing scenarios with metal recovery, missing market demand for recovered minerals, and potential use-phase or end-of-life emissions (not studied so far). Future research shall extend the scope of the prospective LCA (use- and end-of-life) and realization of other climate mitigation strategies in the copper sector for more holistic environmental considerations. Further progress in this direction can help improve assessment quality and increase transparency in tailings-derived product evaluation.

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Chapter 5

Conclusions and outlook

This chapter summarizes the principal conclusions and syntheses of Chapters 2 – 4. Based on these conclusions, scientific relevance and directions for future research are discussed.

Chapter 5: Conclusions and outlook

5.1 Synthesis

This thesis aims to evaluate the environmental implications of reprocessing waste from mining operations using industrial ecology tools, such as life cycle assessment, material flow analysis, and scenario development. The thesis focuses on managing copper mine tailings residues and their emerging resource recovery treatment options. Processing copper tailings is particularly relevant in the context of toxicity footprinting, given its high share of ecotoxicity impacts among global metal commodities (IRP, 2019). In **Chapter 1**, the following research topics were formulated:

RQ 1 How can mine tailings disposal emissions be modeled and improved by considering site-specific factors and characteristics of tailings? Where do global environmental hotspots occur? (**Chapter 2**)

RQ 2 How can small-scale laboratory and pilot experiments to recover materials from mine tailings be used to assess the environmental performance of a future full-scale implementation of reprocessing and valorization technologies? (**Chapter 3**)

RQ 3 What is the potential of recovering minerals and metals from EU mine tailings to produce industrial products? Can alternative tailings management contribute to reducing environmental impacts (and if so, how much)? (**Chapter 4**)

To answer these research questions, first, the site-specific environmental impacts of conventional copper tailings management were assessed in detail as a baseline. Second, the environmental performance of complex, emerging resource-recovery systems for mine tailings reprocessing and valorization was quantified. Third, utilizing the copper industry in the EU as a case study, the large-scale environmental benefits and impacts of implementing alternative tailings management in future scenarios were investigated.

The **first research question** was related to the environmental impacts of tailings landfilling practice, given the large scale and omnipresence of copper tailings globally. The question is: "*How can mine tailings disposal emissions be modeled and improved by considering site-specific factors and characteristics of tailings? Where do global environmental hotspots occur?*"

Employing a combined method to strengthen the usefulness of LCA approaches, the environmental impacts of conventional tailings management were assessed [**Chapter 2** (Adrianto et al., 2022)]. The study focuses on copper tailings generated from primary copper production via pyrometallurgical pathways. Approximately 80% of the world's copper is manufactured using this production method. I built a systematic stepwise approach to generate a global environmental assessment of copper tailings worldwide. The linkages between the copper mining operations at each site and the types of tailings subsequently produced as mine waste are key to this strategy. To facilitate these connections, the systematic stepwise approach includes: i) compilation of copper-active production and ore mineralogy for each production site; ii) process-based approximation of tailings composition using metallurgical process simulation; iii) site- and time-dependent lifecycle inventory modeling of mine tailings emission; and iv) global impact assessment of sulfidic copper tailings over different time horizons.

The study found that 75% of the impacts on long-term ecotoxicity occurred from the deposition of large- and medium-sized volcanogenic massive sulfides and sediment-hosted copper tailings at 431 copper sites. Porphyry copper ores are mainly situated in Americas (i.e., Chile, Peru, and the USA) and Asia (Indonesia and Papua New Guinea), whereas the latter two deposit types are mainly occurring in Canada, Africa (i.e., D.R. Congo and Zambia), Europe (Poland) and Russia. I also discovered that tailings with low buffering capacities generated from volcanogenic massive sulfide and sediment-hosted copper deposits had a higher potential to become hotspot zones in areas with high infiltration rates.

This site-specific evaluation, which results in tailings impacts mapping, helps assessing resource / environmental pressures caused by mining activities and determines where environmental consequences occur. Identifying environmental hotspots helps prioritizing the regions / countries that deserve specific focus and improved environmental protection policy measures. Additionally, the detailed assessment with country or even facility resolution is now possible and creates more granular and accurate emissions estimation for metal tailings. However, many sites may also impose significant risks, not only the identified global toxicity hotspots. In some situations, authorities continue to allow selected sites in Indonesia and Papua New Guinea to perform direct riverine tailings management, which has an immediate adverse impact on freshwater bodies. Capturing these technical details is important to understand the differing pollution impacts of tailings on the local ecosystems. One main takeaway is that the variability of toxicity impacts occurs both, between and within countries and thus country averages might not be very accurate.

In addition to spatial variability, the model allows for temporal sensitivity analysis, i.e., the time horizon considered in the impact assessment can be freely chosen when defining system boundaries. Analyzing temporal sensitivity can enable a more transparent communication of LCA results and reveal the consequences of choosing temporal system boundaries.

Finally, combined with the prospective copper data from other studies conducted by Elshkaki et al. (2018) and Northey et al. (2014), I showed that the global ecotoxicity impacts of copper tailings are expected to increase by up to +70% in 2030 relative to 2020 levels. This is primarily due to increased demand for copper and diminishing grade of sourced copper ore. However, shifting to secondary copper resources can help reduce impacts, thereby lowering total global toxicity levels down to +41% by 2040 and to +9% by 2050 compared to 2020 levels. It is also notable that in the next three decades, the location of hotspots will be almost equally distributed among countries such as Russia, Australia, and D.R. Congo, where strong copper production growth is predicted by mid-century.

One possibility to reduce tailings emissions and, at the same time, make use of the resources included in tailings, is tailings reprocessing and valorization. This strategy recovers metals from tailings and transforms mineral matrices into useful industrial products. Hence, the **second research question** specifically addressed methodological challenges for quantifying the environmental impacts of emerging reprocessing and valorization techniques for copper tailings: "*How can small-scale laboratory and pilot experiments to recover materials from mine tailings be used to assess the environmental performance of a future full-scale implementation of reprocessing and valorization technologies?*"

Mine tailings recovery and valorization technologies are improving over time, allowing for the recovery of metals stranded in waste flows and the productive use of mineral matrices for industrial products. Emerging technologies usually undergo continuous development, which may be difficult to capture by standard LCA studies. In **Chapter 3**, I employed state-of-the-art prospective LCA frameworks to characterize the environmental performance of complex tailings treatment routes (Adrianto, van der Hulst, et al., 2021; Adrianto & Pfister, 2022).

There are numerous options to treat and add value to mine tailings, which differ depending on the feedstock characteristics, technology choices, and end products. In the ETN Sultan project, there are primarily two main solutions regarding reprocessing / valorization schemes: 1) extraction of metals using a combined microwave roasting, leaching, and ion flotation, and 2) transformation of mineral residues rich in pyrite and aluminosilicate fractions into building materials, namely inorganic polymer / geopolymer, calcium sulfoaluminate cement, and ceramics. Drawing on these complex reprocessing pathways, I chose a modular and process-based approach to model the reprocessing and valorizing of copper tailings. This has the advantage of assessing different constellations of processes and value chains. Mass / energy balance and technical assumptions were taken into account for the design of the reprocessing and valorization value chain based on the input of technology experts. The process-based LCA captures the compatibility between processes, which is often overlooked in standard LCA models. All processes were upscaled with a combination of process simulation, engineering-based calculations, and technology proxies to allow for a comparison with other large-scale technologies.

The study found that reprocessing copper tailings and mineral valorization offer environmental benefits compared to typical tailings management. Applying a system expansion allocation approach, I found that the net climate change impacts for four investigated reprocessing routes range from -25 to -930 kg CO_2-eq. per tonne treated copper tailings. A large share of the environmental benefits comes from the substitution credits of secondary building materials, e.g., ceramics and calcium sulfoaluminate cement. However, there are tradeoffs for specific treatment pathways and processes in the value chain. For instance, reprocessing pathways with small climate change benefits can significantly reduce freshwater ecotoxicity impacts, owing to the prevention of tailings deposition, and substitution of primary metal mining that generates tailings in the upstream stage. Indeed, this potential to reduce ecotoxicity stress is crucial, given that the copper industry contributes more than half of the global toxicity impacts among major metal resources (IRP, 2019). Overall, the modular nature of the prospective LCA enables the identification of process bottlenecks, which can be used to highlight improvement potentials through further process optimization.

Identification of the influential modeling parameters is a prerequisite for discussing the environmental implications of alternative mine tailings management. I evaluated the effects of changing substitution ratios and energy mixes for the background inventories. The sensitivity analyses revealed that there would be a deviation of $\pm$ 30% in the various environmental impacts from the baseline environmental performances. The largest sensitivity changes are for energy-related environmental impact indicators such as climate change, fossil depletion potential, and cumulative energy demand.

Chapter 3 has assessed tailings reprocessing and valorization by performing small-scale prospective LCA. However, large-scale deployment of tailings management and forward-looking parameters should be considered in systems modeling. Examples of parameters include dynamics of metal demand, energy systems, technology improvements with time, and tailings reprocessing pathways with the associated recovery products. Therefore, the **third research question** is: *"What is the potential of recovering minerals and metals from EU mine tailings to produce industrial products? Can alternative tailings management contribute to reducing environmental impacts (and if so, how much)?"*

Using established scenarios from previously published metal outlooks and energy perspectives, based on MFA (Ciacci et al., 2020; Elshkaki et al., 2018; Van der Voet et al., 2019), the environmental assessment of various options for the future copper tailings management in the EU was investigated in **Chapter 4**. In addition to predicted metal demand in previous metal outlooks, I complemented the study by adding EU-specific metal demand estimates for supporting low-carbon transition (Gregoir & Van Acker, 2022). I then quantitatively assessed the environmental performance of various scenarios for European copper tailings management, building on earlier research (Adrianto et al., 2023). This analysis helps to discuss the large-scale environmental implications of future mine tailings management across a broad set of impact metrics and the role of improved tailings management to impact mitigation.

Decarbonization in the power sector can help decrease the environmental impacts of the supply chain of tailings reprocessing, as shown in **Chapters 3 and 4**. Yet, the impacts of substituted primary products may also gradually decrease with industry decarbonization—thus limiting the substitution credits that can be generated from product displacement. For example, substituting primary building materials, such as cement in 2020, would differ from substituting the same material manufactured in factories using carbon capture technology and other emission reduction techniques. Moreover, secondary material demand that the market can realistically absorb may be limited. The production of secondary materials from tailings reprocessing depends on the availability of raw ingredients needed, such as alumina and alkali sources, for producing useful materials from tailings. Such considerations may limit the scaling up of building materials production from tailings.

In spite of these restrictions, our study suggests that copper tailings reprocessing and valorization can help cover metal demand in the EU to a certain extent. Implementing resource-recovery strategies as part of improved copper tailings management scenarios could satisfy up to 3% of European base metals demand (i.e., zinc and copper). This finding confirms that anthropogenic resource stocks such as mine heaps and tailings streams should be re-evaluated for their potential contribution to supporting the shift to a circular economy (Lèbre et al., 2017; Winterstetter et al., 2021).

Regarding the environmental performance of future tailings management, our assessment showed that implementing large-scale resource-recovery systems across copper tailings sites in the EU can generate benefits for relevant impact indicators such as climate change and freshwater ecotoxicity. Scenarios that rely solely on mineral residue valorization could yield the greatest climate-change benefits of all management scenarios. Scenarios with simultaneous metals and minerals recovery for building materials have higher climate-change impacts due to energy / resource-intensive metal extraction process blocks but show drastic reductions in ecotoxicity impacts. Such tradeoffs exist between climate change and ecotoxicity impacts. However, the energy-related impacts may be minimized if: (i) new metal extraction technologies with better energy and resource efficiencies are developed and (ii) the product market penetration for secondary mineral products can be increased, such that geopolymer derived from tailings can displace ordinary Portland cement at higher volumes. Similarly, the presence of secondary materials market determines the environmental advantages of tailings reprocessing and valorization. Market intervention could encourage the use of recycled products from tailings, for instance, by prescribing them for green public procurement (GPP) initiatives (European Commission, 2016; UNEP, 2022).

To contextualize the contribution of improved tailings management to climate mitigation, I assessed the total climate benefits of the alternative tailings-management scenarios and compared them against emission targets of the copper sector. Improved tailings management for the European copper sector could help cutting approximately 2 – 3 Mt CO_2-eq. GHG emissions in the year 2050. These amounts correspond to climate mitigation of ~2 % of the 'metal industry' category GHG emissions in the EU. This is a limited contribution compared to climate ambitions in 2050 compatible with 1.5°C global temperature increase limits; hence, the EU copper industry has yet to meet its emission targets by implementing additional measures.

5.2 Relevance of the thesis

5.2.1 Scientific relevance

This thesis contributes to the development of methodologies for assessing the environmental performance of tailings deposition and reprocessing / valorization. Based on the application of these methodologies, recommendations for tailings management with better environmental outcomes are provided.

This thesis provides a scientific contribution to the global assessment of mine tailings by integrating copper metallurgical process models (Reuter et al., 2019) and site-specific tailings emission models into LCA. The developed bottom-up and combined frameworks allow a global coverage with a more systematic and accurate assessment of the environmental impacts of conventional tailings disposal. Tailings emissions are modeled as a function of ore deposit composition, mineral processing technologies, infiltration rate, and heavy metals leaching behaviors. This functionality allows the thesis to specifically model emissions for individual tailings sites and provides an overview of the variability of emissions between tailings sites. This is a notable advantage compared to previous research (Doka, 2018; Turner et al., 2019) and the currently available mine tailings dataset in the common LCA database Ecoinvent v3.8 (2022). This achievement thus may hold the potential for expanding the coverage of the tailings dataset in terms of locations, ore types, and processing. This is another step forward to enable the assessment of accurate environmental impacts of waste treatment processes such as tailings in the overall product supply chain, which was frequently neglected, underestimated, or overestimated in the LCA of metal-containing products.

While the current thesis focused on modeling the emissions of copper tailings, the approach applied by this study can serve as a model also for the assessment of other mine waste types and tailings from other metal production. Waste rock is potentially another large source of environmental footprints, so modeling impacts from waste rock might be possible based on the presented modeling approaches. Besides copper tailings, other tailings from the production of other metal resources such as gold, iron, zinc, and nickel are also lacking in site-specific, temporally resolved environmental assessment and could be assessed by adapting the presented methodology.

When reprocessed using the right technologies, tailings can be regarded as resources that may help to reduce the need for primary mining operations. Such technologies are, however, under development and change with time, requiring modeling of scaling effects if one wishes to assess the environmental impacts of those emerging technologies. Since environmental performance of a full-scale implementation of tailings reprocessing and valorization was rarely clarified in peer-reviewed literature, this thesis fills the gap by applying a technology upscaling framework in a prospective LCA. Another strength of this study is that it was built upon technical and collaborative inputs from the technology designers (e.g., material scientists and geologists), ensuring technical compatibility along the value chain.

Through structured prospective LCA methods, this work offers opportunities to track the environmental performance of upscaled technologies at the modular level, indicating optimization potential with high transparency. The newly created datasets for each new process block, specifically for treating copper tailings using emerging metallurgical technologies, are also valuable additions to the growing literature on sustainable mine waste management and the wider LCA community. The developed LCA models are parametric and may be utilized by other academics and environmental practitioners to examine the properties of various types of tailings and their operating conditions based on future assessment needs.

This thesis additionally contributes to state-of-the-art research of prospective LCA of copper tailings management by integrating scenario analysis / narratives (Ciacci et al., 2020; Elshkaki et al., 2018; Gregoir & Van Acker, 2022) with outcomes from integrated assessment models (Sacchi et al., 2022). This integrated approach allows researchers and practitioners to effectively understand the potential large-scale contribution of alternative tailings management to the mitigation of various environmental impacts in the future. This research suggests there are both secondary resource potential and related environmental benefits, which also require strong partnerships between industry players across the reprocessing / valorization chain. However, one must be mindful of the potential pitfalls, such as the net climate change impacts of tailings reprocessing scenarios with metal recovery.

5.2.2 Practical relevance

This thesis provides practical recommendations and opportunities to lower future environmental impacts of copper tailings. The mining and minerals industry has been using LCA for a long time to assess the environmental impacts of products / services. However, data about tailings impacts were either missing or of low quality (Reinhard et al., 2019). The International Copper Association (ICA) is a global copper industry association that acknowledges the importance of reliable emissions datasets for tailings. For example, during the '2019 Metal Leaching and Acid Rock Drainage Workshop' in Vancouver, Canada, industry practitioners and researchers raised concerns about the reliability of the current tailings data (Rader et al., 2019; Turner, 2019). As a follow-up, there was a couple of exchanges with ICA and Ecoinvent towards more representative copper tailings datasets. This thesis closes this data gap concerning the waste treatment of copper tailings, hence providing opportunities for decision-making support by stakeholders such as policymakers and the metal industry to mitigate tailings impacts on the environment. Since metal consumption is expected to increase significantly in future decades (Elshkaki et al., 2018; Lee et al., 2020), the availability of a site-specific dataset for tailings will be of paramount value for the copper industry, copper-consuming industries, and the public domain. In an industry where a lack of data remains a challenging factor in quantifying environmental impacts, the developed methodology and datasets can help provide transparency to improve the decision-making processes related to tailings management (Kemp et al., 2021). Examples of such implementation on a global level include mapping the impacts / risks of tailings dams accessible through 'Global Tailings' portals by UNEP (GRID-Arendal, 2020) or via the public domain by university researchers (Rana et al., 2021).

Tailings reprocessing and valorization are concepts that governments and policymakers appreciate, but these concepts needs to be assessed for environmental sustainability (Kinnunen & Kaksonen, 2019). This thesis provides methods and data to assess emerging resource recovery technologies for copper mine tailings. The developed frameworks and assessment approaches can be adapted to various resource recovery projects due to the modular structure, and they can also be used in early-stage process development. Furthermore, continuous feedback among LCA practitioners and technology specialists in multidisciplinary contexts enables techno-environmental evaluation with 'big picture' thinking in mind. In practice, this process design accounts for material compatibility and interoperability between processes in the resource recovery systems.

The developed methodology proposed by this thesis has already been successfully applied to provide environmental decision-making recommendations for process improvements to the research on developing mine waste valorization technologies within SULTAN. This also allowed a prospective LCA to be conducted for those emerging technologies to guide the improvement directions. This included: (i) mine waste-derived geopolymer (Adrianto, Niu, et al., 2021; Niu et al., 2021) and (ii) tailings-derived ceramic bricks (Veiga Simão et al., 2022). Beyond these specific studies, recent years have witnessed the growing appetite for tailings recovery projects such as the implementation of Auxilium, EnviroGold, Phoenix Tailings, Circular Mine, and EU Horizon 2020 follow-up projects (e.g., Nemo, Rawmina, Tarantula, and others). Such projects can use the process flow sheeting framework, scale-up procedures, and prospective LCA methods of this thesis to improve the design of their mine waste treatment solutions. This will help with the design and optimization phase of the proposed resource recovery systems when changes can still be made and before pilot plants are put into operation. This may also include exploring innovations in value recovery from currently discarded product flows such as tailings, slags, waste rock, and other waste streams (Reijnders, 2021). Identifying the most suitable technologies to salvage important elements / minerals from these product flows provides additional opportunities for improving the industry's sustainability profile.

Regarding the large-scale deployment of mine tailings reprocessing and valorization solutions in the EU, I found that tailings may offer resource and environmental benefits. However, these benefits are relatively moderate compared to the potential GHG emission savings included in the climate goals planned by the copper industry. The thesis underscores the need for industrial symbiosis, such that the transfer of technology, materials, and knowledge can be fostered throughout the value chain instead of remaining stagnated within the existing siloed process. Similar findings were articulated by the recent collaborative work on iron tailings for material substitutes led by UNEP, the University of Geneva, and the University of Queensland (Golev et al., 2022; UNEP, 2022). Although the materials explored in our study and the findings noted above are different, there are similarities concerning both anticipated market acceptance of secondary products and industry collaboration across the value chain. At the fourth consultation session by United Nations Environment Assembly (UNEA) in March 2019, (i) tailings management and (ii) mine waste recycling and circularity were highlighted as two priority issues relating to the sustainable management of metal and mineral resources (Franks et al., 2022; UNEP, 2019). The assessment methods developed and result presented in this thesis are helpful to evaluate whether such tailings circularity options are indeed environmentally sustainable and deserve to be supported by policy.

5.3 Critical appraisal

This thesis contributes to a better understanding the environmental impacts of mine tailings and reprocessing / valorization technologies. This section discusses the limitations and uncertainties of the methods and results provided.

The regionalized life cycle assessment of mine tailings presented in **Chapter 2** can help identify environmental hotspots worldwide – most notably through detailed site-specific assessment. Another strength of the approach is the ability to model emission releases at each site as a function of time. The proposed stepwise approach can thus be used for time-resolved inventory modeling accounting for local specificities.

Nevertheless, the developed tailings model also comes with several limitations:

- The composition of tailings was approximated using a steady-state process simulation and a single-stage beneficiation circuit for different copper ore classes. Dynamic behaviors at the plant level and their metallurgical process based on actual operations might be different from the simulation model. Also, tailings in every site were modeled as homogeneous impoundment without considering inherent heterogeneity and additional complexity in tailings deposits, such as compositional differences, preferential flows of infiltrating water, and other multi-layer related variabilities.
- The hydrology and geochemistry models and underlying approaches contain assumptions that might not fit perfectly. For instance, the global hydrology model PCR-GLOBWB2 (Sutanudjaja et al., 2018) simulates net infiltration rates at specific grid size resolutions and time horizons. However, these rates may not remain constant over long-term time horizons, as relevant for modeling leaching emissions from tailings. Climate change effects might play a role in a long-term hydrology projection that involves centuries to thousands of years as time steps. Also, PCR-GLOBWB2 models are based on natural soil grain size modeling; hence, infiltration mechanisms might differ from tailings. In addition, geochemical modeling in PHREEQC (Parkhurst et al., 2013) relies on chemical speciation / dissolution reactions in soils, and such modeling might not capture all reactions that may take place in tailings. For instance, the kinetically controlled mechanisms in microbial activities might also affect the leaching conditions, but they were not taken into account in the quasi-equilibrium long-term processes (Chen et al., 2014).
- A general assumption in the modeling was that the standard management of tailings covers and dams could only last for a limited time of less than a century (Rowe & Islam, 2009). After this period, tailings management, including covers and liners, were assumed to break without continued controls thereafter. There is a range of options to minimize acid mine drainage potential from tailings deposits, which include the active maintenance and recirculation of the leachates system even long after surveillance phases and thereby limit toxicity impacts (Park et al., 2019). However, if such approaches are to be implemented, proper accounting of resources and energy needed for the passive / active operation of tailings deposits must be considered in LCA as a way to account for environmental impacts of this management, such as infrastructure, energy, and chemicals.
- All impacts were allocated to the extraction and processing of copper. Copper mining and processing are often associated with the production of other companion metals such as gold, molybdenum, zinc, and other metals. Analyses such as those conducted by Memary et al. (2012), Nuss and Eckelman (2014), and Rötzer and Schmidt (2020) indicate that more than 99% and 95% of impacts are allocated to copper for mass and economic allocation, respectively. However, the site-specific tailings data of this study can be used to apply other allocation principles for further research. Overall, allocation choices are still an open field that can benefit from more harmonization

measures and in-depth future investigations (Stamp et al., 2013; Weidema & Schmidt, 2010).

- The model has only been parameterized for copper tailings from pyrometallurgical copper processing. Waste rock and residues from hydrometallurgical copper processing (heap leaching) were not addressed.

Similar to the tailings, the reprocessing technologies only addressed base metals from copper tailings as the metals of interest in this study, along with derivatives of tailings-based construction products such as cement and ceramics. Other extraction techniques do exist that can recover stranded metals in mine tailings, including bioleaching (Schippers et al., 2013), solvo-metallurgy (Li et al., 2020), and deep-eutectic solvents (Pires et al., 2022). The assessment of these technologies would require additional inventory and LCA models to be developed.

The emergence of tailings reprocessing routes and recovery options also implies a multiple process design for industrial symbiosis. Another limitation imposed in **Chapter 3** pertains to the restricted number of scenarios for tailings reprocessing and valorization. The flows of different goods in such industrial networks theoretically can be optimized according to environmental performance, economic objectives, and other types of objectives in the process design (Klinglmair et al., 2017; Vadenbo et al., 2014). In addition, the life cycle inventory modeling of this study was facilitated mainly by engineering-based calculations and chemical process design calculations for technology upscaling (Parvatker & Eckelman, 2019; Tsoy et al., 2020). These approaches may suffer from simplifications of mineralogy and thermodynamics- and kinetics-based approaches, which can provide better quantification of materials and losses from the system (Reuter et al., 2019; van Schalkwyk et al., 2018). Lastly, despite the presence of other metals in copper tailings, such as rare earth elements, reprocessing and recovery of these elements may require uneconomically high energy and resource inputs, as described in the "metal wheel" concept (van Schaik & Reuter, 2014). While this study performed no sophisticated multi-objective optimization techniques in the process design of reprocessing and valorization, future research may explore this by applying mathematical optimization and complete process simulation-based approaches.

In this thesis, cradle-to-gate LCA models were constructed for reprocessing technologies, whereas the subsequent phases of the waste-derived products, i.e., the use phase and disposal of building products after use, were not modeled explicitly. This is another limitation of the study, since the long-term leaching assessment was conducted for the tailings landfilling but not for the tailings resource recovery systems. Expanding the previous system boundary beyond cradle-to-gate LCA models means considering potential environmental risks during the use and end of life phase of these products. One risk is the leaching of harmful components possibly released by those valorized products, which may impede their widespread market adoption. According to the comprehensive review conducted by Velásquez et al. (2022), contaminant leaching is one crucial consideration regarding the environmental and health implications of recycling mine tailings for construction purposes. In the 'ETN SULTAN' project, three separate leaching tests (EN 12457-2, US EPA's Toxicity Characteristic Leaching Procedure or TCLP, and pH-dependent leaching tests) were performed on the finished materials, which included cement, alkali-activated binders, and ceramic material manufactured from mine tailings (Helser et al., 2022). At the time of this research, the parallel study was not yet completed; hence, detailed scientific exchanges were not made. The gaps in our study can be filled (i) by including leaching results noted above in the cradle-to-grave LCA and in parallel (ii) by refining various techniques that can best capture health / risks assessment for the use of tailings-derived products in different applications.

This thesis assesses the environmental sustainability of tailings reprocessing / valorization from techno-environmental perspectives, using LCA and scenario analysis (**Chapter 3** and **Chapter 4**). In addition to environmental assessments, consideration of the economic feasibility and social consequences of various tailings repurposing routes is necessary. Tools such as techno-economic feasibility and social LCA for emerging technologies can be applied in future studies to account for all aspects of sustainability (Van Schoubroeck et al., 2021).

Figure 5.1 depicts a simplified scheme of tailings valorization connected to challenges and steps toward solutions.

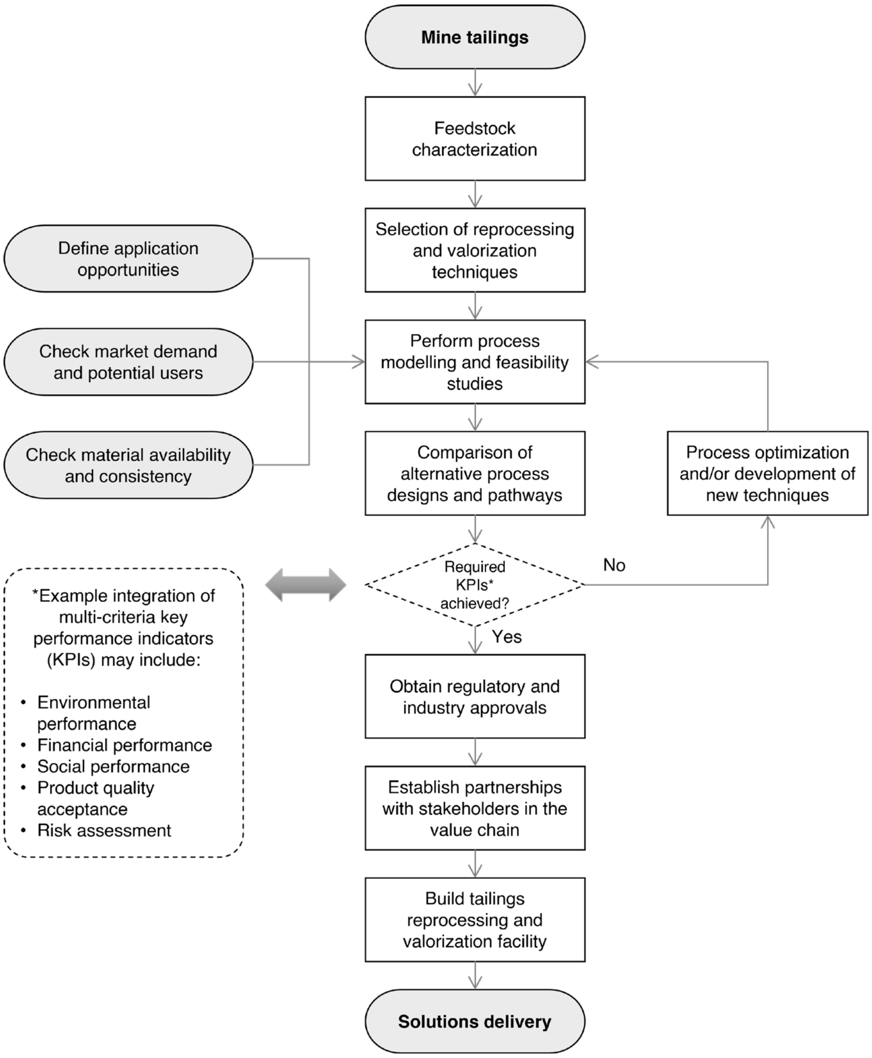

Figure 5.1: Flowchart for mine tailings reprocessing and valorization, from characterization to solution[1]. Modified by combining components from relevant studies (Bardovsky et al., 2021; Sarkkinen et al., 2019; Vuillier & Ingwersen, 2022).

Several practical constraints and challenges for the processing of tailings exist. Examples include (i) difficulty in transportation and logistics for remote mining areas, (ii) lack of a mature market for the repurposing of mine tailings and its derivatives, (iii) inconsistent material properties and availability, (iv) lack of credible track records and identified industrial partnerships [e.g., (Almeida et al., 2020; Kinnunen et al., 2022; Maruthupandian et al., 2021; Sibanda & Broadhurst, 2018)].

Evaluation of emerging technologies involves prospective assessments subject to variability, technological development changes, and uncertainties. The process models and future scenarios developed in **Chapter 4,** specifically rely on the estimates from the industry / market outlooks, established socio-economic scenarios, and LCA database coupled with projections from integrated assessment models / IAMs. These combinations

[1] There may be situations for which the required KPIs are not achieved even after many loops in the decision tree. In this case, the old solution must be discarded, and a new one is proposed.

of inputs provided key insights and parameters for scenario modeling purposes, but their adoption in the simulations, either individually or as a collective, is highly uncertain. Improved scenario models can be utilized in future research to enhance prospective LCAs.

The prospective life cycle inventory modeling depicted in **Chapters 3** and **4** is based on technology upscaling. The new technologies entering the mining sector will evolve and increase their performance, potentially decreasing economic costs and environmental impacts through learning effects (Bergesen & Suh, 2016; Caduff et al., 2012; Thomassen et al., 2020). While technology upscaling is one crucial element to be considered in prospective LCA, learning effects may also contribute to improved future technology performance. However, different learning effects exist, such as learning-by-doing, learning-by-searching, learning-by-interacting, and learning-by-using (Thomassen et al., 2020). The modeling aspects of various learning effects were not considered in this thesis and thus may be investigated by future researchers, when learning paths for different tailings reprocessing technologies are identified.

Regarding technology development, it was assumed that the technologies to reprocess tailings and recover metals and minerals could be scaled up by 2050. Although this time frame is commonly used in many research studies and industry reviews, such as the IEA energy outlook (IEA, 2020) and SSPs (Riahi et al., 2017), there may be deviations. Also, external influences might alter the development trajectory of technologies from predictions. This includes supportive technologies used in the mining industry, such as reprocessing / metallurgical innovations, as well as changes in background systems, such as energy supply. If the timeline matches, the technology is ready to scale up rapidly; thus, alternative mine tailings management could deliver environmental and resource benefits larger than modeled. However, movements in the mining industry market may be slow and diminish the anticipated environmental benefits.

Moreover, environmental benefits gained primarily through material credits might also require updates to the currently implemented substitution modeling approaches, as these approaches borrowed semi-quantitative procedures from consequential LCAs (Weidema et al., 2009). Other dynamic interactions and industry market risks beyond simplified marginal technologies identification, such as accounting for advanced economic models and material trade disruptions, can be integrated into future assessments. Examples of external or industry market risks may include embargos or global disruptions due to trade wars / geopolitical risks, such as recently caused by the Russia-Ukraine conflict, US-China tensions, or the Covid-19 pandemic.

In **Chapter 4**, I used metal demand modeling and projection approaches from Ciacci et al. (2020) and Elshkaki et al. (2018), which did not consider detailed economic constraints in the scenario development. There might be under- / overestimation of projected copper extraction and processing flows in 2050, as the scenarios are based on historical trends and past consumption patterns in a top-down material accounting approach. For instance, in future scenarios, copper consumption for standard applications in Europe was assumed to follow a downward trend based on the dynamic MFA models conducted by Ciacci et al. (2020). This approach may underestimate the future consumption of copper for supporting systems transition. While this thesis tried to solve this issue by adding copper demand for systems transition from the study of Gregoir and van Acker (2022), the storylines derived from mining trends between 2020 and 2050 may differ and thereby necessitate different scenario modeling techniques (Bisinella et al., 2021).

The linking of IAMs with scenario-based LCA is becoming popular in prospective systems analysis. In **Chapter 4**, I used projections data from IAM 'IMAGE' (Stehfest et al., 2014) to forecast the socio-economic changes in background energy systems by 2050, based on shared socio-economic pathways (SSPs). However, there are several alternative SSPs with various radiative concentrating pathways (RCPs) (van Vuuren et al., 2017). In addition to the SSP2-RCP1.9 used in this thesis, one may also implement SSPs in which completely different future trajectories are forecast. Other distinct narratives exist, such as a green growth strategy in SSP1, regional rivalry in SSP3, or a fossil-based economic development in SSP5. Next, in terms of modeling and computational approaches, IMAGE represents one example of well-established IAMs. Numerous choices exist to generate socio-economic forecasts, such as REMIND (Kriegler et al., 2017), MESSAGE (Fricko et al., 2017), and AIM / CGE (Fujimori et al., 2017). Testing the models using different SSPs might also be an interesting way to understand the variability of results caused by different scenarios and forecasting choices. Lastly, the IAM coupling method that I chose was based on the recently developed tool 'premise' for systematic LCA database modifications by Sacchi et al. (2022). Other IAM-LCA linking methods are also available such as using the superstructure approach (Steubing & de Koning, 2021). Various other approaches may emerge to enable the widespread use of future background scenarios in LCA and be used in future research to get more robust scenario assessments.

When calculating the emission budget for the copper mining industry, a relatively straightforward allocation approach or so-called grandfathering was applied for simplicity in **Chapter 4**. The method has its fair share of critics, as past emissions may increase emission entitlements (Knight, 2013). The selection of an accounting approach is a normative choice and still debated (Kulionis et al., 2021). This would imply that the emission target defined in this thesis may change according to different effort-sharing approaches, such as those principles that depend on equality, responsibility, and need / capability (van den Berg et al., 2020). The topic of climate justice remains an open point regarding allocation of the remaining emissions budget (Williges et al., 2022) and will likely affect the copper industry as well.

As should be clear from this section, the actual implications of large-scale mine tailings reprocessing on numerous environmental impacts contain various uncertainties resulting from inputs and methodological choices. Regardless of these complications, it is critical to recognize that the thesis aims to answer research questions from exploratory viewpoints rather than attempting to forecast real events with absolute certainty.

5.4 Outlook

Figure 5.2 presents an overview of the topics covered in this thesis and topics that should be investigated by future research work around mine tailings resource recovery and sustainability assessment.

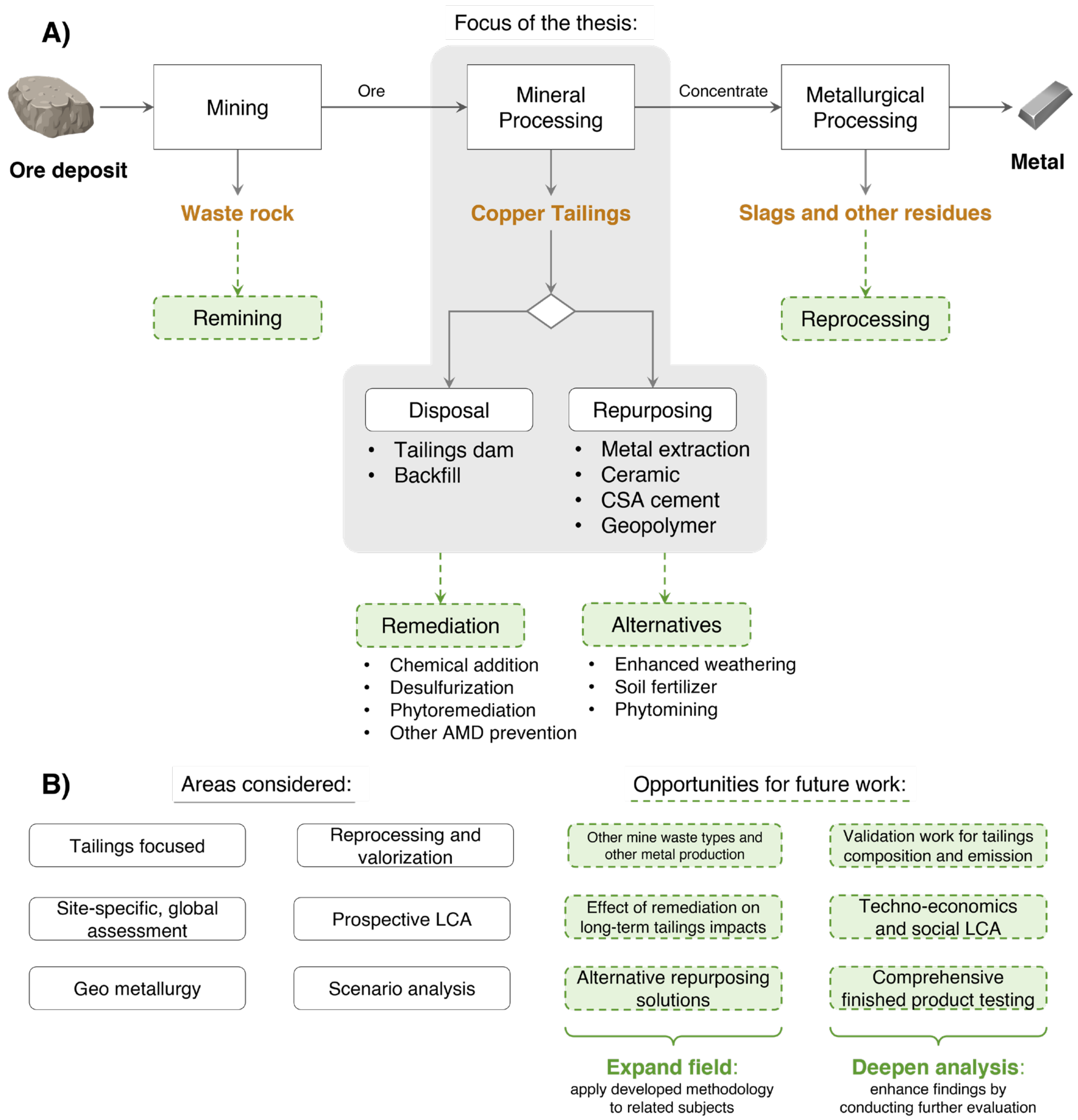

Figure 5.2: A) Structure of the thesis, the focus of the study, and selection of topics for future work (highlighted in dashed green boxes). B) Future research can adapt the developed methodology used in this thesis for other mine waste types, include novel repurposing technologies, and extend the environmental assessment scope. Such integrated analysis can be further enhanced by assessing the socio-economic and techno-environmental impacts of alternative mine waste management options.

The thesis investigated the environmental implications of mine tailings management from a life cycle perspective. The research work conducted in this thesis paves the way for follow-up research projects and provides the initial step for building a comprehensive, global dataset of emission releases and related impacts from metal production (i.e., copper). This allows for the bottom-up regionalized environmental assessment of tailings in copper production. One of the main results from this thesis is a spatial mapping of environmental impacts (mainly toxicity-caused by acid mine drainage potential) to identify the risk management and tailings contamination potential across the globe. Despite recognized limitations, this study's mapping and site-specific assessment can complement analyses of other studies. There has been increasing interest in analytically building maps to characterize mine tailings sites worldwide. Some examples of recent global mapping / database developments are (1) global-scale land use datasets on mining (Maus et al., 2022) and related impacts on biodiversity (Cabernard & Pfister, 2022) and (2) tailings dams risks and failure records (Franks et al., 2021; Islam & Murakami, 2021; Nungesser & Pauliuk, 2022). With the consolidation of high-resolution information for other metals, there are opportunities to extend the contribution of this study for site-specific toxicity assessment of tailings to derive far-reaching insights on mine tailings' potential risks and impacts from a broad environmental perspective.

Although a bottom-up regionalized LCA is a suitable tool for quantifying the environmental impacts of conventional tailings disposal across the globe, other areas should be covered in the future. For example, future research should aim to verify the impact estimation by performing validation work on real-world tailings datasets. These steps are necessary to confirm the prediction performance of the combined approaches of this thesis model. This can be completed in two ways: (1) perform the comparison of the modeled metal concentrations in tailings from this study vs. field measurements, and (2) perform the comparison of the modeled metal concentrations in leachate vs. field measurements. Starting points might be to extract data on mine tailings repositories from available tailings measurement databases, one of which is reported in the extensive geochemistry sampling data of Chilean tailings deposits (SERNAGEOMIN, 2020). Still, one must also recognize the difficulties of data validation involving long-term time horizons. Additionally, the influences of acid mine drainage prevention for different sites during the closure period, should be considered in future studies.

To understand the potential beneficial uses of mine tailings, comparative LCA studies also revealed that innovative reprocessing and valorization technologies could contribute significantly to material supply, thereby potentially leading to environmental benefits. These benefits include reduced environmental impacts of mine tailings management compared to standard landfilling, reduced mine tailings volume, and reduced need for producing virgin materials. These results, assessed through systematic process modeling and prospective LCAs, can be extended to generate additional insights on mine tailings evaluation as potential resources. For instance, the developed methodology can be employed to identify prospects of secondary resources from mine waste, such as the EU Urban Mine Knowledge Platform ProSUM (Huisman et al., 2016). This thesis mainly focuses on copper tailings as the feedstock for further reprocessing, while other types of mine waste, such as waste rock, are also produced in large amounts. While the copper industry globally is responsible for a large generation of tailings, other materials such as gold, iron, and coal rank high in terms of global tailings volume (Oberle et al., 2020). Future research can build on this thesis' assessment methodologies to investigate the life cycle environmental impacts of other relevant mine waste management systems.

Technological solutions to reprocess and valorize copper tailings might change when new, competitive pathways may be considered. Examples of emerging techniques include the reprocessing of tailings and waste rock for soil amendments (Araujo et al., 2022; Swoboda et al., 2022), for carbon sequestering materials (Bullock et al., 2021; Harrison et al., 2013), and for valuable elements via plants phytomining (Akinbile et al., 2021; Corzo Remigio et al., 2020). This wide array of alternative repurposing techniques for different waste types and metal production needs to be assessed in future research in order to find most-suitable solutions to global mining waste problems for specific cases.

Incorporating scenarios into prospective LCA helps to better understand the environmental benefits and tradeoffs of large-scale tailings management in the EU. Implementing alternative tailings management strategies can minimize long-term environmental liabilities in tailings disposal and offer associated environmental benefits through the supply of secondary resources. Yet, the success behind reprocessing tailings and circular utilization of mine tailings is constrained by some limiting factors. One of the main barriers to technology and market diffusion is the limited availability of ingredients for alternative cement production, such as bauxite and sodium silicate (Habert et al., 2020). Another limitation is the market readiness for secondary products made with tailings materials. These challenges would undermine or even prevent rapid and mass adoption of sustainable utilization of mine tailings for resource management and environmental impact mitigation. Industry regulations and initiatives may speed up these practices eventually. However, at the same time, the beneficial uses of mine tailings-based products should be based on sound science and environmental considerations. Future scholars could focus on validating the safe and sustainable use of such materials by expanding the scope of the presented methods in conjunction with relevant regulatory standards and health-based benchmarks. Examples include the critical evaluation of constituents of potential concerns from tailings-based products during the use and disposal phases. One can extend the research work of Helser et al. (2022) to translate their leaching experiment data into life cycle inventories during use and end-of-life stages.

In anticipation of the extensive decarbonization of the mining industry needed to achieve global climate goals, it is imperative to apply interdisciplinary research into sustainable resource management. In the coming decades, sustainable materials supply is expected to play a strategic role in achieving industry-wide climate commitments. While significant material and environmental opportunities are present in copper tailings repurposing, complementary GHG mitigation measures are necessary for closing the emission reduction gaps. Improving resource efficiencies alongside electrification of the power sector and technology improvements may offer sizable contributions to meeting the emission targets. This also confirms that no single solution can solve the sectoral problem; one must look at every step of the processing chain comprising different players to identify levers that reduce impacts and create value. This thesis has illustrated and drawn attention to the importance of this task using case studies and applied industrial ecology research, which may eventually lead to the creation of appropriate policy measures and practices. Overall, this research work can contribute to understanding and quantifying the environmental consequences of tailings management in the metal and mining industries.

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Appendix A

Regional life cycle inventories of global sulfidic copper tailings

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Supplementary information for the following paper:

Regionalized Life Cycle Inventories of Global Sulfidic Copper Tailings in ***Environmental Science & Technology*** (2022). Link.

 Digital SI with spreadsheet tables (referred to as Appendix-2) can be accessed online through this link.

A. Supplementary information for Chapter 2

A.1 Copper sites coverage in this study

Table A1. Global copper deposits coverage in terms of production, ore tonnage, and grades around the world

Ore deposit type	Production in Mt	Cumulative Production (% total of this study)	Nr. of sites	Ranges of Tonnage (median) in Mt	Ranges of Cu-grades (median) in %w	Important countries or regions
Porphyry	11.8	61%	202	0.6 – 21, 277 (288)	0.1 – 1.5 (0.6)	China, North America, Latin America
Sediment-hosted Stratiform/ Strata bound Cu	3.9	20%	45	0.9 – 3,230 (72)	0.46 – 4.38 (1.3)	USA, Poland, Central Africa
Volcanogenic massive sulfide (VMS)	1.6	8%	65	1 – 334 (23)	0.19 – 4.7 (1)	Canada, Australia, Europe
Skarn	1.1	6%	29	0.4 – 10, 060 (72)	0.07 – 2.19 (1.3)	China, Latin America
Iron-oxide Cu-Au (IOCG)	0.5	3%	15	3.2 – 329 (100)	0.07 – 3.09 (0.7)	Australia, Latin America
Magmatic sulfide	0.4	2%	52	1.8 – 3,709 (36)	0.01 – 1.82 (0.4)	China, USA, Central Africa
Epithermal	0.1	1%	20	0.3 – 130 (18)	0.3 – 1.83 (0.7)	Latin America, Asia
Intrusion-related Au	0.02	0.1%	1	152	0.1%	Australia
Orogenic Au	0.02	0.1%	2	2.9 – 63 (33)	0.8 – 0.9 (0.9)	Australia, China
Total copper production collected in this study (2019)	**15.1**	**75% coverage in 2019**	**431**			
Annual global copper production based on S&P Global Market Intelligence – Mining and Metal 2019 data (S&P, 2020)	**20.5**					

Note: A complete table to derive this summary is provided in Appendix-2 of the online manuscript. It comprises a list of 431 project IDs.

A.2 Classification of copper production process

Based on the worldwide copper production practice and its classification (G. M. Mudd et al., 2013), copper production can involve a variety of approaches. Generally, it can be divided into pyrometallurgical and hydrometallurgical routes (Ayres et al., 2002; Norgate & Jahanshahi, 2010), as described in the following:

- **Pyrometallurgical route (the focus of this study)**

Mining-Flotation-Smelting-Refining – an open pit or underground mine extracts Cu ore (with a grade of 0.4-2% Cu), which is then processed in a flotation plant to produce a Cu-rich concentrate (typically 20-25% Cu) which is then smelted to produce Cu anode (say 95-98% Cu), in turn processed in a refinery to produce high purity Cu (>99.9% Cu).

  - Examples: Australia, Canada, USA, Chile, etc.
  - Around 80% of global mined Cu is produced using this technology

- **Hydrometallurgical route**

Mining-Heap Leaching-Refining – an open pit mine extracts Cu ore (the grade of 0.4-1% Cu) which is piled in large heaps, sulfuric acid is irrigated across this heap and the solution percolates through it and is captured at the bottom rich in Cu, where it is pumped to a refinery to produce high purity refined Cu.

  - Examples: USA, Chile, etc.
  - About 20% of global mined Cu is produced this way
  - Heap leach projects are mostly based on oxide ores

Mining-Acid Leaching-Refining – an open pit or underground mine extracts Cu ore (with the grade of 1-4%Cu), which is then processed in a plant using acid to directly dissolve the Cu together with the other metals, and the solutions refined to produce high purity Cu.

  - Examples: Zambia, Laos, Democratic Republic of the Congo (DRC), etc.
  - About 1-2% of global mined Cu is produced this way

A.3 Mineral process modelling in HSC Sim©

HSC Simulation v10 (Metso Outotec, 2020) for nine types of deposit, with simplified flowchart being drawn (Figure A1 and Figure A2). We used similar models as done by Michaux and Reuter's three-model component (Ferreira & Loveday, 2000; Michaux et al., 2019). The flotation parameters and recovery data from handbook (Bulatovic, 2007; King, 2001; Wills & Finch, 2016a) are then supplied to the simulator to obtain results for each deposit type. Figure A3 presents how the tailings properties for specific ore deposits and the calculation is applicable generally.

Appendix A

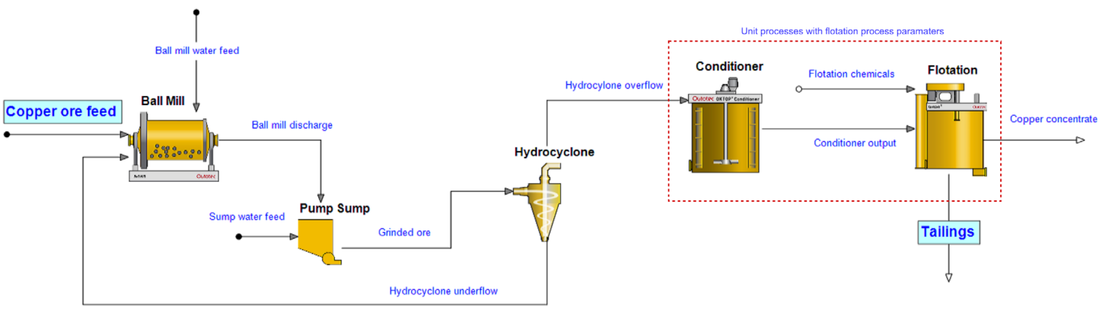


Figure A1. A simplified Outotec© HSC Sim 10 flowsheet for copper beneficiation process with ore deposit as input. Red dashed box indicates the main flotation controls in the simulation.

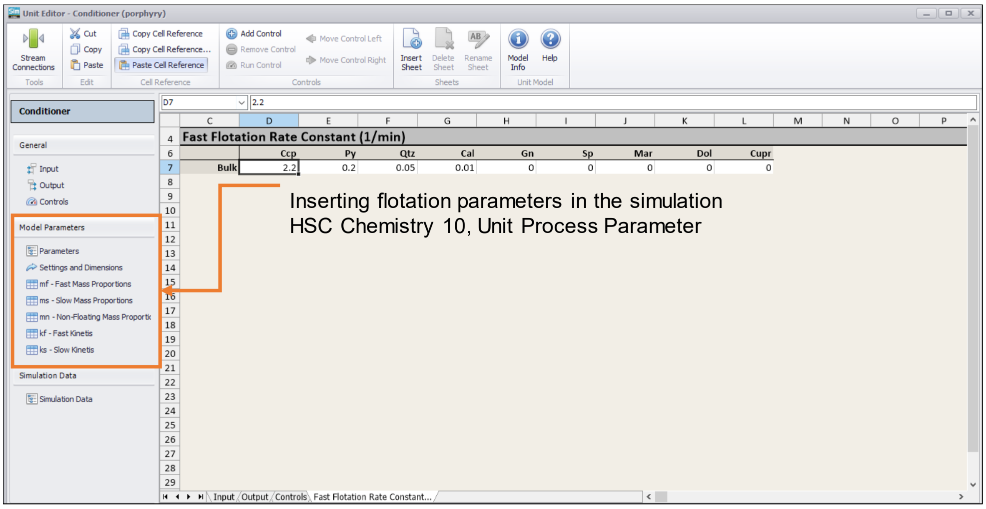


Figure A2. An example of flotation unit module in HSC Chemistry 10. Parameterizations are performed by accessing model tabs on the left.

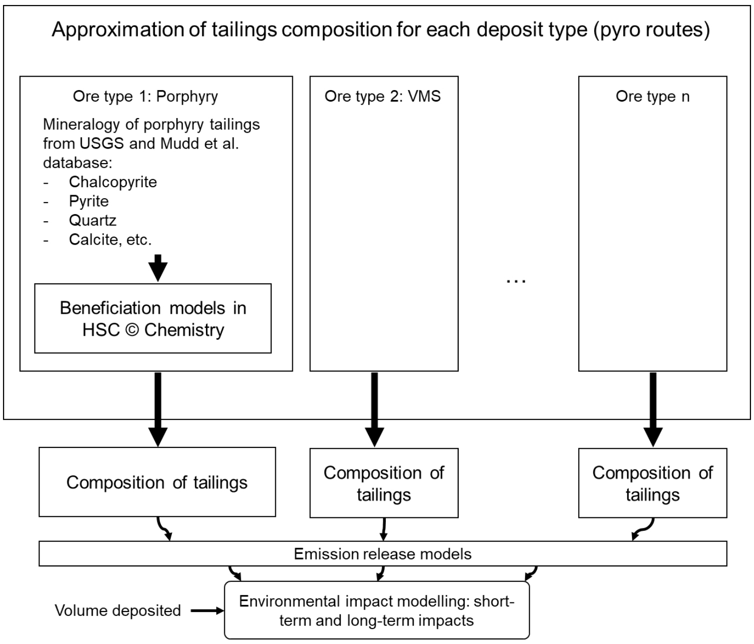


Figure A3. Method for estimating the tailings composition from different ore deposit types

Table A2. Flotation kinetic parameters and data sources used in the beneficiation modelling

Copper deposit type			*Porphyry*	*Sediment-hosted Stratiform/ Strata bound Cu*	*Volcanogenic massive sulfide (VMS)*	*Skarn*	*Iron-oxide Cu-Au (IOCG)*	*Magmatic sulfide*	*Epithermal*	*Intrusion-related Au and orogenic Au*
Sulfide minerals			Chalcopyrite (Ccp)	Chalcopyrite (Ccp)	Chalcopyrite (Ccp)	Chalcopyrite (Ccp)	Cuprite (Cupr)	Chalcopyrite (Ccp)	Chalcopyrite (Ccp)	Chalcopyrite (Ccp)
Gangue sulfide minerals			Pyrite (Py)	Pyrite (Py), Galena (Gn), Sphalerite (Sp), Marcasite (Mar)	Pyrite (Py), Galena (Gn), Sphalerite (Sp)	Pyrite (Py)	-	Pyrite (Py), Galena (Gn), Sphalerite (Sp)	Pyrite (Py), Galena (Gn), Sphalerite (Sp)	Pyrite (Py)
Other gangue minerals			Quartz (Qtz), calcite (Cal)	Quartz (Qtz), calcite (Cal)	Quartz (Qtz), calcite (Cal)	Quartz (Qtz), calcite (Cal)	Quartz (Qtz), dolomite (Dol)	Quartz (Qtz), calcite (Cal)	Quartz (Qtz), calcite (Cal)	Quartz (Qtz), calcite (Cal)
Flotation kinetics parameters (m_n is calculated via mass balance)	m_f	***Ccp***	9.10E-01	1.62E+00	1.71E+00	1.55E+00	-	1.80E+00	1.26E+00	1.09E+00
		Py	1.00E-01	1.36E-01	1.44E-01	1.28E-01	-	8.80E-02	8.00E-02	1.10E-01
		Qtz	6.00E-02	6.50E-02	9.50E-02	8.40E-02	7.20E-02	7.50E-02	1.00E-01	7.20E-02
		Cal	2.00E-02	4.50E-02	5.40E-02	3.80E-02	-	4.80E-02	4.50E-02	2.80E-02
		Gn	-	2.80E-02	3.80E-02	-	-	3.20E-02	3.40E-02	-
		Sp	-	1.18E+00	1.82E+00	-	-	1.46E+00	1.37E+00	-
		Mar	-	1.12E-01	-	-	-	-	-	-
		Cupr	-	-	-	-	1.28E+00	-	-	-
		Dol	-	-	-	-	2.00E-02	-	-	-
	m_s	***Ccp***	4.00E-02	1.12E-01	1.52E-01	6.40E-02	-	1.20E-01	1.52E-01	6.00E-02
		Py	1.50E-02	2.40E-02	3.15E-02	1.65E-02	-	1.95E-02	2.55E-02	1.65E-02
		Qtz	1.00E-02	1.12E-02	1.68E-02	1.40E-02	1.10E-02	1.44E-02	8.00E-03	1.10E-02
		Cal	-	6.40E-03	7.60E-03	1.20E-02	0.00E+00	4.40E-03	5.20E-03	-
		Gn	-	9.00E-03	9.50E-03	-	-	6.50E-03	8.00E-03	-
		Sp	-	7.60E-02	8.40E-02	-	-	7.20E-02	6.40E-02	-
		Mar	8.00E-03	1.80E-02	-	-	-	-	-	9.60E-03
		Cupr	-	-	-	-	6.00E-02	-	-	-
		Dol	-	-	-	-	9.60E-03	-	-	-
	k_f	***Ccp***	2.20E+00	3.84E+00	6.08E+00	3.30E+00	-	5.76E+00	4.16E+00	3.30E+00
		Py	2.00E-01	1.20E-01	2.10E-01	3.20E-01	-	1.90E-01	1.30E-01	3.00E-01
		Qtz	5.00E-02	5.60E-02	7.20E-02	9.00E-02	6.00E-02	5.60E-02	6.00E-02	5.00E-02
		Cal	1.00E-02	1.52E-02	1.60E-02	1.20E-02	-	1.60E-02	1.60E-02	1.20E-02
		Gn	-	9.50E-03	9.50E-03	-	0.00E+00	6.50E-03	8.00E-03	-
		Sp	-	4.62E+00	6.60E+00	-	0.00E+00	5.60E-03	6.30E-03	-
		Mar	-	1.40E-01	-	-	0.00E+00	0.00E+00	0.00E+00	-
		Cupr	-	-	-	-	4.32E+00	-	-	-
		Dol	-	-	-	-	1.20E-02	-	-	-
	k_s	***Ccp***	2.00E-02	1.10E-02	1.80E-02	3.80E-02	-	1.20E-02	2.00E-02	2.60E-02

	Py	1.00E-02	6.50E-03	1.00E-02	1.50E-02	1.50E-02	6.00E-03	1.00E-02	1.40E-02
	Qtz	1.00E-03	6.00E-04	1.00E-03	1.80E-03	-	6.00E-04	6.00E-04	1.30E-03
	Cal	1.00E-02	5.40E-03	6.00E-03	1.90E-02	-	3.60E-03	5.70E-03	1.50E-02
	Gn	-	1.60E-04	2.00E-04	-	-	1.50E-04	1.90E-04	-
	Sp	-	2.40E-02	3.80E-02	-	-	4.00E-02	2.20E-02	-
	Mar	-	7.50E-04	-	-	-	-	-	-
	Cupr	-	-	-	-	3.20E-02	-	-	-
	Dol	-	-	-	-	1.50E-03	-	-	-
Reagents/ ancillaries		Cu recovery ranges from 80 - 92%; Xanthate as the main collector with basic pH conditions	Cu recovery ranges from 78-87%; Reagents for bulk process: CaO 700 g/t, Na2SO3 800 g/t	Cu recovery ranges from 81-95%; Reagents for bulk process: $Na_2S_2O_5$ 2000 g/t, CaO (pH 7.5), HQS 800 g/t	Cu recovery ranges from 80 - 92%; Behaved similar to porphyry and thus modelled as is, as inferred from the literatures	Cu recovery ranges from 75-84%; Xanthate as the main collectors. Used sulfidizers Na_2SiO_3 = 300 g/t to treat dolomitic ores	Cu recovery ranges from 85-95%; Reagents: $Ca(OH)_2$ 600 g/t, Na_2SO_3 500 g/ t	Cu recovery ranges from 82-90%; Reagents: $Ca(OH)_2$ = pH 9.5, NaCN = 300 g/t	Cu recovery ranges from 88-95%; Reagents: Na_2S 30 g/t
Main reference "Handbook of Flotation Reagents" (Bulatovic, 2007), **Mineral Processing Technology** (Wills & Finch, 2016a) **and additional remarks**		1) Handbook of Flotation Reagents chapter 12, 2) Environmental Attributes and Resource Potential of Mill Tailings from Diverse Mineral Deposit Types(Seal & Nadine, 2017)	1) Handbook of Flotation Reagents chapter 15, 2) Other plants data and literatures(Bakalarz, 2019; Martín-Crespo et al., 2020; Nadeif et al., 2019)	1) Handbook of Flotation Reagents chapter 14	1) Handbook of Flotation Reagents chapter 12, 2) Other literatures(Hällström et al., 2018; Misra, 2000)	1) Handbook of Flotation Reagents chapter 19, 2) Other resource(Craw et al., 2015)	1) Handbook of Flotation Reagents chapter 13, 2) Other literatures(Placencia-Gómez et al., 2010; Schulz et al., n.d.; Seal & Nadine, 2017)	1) Handbook of Flotation Reagents chapter 13, 2) Other literatures(Forsythe et al., 2019; Triantafyllidis et al., 2007)	1) Handbook of Flotation Reagents chapter 17, 2) Environmental Attributes and Resource Potential of Mill Tailings from Diverse Mineral Deposit Types(Seal & Nadine, 2017), 3) Other sources(Craw et al., 2015; Velásquez et al., 2020)

Table A3. Generic chemical compositions taken from HSC Geo Database

Elements	**Chalcopyrite**	**Pyrite**	**Quartz**	**Calcite**	**Galena**	**Sphalerite**	**Marcasite**	**Cuprite**
Cu	34.63							88.82
Fe	30.43	46.55					46.55	
S	34.94	53.45			13.4	32.91	53.45	
O			53.26	47.96				11.18
Si			46.74					
C				12				
Ca				40.04				
Pb					86.6			
Zn						67.09		
Mg								

A.4 Additional details for copper compositions

The following examples are the procedures of how the generic composition of porphyry deposits are adjusted to the copper grade. The HSC Sim Element to Mineral Conversion "Geomodule" is used to convert the composition of ore deposit input mineralogy to achieve the target grade (in the example below, Cu-grade of 0.5%).

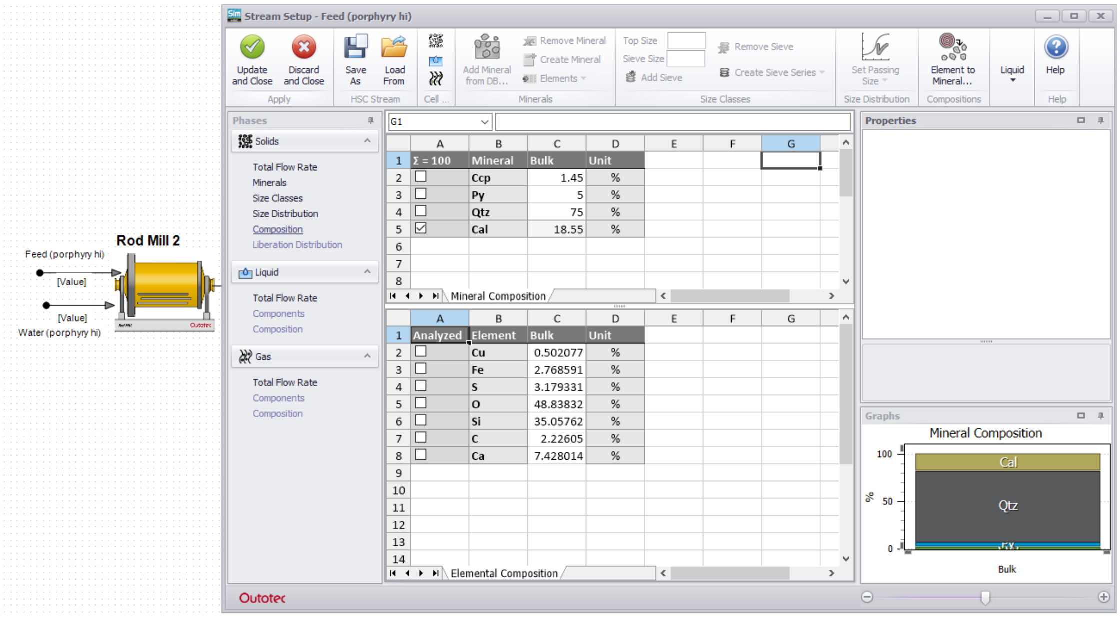


Figure A4. Mineral to Element Conversion in HSC Sim 10

The buffering capacity (i.e., calcite) and other inert mineral (i.e., quartz) are used to equalize the compositions when copper grade increases or decreases (Figure A5). These are reflected in the output tailings after running the flotation processes of the circuit. Based on information from Heinrich and Candela (2013), the growth of ore zone at the center determines the amounts of quartz and pyritic minerals (in this case pyrite and chalcopyrite). It implies that the more copper we have at the core in the sulfidic regions, the larger the composition of quartz, pyrite, and chalcopyrite. This is also in line with the cross-section picture of the deposit, showing that the propylitic layer that contains calcite and other minerals are only present in the outer layer of copper deposit according to the preliminary model of copper deposits by USGS (Berger et al., n.d.). Hence, we assumed that whenever copper grade increases, the buffering minerals will decrease.

	Cu grade = 0.5%		
	Mineral	Content	
		%w	mol/kg
Main	Calcite	18.06	1.80560
	Chalcopyrite	1.45	0.26613
	Pyrite	5.07	0.60739
	Quartz	75.00	4.50000
Traces	Ca(OH)2	0.3000	0.02220
	Cd(OH)2	0.0121	0.00177
	Zn(OH)2	0.0967	0.00967
	Pb(OH)2	0.0011	0.00028
	Cu(OH)2	0.0126	0.00123
	As, on OH sites	0.0026	0.00003
	Secondary minerals		
	CdCO3	Secondary mineral	
	ZnCO3	Secondary mineral	
	CuCO3	Secondary mineral	
	PbCO3	Secondary mineral	
	CdO	Secondary mineral	
	PbO	Secondary mineral	
	ZnO	Secondary mineral	
	Cu2O	Secondary mineral	
	constant, assumed		

down

	Cu grade = 0.1%		
	Mineral	Content	
		%w	mol/kg
Main	Calcite	25.23	1.91200
	Chalcopyrite	0.29	0.00734
	Pyrite	4.77	0.57145
	Quartz	69.4	4.53000
Traces	Ca(OH)2	0.3000	0.02220
	Cd(OH)2	0.0003	0.00005
	Zn(OH)2	0.0027	0.00027
	Pb(OH)2	0.0002	0.00001
	Cu(OH)2	0.0025	0.00351
	As, on OH sites	0.0026	0.00003
	Secondary minerals		
	CdCO3	Secondary mineral	
	ZnCO3	Secondary mineral	
	CuCO3	Secondary mineral	
	PbCO3	Secondary mineral	
	CdO	Secondary mineral	
	PbO	Secondary mineral	
	ZnO	Secondary mineral	
	Cu2O	Secondary mineral	

up

	Cu grade = 1%		
	Mineral	Content	
		%w	mol/kg
Main	Calcite	8.60	1.07700
	Chalcopyrite	2.90	0.08076
	Pyrite	5.34	0.63973
	Quartz	82.8	4.96800
Traces	Ca(OH)2	0.3000	0.02220
	Cd(OH)2	0.0037	0.00054
	Zn(OH)2	0.0293	0.00293
	Pb(OH)2	0.0023	0.00001
	Cu(OH)2	0.0252	0.00351
	As, on OH sites	0.0026	0.00003
	Secondary minerals		
	CdCO3	Secondary mineral	
	ZnCO3	Secondary mineral	
	CuCO3	Secondary mineral	
	PbCO3	Secondary mineral	
	CdO	Secondary mineral	
	PbO	Secondary mineral	
	ZnO	Secondary mineral	
	Cu2O	Secondary mineral	

Figure A5. The changes of composition for porphyry copper deposits feed when the grades vary (an example for illustrative purposes)

For porphyry copper deposits, they are assumed to contain similar mineralogy as skarn (Misra, 2000), iron oxide (Craw et al., 2015), intrusion related Au (Craw et al., 2015) deposits. These are simplifications to include the major primary minerals that are often present in various deposit types and are reflected in the flotation handbook by Bulatovic (2007). There, volcanogenic massive sulfide, sediment-hosted, magmatic sulfide and epithermal may contain additional minerals that might affect the overall beneficiation schemes and flotation mechanisms due to the presence of galena (Pb-containing), sphalerite (Zn-containing), and marcasite (sources of FeS) in the ore growth zone. We repeated the similar procedures to the other deposits as shown in Figure A5, Figure A5, and HSC Mineral to Element Conversion in "Geomodule" by also adding the presence of the previously mentioned minerals.

A.5 Further descriptions of PHREEQC modelling and input parameters

We used the coupled transport and reaction model PHREEQC and WATEQ4F (Ball & Nordstrom, 1991; Parkhurst & Appelo, 2013) to model heavy metal emissions from tailings deposits as a function of time. The parameters for the models are tabulated in Table A4, whereas the chemical equations are listed in Table A13. The model predicts the emissions of metals over time, following the development of pH and mineralogy changes as the solid fractions interact with the infiltrating water and simulating simple percolation scenarios. In order to do this, 1 kg of initial mineral assemblages containing a number of solubilities controlling minerals were defined. A single-cell column containing the mineral assemblage was set-up and water equilibrated with atmospheric CO_2 was shifted through the column to simulate percolation. For simplicity, only 1-D transport of eluent/ forward flow was used, while neither dispersivity nor dual-porosity were considered. Detailed explanations about this approach can be found in other prediction-based and long-term leaching studies (Hyks, 2008; Hyks et al., 2009; Sieland et al., 2016; Tiruta-Barna, 2008).

Minerals such as calcite control the rate of heavy metal leaching due to its buffering mechanism. Thus, majority of metal emissions in this study was limited by mineral dissolution and precipitation processes. The other kinetically controlled mechanisms such as microbial activities and pyrite oxidations are therefore excluded for such a long-term assessment (Chen et al., 2014). However, arsenic was modelled differently following the double-layer complexation and strong sorption on iron oxide surfaces as prescribed by Dzombak and Morel (Dzombak & Morel, 1990) and Appelo et al. (Appelo et al., 2002). Activity coefficients were adjusted for an ionic strength calculated from the component's concentrations according to Debye-Hueckel (Parkhurst et al., 2013). The initial mineralogical compositions or assemblages of the tailings are shown in Table A5-Table A12. We compiled a set of relevant reactions (e.g., mineral dissolution/ precipitation and surface complexation) from the main PHREEQC database, complemented with the other geochemical database such as WATEQ4F and Dzombak-Morel layer model. It was not intended to model the complete mineral composition but to include some major species relevant for heavy metal solubility. The base sealing and the drainage system of the storage facilities are generally estimated to endure less than 100 years (Rowe & Islam, 2009). Therefore, the technical barrier is only relevant for short-term emissions and will not prevent a large fraction of long-term emissions from entering the subsoil and the groundwater compartment. Hence, the technical barrier system was neglected in the present analysis.

Table A4. Parameters used in PHREEQC simulations

Parameter	**Value**	**Unit**
Infiltration rate	Annual groundwater recharge from PCR-GLOBWB (see section 3.5)	l/m2*a
Partial pressure of CO_2	$10^{-3.5}$	atm
Time step	1	year
Porosity	Ranges of values between 40 – 55 based on literatures (Gitari et al., 2018; Gorakhki & Bareither, 2017; Mahmood & Elektorowicz, 2020; Shamsai et al., 2007). 50 is chosen for the simulations	%
Tailings thickness (d)	20 – average: sources of data (Lyu et al., 2019; Porter & Bleiwas, 2003)	m
Tailings density (ρ)	2200 – average: primary industry's (SULTAN, 2019) data and literature sources (Hu et al., 2017; Shamsai et al., 2007)	kg/m3
Solution pH	5	
Temperature	25 – median for stored tailings: partner's data (SULTAN, 2019)	

Table A5. Initial composition of tailings from porphyry deposit

Mineral	**Content**	
	Wt %	**(mole/kg)**
Calcite	1.60E+01	1.60E+00
Chalcopyrite	1.56E-01	2.86E-02
Pyrite	5.07E+00	6.07E-01
Quartz	7.85E+01	4.71E+00
$Ca(OH)_2$	3.00E-01	2.22E-02
$Cd(OH)_2$	1.30E-03	1.90E-04
$Zn(OH)_2$	1.04E-02	1.04E-03
$Pb(OH)_2$	1.13E-04	2.73E-05
$Cu(OH)_2$	1.36E-03	1.32E-04
As, on hydroxide sites	2.58E-03	3.45E-05
$CdCO_3$, $ZnCO_3$, $CuCO_3$, $PbCO_3$, CdO, PbO, ZnO, Cu_2O	secondary mineral	

Compositions taken from HSC Sim tails output

Table A6. Initial composition of tailings from volcanogenic massive sulfide deposit

Mineral	**Content**	
	Wt %	**(mole/kg)**
Calcite	6.19E+00	6.19E-01
Chalcopyrite	4.28E-01	7.85E-02
Galena	5.57E+00	1.34E+00
Pyrite	1.05E+01	1.26E+00
Quartz	7.66E+01	4.60E+00
Sphalerite	4.00E-01	3.88E-02
$Ca(OH)_2$	2.50E-01	1.85E-02
$Cd(OH)_2$	7.44E-05	1.09E-05
$Zn(OH)_2$	8.67E-03	8.67E-04
$Pb(OH)_2$	1.64E-03	3.95E-04
$Cu(OH)_2$	3.72E-03	3.63E-04
As, on hydroxide sites	3.24E-03	4.32E-05
$CdCO_3$, $ZnCO_3$, $CuCO_3$, $PbCO_3$, CdO, PbO, ZnO, Cu_2O	secondary mineral	

Compositions taken from HSC Sim tails output

Table A7. Initial composition of tailings from skarn deposit

Mineral	Content	
	Wt %	(mole/kg)
Calcite	1.26E+01	1.26E+00
Chalcopyrite	1.65E-01	3.02E-02
Pyrite	5.90E+00	7.07E-01
Quartz	8.11E+01	4.87E+00
$Ca(OH)_2$	1.80E-01	1.33E-02
$Cd(OH)_2$	6.22E-04	9.10E-05
$Zn(OH)_2$	4.51E-02	4.51E-03
$Pb(OH)_2$	2.27E-03	5.48E-04
$Cu(OH)_2$	2.50E-02	2.44E-03
As, on hydroxide sites	4.63E-04	6.17E-06
$CdCO_3$, $ZnCO_3$, $CuCO_3$, $PbCO_3$, CdO, PbO, ZnO, Cu_2O	secondary mineral	

Compositions taken from HSC Sim tails output

Table A8. Initial composition of tailings from sediment hosted deposit

Mineral	Content	
	Wt %	(mole/kg)
Calcite	1.47E+01	1.47E+00
Chalcopyrite	7.02E-01	1.29E-01
Galena	2.63E+00	6.31E-01
Marcasite	3.74E+00	4.49E-01
Pyrite	9.88E+00	1.18E+00
Quartz	6.50E+01	3.90E+00
Sphalerite	2.72E+00	2.64E-01
$Ca(OH)_2$	1.60E-01	1.18E-02
$Cd(OH)_2$	6.11E-03	8.94E-04
$Zn(OH)_2$	4.39E-01	4.39E-02
$Pb(OH)_2$	2.78E-03	6.70E-04
$Cu(OH)_2$	6.11E-03	5.96E-04
As, on hydroxide sites	4.97E-03	6.62E-05
$CdCO_3$, $ZnCO_3$, $CuCO_3$, $PbCO_3$, CdO, PbO, ZnO, Cu_2O	secondary mineral	

Compositions taken from HSC Sim tails output

Table A9. Initial composition of tailings from magmatic sulfide deposit

Mineral	**Content**	
	Wt %	**(mole/kg)**
Calcite	7.29E+00	7.29E-01
Chalcopyrite	1.76E-01	3.22E-02
Galena	1.06E+01	2.54E+00
Pyrite	5.00E+00	5.99E-01
Quartz	7.20E+01	4.32E+00
Sphalerite	4.83E+00	4.69E-01
$Ca(OH)_2$	1.20E-01	8.88E-03
$Cd(OH)_2$	1.97E-03	2.89E-04
$Zn(OH)_2$	4.64E-02	4.64E-03
$Pb(OH)_2$	6.94E-04	1.67E-04
$Cu(OH)_2$	1.53E-03	1.49E-04
As, on hydroxide sites	1.56E-05	2.08E-07
$CdCO_3$, $ZnCO_3$, $CuCO_3$, $PbCO_3$, CdO, PbO, ZnO, Cu_2O	secondary mineral	

Compositions taken from HSC Sim tails output

Table A10. Initial composition of tailings from iron oxide deposit

Mineral	**Content**	
	Wt %	**(mole/kg)**
Calcite	1.30E+01	1.30E+00
Chalcopyrite	2.44E-01	4.47E-02
Pyrite	2.97E+00	3.56E-01
Quartz	8.36E+01	5.01E+00
$Ca(OH)_2$	1.20E-01	8.88E-03
$Cd(OH)_2$	2.17E-03	3.18E-04
$Zn(OH)_2$	7.86E-02	7.86E-03
$Pb(OH)_2$	2.35E-04	5.68E-05
$Cu(OH)_2$	2.12E-03	2.07E-04
As, on hydroxide sites	1.77E-03	2.35E-05
$CdCO_3$, $ZnCO_3$, $CuCO_3$, $PbCO_3$, CdO, PbO, ZnO, Cu_2O	secondary mineral	

Compositions taken from HSC Sim tails output

Table A11. Initial composition of tailings from intrusion related deposit

Mineral	Content	
	Wt %	(mole/kg)
Calcite	1.06E+01	1.06E+00
Chalcopyrite	1.53E-01	2.80E-02
Pyrite	2.91E+00	3.49E-01
Quartz	8.62E+01	5.17E+00
$Ca(OH)_2$	1.50E-01	1.11E-02
$Cd(OH)_2$	9.78E-04	1.43E-04
$Zn(OH)_2$	5.26E-02	5.26E-03
$Pb(OH)_2$	1.28E-04	3.10E-05
$Cu(OH)_2$	1.33E-03	1.29E-04
As, on hydroxide sites	1.21E-03	1.61E-05
$CdCO_3$, $ZnCO_3$, $CuCO_3$, $PbCO_3$, CdO, PbO, ZnO, Cu_2O	secondary mineral	

Compositions taken from HSC Sim tails output

Table A12. Initial composition of tailings from epithermal deposit

Mineral	Content	
	Wt %	(mole/kg)
Calcite	5.87E+00	5.87E-01
Chalcopyrite	1.65E-01	3.03E-02
Galena	5.44E+00	1.31E+00
Pyrite	1.03E+01	1.23E+00
Quartz	7.72E+01	4.63E+00
Sphalerite	4.00E-01	3.88E-02
$Ca(OH)_2$	1.10E-01	8.14E-03
$Cd(OH)_2$	5.96E-03	8.73E-04
$Zn(OH)_2$	1.52E-01	1.52E-02
$Pb(OH)_2$	2.77E-01	6.69E-02
$Cu(OH)_2$	8.94E-02	8.72E-03
As, on hydroxide sites	2.37E-03	3.16E-05
$CdCO_3$, $ZnCO_3$, $CuCO_3$, $PbCO_3$, CdO, PbO, ZnO, Cu_2O	secondary mineral	

Compositions taken from HSC Sim tails output

Table A13. Geochemical thermodynamic reactions included in the simulation from PHREEQC database (Parkhurst et al., 2013) and WATEQ4F database (Ball & Nordstrom, 1991)

Solid phases (bold letters)	
Chemical equilibrium of precipitation / dissolution reactions	log K_{s0}
$CaCO_3$ = Ca^{2+} + CO_3^{2-}	-8.48
$CaSO_4.2H_2O$ = Ca^{2+}+ SO_4^{2-} + 2 H_2O	-4.58
$Ca(OH)_2$ + 2 H^+ = Ca^{2+} + 2 H_2O	22.8
$Cd(OH)_2$ + 2 H^+ = Cd^{2+} + 2 H_2O	20.19
$Zn(OH)_2$ + 2 H^+ = Zn^{2+} + 2 H_2O	17.82
$Cu(OH)_2$ + 2 H^+ = Cu^{2+} + 2 H_2O	16.2
$Pb(OH)_2$ + 2 H^+ = Pb^{2+} + 2 H_2O	16.94
$Al(OH)_3$ + 3 H^+ = Al^{3+} + 3 H_2O	8.11
$CuCO_3$ = Cu^{2+} + CO_3^{2-}	-9.63
$ZnCO_3$ = Zn^{2+} + CO_3^{2-}	-10.0
$CdCO_3$ = Cd^{2+} + CO_3^{2-}	-12.1
$PbCO_3$ = Pb^{2+} + CO_3^{2-}	-13.1
CdO + 2 H^+ = Cd^{2+} + H_2O	13.8
PbO + 2 H^+ = Pb^{2+} + H_2O	12.9
ZnO + 2 H^+ = Zn^{2+} + H_2O	11.1
Pyrite reactions	
FeS_2 + $\frac{7}{2}$ O_2 + H_2O = Fe^{+2} + 2 SO_4^{2-} + 2 H^+	
FeS_2 + 2 H^+ + 2 $^{e-}$ = Fe^{+2} + 2 HS^-	-18.48
Arsenic complexation on HFO (Dzombak & Morel, 1990)	
Acid base reactions	
$AsO4^{3-}$ + H^+ = $HAsO_4^{2-}$	11.6
$AsO4^{3-}$ + $2H^+$ = $H_2AsO_4^-$	18.35
$AsO4^{3-}$ + $3H^+$ = H_3AsO_4	20.6
Surface reactions	
SurfOH + H^+ = $SurfOH_2^+$	7.29
SurfOH = SurfO- + H^+	-8.93
SurfOH + AsO_4^{3-} + $3H^+$ = $SurfH_2AsO_4$ + H_2O	29.31
SurfOH + AsO_4^{3-} + $2H^+$ = $SurfHAsO_4^-$ + H_2O	23.51
SurfOH + AsO_4^{3-} = $SurfOHAsO_4^{3-}$	10.58

The following figures show the model output of simulations for the various types of tailings. It can be seen that some metals like Zn and Cu (and partially Pb) in the leachate surpass threshold concentrations and hence represent a risk to the environment. We also display the threshold values in Figure A6-Figure A13, taken from EU groundwater leachate discharge values (European Commission, 2009).

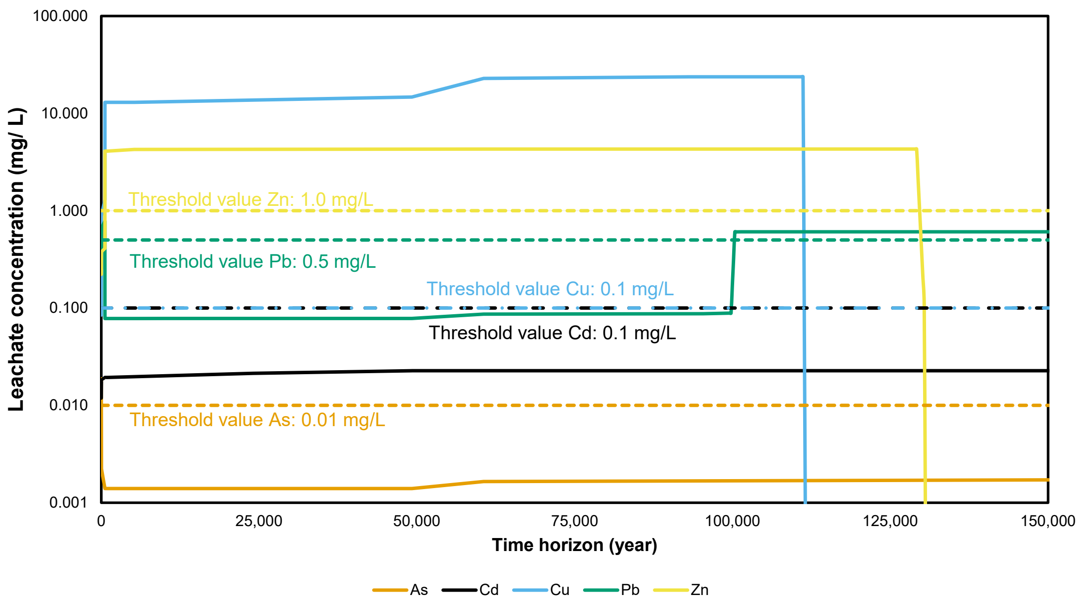


Figure A6. Porphyry tailings: Modeled leachate concentrations of As, Cd, Cu, Pb and Zn, according to the geochemical model up to 150k years after disposal of porphyry tailings. EU groundwater leachate discharge threshold values *(European Commission, 2009)* are displayed for comparison

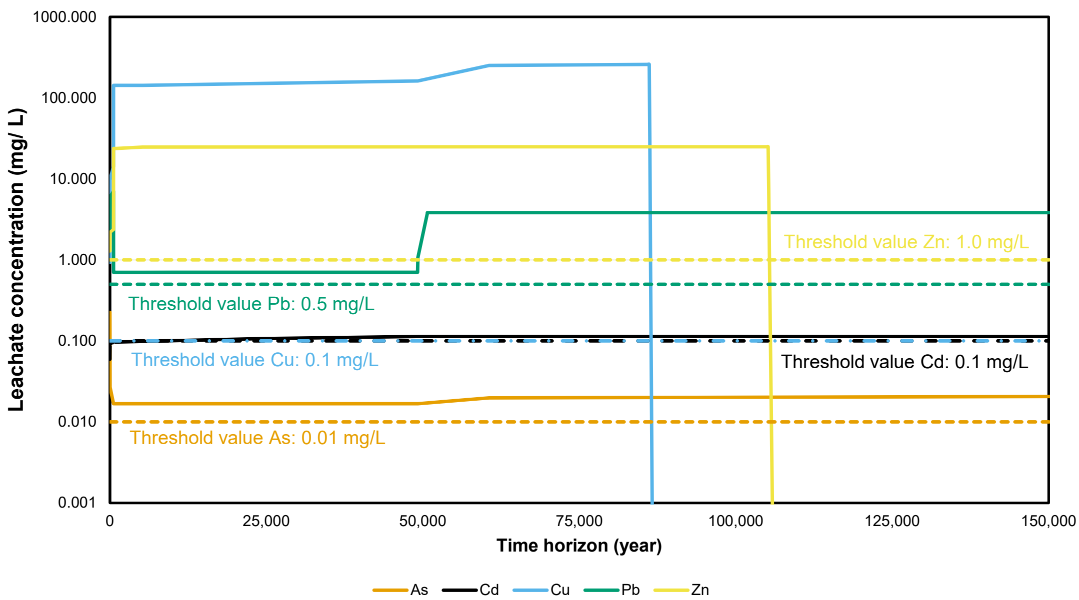


Figure A7. Volcanogenic massive sulfide tailings: Modeled leachate concentrations of As, Cd, Cu, Pb and Zn, according to the geochemical up to 150k years after disposal of volcanogenic massive sulfide tailings. EU groundwater leachate discharge threshold values *(European Commission, 2009)* are displayed for comparison

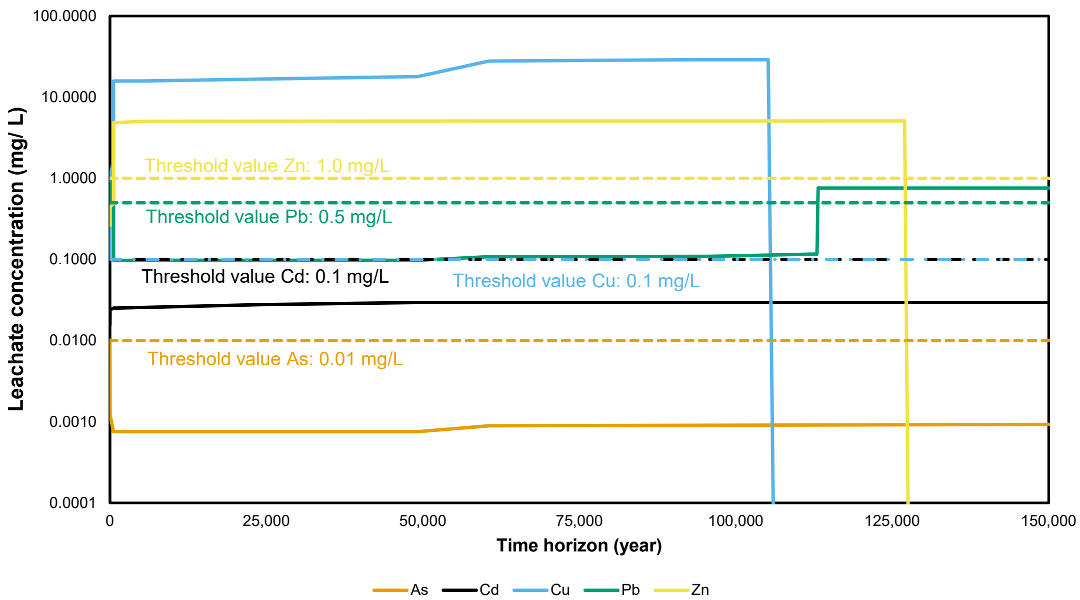


Figure A8. Skarn tailings: Modeled leachate concentrations of As, Cd, Cu, Pb and Zn, according to the geochemical up to 150k years after disposal of skarn tailings. EU groundwater leachate discharge threshold values *(European Commission, 2009)* are displayed for comparison

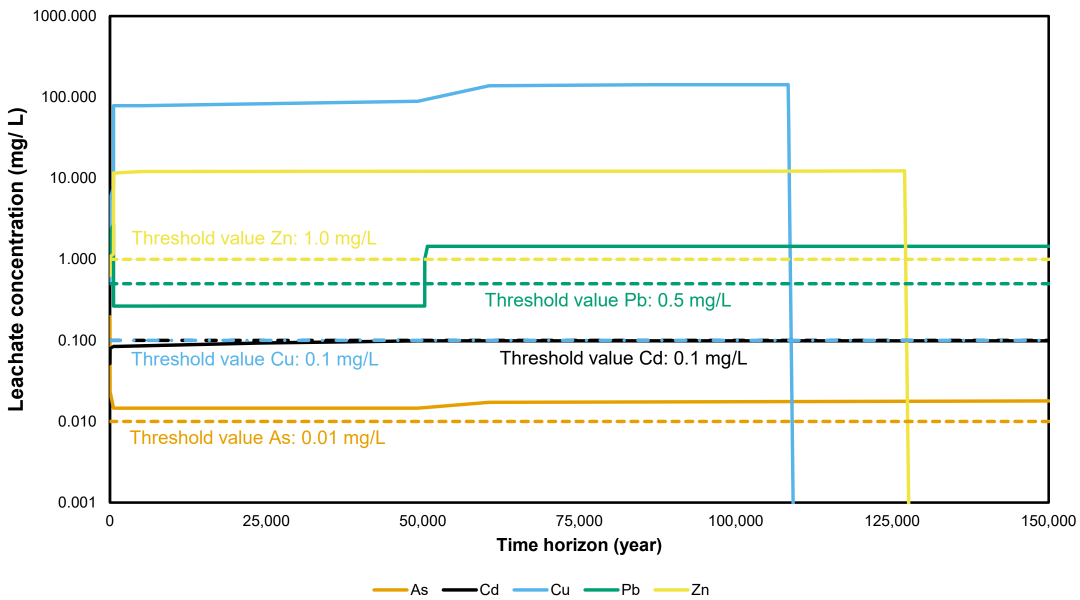


Figure A9. Sediment hosted tailings: Modeled leachate concentrations of As, Cd, Cu, Pb and Zn, according to the geochemical up to 150k years after disposal of sediment hosted tailings. EU groundwater leachate discharge threshold values *(European Commission, 2009)* are displayed for comparison

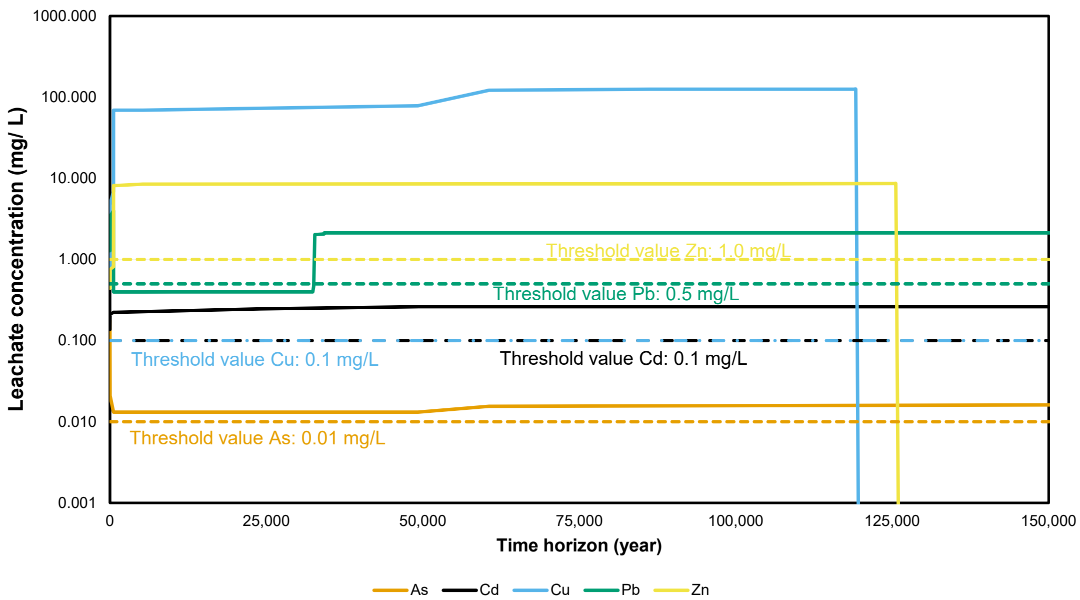


Figure A10. Magmatic sulfide tailings: Modeled leachate concentrations of As, Cd, Cu, Pb and Zn, according to the geochemical up to 150k years after disposal of magmatic sulfide tailings. EU groundwater leachate discharge threshold values *(European Commission, 2009)* are displayed for comparison

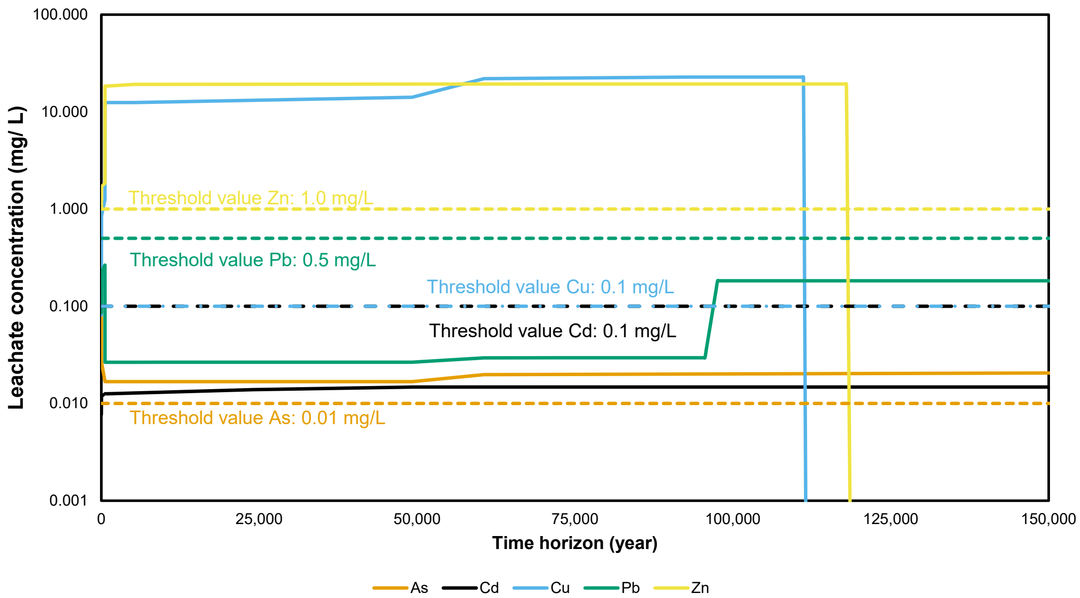


Figure A11. Iron oxide tailings: Modeled leachate concentrations of As, Cd, Cu, Pb and Zn, according to the geochemical up to 150k years after disposal of iron oxide tailings. EU groundwater leachate discharge threshold values *(European Commission, 2009)* are displayed for comparison

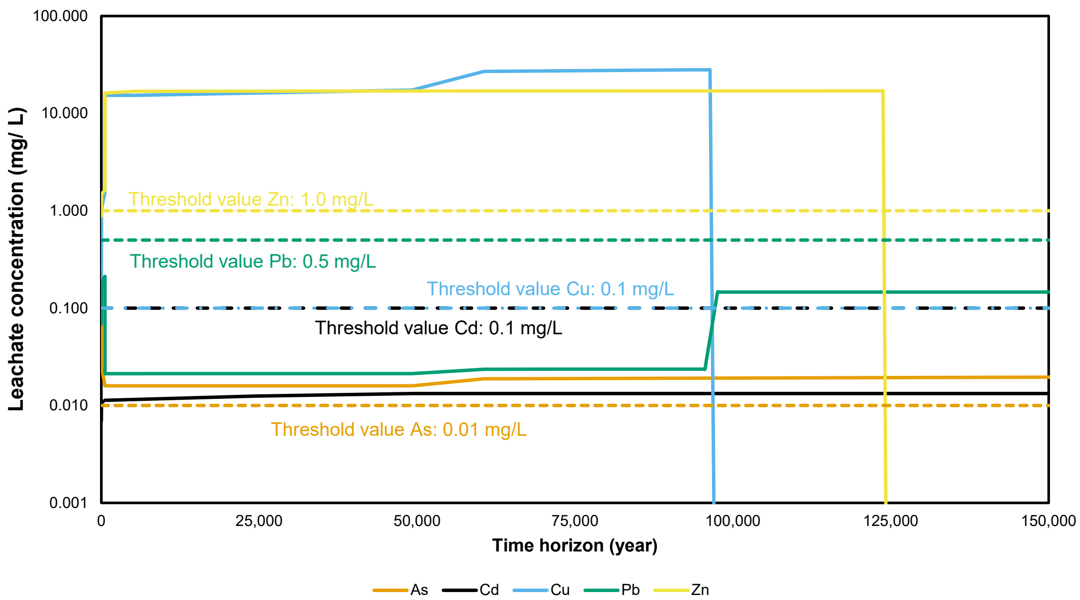


Figure A12. Intrusion-related tailings: Modeled leachate concentrations of As, Cd, Cu, Pb and Zn, according to the geochemical up to 150k years after disposal of intrusion-related tailings. EU groundwater leachate discharge threshold values *(European Commission, 2009)* are displayed for comparison

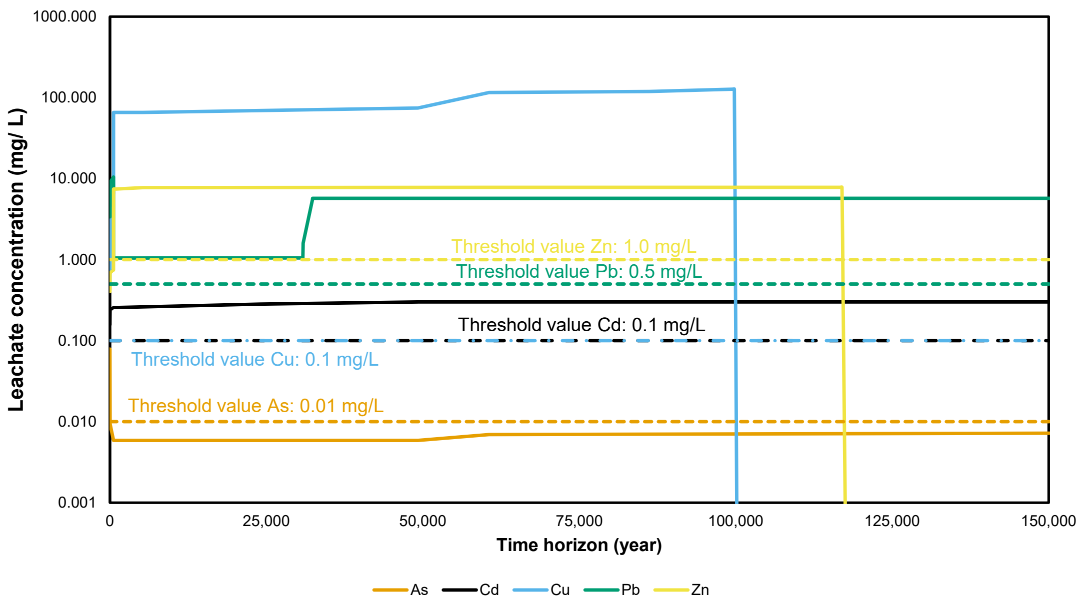


Figure A13. Epithermal tailings: Modeled leachate concentrations of As, Cd, Cu, Pb and Zn, according to the geochemical up to 150k years after disposal of epithermal tailings. EU groundwater leachate discharge threshold values *(European Commission, 2009)* are displayed for comparison

A.6 Net infiltration map (groundwater recharge) using GLOBWB model

To account for site-specific net infiltrations, we took the values from PCR GLOBWB (Sutanudjaja et al., 2018; Wada et al., 2010) (for further descriptions, check the mentioned references). The schematic water flows and the annual net infiltration (or groundwater recharge) map are displayed in Figure A14.

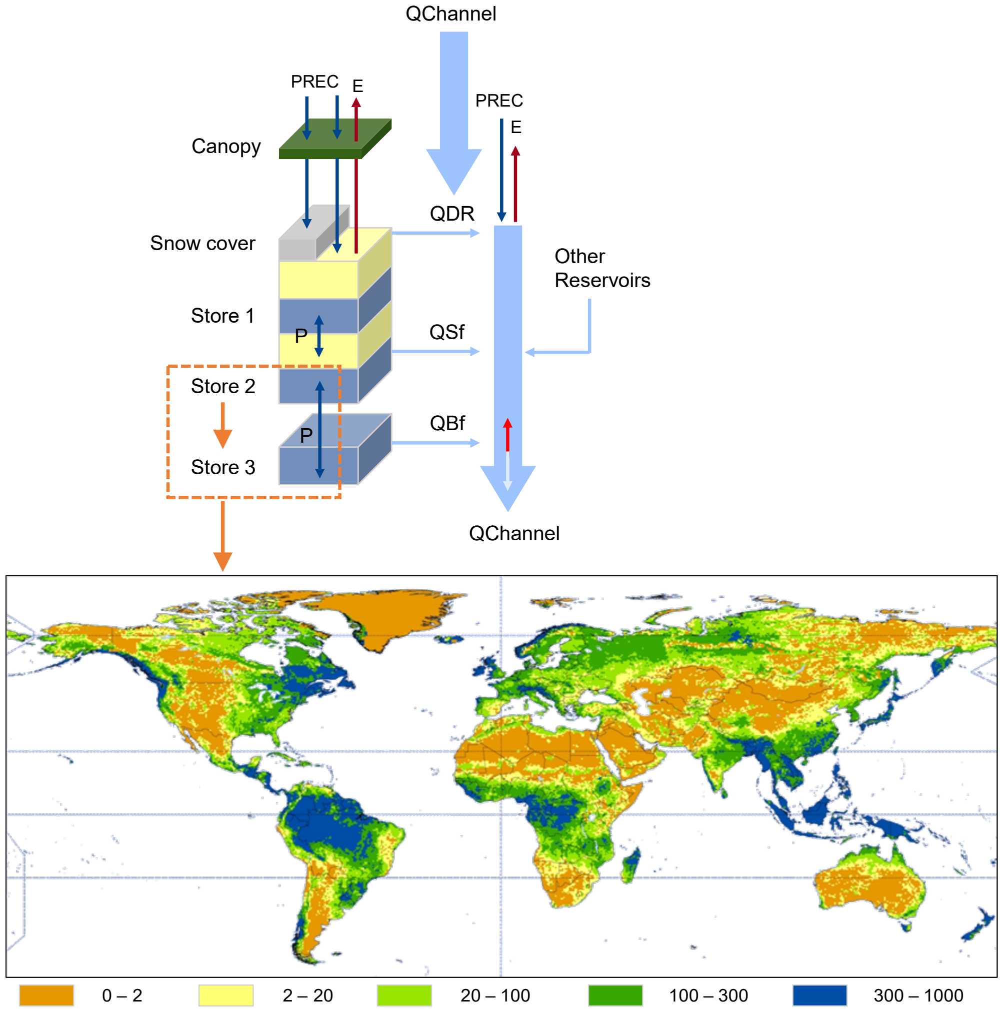


Figure A14. Top: The conceptual water flows in PCR-GLOBWB, redrawn from Sutanudjaja et al.*(2018)*. Bottom: The annual groundwater recharge, taken from the results of PCR-GLOBWB in mm per year *(Sutanudjaja et al., 2018; Wada et al., 2010)*. Abbreviations: PREC= Precipitation, E= Evaporation, P= Percolation/ capillary rise, QDR= Direct run-off, QSf= Interflow or subsurface stormflow, QBf= Baseflow, QChannel= Discharge along the channel

A.7 Baseline scenario: Environmental hotspots

This section explains how we derived the grouping of datasets and performed global LCA for copper tailings. The gray area in Figure A15 is the focus of this study, where we want to improve the granularity of tailings disposal dataset before they are matched with the other downstream processes (indicated in orange area). Due to difference in resolutions, we classified the datapoints (n = 431) of tailings disposal with the other six datasets that represent copper smelter/ refinery processes across the world. Meanwhile, Figure A16 shows how we grouped the simulated results in our study and eventually the basis of comparison in Figure 5 in the main paper.

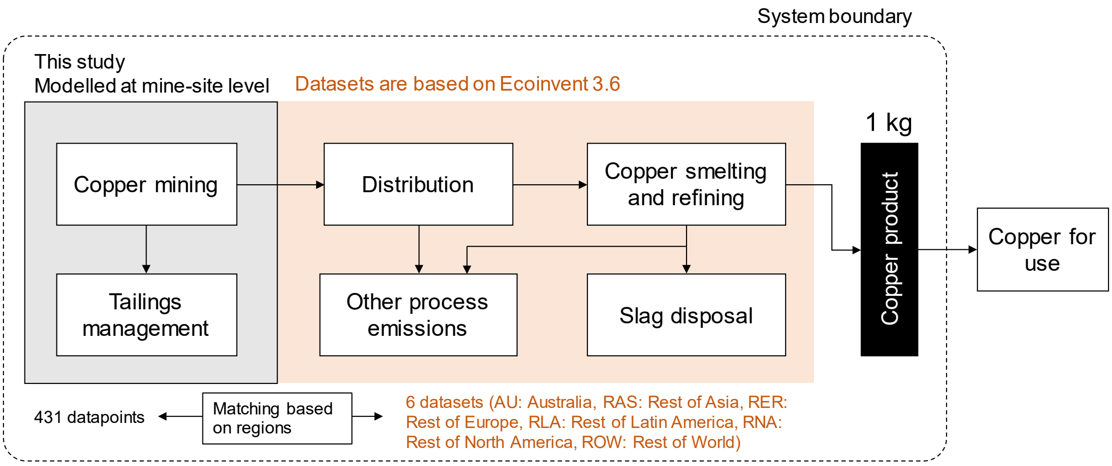


Figure A15. LCA system boundary to produce 1 kg copper

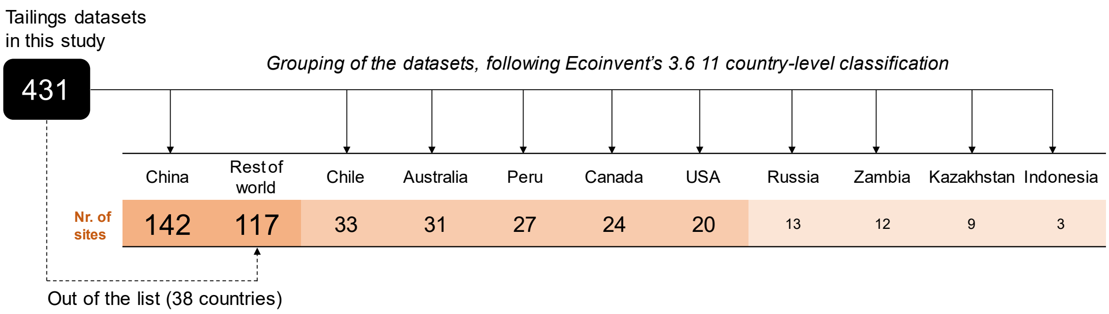
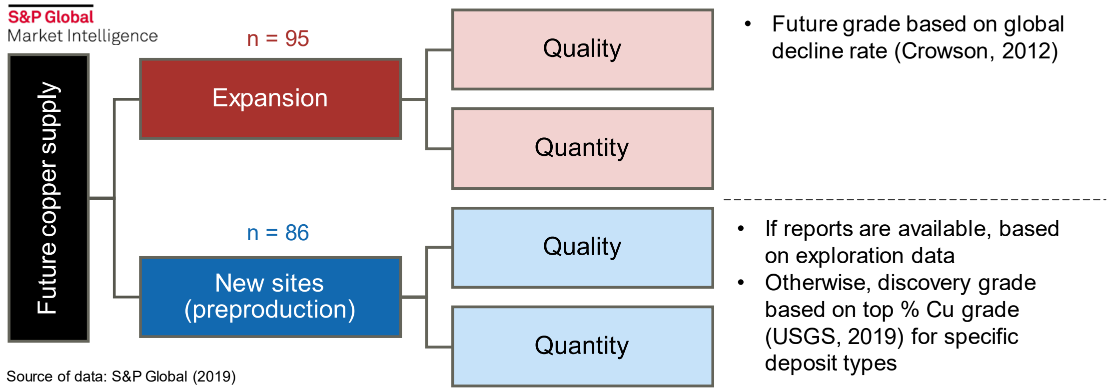


Figure A16. The procedure to systematically compare generated tailings inventory data (this study) with the available ones in LCA database

A.8 Primary copper production data for 2019 – 2050

The data for copper supply up to mid of the 21st century are mainly collected from Elshkaki et al. and Northey at al. studies (Elshkaki et al., 2016, 2018; Northey et al., 2014). Where gaps are found, we used assumptions to estimate the volume of copper produced in respective countries by subtraction. In addition to complete descriptions of this approach in the main paper, here we presented in Figure A17 and Figure A18 the schematic of our study scopes.



$$Primary\ supply_{year\ t} = \sum Primary\ supply_{year\ t-1} + \sum (Supply\ expansion + New\ expansion)_{year\ t}$$

Figure A17. Future copper supply estimation and data compilation

In this study, we considered only the preproduction as the data already represent sites with higher certainty to supplying copper. However, three preproduction categories were obtained from the market intelligence platform (S&P, 2020) that indicates the active operational phase:

- Commissioning, hence, it will be active in 1 year
- Construction stages. In these subcases, we assumed it will take 3 years for those already in track while it will take 5 years for those sites in plan. Further details can be seen in the Appendix-2, S2.7 – S2.8 (i.e., the list of all projects, including site expansion: 95 sites and new site opening: 86 sites).

Based on the latest available data on S&P Market Intelligence (S&P, 2020), we narrowed down the future copper sites on phase 2 and 3 (see Figure A18). The data on this platform is somewhat limited to account for abandoned sites and the appearance of sites with its copper throughputs. Therefore, we ultimately merged the data gathered from this online source with the other forecasting studies (Elshkaki et al., 2016, 2018; Northey et al., 2014) to obtain complete information for future supplies.

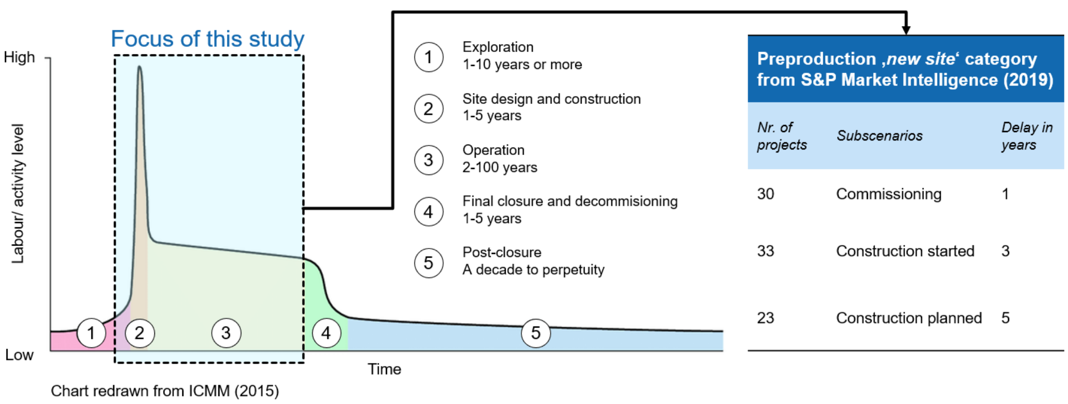


Figure A18. The cases for preproduction sites with three different categories of greenfield mining. Adapted after ICMM (2015)

A.9 Freshwater ecotoxicity impacts from baseline year 2019 to 2050

Table A14. Summary of results for the ecotoxicity projection from 2019 to 2050 in all countries (Unit: CTUe per year, method: USETox)

Cases	Argentina	Armenia	Australia	Azerbaijan	Bolivia	Botswana	Brazil	Bulgaria	Canada	Chile	China	Colombia	Cyprus	Other countries	TOTAL all countries	% - changes relative to baseline	Effect of demand growth	Effect of ore decline
Ecoinvent 3.6 (2019)	1.20E+11	2.10E+11	3.38E+12	7.46E+09	6.83E+09	1.64E+10	1.72E+12	6.52E+11	2.74E+12	2.58E+13	5.48E+12	1.29E+10	2.60E+10	...	**8.50E+13**	124%		
This study (2019)	1.98E+10	2.65E+10	2.38E+12	8.36E+09	6.61E+09	1.92E+10	1.84E+12	1.68E+11	3.92E+12	6.39E+12	3.12E+12	7.43E+10	4.92E+09	...	**6.83E+13**	100%		
2030 - same ore grade	5.98E+09	8.00E+09	3.89E+12	2.53E+09	2.00E+09	5.79E+09	1.38E+12	5.08E+10	2.93E+12	1.15E+13	6.03E+12	2.25E+10	1.49E+09	...	**1.14E+14**		87%	
2030 - with ore grade decline	6.85E+09	9.16E+09	4.46E+12	2.89E+09	2.29E+09	6.63E+09	1.58E+12	5.82E+10	3.36E+12	1.32E+13	6.91E+12	2.57E+10	1.70E+09	...	**1.31E+14**	192%		13%
2040 - same ore grade	1.78E+10	2.39E+10	4.15E+12	7.54E+09	5.96E+09	1.73E+10	1.77E+12	1.52E+11	3.76E+12	1.15E+13	4.49E+12	6.70E+10	4.43E+09	...	**9.61E+13**		77%	
2040 - with ore grade decline	2.31E+10	3.09E+10	5.38E+12	9.77E+09	7.72E+09	2.24E+10	2.29E+12	1.97E+11	4.87E+12	1.49E+13	5.82E+12	8.69E+10	5.75E+09	...	**1.25E+14**	182%		23%
2050 - same ore grade	1.71E+10	2.28E+10	4.12E+12	7.21E+09	5.70E+09	1.65E+10	1.69E+12	1.45E+11	3.59E+12	1.08E+13	2.55E+12	6.41E+10	4.24E+09	...	**7.43E+13**		68%	
2050 - with ore grade decline	2.50E+10	3.34E+10	6.03E+12	1.06E+10	8.35E+09	2.42E+10	2.48E+12	2.12E+11	5.27E+12	1.59E+13	3.74E+12	9.39E+10	6.21E+09	...	**1.09E+14**	159%		32%
														See SI for complete list		Value for demand growth = Sum without decline divided by sum with ore grade decline		Value = 100% – effect of demand growth

*Other results are available in the Appendix-2 document (S2.6a supply projection and S2.6b the exhaustive table with all countries)

A.10 Compare tailings characteristics from model prediction vs. tailings characteristics from actual data

We compared tailings characteristics from model prediction vs. tailings characteristics from actual data (on-site, taken from an operating facility (SULTAN, 2019)). The results of the sensitivity analysis for low efficiency and high efficiency in ore beneficiation are also presented in both Table A15 and Figure A19.

Table A15. Comparison of approximated results for three flotation cases vs. plant data

	Simulated			Plant data						
Metals (in mg/ kg)	Low limit Hi- efficiency	Base	High limit Lo-efficiency	Tailings sample A	Tailings sample B	Tailings sample C	Average	% diff with low	% diff with base	% diff with high
Zn	5,269	8,130	11,019	7,400	10,529	8,535	8,821	-67%	-8%	20%
Cu	2,815	4,436	6,005	6,600	4,721	4,480	5,267	-87%	-19%	12%
As	2,424	3,819	5,170	5,095	4,073	4,952	4,707	-94%	-23%	9%
Pb	1,379	2,528	3,603	2,500	5,896	3,300	3,899	-183%	-54%	-8%
Cd	18	29	39	NA	31	NA	31	-70%	-8%	20%

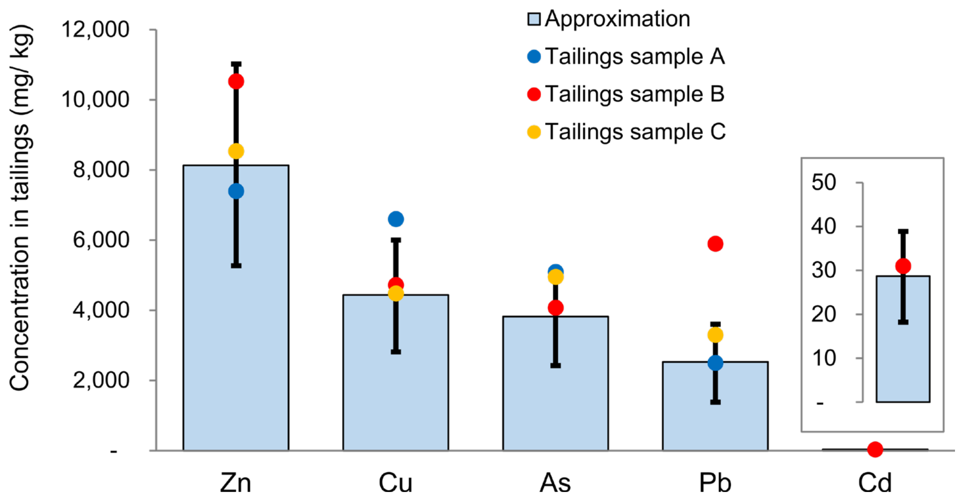


Figure A19. Comparison of metal compositions from simulated results vs. collected data from mining partners. The upper limit of the whisker represents worst case with worse flotation performance, and the lower limit represents better flotation performance.

We additionally conducted comparative analysis of tailings composition from our model vs. data from field measurements in Chilean tailings. For that, we rely on the compilation of data by the Chilean Geological Survey 'SERNAGEOMIN' (SERNAGEOMIN, 2020), which provide data for heavy metal compositions of major sites in Chile. Two sites, namely Escondida and Los Pelambres were chosen as cases because of the data availability (n>2). Copper and cadmium, among other heavy metals, have a large variability even in a single site due to multiple factors (Hervé et al., 2020; Padilla Garza et al., 2001): ore deposits, location of sampling, weathering, and other uncertainties in the data collection. The sensitivity in the model input also shows that while there is disparity between simulation and reality, the values are generally in good agreement within the same order of magnitude. However, more validations are still needed to confirm this finding for other sites and, in the same line, for recognizing uncertainties within input characteristics and tailings/ ore deposit geochemistry.

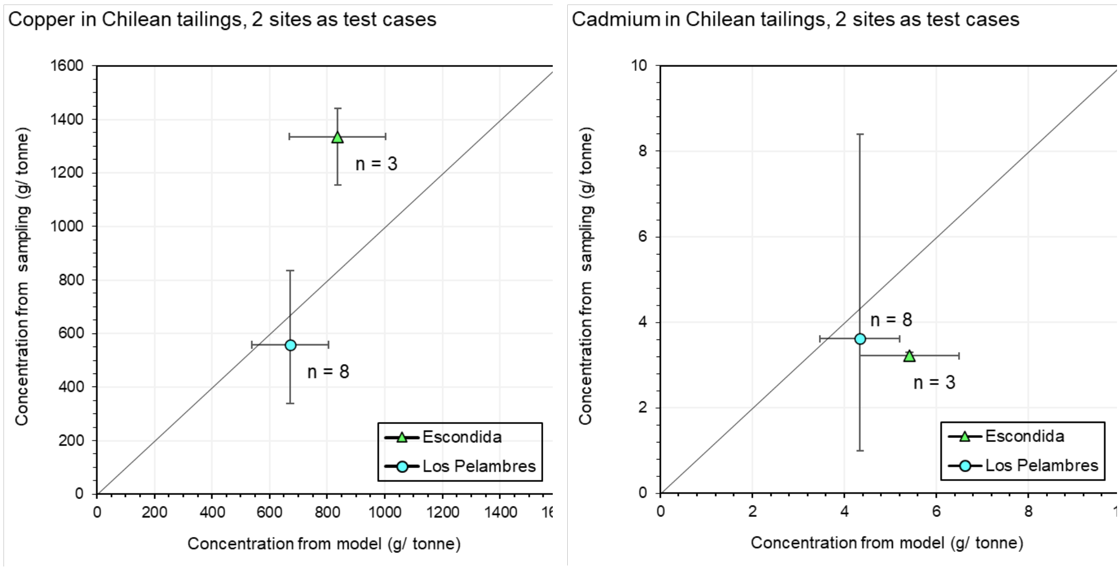


Figure A20. Comparison of metal compositions from simulated results x-axis vs. tailings data from Chilean sites y-axis. Blue circles represent results from base calculation, while green triangles are averaged sampling data from survey. Whiskers represent the highest and lowest value in the dataset (vertical) and simulated results (horizontal), respectively.

In average, copper yield in the beneficiations is ranging from 80 – 95% from our simulations. As concluding remarks, improving flotation processes in general (Asghari et al.; Guang-Yi et al., 2011) is a must when the yield is comparably lower according to the previously mentioned range. However, the efficiency of flotation in general is already reaching the mature performance according to the literature, even in the last couple of decades, so the efficiency can only improve slightly by upgrading the beneficiation facility (Alcalde et al., 2018).

A.11 Aggregated results for long term horizon

Table A16 contains aggregated freshwater ecotoxicity results caused by tailings deposition, in a long-term perspective (60,000 years). In addition, we also presented a graphical representation of this result in Figure A21 as tree map chart for a hi-level overview.

Table A16. The summary of aggregated freshwater ecotoxicity (long-term horizon) results in this study. The data is also derived from previous figures.

Continents	Country	Aggregated Freshwater Eco-toxicity (CTUe)	Contribution
Africa	Dem. Rep. Congo	5.72E+12	8%
Africa	Zambia	4.83E+12	7%
Africa	Morocco	1.56E+11	0%
Africa	South Africa	7.41E+10	0%
Africa	Mauritania	4.13E+09	0%
Africa	Eritrea	4.29E+10	0%
Africa	Namibia	2.02E+10	0%
Africa	Zimbabwe	2.77E+10	0%
Africa	Botswana	1.92E+10	0%
Asia and Oceania	Australia	2.38E+12	3%
Asia and Oceania	China	3.12E+12	5%
Asia and Oceania	Indonesia	3.99E+12	6%
Asia and Oceania	Kazakhstan	9.67E+11	1%
Asia and Oceania	Mongolia	1.06E+10	0%
Asia and Oceania	Iran	4.58E+10	0%
Asia and Oceania	Laos	5.86E+11	1%
Asia and Oceania	Papua New Guinea	5.80E+11	1%
Asia and Oceania	Saudi Arabia	1.11E+10	0%
Asia and Oceania	Uzbekistan	1.36E+10	0%
Asia and Oceania	Philippines	2.83E+11	0%
Asia and Oceania	India	4.27E+10	0%
Asia and Oceania	Myanmar	8.34E+10	0%
Asia and Oceania	Kyrgyzstan	6.99E+08	0%
Asia and Oceania	Pakistan	4.36E+09	0%
Europe	Russia	1.17E+13	17%
Europe	Poland	3.34E+12	5%
Europe	Spain	3.02E+12	4%
Europe	Portugal	7.40E+11	1%
Europe	Finland	5.40E+11	1%
Europe	Turkey	1.22E+11	0%
Europe	Bulgaria	1.68E+11	0%
Europe	Sweden	3.82E+11	1%
Europe	Georgia	3.30E+11	0%
Europe	Serbia	2.41E+10	0%
Europe	Armenia	2.65E+10	0%
Europe	Macedonia	2.64E+10	0%
Europe	Azerbaijan	8.36E+09	0%
Europe	Cyprus	4.92E+09	0%
Europe	Romania	1.92E+08	0%
Latin America	Chile	6.39E+12	9%
Latin America	Peru	9.23E+12	14%
Latin America	Mexico	7.67E+11	1%
Latin America	Brazil	1.84E+12	3%
Latin America	Colombia	7.43E+10	0%

Latin America	Dominican Republic	5.22E+10	0%
Latin America	Argentina	1.98E+10	0%
Latin America	Bolivia	6.61E+09	0%
North America	USA	2.56E+12	4%
North America	Canada	3.92E+12	6%
TOTAL		8.87E+13	6.83E+13

Note: Other results in different time horizons and human-toxicity impacts can be seen in the online Appendix-2.

Aggregated at higher level, Figure A21 shows how each continent's ecotoxicity contribution in a tree map-like chart. Obviously, Latin America leads due to the large production capacity and the amount of copper tailings being disposed. Europe and Africa, however, are ranked at the first and third despite having fewer copper site units and a limited number of copper-extracting countries. While this analysis is conducted for 2019, an in-depth discussion about future primary mining operations, especially for countries where ramping-up and brownfield sites are expected, is included in section 3.7.

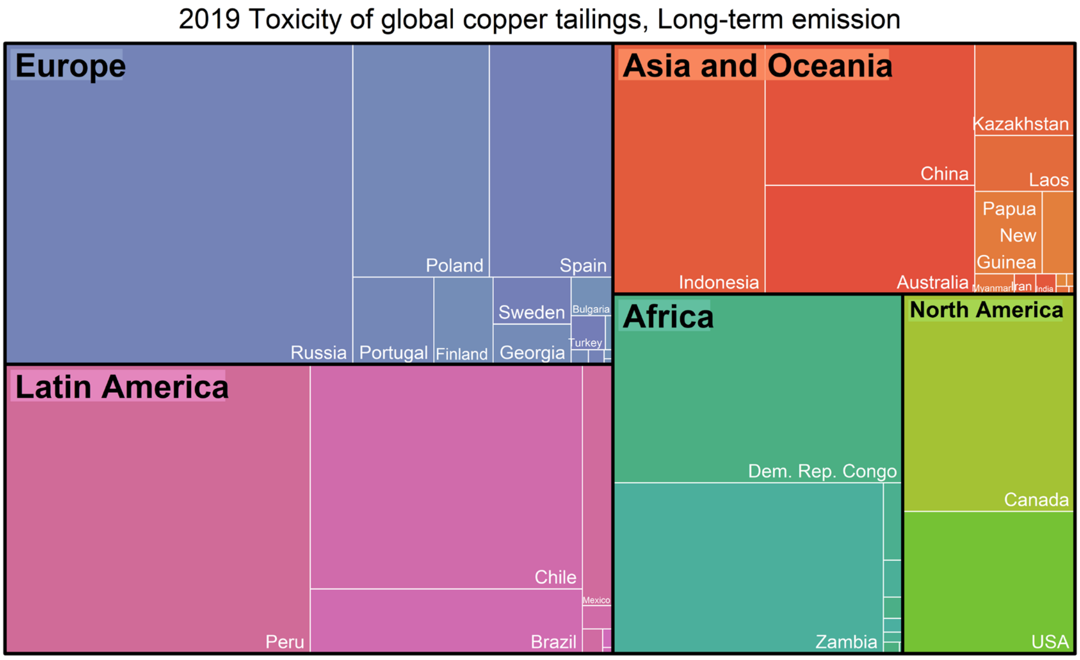


Figure A21. Contribution to global ecotoxicity impacts in 2019 per continent and country, illustrated in tree map chart. The area of each region and country is proportional to the estimated emissions.

A.12 Impacts of climate conditions and ore deposit types (complementary results)

Figure A22 presents a boxplot chart to analyze the distribution of freshwater ecotoxicity across different deposit types and infiltration rates. Another way to see the results is shown in Figure A23, where we rank the highest toxicity results from the top based on median values of each group.

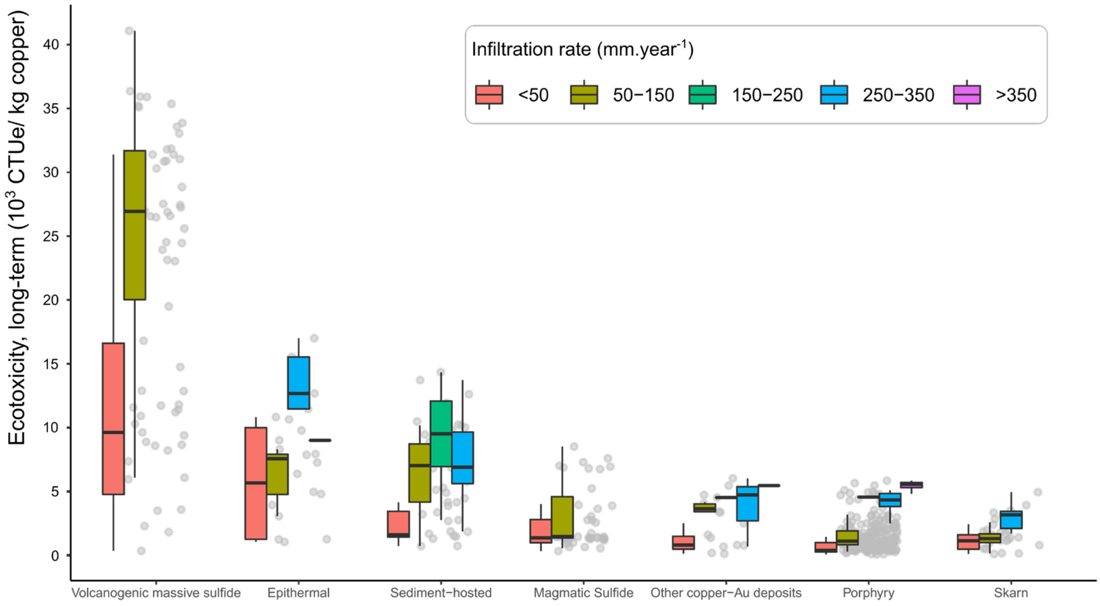


Figure A22. Distribution of ecotoxicity, grouped by net infiltration. The x-axis (copper deposit type) is ordered, based on the overall median of each class.

Moreover, we built two regression models below to see the influence of ore grades (Figure A24) and infiltration rate (Figure A25).

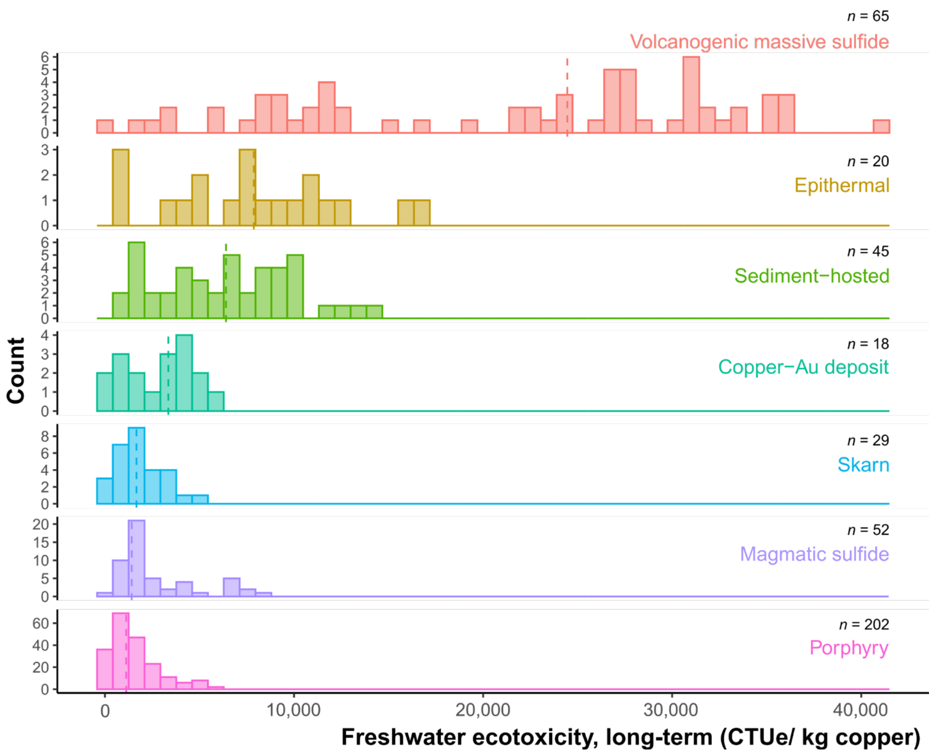


Figure A23. Histogram, shared x-axis (toxicity), grouped by deposit types. Dashed lines indicate median.

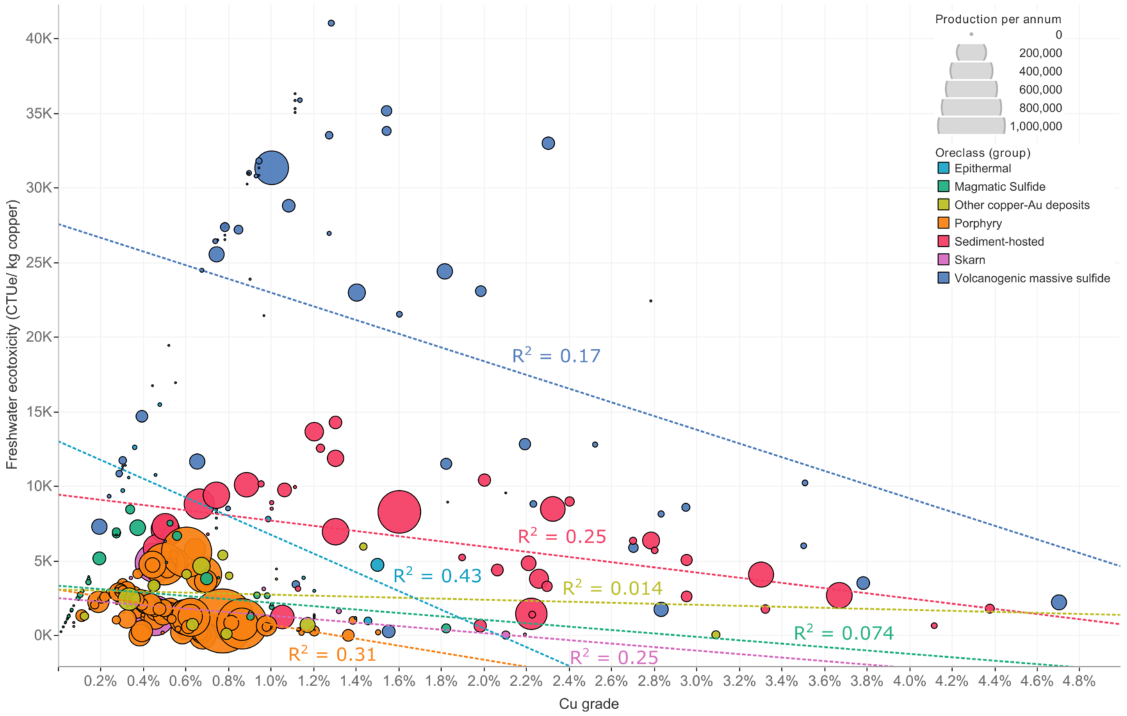


Figure A24. Regression Line, Cu grade vs. Toxicity per deposit type (left) and overall (right)

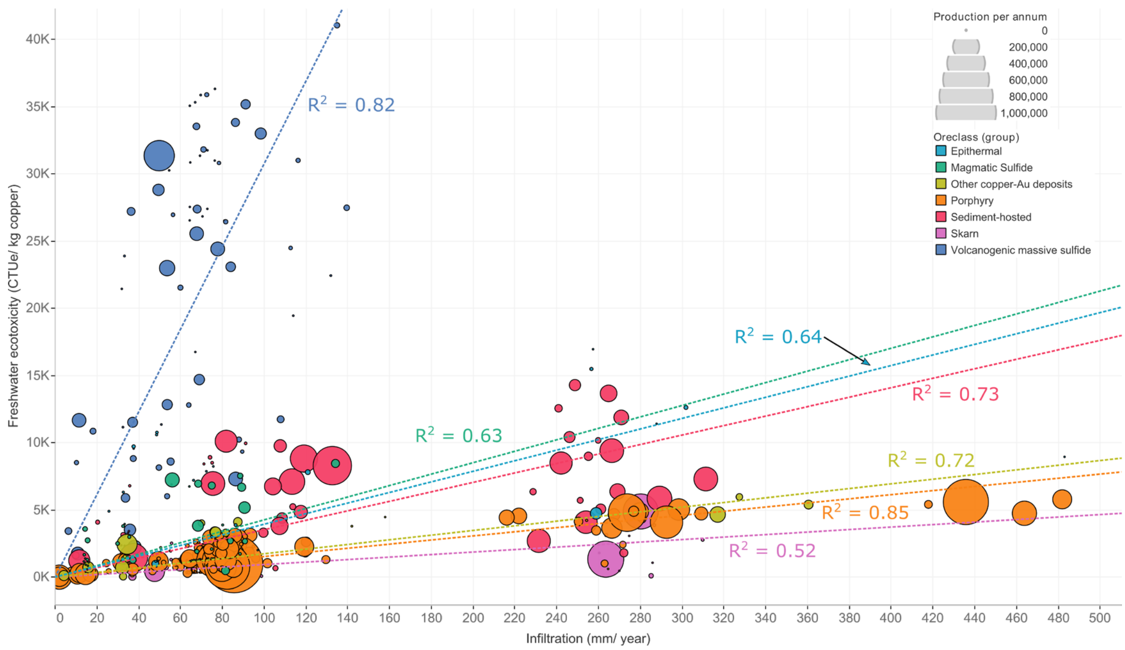


Figure A25. Scatter plots with regression lines: Net infiltration vs. Toxicity

Short summary of potential beneficiation upgrades:

Studies and field testing from sites have shown that flotation processes depend highly on the optimum size range of the ground ores, and is best represented by the "elephant curve" (Lynch, 1981). This phenomenon describes that recovery of minerals follows an inverted U-shape curve, and it will suffer whenever the particle size entering flotation stage falls outside this recommended range. It is a well-recognized fact that flotation only works best when the floating particles can be liberated freely under ideal pulp-froth interactions, which in turn improve the area, kinetics, and overall recovery. In this study, the process simulator HSC Sim is set by default to minimize energy consumption in the single grinding stage at the upstream without maximizing the recovery of minerals. It implies that the single grinding stage will automatically lower its efforts to crush the higher-grade ore and as the consequence, it will also decrease the overall recovery (Pease et al., 2006). We recognize this as one of the limitations in the mineral processing of our study. To overcome this setback, one can equip the standard copper recovery circuits with advanced grinding techniques (Kohmuench et al., 2018) (i.e. installing multiple re-grinding stages to extend the liberation curve at optimum mineral recovery). However, this is usually not done to all common processes since we maintain standard copper recovery circuits in our simulations.

A.13 Compiled copper production data for 2019

The main source of copper active sites are compiled from SNL Metal & Mining Market Intelligence data, study of Mudd et al., and USGS Mineral Deposits of the world (main features recorded: coordinates, tonnage, annual production capacity, and class of ore deposits). Sources: (G. M. Mudd & Jowitt, 2018; S&P, 2020; USGS, 2019).

Table A17. Copper production from sulfides in 2019, compiled (only listed as an illustration in this document. For complete list, see Appendix-2)

Mine ID	Annual production (Mt)	Longitude	Latitude	Tonnage (Economically mineable) in Mt	Cu-grade in %-w	Country	Continent	Ore deposit type
Escondida	904800	-69.1	-24.3	11,158.0	0.77%	Chile	Latin America	Porphyry
Collahuasi	559100	-68.7	-21.0	3,100.0	0.86%	Chile	Latin America	Porphyry
Grasberg	556557	137.2	-3.8	4,000.0	0.60%	Indonesia	Asia and Oceania	Porphyry
Cerro Verde	385818	-71.6	-16.5	2,528.0	0.50%	Peru	Latin America	Porphyry
El Teniente	320878	-70.5	-34.1	20,731.0	0.62%	Chile	Latin America	Porphyry
Morenci	129274	-109.4	33.1	6,470.0	0.52%	USA	North America	Porphyry
Las Bambas	285121	-72.3	-14.2	114.0	0.68%	Peru	Latin America	Porphyry
Los Bronces	284500	-70.3	-33.1	16,816.0	0.60%	Chile	Latin America	Porphyry
Los Pelambres	230240	-70.5	-31.7	7,458.0	0.62%	Chile	Latin America	Porphyry
Radomiro Tomic	232867	-68.9	-22.3	21,277.0	0.59%	Chile	Latin America	Porphyry
Chuquicamata	230936	-68.7	-21.0	3,100.0	0.86%	Chile	Latin America	Porphyry
Andina Division	131006	-70.5	-31.7	7,458.0	0.62%	Chile	Latin America	Porphyry
Spence	56448	-69.3	-22.8	497.0	0.92%	Chile	Latin America	Porphyry
Sarcheshmeh	120558	55.9	30.0	1,538.0	0.58%	Iran	Asia and Oceania	Porphyry
Toquepala	127200	-70.6	-17.2	2,320.0	0.55%	Peru	Latin America	Porphyry
Batu Hijau	161352	116.9	-9.0	1,640.0	0.44%	Indonesia	Asia and Oceania	Porphyry
Cuajone	112405	-70.7	-17.0	1,630.0	0.69%	Peru	Latin America	Porphyry
Oyu Tolgoi	159100	106.9	43.0	3,107.1	0.68%	Mongolia	Asia and Oceania	Porphyry
Centinela Sulfide	115070	-69.2	-23.2	519.0	0.41%	Chile	Latin America	Porphyry
...	...	...	...	...	...	...	...	...

Note: More exhaustive tables are provided in the spreadsheet of Appendix-2 (A2.1).

A.14 Freshwater ecotoxicity results across continents and countries

We aggregated the data at mine-site level to a continental-level by weighting the toxicity values (in CTUe, USEtox method) with the production capacity. The results for both short-term and long-term are presented in Table A18 and Figure A26. As for a more detailed analysis, the spread of toxicity at every mine site is included in the Appendix-2 (A2.10 – A2.11).

Table A18. Comparison of ecotoxicity (method: USEtox) results: Our study vs. Ecoinvent 3.6, aggregated following regions in the copper datasets

Short-term								
Region	Description	Tailings (our data) - weighted average	Tailings (ecoinvent 3.6)	Copper primary	Slag	Other processes	Total incl. tailings (Ecoinvent 3.6)	Total incl. tailings (our data)
AU	Australia	0	4	118	23	3	148	145
RAS	Rest of Asia	1	33	119	26	8	186	153
RER	Rest of Europe	2	3	10	25	2	40	40
RLA	Rest of Latin America	1	7	118	24	4	154	147
RNA	Rest of North America	2	32	10	25	8	74	44
ROW	Rest of World	1	33	118	26	5	182	150
Long-term								
Region	Description	Tailings (our data) - weighted average	Tailings (ecoinvent 3.6)	Copper primary	Slag	Other processes	Total incl. tailings (Ecoinvent 3.6)	Total incl. tailings (our data)
AU	Australia	2739	6596	118	54	86	6854	2997
RAS	Rest of Asia	3217	23084	119	62	107	23372	3505
RER	Rest of Europe	13359	2618	10	60	15	2703	13443
RLA	Rest of Latin America	2576	14256	118	57	81	14512	2832
RNA	Rest of North America	4764	25367	10	58	162	25597	4994
ROW	Rest of World	6052	15453	118	61	35	15666	6266

Note: The Appendix-2 (A2.10 – 11) document stores all the site-level results to obtain summarized values in this table.

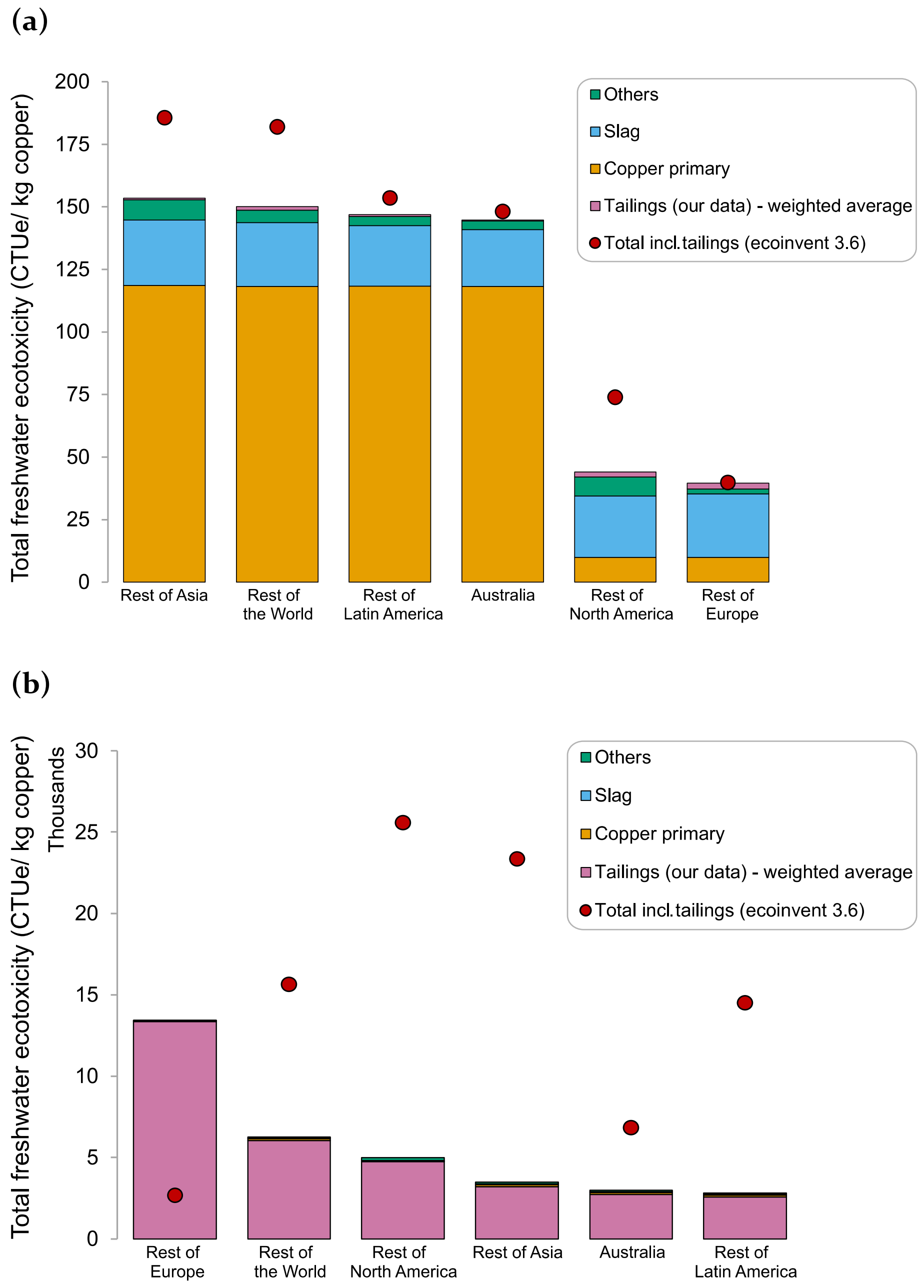


Figure A26. LCA of copper production (method: USEtox)—a) short-term and b) long-term in different regions. Data to generate this chart is available in Table A3.

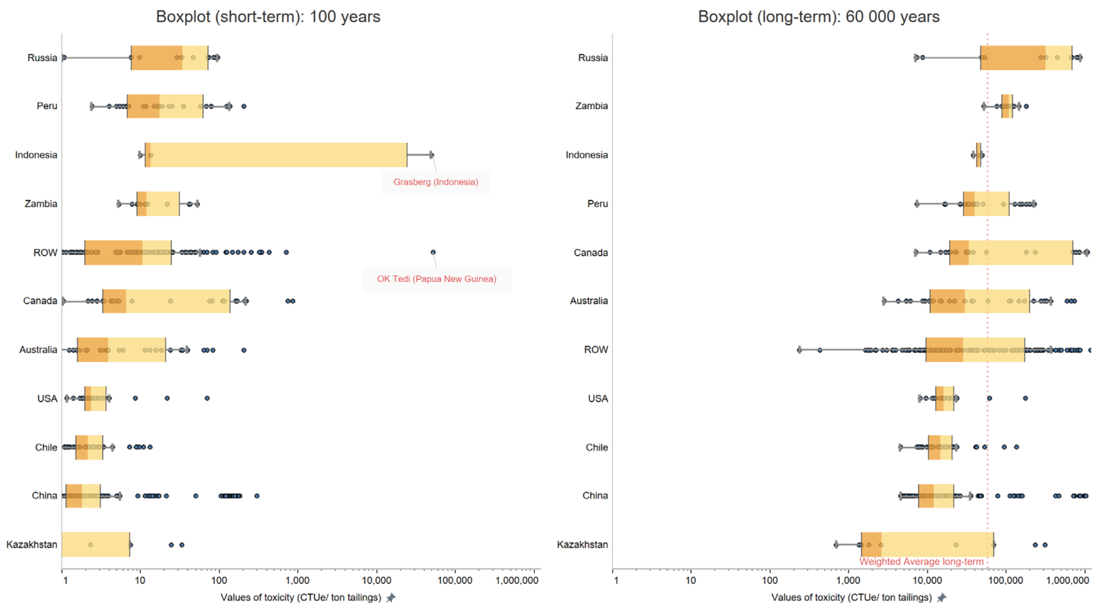
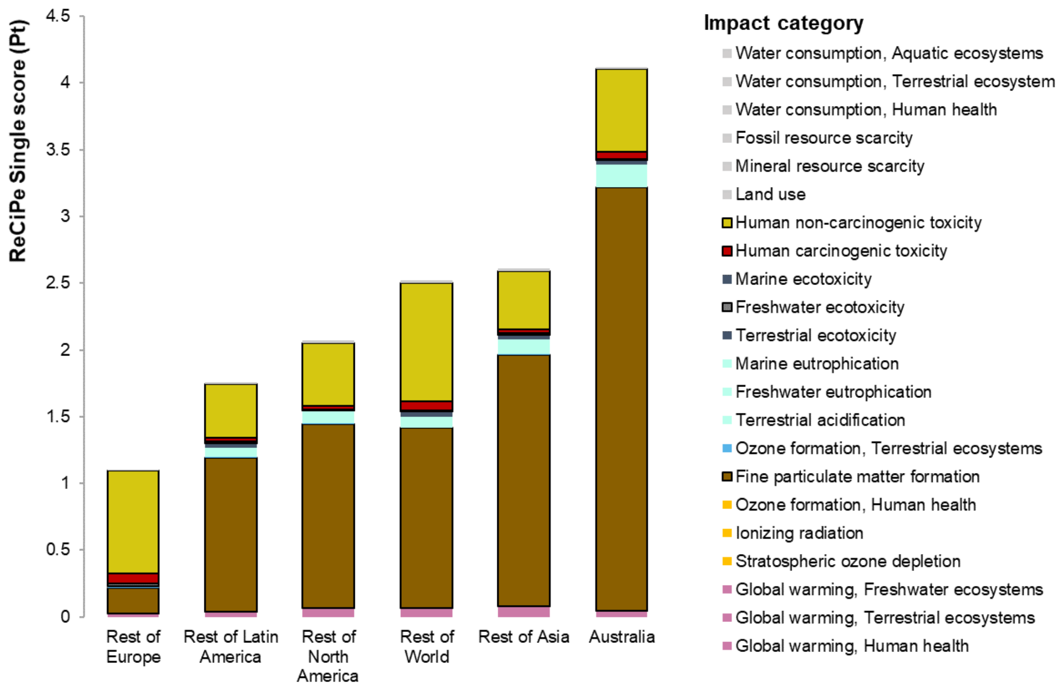


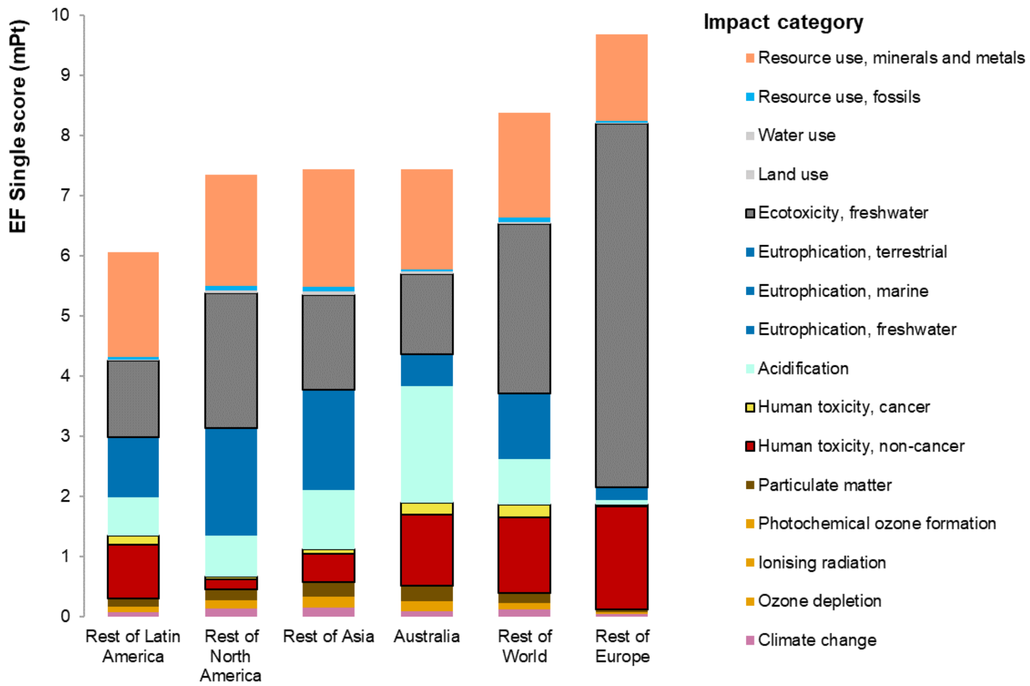
Figure A27. Freshwater ecotoxicity per ton of tailings in log-scale for short-term and long-term horizons

A.15 Complementary LCA results: 1) USEtox human toxicity, midpoint, 2) ReCipe, endpoint single score, 3) EF 3.0, endpoint single score

The toxicity analyses in the main paper are based on the recommended UNEP-SETAC LCIA method, which is USEtox (Rosenbaum et al., 2008). The human toxicity results are documented in Appendix-2 A2.12 – A2.13. In general, the ecotoxicity and human toxicity assessments are almost the same, except that in the human-toxicity cancer category, Cu and Zn lack characterization factors. Furthermore, we also presented in Figure A28 the results for single score ReCipe endpoint methods and EF endpoint methods (Fazio et al., 2018). This is to provide alternative views of the results in ranges of impact categories and to double check the consistencies across different methods. Compared to USEtox methods, which are behaving similarly between freshwater ecotoxicity and human toxicity, the results using ReCipe method are different. ReCipe shows that particulate matter formation and human toxicity as the major contributors in the whole system to produce 1 kg of copper. Furthermore, the weighting factors in ReCiPe are more spread to processes outside beneficiation systems (i.e., copper smelter and refinery), and therefore the emissions from these processes in the particulate matter formation category are high (in average 65% contribution to overall impacts across all continents due to sulfur dioxide and particulates). Single scores obtained from the EF methods, however, express the damages to wider categories like eutrophication and resource use, with ecotoxicity and human toxicity have 47% average contribution combined. Despite differences in the identification of priority life cycle stages of various LCIA methods, we can still conclude that metal leaching in the upstream stage is shown to be relevant for all methods (i.e., tailings long-term emission, demonstrated by Cu and Zu emissions in top process contributors in all continents as well as in the water compartment in Appendix-2 A2.14). Other metal flows such as lead, arsenic, and cadmium are also present in the top flows that leach to the water compartment.



(A)



(B)

Figure A28. LCA results of 1 kg copper production. (A) ReCiPe single score, endpoint, incl. long-term emissions and (B) EF single score, endpoint, incl. long-term emissions)

A.16 Further discussions and limitations on main waste streams in mining, rehabilitation, and tailings management

Besides discussions of data and limitations elaborated in the main manuscript, other aspects that require future improvements or care when interpreting the results are: (i) impacts from waste rock, overburden and other metallurgical slags/ residues, (ii) short term or continuous rehabilitation of tailings sites, and (iii) tailings disposal methods.

i) **Other main waste streams such waste rock, overburden, slags, and other residues**: We need better and in-depth study to estimate the actual environmental impacts caused by its deposition. Acid mine drainage (AMD) could be a source of toxicity that can arise from the landfilling/ backfilling of overburdens. In our study, we applied cut-off to these wastes assuming its relatively benign characteristics with lower metal contents compared to tailings. Normally, the waste rock and overburden are mixed with cement paste for the making of backfill materials, rendering them stable. Thus, the ecotoxicity impacts from overburden/ waste rock are assumed to be negligible, as similarly implemented in the life cycle Ecoinvent database (Classen et al., 2009). We are aware of this limitation and thus recommend a better quantification and higher resolution of overburden/ waste rock impacts life cycle inventories in future research (besides limited coverage from reporting from companies (G. Mudd, 2014)). As for slags and other residues, Gordon (2002) estimated the ratio of copper slags/ residues to metal in the US, which is equivalent to 2.1:1, compared to ~100:1 for tailings to metal ratio. To give an emphasis to the waste stream in this study and other important waste streams in the value chain, we present Figure A29.

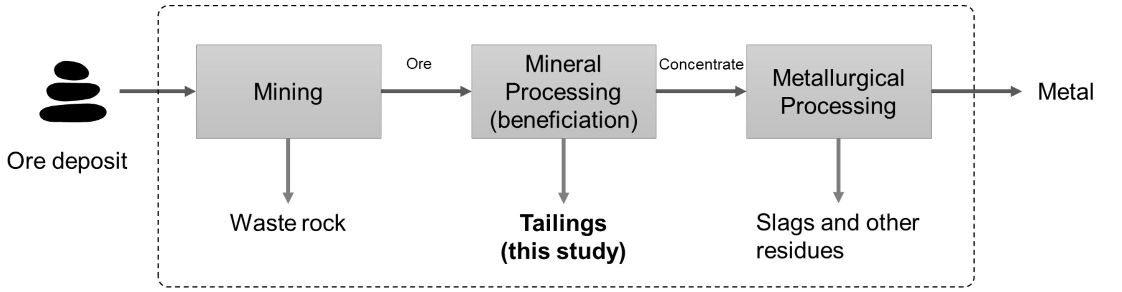


Figure A29. Simplified mining process flowsheets with the source of main waste flows (adapted from Lebre et al. (Lèbre et al., 2017))

ii) **Dust emissions**: Tailings are all assumed to enter soil and ultimately groundwater as the receiving compartment. In some cases, tailings can be carried away by wind and resulting in air/ dust emissions, sometimes the cases for the extremely arid regions. The environmental assessment studies for this case is somewhat limited; only one study tries to include the impacts through life cycle study from company reporting (Beylot & Villeneuve, 2017). From their study, it is found that dust emissions would have in average 4% contribution to all toxicity indicators in short term assessment. While we did not include explicitly the modeling of dust in our study, future studies could investigate thoroughly the effects of local environments to understand the actual impacts of dust in the overall system.

iii) **Rehabilitation**: we assume estimate of leaching over long-term using cut-off temporal approach for disposal of waste, following what is done for other long-term waste disposals (Doka & Hischier, 2005). We use this as a default choice and aim to estimate the impacts of tailings on standard storage facility on land, except for notable sites where storage/ embankment structure is not feasible like in Indonesia and Papua New Guinea (more details for these sites or aqueous disposal is described below). In short term or during surveyable period, leaching of heavy metals does not influence the results and can be relatively ignored in the entire life cycle of metal production but the results would turn the complete opposite when long-term horizon is taken into account. We then suggest further studies could expand the research in carrying out comparative analysis of different remediation or rehabilitation systems, either via active or passive mechanisms. This would also imply performing comparative analyses for continuous maintenance and operation for the duration of rehabilitation so that it can be used to select the optimized option and avoid burden shifting.

iv) **Tailings disposal and management**. There are various disposal methods for depositing tailings storage (Ellis, 1996). Most of the cases, tailings are stored on land within dams with different types of structure. We choose this type of storage for all of the tailings landfill modeled in the present study, except for notable mines where direct disposals are practiced. Besides the standard impoundment behind a dam on land, common alternatives to conventional tailings dams are (Wills & Finch, 2016b) (readers are invited to refer to the previous document for more information on tailings disposals):

a. Backfilling: tailings are usually combined with binders like cement, then used to fill voids in underground operations. With this method, tailings are returned to their source. After a curing period, the backfill can act as the ground structure support and allow recovering ore from adjacent stope. Special type of backfills called cemented paste backfill—a mixture of tailings, water, and binder (e.g., Portland cement)—gained popularity owing to its mechanical strength and economics. However, as the volume of rock increases following milling, not all tailings can be returned to the pit or underground. There is still rooms for potentials for an improved backfill operation, for instances by using by-products from other industries such as fly ash and blast furnace slag as cement replacement (Peyronnard & Benzaazoua, 2011).

b. Filtered tailings or sometimes called dry stack: tailings that have a reduced water content, or “filtered”, after mill processing. The resulting tailings are like a moist soil and can be stacked in piles and compacted. However, filtered tailings can still lead to water contamination under certain circumstances.
c. Aqueous dumping and submarine disposal: tailings are deposited directly into oceans, rivers, or streams. While there are many rationales such as unstable mountainous and extreme rainfalls for building proper storage facility (Vogt, 2013), this direct disposal method is extremely destructive, polluting water, destroying ecosystems, and ruining livelihoods. Only a few jurisdictions (Vogt, 2012) allow this choice of disposal due to extensive environmental loads to the environment. An analysis conducted by Earthworks in 2012 shows that million tons of mining waste are still discharged directly into rivers and seas, which prove to be irresponsible because of heavy metal contamination and milling chemicals. The extent of environmental impacts and its wide implications to the natural habitats and people’s livelihood has been extensively discussed elsewhere (Mckinnon, 2002; Vare et al., 2018).

A.17 By-products and allocation in metal mining systems

The production of metals typically involves more than one product as the output. For instance, copper production is also linked with the other by-products such as zinc, gold, silver, cobalt, nickel, etc. In life cycle assessment (LCA), there are different ways to deal with multiple product systems: system expansion and allocation based on physical relationships or economic concepts (Santero & Hendry, 2016). System expansion is seldom feasible for metal production because of the nonexistent single production or mono-output systems for the co-metals. Similarly, allocation due to physical relationships is not applicable in most cases. The alternative suggested approaches are thus economic allocation or allocation based on physical properties (e.g., mass). On one hand, economic allocation suffers from the fact that metal prices may fluctuate over time, revealing dynamics that are not easily captured. The temporal scope chosen for prices can significantly affect the economic proportion and, therefore, the environmental impacts of metals and co-metals under study. On the other hand, physical allocation suffers from the fact that masses may be very disproportionate (e.g., very expensive low-concentration metals may be the reason for the mining activity, while less-valuable but higher concentration metals would be assigned the majority of the burdens). Thus, mass allocation does not reflect the economic reasons for doing the mining operation.

Studies such as those conducted by Memary et al. (2012), Nuss and Eckelman (2014), and Rötzer and Schmidt (2020) show that more than 99% and 95% of impacts are allocated to copper, for mass and economic allocation respectively. Therefore, in this paper we simplified the analysis by allocating all impacts to copper. However, the data can be used to apply other allocation principles in future research. Overall, allocation choices and the effects of applying specific methods in the LCA of metals are still an open field that can benefit from more harmonization measures and in-depth future investigations (B. Weidema, 2000; B. P. Weidema & Schmidt, 2010).

In the case of deposits where geochemical zoning occurs, like VMS ore deposits, the processing configurations are set up to maximize the extraction of rich metals from the specific lithology. VMS deposits are compositionally zoned such that different areas or metal-laden zones are present with clear, distinctive characteristics (Hannington, 2013): Cu- and Fe-rich sulfide being in the center and Zn- and Pb-rich sulfides concentrated at the outer margins (Figure A30).

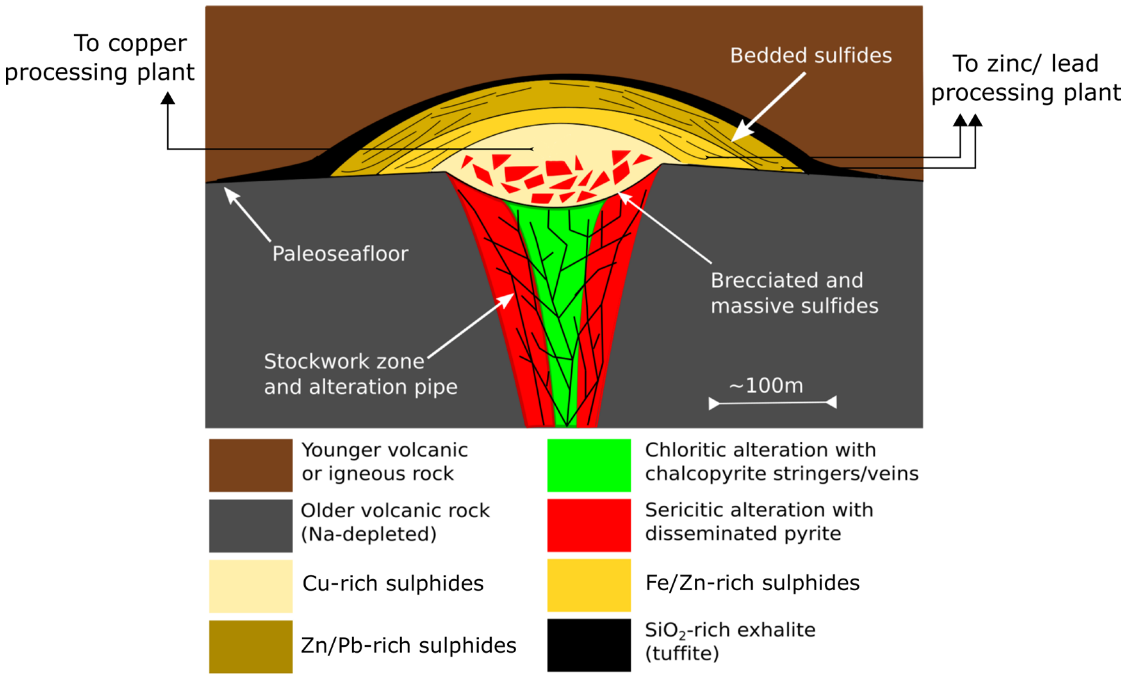


Figure A30. A cross-section of typical volcanogenic massive sulfide (VMS) as seen in the sedimentary record (Hannington, 2013), image adapted from VMS webpage (Wikipedia, 2020)

Due to this geochemical zoning, VMS deposits were exploited at different treatment facilities, designed for extracting particular metal of interest, either copper or zinc/ lead. Taking the case of Iberian Pyrite Belt deposits, for instance, the mining and beneficiation of ore deposits comprise more than a single train to treat both copper-rich and zinc-rich mined materials. Copper deposits undergo mineral processing in the first unit that specifically extracts copper as the primary metal, while the second unit aims at primarily to treat zinc. Hence, these configurations already separate two metals (i.e., copper and zinc) in a different processing plant where each has its own isolated systems. This suggests that environmental impacts of copper, in the first processing plant, must be allocated with the emissions and energy/ material consumption for this dedicated treatment plant only. This entire process is thus focused solely in producing concentrates with more than 96%

copper volume in the same mine location; the rest is co-produced along with the zinc/lead in the Zn-Pb processing plant. The common parts of the production system include the waste rock and tailings management.

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Appendix B

Prospective environmental assessment of reprocessing and valorization alternatives for sulfidic copper tailings

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Supplementary information for the following paper:

Prospective environmental assessment of reprocessing and valorization alternatives for sulfidic copper tailings in ***Resources, Conservation and Recycling*** (2022). Link.

 Digital SI with spreadsheet tables (referred to as Appendix-3) can be accessed online through this link.

B. Supplementary information for Chapter 3

B.1 Feed tailings characteristics

We base the characteristics of waste input on the recently published work (Xanthopoulos et al., 2021) for the same material. The chemical composition of tailings is presented in Table B1. The base metals of interests and included in the model are highlighted.

Table B1. The elemental composition (%) of sulfidic tailings from (Xanthopoulos et al., 2021)

Element	Concentration in %-w
Al	3.12
As	0.46
Ba	0.01
Be	n.d.
Ca	0.60
Cd	0.00
Co	0.02
Cr	0.00
Cu	0.46
Fe	25.50
K	0.42
Mg	0.99
Mn	0.08
Mo	n.d
Na	0.20
Ni	0.00
P	0.02
Pb	0.38
S	23.70
Sb	0.02
Se	0.01
Si	11.12
Sn	0.01
Sr	0.00
Zn	0.92

B.2 Life cycle system boundary, inventory modeling, and data collection

B.2.1 System boundaries

Following the 'basket of products' approach implemented in this LCA study, the functional units (FUs) of the systems are the sum up of all functions/ services provided. In summary, the FUs include:

- The disposal of 1 tonne sulfidic tailings,
- The production of 1.56 tonne CSA cement,
- The production of 4.1 tonne ceramics,
- The production of 0.69 geopolymer (equiv. to ordinary Portland cement),
- The production of 2.9 kg copper and 7.5 kg zinc,
- The production of 110 kg sulfuric acid,
- The production of 182 MJ heat energy,

As a result of this selected FU, the reference case should be able to fulfill all other materials above from primary production sources, in addition to the disposal of 1 tonne sulfidic copper tailings. For the other reprocessing routes, the environmental impacts of additional primary products besides secondary materials already produced from such routes should be considered to enable comparison with all routes. Calculating net environmental performances (benefits) between alternative reprocessing routes vs. the reference route becomes the difference of impacts between those two routes. We give the calculation examples below to illustrate the comparison of route A-1 against the reference route.

$$\begin{aligned}
&Net\ environmental\ performances\ of\ route\ A.1\ vs.reference \\
&\qquad = Total\ impacts\ of\ route\ A.1 - Total\ impacts\ of\ reference = \\
&\sum total\ reprocessing\ impacts\ of\ route\ A.1 \\
&\qquad + \sum total\ impacts\ of\ additional\ primary\ products\ for\ route\ A.1 \\
&\qquad - \sum total\ impacts\ of\ primary\ products\ for\ reference\ route \\
&\qquad - total\ impacts\ due\ to\ tailings\ landfilling
\end{aligned}$$

For route A.1, CSA cement and ceramics are produced as secondary materials. We still need to add other products from primary sources such as geopolymer, copper, zinc, and others, such that all FUs are satisfied. These additional impacts of primary products for route A.1 will cancel out with the impacts of primary products for the reference route, except for the secondary materials already generated from route A.1.

The subtraction of primary products between route A.1 and reference route leads to:

$$\sum total\ impacts\ of\ additional\ primary\ products\ for\ route\ A.1$$
$$- \sum total\ impacts\ of\ primary\ products\ for\ reference\ route =$$
$$- (impacts\ of primary\ CSA\ cement\ production$$
$$+ impacts\ of\ primary\ ceramics\ production)$$

If we substitute the terms above to the previous equation, the net environmental impacts of A.1 vs. reference route are:

$$\boxed{\begin{aligned} &Net\ environmental\ performances\ of\ route\ A.1\ vs. reference \\ &= \sum total\ reprocessing\ impacts\ of\ route\ A.1 \\ &- impacts\ of primary\ CSA\ cement\ production \\ &- impacts\ of\ primary\ ceramics\ production \\ &- total\ impacts\ due\ to\ tailings\ landfilling \end{aligned}}$$

We perform similar calculation procedures above to other routes (i.e., A-2, B-1, and B-2) evaluated in this study to arrive at the net environmental impacts in the results section.

B.2.2 Inventory modelling and approaches

As mentioned in the methods section, the basis of our study combines life cycle inventory data from multiple upscaling approaches and recommendations for prospective life cycle assessments (LCA) (Arvidsson et al., 2018; Parvatker & Eckelman, 2019; Piccinno et al., 2016; Thonemann et al., 2020; Tsoy et al., 2020). The life cycle inventory and technical parameters are collected in the APPENDIX-3, which users can import into their systems to simulate and obtain the results of the study. In total, there are seven processing steps in our study, which are modeled separately and explicitly to complete the missing data gaps. Section B4 – B10 describe the details for each process and technical parameters/ assumptions involved in the life cycle inventory modeling. The approaches selected for each process are summarized in Table B2.

Table B2. The approaches to complete life cycle inventory data in the study

Foreground unit process modelled in this study	Life cycle data reconciliation and upscaling methods	Data source for inventory modeling
Flocculation-flotation	Industrial proxy data, coupled with inhouse experiments.	Standard flotation process (Norgate & Haque, 2010) in beneficiation with focus on desulfurization of fresh tailings (Broadhurst et al., 2015).

Microwave roasting and leaching	Microwave heating energy based on other large-scale studies, chemical engineering process design calculation for drying and leaching steps.	The process scheme for roasting, leaching, and ion flotation is based on own experiments, conducted by (Xanthopoulos et al., 2021). Microwave heating and roasting energy data is adopted from other upscaling study (Bermúdez et al., 2015). For chemical equipment: Process design calculations, following the approach of (Piccinno et al., 2016).
Ion flotation and precipitation	Chemical engineering calculation for reactors (i.e., conditioning and precipitation). Recovery of solvent via distillation is chosen. Zinc precipitation is assumed to mimic industrial dissolved air flotation unit (DAF). Reagents consumption are upscaled based inhouse experiments.	For chemical equipment and reagents/ chemicals consumptions: Following the approach of (Piccinno et al., 2016). Life cycle inventory of solvent recovery is estimated using Ecosolvent tool (Capello et al., 2005, 2007). DAF data are obtained from field measurements of wastewater treatment and industrial effluent plant (O'Connor et al., 2014; Pokhrel & Viraraghavan, 2004).
Sulfuric acid production	Industrial proxy data, supplemented with Best-Available-Technology Reference Document (BREF) of pyrite roasting and sulfuric acid production in Europe.	Large scale combined pyrite roasting-sulfuric acid industrial data from the study of (Runkel & Sturm, 2009) and BREF document Joint Research Centre (JRC) (European Commission, 2007), which reports average performance of sulfuric acid plants.
Calcium sulfoaluminate (CSA) cement production	Industrial proxy data coupled with inhouse experiments for tailings mix design in the raw milling step. All background related data are adapted to Europe in the original dataset.	Mix designs are taken from the study of Pires Martins for the particular pyritic material (Martins et al., 2020). Average CSA cement plants in China, both for conventional (from virgin raw materials) and alternative production (derived from industrial/ mine waste residues) (Ren et al., 2017).
Ceramic tile production	Industry data already available in standard Ecoinvent 3.7.1 LCA database. We adapted to our case for tailings mix design in the first step (i.e., substituting virgin clay, sand, and filler inputs).	Ecoinvent 3.7.1 (Ecoinvent, 2021) ceramic tile production (region: Switzerland, CH), modified with mix designs from (Veiga Simão et al., 2021)
Geopolymer binder production	Due to unavailability of installed operations worldwide, we used metallurgical process simulation to generate life cycle inventories for this process.	Specific tool used: HSC Chemistry 10 process simulator (Metso Outotec, 2020), as implemented in the study of (Adrianto et al., 2021) and (Niu et al., 2021). The optimized geopolymer case in the previous work is used in our study.

B.3 Life cycle impact categories in the study

In the visualizations of results, we only select certain categories that show high relevance in the single score ReCiPe (H) methods (Figure B9). Notwithstanding these deliberate choices, complete results for other indicators are presented in the spreadsheet (online copy).

Table B3. The list life cycle impact assessment (LCIA) methods

Impact category, abbreviation if any	**Unit**
ReCiPe Midpoint (H) V1.13, agricultural land occupation, ALOP	square meter-year
ReCiPe Midpoint (H) V1.13, climate change, GWP/ CC	kg CO_2-Eq
ReCiPe Midpoint (H) V1.13, fossil depletion, FDP	kg oil-Eq
ReCiPe Midpoint (H) V1.13, freshwater ecotoxicity, FETP	kg 1,4-DC.
ReCiPe Midpoint (H) V1.13, freshwater eutrophication, FEP	kg P-Eq
ReCiPe Midpoint (H) V1.13, human toxicity, HTP	kg 1,4-DC.
ReCiPe Midpoint (H) V1.13, ionizing radiation, IRP	kg U235-Eq
ReCiPe Midpoint (H) V1.13, marine ecotoxicity, METP	kg 1,4-DB.
ReCiPe Midpoint (H) V1.13, marine eutrophication, MEP	kg N-Eq
ReCiPe Midpoint (H) V1.13, metal depletion, MDP	kg Fe-Eq
ReCiPe Midpoint (H) V1.13, natural land transformation, NLTP	square meter
ReCiPe Midpoint (H) V1.13, ozone depletion, ODP	kg CFC-11.
ReCiPe Midpoint (H) V1.13, particulate matter formation, PMFP	kg PM10-Eq
ReCiPe Midpoint (H) V1.13, photochemical oxidant formation, POFP	kg NMVOC-.
ReCiPe Midpoint (H) V1.13, terrestrial acidification, TAP	kg SO_2-Eq
ReCiPe Midpoint (H) V1.13, terrestrial ecotoxicity, TETP	kg 1,4-DC.
ReCiPe Midpoint (H) V1.13, urban land occupation, ULOP	square meter-year
ReCiPe Midpoint (H) V1.13, water depletion, WDP	m^3 water-.
USETox, freshwater ecotoxicity*	CTUe
USETox, human-toxicity total*	CTUh
Cumulative energy demand (CED), fossil*	MJ-eq
ReCiPe, Endpoint (H), total aggregated single score, ReCiPe (H) SS	Points

*USETox and CED are not part of the ReCiPe methodology and were calculated additionally

B.4 Flocculation-flotation

We rely on the data of standard ore sulfide flotation process (Norgate & Haque, 2010) in the industry and adapt it to our case by adding flocculation process in the upstream (Broadhurst et al., 2015; Da Rosa & Rubio, 2005). The process is principally a desulfurization treatment to separate sulfur-rich stream (i.e., pyrite) from gangue materials, such as quartz (Figure B1). We collected optimum operating conditions from the work of Ana Luiza et al. (2021, under review). However, the collector data for desulfurization purpose (i.e., Xanthate) is not available in the standard database, and hence is adopted from other life cycle inventory study based on large scale operations in South Africa from (Kunene, 2014). Dewatering processes are achieved using belt filter press equipment, with its specific energy consumption of ~20 kWh/ ton input flow (Suez, 2019) as the average values for this purpose.

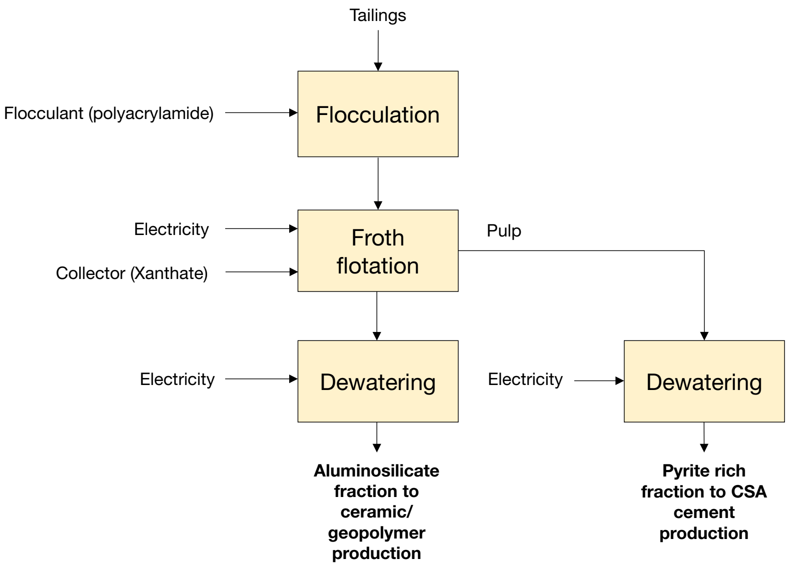


Figure B1. Process flowchart: Flocculation-flotation

Table B4. Process parameters used in constructing life cycle inventory data of flocculation-flotation process (Route A)

Process parameters	Values	Unit	References	Comments
Mass of tailings	1000	kg	Default	Functional unit (FU)
Moisture	30%		(McPhail et al., 2019)	Assuming from the thickened paste on site
Solid content tailings	700	kg	Calculated	Average data from samples
The amount of floccculant	0.025	kg/ ton tailings	Inhouse data	Collected from partners
Electricity for flotation	5.625	kWh/ton tailings	(Broadhurst et al., 2015)	Flotation and flocculation pumping (25% extra for latter purpose)
The amount of collector	0.1	kg/ ton tailings	(Broadhurst et al., 2015)	Consumption based on desulfurization studies

Electricity for dewatering	20	kWh/ton tailings	(Norgate & Haque, 2010)	Average values from belt filter energy consumption
Fraction to CSA cement	0.41	-	Inhouse data	Collected from partners
Fraction to Ceramic/ geopolymer	0.59	-	Inhouse data	Collected from partners

Table B5. Life cycle inventory of the treatment of 1 tonne tailings via flocculation-flotation (Route A)

LCI of flocculation-flotation				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Polyacrylamide	0.03	kg	Market for polyacrylamide, GLO, Ecoinvent 3.7.1
	Electricity total	25.63	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1
	Xanthate	0.10	kg	(Kunene, 2014)
Technosphere output	Si-Al fraction	413.00	kg	Calculated
	Pyrite rich fraction	287.00	kg	Calculated
Waste output	Water disposal	300.00	kg	Treatment of wastewater, Europe, Ecoinvent 3.7.1

Table B6. Life cycle inventory of xanthate production based on (Kunene, 2014))

LCI of xanthate production (FU = 1 tonne xanthate)					
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data from Ecoinvent 3.7.1**	**Region**
Technosphere	CS2 new process	0.53	tonne	CS2 new process	GLO
	electricity, medium voltage	163.9	kWh	market group for electricity, medium voltage	Europe without Switzerland
	tap water	1580	kg	market for tap water	Europe without Switzerland
	sodium hydroxide, without water, in 50% solution state	280	kg	market for sodium hydroxide, without water, in 50% solution state	GLO
	ethanol, without water, in 99.7% solution state, from ethylene	320	kg	market for ethanol, without water, in 99.7% solution state, from ethylene	RER
	nitrogen, liquid	27.52	kg	market for nitrogen, liquid	RER

Table B7. Life cycle inventory of CS2 production based on (Kunene, 2014))

LCI of carbon disulfide CS2 new process production (FU = 1 tonne CS2)					
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data from Ecoinvent 3.7.1**	**Region**
Technosphere	electricity, medium voltage	225	kWh	market group for electricity, medium voltage	Europe without Switzerland

	heat, district or industrial, natural gas	7510	MJ	market group for heat, district or industrial, natural gas	RER
	heat, from steam, in chemical industry	6540	MJ	market for heat, from steam, in chemical industry	RER
	tap water	1380	kg	market group for tap water	RER
	natural gas, low pressure, vehicle grade	224.02	kg	natural gas production, low pressure, vehicle grade	RoW
	sulfur	881.78	kg	sulfur production, petroleum refinery operation	Europe without Switzerland
	wastewater, average	-0.56	cubic meter	treatment of wastewater, average, capacity 1E9l/year	Europe without Switzerland
	municipal solid waste	-1.86	kg	market group for municipal solid waste	Europe without Switzerland
	refinery sludge	-2.4	kg	treatment of refinery sludge, hazardous waste incineration	Europe without Switzerland
	nitrogen, liquid	16.33	kg	market for nitrogen, liquid	RER
Types of inputs/ outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**	**Compartment**
Biosphere	Sulfur dioxide	40.5	kg	biosphere3	air
	Carbon dioxide, fossil	362.36	kg	biosphere3	air

B.5 Microwave-roasting and chemical leaching

In the beginning, a dewatering and drying step is important to remove moisture in the feed. We apply the process design calculation methods for the drying part, as suggested by (Piccinno et al., 2016) to calculate the energy consumption. Next, the crucial part of this process block is the microwave (MW) heating and roasting. It increases the activation of tailings as well as the removal of sulfur species in the original tailings material (Haque, 1999). It follows pyrite oxidation and mineralogical transformation as reported in the experiment (Xanthopoulos et al., 2021) and other study by (Ozer et al., 2017):

First reactions (pyrite oxidation) causing mass loss occurs at 380 – 500 °C

- $2FeS_2 \rightarrow 2FeS + S_2$ (1)
- $S_2 + 2O_2 (g) \rightarrow 2SO_2 (g)$ (2)
- $FeS_2 + O_2 (g) \rightarrow FeS + SO_2 (g)$ (3)
- $FeS + 2O_2 (g) \rightarrow FeSO_4$ (4)

Second reaction (hematite formation) causing another mass loss occurs at 500 – 730 °C

- $2FeSO_4 \rightarrow Fe_2O_3 + SO_2 (g) + SO_3 (g)$ (5)

Moreover, the oxidation of sulfur also helps increase the effectiveness of leaching process and metal recoveries in the next reprocessing steps. Worldwide, there is a lack of data to

MW heating in general, and therefore the estimation for energy is adopted from scaling up study of similar microwave heating processes (Bermúdez et al., 2015). Lastly, the inputs (chemical, heat, and electricity) for chemical leaching process follow the same procedures as described by (Piccinno et al., 2016) for 1000 L reactor capacity.

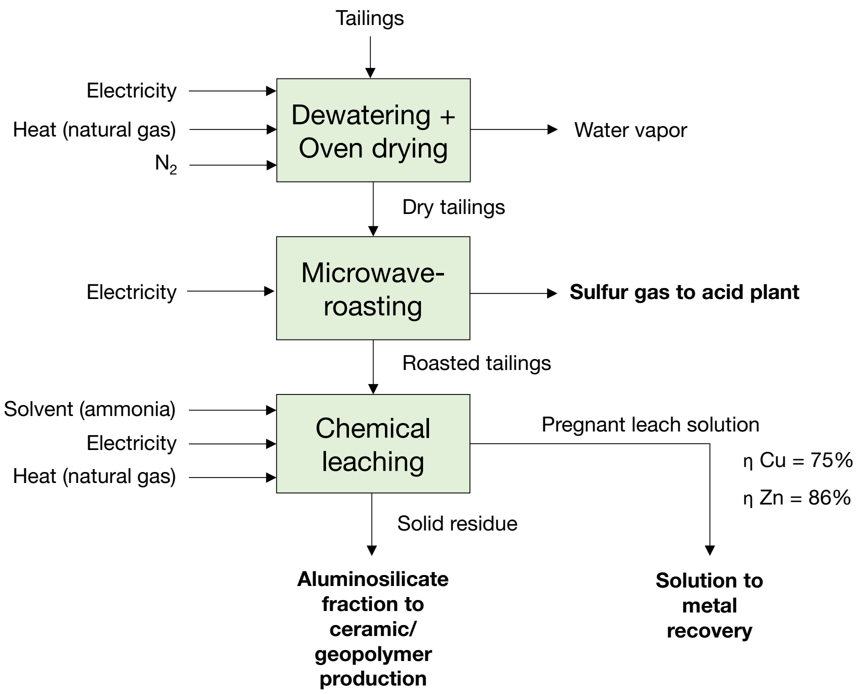


Figure B2. Process flowchart: Microwave roasting and chemical leaching

Table B8. Process parameters used in constructing life cycle inventory of MW-roasting and leaching process (Route B)

Process parameters	Values	Unit	References	Comments
Mass of tailings	1000	kg	Default	Functional unit (FU)
Moisture	30%		(McPhail et al., 2019)	Assuming from the thickened paste on site
Solid content tailings	700	kg	Calculated	Average data from samples
Cu content in tailings	4.6	kg/ ton tailings	Sample data	Average data from samples
Zn content in tailings	9.2	kg/ ton tailings	Sample data	Average data from samples
Pb content in tailings	3.8	kg/ ton tailings	Sample data	Average data from samples
N_2 for oven drying	7.508	kg/ ton tailings	Inhouse data	Purging purpose, based on (Xanthopoulos et al., 2021). Reductions to 40% of lab scale consumption are assumed
Water vapor from drying	100%		Calculated	Assuming complete drying
Dewatering energy	3.5	kWh/ ton tailings	Calculated	Assuming 5kWh/ dry materials (Piccinno et al., 2016)
Heat for oven drying	45.49	MJ/ ton tailings	Calculated	Energy for heating up and water evaporation - See

				procedure 1. Half of the heat is supplied from sulfuric acid plant
Electricity for MW-roasting	25.00	kWh/ ton tailings	Calculated	Specific energy consumption based on (Bermúdez et al., 2015)- see procedure 2
Mass loss due to pyrite oxidation during roasting	11%		Inhouse data	Oxidation of pyrite causes SO_2 emissions in the first reaction (Xanthopoulos et al., 2021)). Following reactions: $FeS_2 + O_2 (g) \rightarrow FeS + SO_2 (g)$
Mass loss due to mineralogy transformation	5.5%		Inhouse data	Conversion of Fe (II) sulfate to hematite (Xanthopoulos et al., 2021(Xanthopoulos et al., 2021)). $2FeSO_4 \rightarrow Fe_2O_3 + SO_2 (g) + SO_3 (g)$
Mole of FeS2, first mass loss	0.875	kmol	Calculated	Stoichiometric reactions
Mole of $FeSO_4$, second mass loss	0.25	kmol	Calculated	Stoichiometric reactions
Roasted tailings	584.5	kg	Calculated	Mass after loss of sulfur and mineralogy changes
Liquid-to-solid ration (L/S) of leaching process	10	L/ kg roasted tailings	Inhouse data	Based on Xanthopoulos et al., 2021(Xanthopoulos et al., 2021)
Water consumption for leaching	584.5	L	Inhouse data	Ammonia leaching, based on (Xanthopoulos et al., 2021). The consumption is 90% less than the lab scale, using framework of (Piccinno et al., 2016)
Ammonia to ammonium carbonate ratio	2		Inhouse data	(Xanthopoulos et al., 2021)
Lixiviant concentration	4	mol/ L solvent	Inhouse data	(Xanthopoulos et al., 2021)
Total mole of lixiviant (NH_3 NH_4+)	175.35	mol	Calculated	Assuming becomes 7.5% of lab scale consumption, thanks to continuous processes "95% close loop" and recycling of solvent. Make up of 2.5% is still required.
Mole of ammonia	116.9	mol	Calculated	(Xanthopoulos et al., 2021)
Mole of ammonium carbonate	58.45	mol	Calculated	(Xanthopoulos et al., 2021)
Electricity for chemical leaching reactor (stirring)	0.031	kWh/ ton tailings	Calculated	Assuming continuous stirred tank reactor (reactor capacity of 1000L), homogeneously mixed. Energy consumption of reactors is based on (Piccinno et al., 2016) – see procedure 3
Heat for leaching reactor	183.29	MJ/ ton tailings	Calculated	Same as above. Half of the heat is supplied from sulfuric acid plant.
Cu leaching efficiency	75%		Inhouse data	Optimum conditions, based on (Xanthopoulos et al., 2021)
Zn leaching efficiency	86%		Inhouse data	Optimum conditions, based on (Xanthopoulos et al., 2021)
Cu in PLS	3.4	kg	Calculated	Cu in solution for further processing

Zn in PLS	7.9	kg	Calculated	Zn in solution for further processing
Fraction to pregnant leach solution	0.41	-	Inhouse data	Collected from partners
Fraction to Ceramic/ geopolymer	0.59	-	Inhouse data	Collected from partners

Table B9. Calculation procedure and data for oven drying heat energy estimation

Procedure 1 scaling up energy for oven drying			
Parameters	**Values**	**Unit**	**Comments**
Drying efficiency	85%		Standard dryer equipment efficiency
Cp liquid	4.184	kJ/ kg °C	Properties of water at standard condition
Hv liquid	2264	kJ/ kg	
Mass liquid	15	kg	Output of belt filter, only 5% original moisture
T boil	100	deg C	
T o	25	deg C	
Q dry	45.49	MJ	$Q_{dry} = \frac{C_{p,liq} * m_{liq} * (T_{boil} - T_0) + \Delta H_{vap} * m_{vap}}{\eta_{dry}}$

Table B10. Calculation procedure and data for microwave roasting energy estimation

Procedure 2 scaling up energy for microwave roasting			
Parameters	**Values**	**Unit**	**Comments**
Roasting efficiency	85%		Assumed
Specific energy consumption at lab scale	16.6	kWh / kg	At temperature of 600 °C
Mass solid input	700	kg	Only solid fraction of tailings
Specific energy consumption at large scale, normalized to lab scale value above	0.182	%	Using correlation equation ($365x^{-1.1}$) from (Bermúdez et al., 2015). Microwave heating at x = 500, assuming asymptotic values are reached at this scale
Electricity consumption at large scale	25.00	kWh	Calculated using large scale energy consumption

Table B11. Calculation procedure and data for chemical leaching energy estimation

Procedure 3 scaling up energy chemical leaching (Properties of ammonia-ammonium solution are taken from Ammonia Handbook ("General Ammonia Information," 2006))			
Parameters	**Values**	**Unit**	**Comments**
Heating efficiency	80%		Assumption
Leaching temperature (T r)	90	deg C	From own experiments, optimum conditions for ammonia leaching
Cp liquid	3.77	kJ/ kg K	Assuming constant, as ammonia solution
Mass liquid + reactants	592.09	kg	Mass of water + ammonia + ammonium carbonate
T o	25	deg C	From own experiments
Reaction time	2	h	From own experiments, assuming 60% time is saved due to scale up

Q heat	183.29	MJ	$Q_{react(1000L)} = \frac{C_p * m_{mix} * (T_r - 298) + 3.303\frac{W}{K} * (T_r - 298)}{\eta_{heating}}$
Density of liquid	880	kg/ m^3	Density of ammonium solution in water
E stir (1000 L)	0.11	MJ	$E_{stir(1000L)} = 0.018\ m^5s^{-3} * \rho_{mix} * t$

Table B12. Life cycle inventory of the treatment of 1 tonne tailings via MW-roasting and leaching (Route B)

LCI of MW-roasting and leaching				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Electricity total	28.54	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1
	Heat, natural gas, total	228.79	MJ	Market group for heat, district or industrial, natural gas, Ecoinvent 3.7.1
	Ammonia	1.99	kg	Market for ammonia, anhydrous liquid, RER, Ecoinvent 3.7.1
	Ammonium carbonate	5.61	kg	Market for ammonium carbonate, RER, Ecoinvent 3.7.1
	Nitrogen	7.51	kg	Market for liquid nitrogen, at plant, Ecoinvent 3.7.1
	Water	584.50	kg	Market for tap water, RER, Ecoinvent 3.7.1
Technosphere output	Si-Al fraction	344.86	kg	Calculated
	PLS fraction	239.65	kg	Calculated
	SO_2	68.97	kg	Calculated according to mass loss reactions during roasting, assuming 80% to SO_2 - alternatively continues to sulfuric acid production path
	SO_3	4.05	kg	Calculated according to mass loss reactions during roasting, 20% converted to SO_3
Waste output	Water vapor Ammonia Ammonium carbonate	300.00 0.0040 0.0112	kg kg kg	Direct emissions to biosphere Fugitive emissions to air, biosphere Fugitive emissions to air, biosphere

B.6 Ion flotation and precipitation

Ion flotation studies and foam separations of this work are based on project partner's experiments in the study of (Xanthopoulos et al., 2021). Pregnant leach solution (PLS) from the previous MW-roasting and leaching process enters conditioning reactor in this scheme, where it is mixed homogenously with surfactant (selected chemicals: sodium dodecyl sulfate or often also known as sodium lauryl sulfate). The stirring process takes approximately 10 – 15 minutes at low agitation speed to avoid the formation of foam. In the ion flotation setup, other frothing agent (in this case ethanol) is also added to promote the foam that physically collapses. This phenomenon helps separate the aqueous parts that contain zinc from the PLS mixture in the later steps. The first separated stream continues to dissolution and centrifugation steps that aims to extract copper in the mixture (85% recovery efficiency). Meanwhile, the second stream enriched with zinc, is fed into a dissolved air flotation (DAF) unit to separate solid and liquid fraction. This is the ideal unit to handle inputs that are already dilute in compositions but requiring high removal efficiency of solid materials (i.e., zinc). Thus, we can practically equip and adjust the operation targets of the DAF unit to maximize the recovery of secondary zinc. One adjustment is by controlling the air to solid ratio, that influences the overall performances of the DAF unit. Our experiments suggest that the yield of zinc after aeration process could reach as high as 95%. To recycle reagents consumed in the entire process, we assume there is another distillation treatment unit next to the main operation, which returns 90% of the streams to the initial conditioning and ion flotation process. The inventory modeling of this proposal makes use of Ecosolvent tool from (Capello et al., 2007) such that required external inputs are supplied to perform solvent distillation treatments. Apart from that, the complete technical parameters and descriptions are indicated in Table B13.

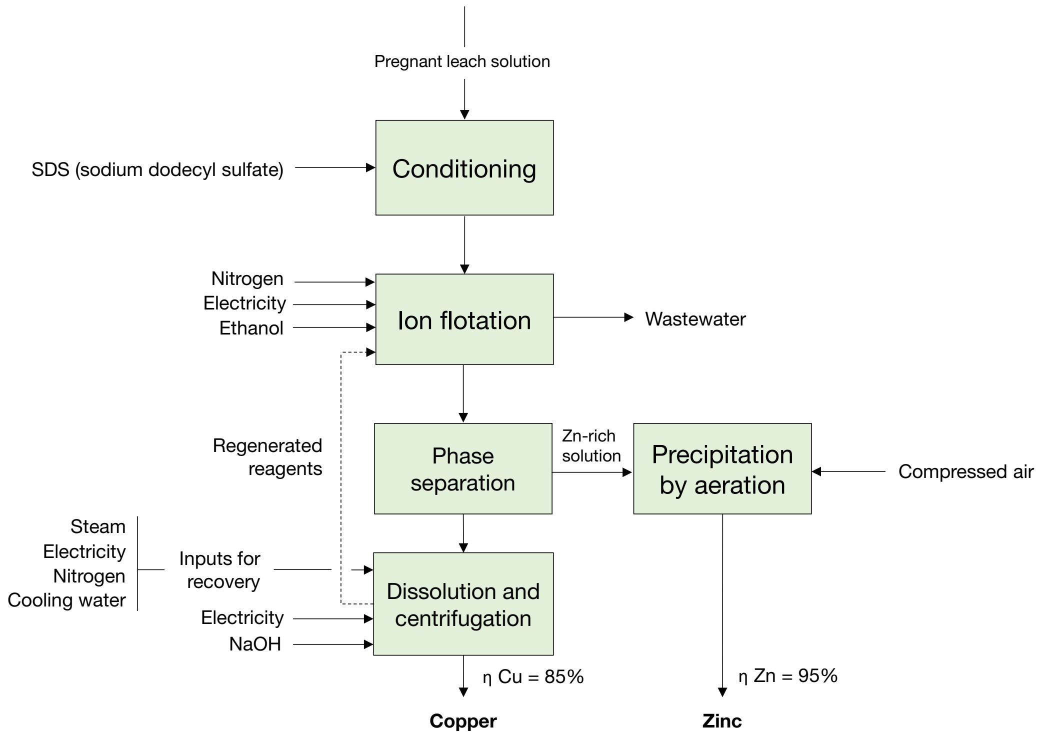


Figure B3. Process flowchart: Ion flotation and precipitation

Table B13. Process parameters used in constructing life cycle inventory of ion-flotation and metal precipitation

Process parameters	**Values**	**Unit**	**References**	**Comments**
Mass of pregnant leach solution	6084.645	kg	Calculated	Output of W-roasting and leaching
SDS consumption	0.005	mol/ L	Inhouse data	(Xanthopoulos et al., 2021)
SDS per FU	0.438	kg/ ton tailings	Calculated	SDS are 95% recycled, thanks to close-loop scheme as suggested by (Doyle, 2003) for

				ion flotation scale up potential scheme
Energy for conditioning (stirring)	0.018	kWh/ ton tailings	Calculated	Assuming continuous stirred tank reactor (reactor capacity of 10 000L), homogeneously mixed. Energy consumption of reactors is based on (Piccinno et al., 2016) – see procedure 4
N_2 for ion flotation	4.932	kg/ ton tailings	Inhouse data	Floating purpose, based on (Xanthopoulos et al., 2021). Reductions to 40% of lab scale consumption
Energy for ion flotation	1.078	kWh/ ton tailings	Broadhurst et al, 2015(Broadhurst et al., 2015). Accounting only the solid fraction of PLS as "ore feed"	Flotation and flocculation pumping (25% extra for latter purpose)
Wastewater	313.459	kg	Calculated	Assuming all liquid fraction from PLS that are no longer recycled and unused PLS after centrifugation and aeration
Ethanol specific consumption	0.005	v/ V total	Inhouse data	(Xanthopoulos et al., 2021)
Ethanol consumption per FU	2.282	kg/ ton tailings		Ethanol for frother, based on (Xanthopoulos et al., 2021). Assuming becomes 7.5% of lab scale consumption, thanks to continuous processes "95% close loop" and recycling of solvent. Make up of 2.5% is still required.
Copper recovery, efficiency	0.850		Inhouse data	Optimum conditions, based on (Xanthopoulos et al., 2021)
Zinc recovery, efficiency	0.950		Inhouse data	Optimum conditions, based on (Xanthopoulos et al., 2021)
Cu recovered	2.917	kg/ ton tailings	Calculated	Secondary Cu recovered
Zn recovered	7.480	kg/ ton tailings	Calculated	Secondary Zn recovered
Centrifugation energy	10.000	kWh/ ton input	Calculated	Assuming 10kWh/ input according to (Piccinno et al., 2016)
Cu-rich solution, trf coeff.	0.281		Inhouse data	Based on (Xanthopoulos et al., 2021)
Zn-rich solution, trf coeff.	0.719		Inhouse data	Based on (Xanthopoulos et al., 2021)
Mass flow to Cu dissolution/ centrifugation	87.938	kg	Calculated	Based on (Xanthopoulos et al., 2021)
Mass flow to dissolved air flotation	225.521	kg	Calculated	Based on (Xanthopoulos et al., 2021)
Energy for centrifugation	3.135	kWh/ ton tailings	Calculated	Calculated
Electricity for solvent recovery (distillation)	6.269	kWh/ ton tailings	Simulated	Ecosolvent tool is used, based on the work of (Capello et al., 2007)

Steam for solvent recovery (distillation)	188.075	kg/ ton tailings	Simulated	Ecosolvent tool is used, based on the work of (Capello et al., 2007)
Nitrogen for solvent recovery (distillation)	0.588	kg/ ton tailings	Simulated	Ecosolvent tool is used, based on the work of (Capello et al., 2007)
Cooling water for solvent recovery (distillation)	12.538	kg/ ton tailings	Simulated	Ecosolvent tool is used, based on the work of (Capello et al., 2007)
NaOH consumption	0.276	kg/ ton tailings	Calculated	Following stoichiometric reactions: $M(OH)_2$ (s) → M^{2+} (aq) + 2 OH^-(aq) M = Copper metal Close loop system that requires 7.5% makeup
Air to solid ratio, dissolved air flotation	0.020	kg air/ kg inputs	Standard value	Typical values for DAF unit. Air at 5 bar, density = 6.9 kg/m3 at standard temperature. Proxy industrial data from the LCA of industrial effluent, based on (O'Connor et al., 2014) study
Compressed air consumption	0.632	m^3 air/ ton tailings	Calculated	Proxy industrial data from the LCA of industrial effluent, based on (O'Connor et al., 2014) study
Electricity for clarification (DAF unit)	0.380	kWh/ m^3	O'Connor et al., 2014(O'Connor et al., 2014)	Collection of average industrial data and measurements. The DAF unit has a capacity of 5 m^3, assumed.
Electricity for DAF	1.900	kWh/ ton tailings	Calculated	Based on proxy data (O'Connor et al., 2014)
Fugitive emissions of ethanol	0.005	kg/ ton tailings	Calculated	0.2% of input, recommendations of chemicals LCI development by (Jiménez-González et al., 2000)
Fugitive emissions of SDS	0.001	kg/ ton tailings	Calculated	0.2% of input, recommendations of chemicals LCI development by (Jiménez-González et al., 2000)
Fugitive emissions of sodium hydroxide	0.001	kg/ ton tailings	Calculated	0.2% of input, recommendations of chemicals LCI development by (Jiménez-González et al., 2000)

Table B14. Calculation procedure and data for conditioning energy estimation

Procedure 4 scaling up energy, conditioning process			
Parameters	**Values**	**Unit**	**Comments**
Np	0.79	-	Power nr of impellers (constants)
Density of liquid	880	kg/ m^3	Density of ammonium solution in water
N	0.658	1/s	Rotational speed of agitator
d	0.803	m	Impeller diameter
t	900	sec	15 mins, based on (Xanthopoulos et al., 2021)

Eff stir	90%		Assumed
E stir (10 000 L)	0.066	MJ	$E_{stir} = \frac{N_p * \rho_{mix} * N^3 * d^5 * t}{\eta_{stir}}$

Table B15. Life cycle inventory of the treatment of PLS through ion flotation and precipitation

LCI of ion flotation-precipitation				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Electricity total	12.40	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1
	Ethanol	2.28	kg	Market group for heat, district or industrial, natural gas, Ecoinvent 3.7.1
	SDS (surfactant)	0.44	kg	Market for non-ionic surfactant, GLO, Ecoinvent 3.7.1
	NaOH	0.28	kg	Market for sodium hydroxide, RER, Ecoinvent 3.7.1
	Nitrogen	5.52	kg	Market for liquid nitrogen, at plant, Ecoinvent 3.7.1
	Compressed air	0.63	m^3	Market for compressed air, 600 kPa gauge, Ecoinvent 3.7.1
	Steam	188.075	kg	Market for steam, in chemical industry, Ecoinvent 3.7.1
	Cooling water	12.6	kg	Natural resource, water from lake, biophere3
Technosphere output	Secondary Cu	2.92	kg	Calculated
	Secondary Zn	7.48	kg	Calculated
Waste output	Wastewater	313.46	kg	Treatment of wastewater, Europe, Ecoinvent 3.7.1
	Ethanol	0.0046	kg	Direct emissions to biosphere, air
	SDS (surfactant)	0.0009	kg	Direct emissions to biosphere, air
	Sodium hydroxide	0.0006	kg	Direct emissions to biosphere, air

B.7 Sulfuric acid production

The production of sulfuric acid in this study is based on the primary industrial data (e.g., pyrite roasting) (Runkel & Sturm, 2009), which converts pyrite as the source of sulfur at large scale production. This process has become the proven method to deal with sulfur dioxide gas in the industry, solving air pollution problems at the stack and potentially generating by-products if the sulfuric acid or the derivatives market is available nearby. At present, sulfuric acid production from off gas contributes to around 30% of market needs globally (King et al., 2013). It makes use of sulfur SO_2 off gas from the roasting process as the main input to produce acid, via wet gas cleaning steps, chemical reactions with catalysts, and water additions in the absorption tower. In this work, the life cycle inventory is developed as a generic model, as there is a lack of technical sheet showing the detailed performances for each processing steps. The data are principally taken from the reports on fertilizer production and obtained from the summary of best available technology (BAT) document for sulfuric acid production in Europe (Europe Fertilizers, 2000; European Commission, 2007). Nevertheless, this aggregated block of processes is still representative and do not vary significantly in terms of foreground unit processes, given that sulfuric acid production is one of the most mature chemical technologies. Hence, there is no upscaling nor modification to the performances in the model as opposed to other life cycle inventories we constructed in this study.

The main reactions in the modeled sulfuric acid plant (with conversion efficiency between 98 – 99.8%).

- Combustion: $2\ H_2S + 3\ O_2 \rightleftharpoons 2\ H_2O + 2\ SO_2 + 518$ kJ/mol
- Oxidation: $2\ SO_2 + O_2 \rightleftharpoons 2\ SO_3 + 99$ kJ/mol

  (With the presence of a vanadium oxide catalyst)
- Hydration: $SO_3 + H_2O \rightleftharpoons H_2SO_4$ (g) + 101 kJ/mol
- Condensation: H_2SO_4 (g) $\rightleftharpoons$ H_2SO_4 (l) + 90 kJ/mol

The overall reactions are highly exothermic; As a result, it is normally the case that sulfuric acid plant generates heat that can be consumed internally or exported to third parties. In our study, we transfer some portion of the heat to the other processes that demand energy (heat or steam) that would otherwise require natural gas from external sources. This surplus heat generally saves resource and energy consumption in the value chain (i.e., in our study for heating reactors) or downstream chains for the adjacent phosphoric acid plant.

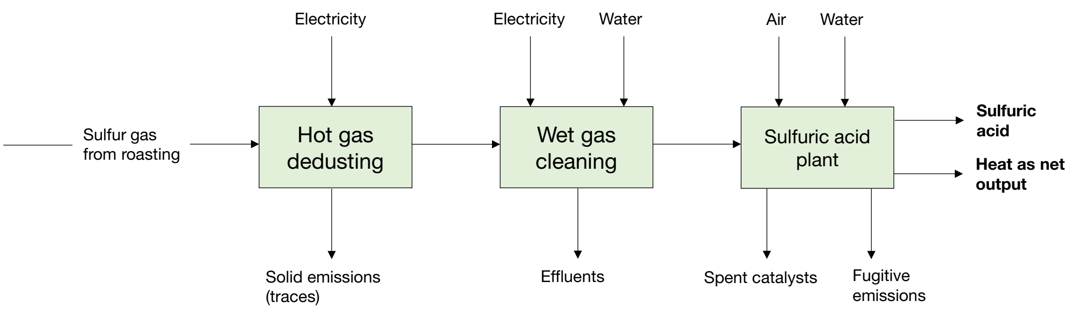


Figure B4. Process flowchart: Sulfuric acid production

Table B16. Process parameters in constructing life cycle inventory of sulfuric acid plant

Process parameters	Values	Unit	References	Comments
Mass flow of SO_2 from roasting	68.968	kg/ ton tailings	Modeled	From MW-roasting process
Mass flow of SO_3 from roasting	4.053	kg/ ton tailings	Modeled	From MW-roasting process
Total electricity input for the entire process	40	kWh/ tonne H_2SO_4	(European Commission, 2007)	Assumed values, taken from a range of collected plant data in the document

Recoverable heat from the entire acid plant	2400	MJ/ tonne H_2SO_4	(European Commission, 2007)	Single value from pyrite roasting plant data. 50% is internally transferred to reactors heating in MW-roasting & leaching units
Conversion rate to sulfuric acid	99.80%		(European Commission, 2007)	The minimum conversion efficiency of sulfur source to sulfuric acid is 99.6%, according to best available technology document (BAT). We take 99.8% as the average values in this sector.
Sulfuric acid produced	110.35	kg/ ton tailings	(European Commission, 2007)	A series of exothermic reactions: Oxidation: $2\ SO_2 + O_2 \rightleftharpoons 2\ SO_3$ Hydration: $SO_3 + H_2O \rightleftharpoons H_2SO_4$ (g) Condensation: H_2SO_4 (g) $\rightleftharpoons$ H_2SO_4 (l)
Air consumption	81.94	kg/ ton tailings	(European Commission, 2007)	Using above stoichiometric reactions, assuming air contains 21% oxygen composition
Wastewater produced	38.22	kg/ ton tailings	(Daniel Mussatti, 2002)	Calculated water consumed in the ratio of 10 gal/ 1000ft$_3$ gas input
Water consumption	62.54	kg/ ton tailings	(European Commission, 2007)	Assumed following stoichiometric reactions +20% and for scrubbers
Total electricity input for the entire process	4.41	kWh/ ton tailings	Calculated	For internal consumption
Total heat export from the entire process	496.58	MJ/ ton tailings	Calculated	For internal consumption
SO2 Emissions into air	0.331	kg/ ton tailings	(European Commission, 2007)	Data taken as is, from pyrite roasting plant
SO3 Emissions into air	0.022	kg/ ton tailings	(European Commission, 2007)	Data taken as is, from pyrite roasting plant
NOx Emissions into air	0.00021	kg/ Nm^3	(European Commission, 2007)	Data taken as is, from pyrite roasting plant
Solid emissions (spent catalyst)	4.414	kg/ ton tailings	(European Commission, 2007)	Data taken as is, from pyrite roasting plant
Dust emissions as pyrite	0.003	kg/ ton tailings	Calculated	BAT report states that around 5% of dust will be trapped in the dedusting process. 0.1% emitted to air.

Table B17. Life cycle inventory of the sulfuric acid plant

LCI of sulfuric acid production				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Electricity	4.41	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1

	Air	11.88	m^3	Resource from nature
	Water	62.54	kg	Market for tap water, RER, Ecoinvent 3.7.1
Technosphere output	Sulfuric acid	110.35	kg	Calculated
	Heat natural gas	182.74	MJ	Calculated (based on 92% recovery from absorption process and 75% heat exchanger efficiency)
Waste output	Wastewater	38.22	kg	Treatment of wastewater, Europe, Ecoinvent 3.7.1
	SO_2	0.33	kg	Direct emissions to biosphere, air
	SO_3	0.0221	kg	Direct emissions to biosphere, air
	NO_x	0.0002	kg	Direct emissions to biosphere, air
	Solid emissions	4.41	kg	Landfill, hazardous waste
	Dust emissions	0.003	kg	Direct emissions to biosphere, air (as iron to air)

B.8 Calcium sulfoaluminate cement (CSA) production

Currently, CSA cement is considered a niche product compared to standard Portland cement with its main producer and market in China (Naqi & Jang, 2019). Given its ability to rapidly cure with high early strength, there is a growing interest to use such product for specialized applications. According to the review of (Habert, 2013), there are several reasons why CSA cements could be considered as greener alternatives to traditional cement, namely: 1) the reduced amount original raw materials (limestone) that would result in less energy consumed for calcination, 2) lower calcination temperature at around 1200 – 1300 °C, 3) possibilities to incorporate secondary raw materials to enhance the final properties of cement products. In order to construct the life cycle inventory modeling for this study, we based our process parameters from the research partners (Martins et al., 2020) (i.e., ratio of secondary pyrite fraction in the overall clinker formation), supplemented in the subsequent steps by adding relevant large-scale primary data from cement plants in China. The lab protocols originally investigated by partners primarily use small dose of chemicals that have been replaced accordingly in the life cycle inventory models (i.e., replacement of aluminum oxide with bauxite). The secondary life cycle data represents direct data acquisition from private partners and company reports, as evaluated by Ren et al (2017). Therefore, we updated the entire inventory by linking the background datasets the authors use in their research with the appropriate input flows in Europe as our plant location. Lastly, the emission factors for the clinkerization processes and other technical parameters are gathered from the review of CSA cement at industrial scale (Gartner, 2004).

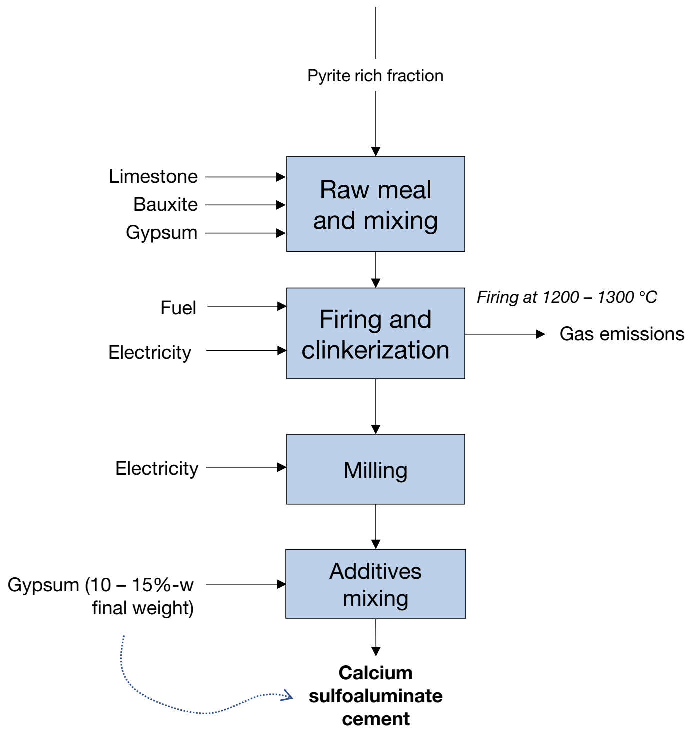


Figure B5. Process flowchart: Calcium sulfoaluminate production

Table B18. Process parameters in constructing life cycle inventory of CSA cement production

Process parameters	Values	Unit	References	Comments
Mass flow of pyrite rich fraction	287.00	kg	Calculated	Taken from flocculation-flotation unit process
Ratio of pyrite fraction in raw meal	0.14		(Martins et al., 2020)	Collected from partners
Ratio of limestone in raw meal	0.36		(Martins et al., 2020)	Collected from partners
Ratio of bauxite in raw meal	0.36		(Martins et al., 2020)	Collected from partners
Ratio of gypsum in raw meal	0.14		(Martins et al., 2020)	Collected from partners
Total mass after mixing	2045.79	kg	Calculated	Using ratios from project's data
Limestone input	735.90	kg	Calculated	Using ratios from project's data
Bauxite input	735.90	kg	Calculated	Using ratios from project's data
Gypsum input	287.00	kg	Calculated	Using ratios from project's data
Produced CSA clinker	1329.77	kg clinker	Calculated	Using ratios from project's data
CSA cement after additives	1564.43	kg CSA/ ton tailings	Calculated	Using ratios from project's data
Gypsum as additives	234.66	kg/ ton tailings	Calculated	Using ratios from project's data
Total electricity consumption	51.91	kWh/ tonne of cement	(Ren et al., 2017)	Large scale CSA cement production based on mine waste in China

Coal consumption	107.35	kg/ tonne of cement	(Ren et al., 2017)	Large scale CSA cement production based on mine waste in China
Diesel consumption	0.08	kg/ tonne of cement	(Ren et al., 2017)	Large scale CSA cement production based on mine waste in China
Water consumption	43.02	kg/ tonne of cement	(Ren et al., 2017)	Large scale CSA cement production based on mine waste in China
Electricity per FU	81.21	kWh/ ton tailings	Calculated	Based on the alternative CSA production, industrial data study of (Ren et al., 2017)
Coal consumed per FU	167.94	kg/ ton tailings	Calculated	Based on the alternative CSA production, industrial data study of (Ren et al., 2017)
Diesel consumed per FU	0.12	kg/ ton tailings	Calculated	Based on the alternative CSA production, industrial data study of (Ren et al., 2017)
Water consumed per FU	67.30	kg/ ton tailings	Calculated	Based on the alternative CSA production, industrial data study of (Ren et al., 2017)

Table B19. Life cycle inventory of the CSA cement production

LCI of CSA cement				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Electricity	81.21	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1
	Coal	167.94	kg	Market for hard coal, Europe, Ecoinvent 3.7.1
	Pyrite	287.00	kg	Flows from flocculation-flotation process
	Limestone	735.90	kg	Market for limestone, crushed for mill, CH, Ecoinvent 3.7.1
	Bauxite	735.90	kg	Market for bauxite, GLO, Ecoinvent 3.7.1
	Diesel	0.08	kg	Market for diesel, Europe, Ecoinvent 3.7.1
	Gypsum	521.66	kg	Market for gypsum, RER, Ecoinvent 3.7.1
	Water	67.30	kg	Market for tap water, RER, Ecoinvent 3.7.1
Technosphere output	CSA cement	1564.43	kg	Calculated
Waste output	Gas and other emissions	See sources	-	Assumed to be similar to clinker and cement production data. Best available technologies applied at industrial scale in Europe. Clinker production, CH, Ecoinvent 3.7.1. CO_2 emissions are adjusted to CSA cement following the values from (Gartner, 2004)

B.9 Ceramic tile production

Ceramic tile production data are based on actual scale experiments, supplemented by project's partner data carried out by our in-house collaborators(Veiga Simão et al., 2021). The mix designs from mini scale production were adapted linearly in the efforts to upscale the data for large scale tile production. In general, up to 10% of tailings can be introduced into the finished product, with a higher proportion possible whenever sulfur content is low. Apart from that, the life cycle data such as electricity and heat consumption for tiles production are also completed by harmonizing our inventories with the other large-scale tiles manufacturing in Switzerland already present in Ecoinvent 3.7.1 (Ibáñez-Forés et al., 2011). The decisions are solely based on the better life cycle performance when we compare Swiss ceramic manufacturers with the average European data. This implies the former's inventory are updated and represent best available practices in the industry. We updated, however, the dataset to region-specific case in our study, that is specifying the source of technosphere to Europe in Ecoinvent. If this is not possible due to lack of data, the global 'GLO' or rest of the world 'ROW' dataset were used as the last choice. To sum up, the general approach implemented herein is similar to what has been done in comparative LCAs of tiles in other regions by (Maia de Souza et al., 2016).

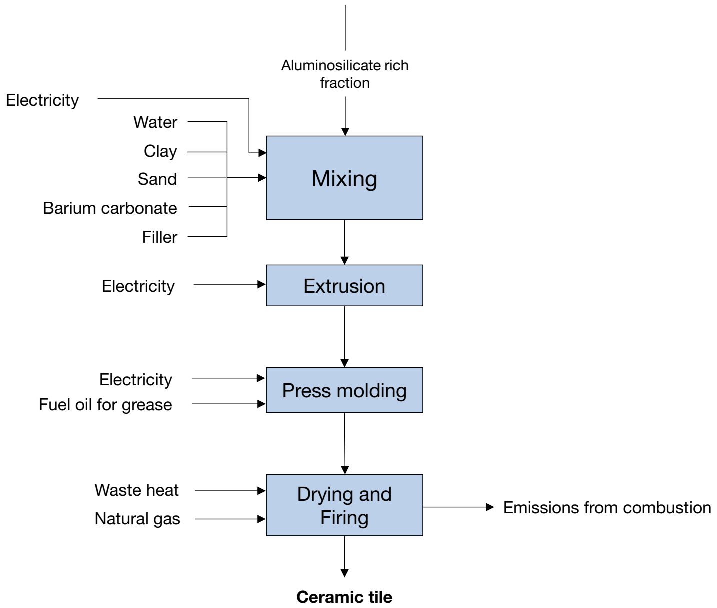


Figure B6. Process flowchart: Ceramic tile production

Table B20. Process parameters in constructing life cycle inventory of ceramic tile production

Process parameters	Values	Unit	References	Comments
Mass flow of Si-Al fraction, route A	413.00	kg	Calculated	From beneficiation stage
Mass flow of Si-Al fraction, route B	344.85	kg	Calculated	From extraction stage

Ratio of Si-Al fraction in mix	10%		(Veiga Simão et al., 2021)	Project collaborators data, linearly upscaled
Ratio of barium carbonate in mix	0.50%		(Veiga Simão et al., 2021)	Project collaborators data, linearly upscaled
Ratio of sand in mix	14.63%		(Veiga Simão et al., 2021)	Project collaborators data, linearly upscaled
Ratio of clay in mix	42.10%		(Veiga Simão et al., 2021)	Project collaborators data, linearly upscaled
Ratio of filler in mix	33.20%		(Veiga Simão et al., 2021)	Project collaborators data, linearly upscaled
Other material inputs	-	-	Ecoinvent 3.7.1	Based on Ecoinvent 3.7.1 data, best available technologies in Europe were taken from Swiss manufacturers with adaptations
Electricity input	0.31	kWh/ kg ceramic	Ecoinvent 3.7.1	Same as above
Heat input	5.57	MJ/ kg ceramic	Ecoinvent 3.7.1	Same as above

Table B21. Life cycle inventory of the ceramic tile production (Route A)

LCI of Ceramic from tailings, route A				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Electricity	1371.16	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1
	Heat	23030.95	MJ	Market group for heat, district or industrial, natural gas, Ecoinvent 3.7.1
	Barium carbonate	22.96	kg	Market for barium carbonate, GLO, Ecoinvent 3.7.1
	Sand	631.89	kg	Market for silica sand, GLO, Ecoinvent 3.7.1
	Clay	1817.61	kg	Market for clay, CH, Ecoinvent 3.7.1
	Filler (feldspar)	1434.76	kg	Market for feldspar, GLO, Ecoinvent 3.7.1
	Other material inputs	Tap water, lime, etc.	-	Assumed to be similar to large scale ceramic technologies applied at industrial scale in Europe.
Technosphere output	Ceramic tile	4130.00	kg	Ceramic tile production, CH, Ecoinvent 3.7.1.
Waste output	Particulate and other emissions and wastewater treatment	See sources	-	Calculated, 10% containing Si-Al fraction

Table B22. Life cycle inventory of the ceramic tile production (Route B)

LCI of Ceramic from tailings, route B				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Electricity	1079.40	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1
	Heat	19230.84	MJ	Market group for heat, district or industrial, natural gas, Ecoinvent 3.7.1
	Barium carbonate	19.17	kg	Market for barium carbonate, GLO, Ecoinvent 3.7.1
	Sand	527.63	kg	Market for silica sand, GLO, Ecoinvent 3.7.1
	Clay	1517.71	kg	Market for clay, CH, Ecoinvent 3.7.1
	Filler (feldspar)	1198.03	kg	Market for feldspar, GLO, Ecoinvent 3.7.1
	Other material inputs	Tap water, lime, etc.	-	Assumed to be similar to large scale ceramic technologies applied at industrial scale in Europe.
Technosphere output	Ceramic tile	3448.55	kg	Ceramic tile production, CH, Ecoinvent 3.7.1.
Waste output	Particulate and other emissions and wastewater treatment	See sources	-	Calculated, 10% containing Si-Al fraction

B.10 Geopolymer binder production

Geopolymer production data are based on the experimental research of Niu et al(Niu et al., 2020), which are scaled up in the metallurgical process simulation environment of HSC (Metso Outotec, 2020) Chemistry 10 © as implemented in the previous study of (Adrianto et al., 2021) and (Niu et al., 2021). The process mainly comprises two steps(Hassan et al., 2019) to mechanically activate clinochlore and muscovite (rich in Si and Al-based minerals) materials in the feed stream. One main difference that we adopted in this study is that we maximize the use rate of Si-Al fraction present in tailings. 0.5%-w amount of lithium chloride is added in the milling equipment (HIG© Mill setup) that helps decreasing the grinding energy during activation. We specifically choose the most optimized process path from the previous study that has higher potential in saving water and electricity consumption. In the subsequent step, the precursor or the materials are mixed with alkali activators (sodium silicate, sodium hydroxide, and water) with specific formulation developed in the experimental work. The proportion of this formulation is kept the same for the upscaled scenarios, assuming that the chemistry remains equal for both scales irrespective of the processing capacity. Other important parameters to construct the life cycle inventory and modeling are described in detail in the work of (Niu et al., 2021) and (Adrianto et al., 2021). The latter uses the same basis, except that the product generated can be further processes to manufacture geopolymer concrete for Portland cement concrete replacement.

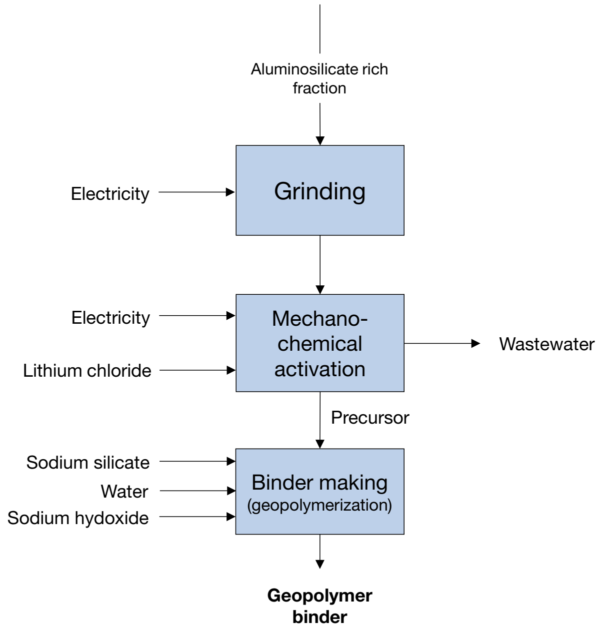


Figure B7. Process flowchart: Geopolymer production

Table B23. Process parameters in constructing life cycle inventory of geopolymer production

Process parameters	**Values**	**Unit**	**References**	**Comments**
Mass flow of Si-Al fraction, route A	413.0	kg	Calculated	From beneficiation stage
Mass flow of Si-Al fraction, route B	344.9	kg	Calculated	From extraction stage
Electricity input for precursor making	0.078	kWh/ kg precursor	Niu et al., 2021	Obtained from process simulation, data upscaled from lab experiments and parameters
Water consumption for precursor	1.48E-01	kg/ kg precursor	Same as above	Same as above
Lithium chloride	2.37E-02	kg/ kg precursor	Same as above	Same as above
Wastewater to treatment	5.10E-05	m^3/ kg precursor	Same as above	Same as above
Sodium silicate consumption	7.22E-02	kg/ kg binder	Same as above	Same as above
Sodium hydroxide consumption	1.45E-02	kg/ kg binder	Same as above	Same as above
Water for geopolymerization	3.04E-01	kg/ kg binder	Same as above	Same as above
Precursor consumption	6.10E-01	kg/ kg binder	Same as above	Same as above
Precursor, route A	422.8	kg/ ton tailings	Calculated	All follow the same ratio in the upscaling part, except for electricity using default 0.7 power law from 100ton to 1ton capacity, as applied in the

				emerging tech study of (van der Hulst et al., 2020)
Electricity for precursor, route A	32.98	kWh/ ton tailings	Calculated	-
Wastewater effluent, route A	2.16E-02	m^3/ ton tailings	Calculated	-
Lithium chloride, route A consumption	10.02	kg/ ton tailings	Calculated	-
Binder generated, route A	693.57	kg/ ton tailings	Calculated	-
Sodium silicate, route A consumption	50.07	kg/ ton tailings	Calculated	-
Sodium hydroxide, route A consumption	10.06	kg/ ton tailings	Calculated	-
Water for precursor, route A consumption	62.57	kg/ ton tailings	Calculated	-
Water, route A consumption	210.66	kg/ ton tailings	Calculated	-
Precursor, route B	353.0	kg/ ton tailings	Calculated	-
Electricity for precursor, route B	27.54	kWh/ ton tailings	Calculated	-
Wastewater effluent, route B	1.80E-02	m^3/ ton tailings	Calculated	-
Lithium chloride, route B consumption	8.37	kg/ ton tailings	Calculated	-
Binder generated, route B	579.13	kg/ ton tailings	Calculated	-
Sodium silicate, route B consumption	41.81	kg/ ton tailings	Calculated	-
Sodium hydroxide, route B consumption	8.40	kg/ ton tailings	Calculated	-
Water, route B consumption	175.90	kg/ ton tailings	Calculated	-
Water for precursor, route B consumption	52.25	kg/ ton tailings	Calculated	-

Table B24. Life cycle inventory of the geopolymer binder production (Route A)

LCI of CSA cement from tailings, route A				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Water	273.23	kg	Market for tap water, RER, Ecoinvent 3.7.1
	Lithium chloride	10.02	kg	Market for lithium chloride, GLO, Ecoinvent 3.7.1
	Electricity	32.98	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1
	Sodium silicate	50.07	kg	Market for sodium silicate, solid, RER, Ecoinvent 3.7.1
	Sodium hydroxide	10.06	kg	Sodium hydroxide, without water, RER, Ecoinvent 3.7.1

Technosphere output	Geopolymer binder	693.57	kg	Calculated
Waste output	Wastewater	0.02	m^3	Treatment of wastewater, Europe, Ecoinvent 3.7.1

Table B25. Life cycle inventory of the geopolymer binder production (Route B)

LCI of CSA cement from tailings, route B				
Type of inputs/outputs	**Flows**	**Values per FU**	**Unit**	**Sources of LCI data**
Technosphere input	Water	228.15	kg	Market for tap water, RER, Ecoinvent 3.7.1
	Lithium chloride	8.37	kg	Market for lithium chloride, GLO, Ecoinvent 3.7.1
	Electricity	27.54	kWh	Market for electricity, Portugal, Medium Voltage, Ecoinvent 3.7.1
	Sodium silicate	41.81	kg	Market for sodium silicate, solid, RER, Ecoinvent 3.7.1
	Sodium hydroxide	8.40	kg	Sodium hydroxide, without water, RER, Ecoinvent 3.7.1
Technosphere output	Geopolymer binder	579.13	kg	Calculated
Waste output	Wastewater	0.018	m^3	Treatment of wastewater, Europe, Ecoinvent 3.7.1

B.10 Life cycle inventory data for substituted primary products/ avoided services

Table B26. Used Ecoinvent 3.7.1 processes for the substituted primary materials production/ avoided services (including building materials, energy, and other credits)

Primary material production	**Ecoinvent 3.7.1 process**	**Location**	**Unit**	**Remark**
CSA cement	NA (Built from secondary LCA data of others, see section 8) *	RER	kg	Adapted primary production inventory based on secondary data
Ceramic tile	Ceramic tile production *	CH (for typical European representation)	kg	Adapted primary production inventory based on own calculation data
Portland cement	Cement production, Portland	Europe without Switzerland	kg	Only primary production
Sulfuric acid	Market for sulfuric acid	RER	kg	Only primary production
Heat	Market group for heat, district or industrial, natural gas	RER	MJ	Recent market data
Cu	Market for copper	GLO	kg	Recent market data
Zn	Market for zinc	GLO	kg	Recent market data
Treatment of sulfidic tailings (via landfill)	NA (created by modifying emissions data of (Doka, 2017) in Ecoinvent)	Portugal (site-specific)	kg	Adapted waste treatment inventory based on recent studies on regionalized mine tailings model (Adrianto et al., 2022)

*Adapted processes, with calculation methods described in the respective sections

B.11 Additional LCA results

Table B27. Complete LCA results according to all assessed indicators for all routes in base cases, including breakdown for each reprocessing step (also presented in Appendix-3 Sheet 9). Abbreviation for impact categories can be found in Table B3. Color coding is grouped per column: from green as lowest, to red as highest values.

Route	ReCiPe (H) midpoint indicators																		3 additional categories			Endpoint
A-1 Steps	**ALOP**	**GWP**	**FDP**	**FETP**	**FEP**	**HTP**	**IRP**	**METP**	**MEP**	**MDP**	**NLTP**	**ODP**	**PMFP**	**POFP**	**TAP**	**TETP**	**ULOP**	**WDP**	**CED**	**USEtox, ecotoxicity**	**USETox, human toxicity**	**ReCiPe (H), SS**
Flocculation-flotation	0.56	9.89	3.32	0.16	3.83E-03	2.69	0.64	0.14	8.27E-03	0.1	6.56E-04	4.89E-07	0.02	0.03	0.06	3.54E-04	0.06	0.08	131.98	45.74	3.48E-06	1.01
CSA cement production	11.34	589.29	143.87	5.32	2.49E-01	201.70	10.53	5.05	1.68E-01	18.0	3.61E-02	1.12E-05	1.45	3.09	2.89	1.78E-02	3.14	1.21	6186.50	1517.09	1.81E-04	56.99
Ceramic tile production	71.59	1923.52	863.16	14.32	2.73E-01	280.39	61.38	13.46	1.99E-01	20.5	1.33E-01	1.93E-04	37.81	3.94	5.17	5.06E-02	8.43	5.01	31207.58	4470.33	2.71E-04	384.45
Substituted CSA cement	-27.90	-1034.58	-176.64	-4.79	-1.18E-01	-130.82	-15.54	-4.43	-1.50E-01	-22.0	-4.65E-02	-3.15E-05	-1.67	-3.43	-3.37	-2.87E-02	-4.77	-2.20	-7678.06	-1798.99	-1.66E-04	-83.22
Substituted ceramic tile	-85.06	-2011.30	-804.66	-20.17	-5.02E-01	-394.23	-149.80	-17.39	-2.77E-01	-26.2	-1.43E-01	-1.40E-04	-39.16	-4.56	-5.29	-8.77E-02	-10.59	-7.93	-29741.07	-5825.44	-3.70E-04	-396.45
Avoided tailings landfilling	-0.394	0	0	-234.4	-0.21466	-2629.8	0	-200	-0.00512	0.0	0.004545	0.00E+00	0	0	0	-0.00022	-0.591	0	0	-118457.78	-0.008	-36.612
Net environmental impacts	-29.87	-523.18	***29.05***	-240	-0.31	-2670	-92.79	-203	-0.06	-9.6	-0.02	***3.30E-05***	-1.55	-0.93	-0.53	-0.05	-4.32	-3.83	***106.93***	-120049.05	-0.01	-73.83
A-2 Steps																						
Flocculation-flotation	0.56	9.89	3.32	0.16	3.83E-03	2.69	0.64	0.14	8.27E-03	0.1	6.56E-04	4.89E-07	0.02	0.03	0.06	3.54E-04	0.06	0.08	131.98	45.74	3.48E-06	1.01
CSA cement production	11.34	589.29	143.87	5.32	2.49E-01	201.70	10.53	5.05	1.68E-01	18.0	3.61E-02	1.12E-05	1.45	3.09	2.89	1.78E-02	3.14	1.21	6186.50	1517.09	1.81E-04	56.99
Geopolymer production	10.71	98.16	26.33	4.03	4.91E-02	41.03	9.37	3.61	7.14E-02	6.8	1.61E-02	1.56E-05	0.19	0.31	0.51	1.05E-02	1.36	1.48	1062.37	1190.53	5.36E-05	10.49
Substituted CSA cement	-27.90	-1034.58	-176.64	-4.79	-1.18E-01	-130.82	-15.54	-4.43	-1.50E-01	-22.0	-4.65E-02	-3.15E-05	-1.67	-3.43	-3.37	-2.87E-02	-4.77	-2.20	-7678.06	-1798.99	-1.66E-04	-83.22
Substituted Portland cement	-5.76	-590.37	-49.22	-2.70	-6.02E-02	-56.24	-23.15	-2.45	-5.19E-02	-4.4	-1.24E-02	-1.42E-05	-0.42	-1.05	-1.03	-8.11E-03	-1.31	-1.18	-2101.46	-782.39	-5.84E-05	-36.80
Avoided tailings landfilling	-0.394	0	0	-234.4	-0.21466	-2629.8	0	-200	-0.00512	0.0	0.004545	0.00E+00	0	0	0	-0.00022	-0.591	0	0	-118457.78	-0.008	-36.612
Net environmental impacts	-11.44	-927.61	-52.35	-232	-0.09	-2571	-18.15	-198	***0.04***	-1.4	0.00	-1.84E-05	-0.43	-1.05	-0.94	-0.01	-2.12	-0.61	-2398.67	-118285.80	-0.01	-88.14
B-1 Steps																						
MW-roasting and leaching	1.30	42.15	17.81	0.61	9.68E-03	7.60	3.19	0.55	4.77E-02	0.8	1.37E-03	3.62E-06	0.04	0.08	0.13	1.57E-03	0.14	0.78	648.53	171.86	7.95E-06	4.29
Sulfuric acid production	0.18	2.94	0.97	0.09	1.58E-03	1.13	0.55	0.08	1.45E-03	0.1	1.95E-04	1.46E-07	0.07	0.04	0.35	1.12E-04	0.04	0.02	38.88	25.58	1.16E-06	0.65
Ion flotation and precipitation	2.80	65.11	27.72	0.45	1.08E-02	8.09	3.69	0.38	1.75E-02	0.4	1.26E-03	6.91E-06	0.06	0.14	0.19	3.84E-01	0.16	0.28	1053.85	120.33	8.94E-06	6.89
Ceramic tile production	59.78	1606.12	720.73	11.95	2.28E-01	234.12	51.25	11.24	1.66E-01	17.1	1.11E-01	1.61E-04	31.57	3.29	4.32	4.22E-02	7.04	4.18	26057.95	3732.67	2.26E-04	321.01
Substituted copper	-3.29	-18.97	-5.49	-66.17	-1.32E-01	-215.06	-2.15	-57.17	-1.71E-02	-110.7	-2.02E-02	-8.74E-07	-0.20	-0.24	-0.38	-7.81E-03	-2.48	-0.41	-223.34	-18126.45	-3.00E-04	-11.71
Substituted zinc	-1.55	-20.41	-6.24	-4.14	-2.54E-02	-73.02	-4.95	-3.71	-9.74E-03	-19.7	-6.43E-03	-1.13E-06	-0.06	-0.13	-0.14	-1.98E-02	-0.62	-0.62	-251.40	-2418.04	-1.64E-04	-4.07
Substituted sulfuric acid	-1.05	-13.05	-16.54	-1.43	-6.26E-03	-9.76	-2.11	-1.25	-3.19E-03	-2.4	-1.84E-03	-1.22E-06	-0.19	-0.13	-0.88	-1.13E-02	-0.36	-2.23	-596.54	-455.92	-4.55E-05	-4.27
Substituted ceramic tile	-71.03	-1679.41	-671.88	-16.84	-4.19E-01	-329.17	-125.08	-14.52	-2.31E-01	-21.9	-1.19E-01	-1.17E-04	-32.70	-3.80	-4.42	-7.32E-02	-8.84	-6.62	-24833.43	-4864.17	-3.09E-04	-331.03
Substituted heat (natural gas)	-0.01	-9.17	-4.82	-0.01	-1.10E-04	-0.10	-0.08	-0.01	-2.88E-04	0.0	-3.49E-05	-1.18E-06	0.00	-0.01	-0.01	-4.53E-05	0.00	-0.01	-166.83	-3.71	-2.26E-07	-0.92
Avoided tailings landfilling	-0.394	0	0	-234.4	-0.21466	-2629.8	0	-200	-0.00512	0.0	0.004545	0.00E+00	0	0	0	-0.00022	-0.591	0	0	-118457.78	-0.008	-36.612
Net environmental impacts	-13.26	-24.68	***62.26***	-310	-0.55	-3006	-75.69	-264	-0.03	-136.3	-0.03	***5.03E-05***	-1.41	-0.76	-0.83	***0.32***	-5.52	-4.62	***1727.66***	-140275.63	-0.01	-55.78
B-2 Steps																						
MW-roasting and leaching	1.30	42.15	17.81	0.61	9.68E-03	7.60	3.19	0.55	4.77E-02	0.8	1.37E-03	3.62E-06	0.04	0.08	0.13	1.57E-03	0.14	0.78	648.53	171.86	7.95E-06	4.29
Sulfuric acid production	0.18	2.94	0.97	0.09	1.58E-03	1.13	0.55	0.08	1.45E-03	0.1	1.95E-04	1.46E-07	0.07	0.04	0.35	1.12E-04	0.04	0.02	38.88	25.58	1.16E-06	0.65
Ion flotation and precipitation	2.80	65.11	27.72	0.45	1.08E-02	8.09	3.69	0.38	1.75E-02	0.4	1.26E-03	6.91E-06	0.06	0.14	0.19	3.84E-01	0.16	0.28	1053.85	120.33	8.94E-06	6.89
Geopolymer production	8.95	81.98	21.99	3.37	4.10E-02	34.27	7.82	3.02	5.96E-02	5.7	1.35E-02	1.30E-05	0.16	0.26	0.43	8.78E-03	1.13	1.24	887.24	994.29	4.48E-05	8.76
Substituted copper	-3.29	-18.97	-5.49	-66.17	-1.32E-01	-215.06	-2.15	-57.17	-1.71E-02	-110.7	-2.02E-02	-8.74E-07	-0.20	-0.24	-0.38	-7.81E-03	-2.48	-0.41	-223.34	-18126.45	-3.00E-04	-11.71
Substituted zinc	-1.55	-20.41	-6.24	-4.14	-2.54E-02	-73.02	-4.95	-3.71	-9.74E-03	-19.7	-6.43E-03	-1.13E-06	-0.06	-0.13	-0.14	-1.98E-02	-0.62	-0.62	-251.40	-2418.04	-1.64E-04	-4.07
Substituted sulfuric acid	-1.05	-13.05	-16.54	-1.43	-6.26E-03	-9.76	-2.11	-1.25	-3.19E-03	-2.4	-1.84E-03	-1.22E-06	-0.19	-0.13	-0.88	-1.13E-02	-0.36	-2.23	-596.54	-455.92	-4.55E-05	-4.27
Substituted Portland cement	-4.81	-492.95	-41.10	-2.25	-5.03E-02	-46.96	-19.33	-2.04	-4.34E-02	-3.7	-1.03E-02	-1.19E-05	-0.35	-0.88	-0.86	-6.77E-03	-1.10	-0.99	-1754.72	-653.29	-4.88E-05	-30.73
Substituted heat (natural gas)	-0.01	-9.17	-4.82	-0.01	-1.10E-04	-0.10	-0.08	-0.01	-2.88E-04	0.0	-3.49E-05	-1.18E-06	0.00	-0.01	-0.01	-4.53E-05	0.00	-0.01	-166.83	-3.71	-2.26E-07	-0.92
Avoided tailings landfilling	-0.394	0	0	-234.4	-0.21466	-2629.8	0	-200	-0.00512	0.0	0.004545	0.00E+00	0	0	0	-0.00022	-0.591	0	0	-118457.78	-0.008	-36.612
Net environmental impacts	***2.13***	-362.37	-5.70	-304	-0.37	-2924	-13.37	-260	***0.05***	-129.5	-0.02	***7.42E-06***	-0.47	-0.87	-1.17	***0.35***	-3.68	-1.94	-364.33	-138803.14	-0.01	-67.72

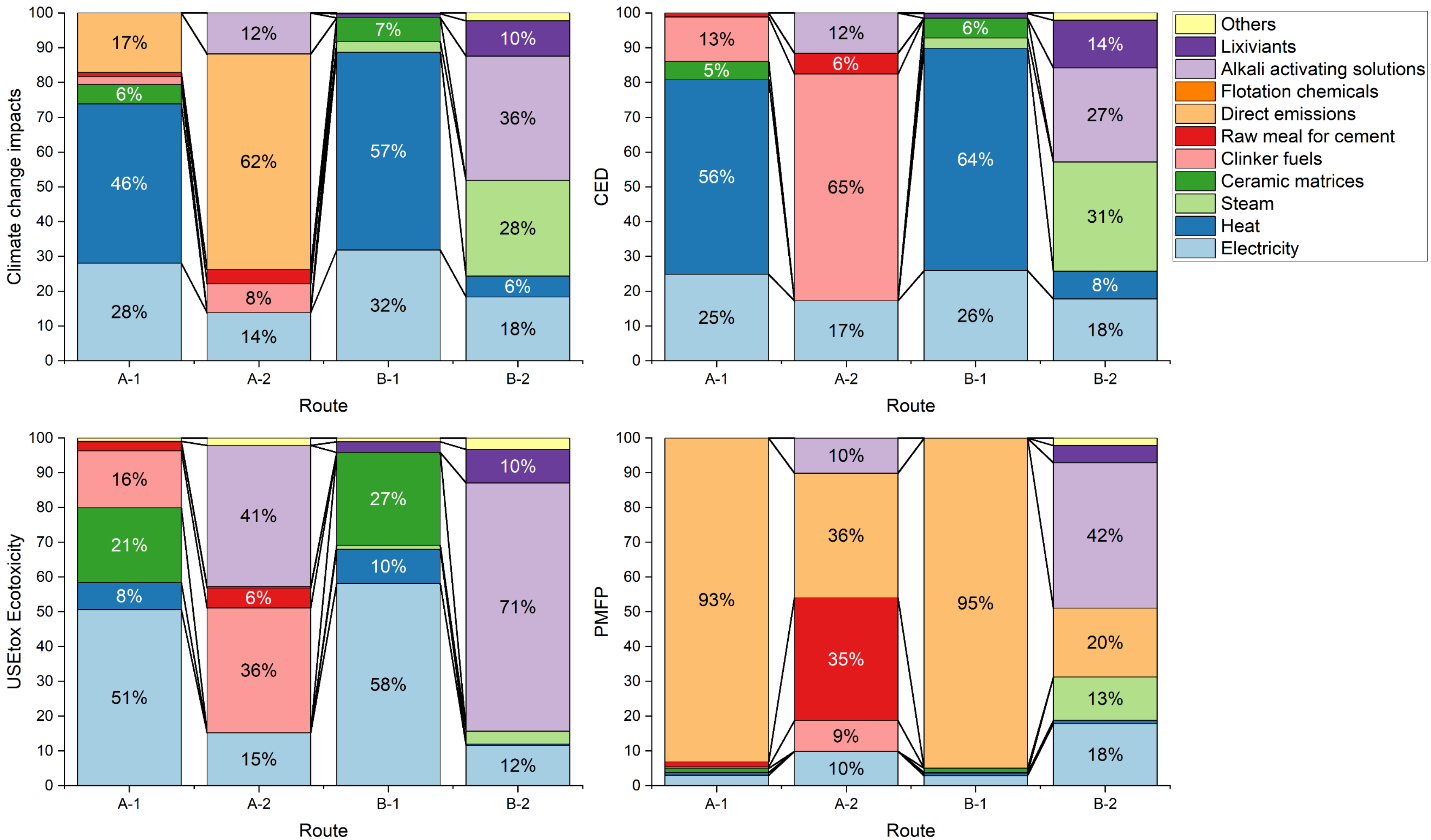
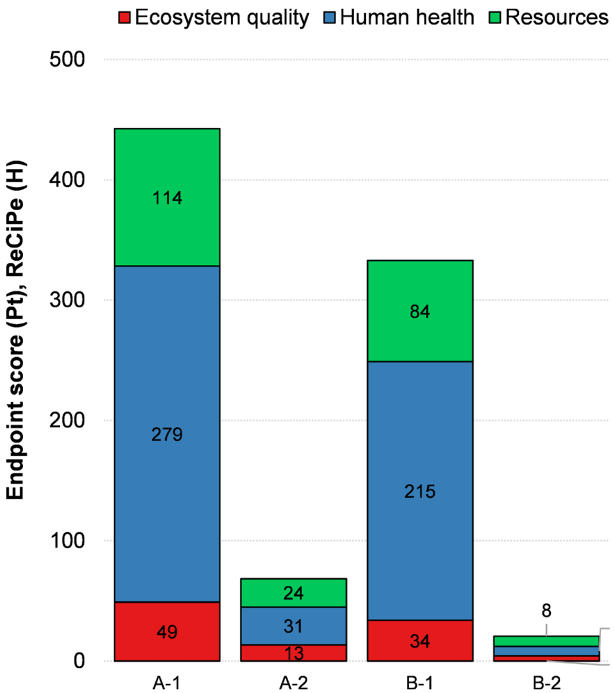


Figure B8. LCA results: contribution analyses by exchanges with technosphere % for all routes and 4 impact categories. Direct emissions include GHG and particulate matters. Data provided in Appendix-3.



Endpoint area of protection	Midpoint categories	Share of the total impact*				Relevance
		A-1	A-2	B-1	B-2	
Ecosystem quality	Agricultural land occupation	0%	1%	1%	2%	**High (> 20%)**
	Climate change, ecosystems	10%	18%	9%	16%	Moderate (1-20%)
	Freshwater ecotoxicity	0%	0%	0%	0%	Low (<1%)
	Freshwater eutrophication	0%	0%	0%	0%	
	Marine ecotoxicity	0%	0%	0%	0%	
	Natural land transformation	0%	0%	0%	1%	
	Terrestrial acidification	0%	0%	0%	0%	
	Terrestrial ecotoxicity	0%	0%	0%	1%	
	Urban land occupation	0%	1%	0%	1%	
Human health	Climate change, human health	16%	**28%**	14%	**26%**	
	Human toxicity	2%	5%	1%	4%	
	Ionizing radiation	0%	0%	0%	0%	
	Ozone depletion	0%	0%	0%	0%	
	Particulate matter formation	**46%**	12%	**49%**	9%	
	Photochemical oxidant formation	0%	0%	0%	1%	
Resources	Fossil depletion	**24%**	**31%**	**24%**	**36%**	
	Metal depletion	2%	4%	1%	4%	

* Total impact is the sum of the normalized impact of all the midpoint impact categories. See absolute values data in Appendix-3.

Figure B9. Share of the total impact for all routes for the ReCiPe impact categories considered in the single score weighted aggregation. Only reprocessing impacts are considered, without avoided burdens.

B.11 Data and input for model sensitivity

1) Low-case and high-case scenarios

Table B28 provides a summary of sensitivity implemented in the model for higher and lower-case values. New technologies such as desulfurization (beneficiation), MW-roasting, ion flotation, geopolymer, among others, are assumed to have larger variability compared to more mature technologies such as ceramic and CSA cement plant. Supplementary results to Fig 4.3 in the thesis are displayed in Figure B10 and Figure B11.

Table B28. Scenarios constructed by varying technical parameters in the system

Parameters	Base case	Lower values case (best)	Higher values case (worse)	References
Electricity use for dewatering and flotation	Default flotation	-25%	+50%	Author's Assumption, supported by scale-up frameworks (Piccinno et al., 2016)
Electricity use for MW-roasting and leaching	Heating efficiency (85%)	Same as base case	Lower heating efficiency (60%) and the specific energy consumption was multiplied by 4	Author's Assumption, supported by scale-up frameworks (Piccinno et al., 2016)
Thermal energy use for drying and reactor heating	Heating efficiency (85%)	Same as base case	+50%	Author's Assumption, supported by scale-up frameworks (Piccinno et al., 2016)
Leaching and flotation efficiencies	Optimized recovery values. These values are already multiplied. Cu: 64%; Zn: 82%	95% overall metal recovery	50% overall metal recovery	Other optimum experiments(Kuo et al., 2005) and common industrial leaching data (Schlesinger et al., 2011) and LCIs for new chemical guidelines(Hischier et al., 2005)
Solvent recycling efficiency	95%	Same with base case	50%	Assumption, based on (Capello et al., 2007) and LCIs for new chemical guidelines (Hischier et al., 2005)

Solvent recycling energy	Directly taken from Ecosolvent for base case	-25%	+50%	Assumption, based on (Capello et al., 2007) and LCIs for new chemical guidelines (Hischier et al., 2005)
Cement production (fuel and electricity)	Default from CSA cement	-10%	+25%	Assumption, based on cement improvement efforts (Boesch & Hellweg, 2010)
Alkali activators preparation	Default from inhouse recipe	-25%	+50%	Author's Assumption
Energy-consumption at ceramic plant	Default from ceramic plant	-10%	+25%	Assumption based on waste heat recovery from (Oliveira et al., 2020)
Particulate matter abatement technologies	Default from ceramic plant	90% reduction	Default from ceramic plant	More stringent PM controls in EU ceramic industry, values from BAT (European Comission, 2007)
Modified values in the process inventories	**Base**	**Low values**	**High values**	**See motivations above**
Electricity use for dewatering and flotation [kWh]	19.50	14.6	29.3	
Electricity use for MW-roasting and leaching [kWh]	28.54	28.54	211.6	
Heat use for MW-roasting and leaching [MJ]	228.79	228.79	343.19	
Cu recovered [kg]	2.92	4.34	2.28	
Zn recovered [kg]	7.48	8.69	4.57	
Solvent recycling efficiency	95%	Same with base case	50%	
Ammonia input [kg]	2.0	2.0	13.9	
Ammonium carbonate input [kg]	5.6	5.6	39.3	
Solvent recycling energy	Base case	-25%	50%	
Electricity [kWh]	12.3	9.3	18.5	
Steam [kg]	186.8	140.1	280.2	
Cement production (fuel and electricity)	Default from CSA cement	-10%	25%	
Hard coal [kg]	167.94	151.1	209.9	
Electricity [kWh]	81.21	73.1	101.5	
Alkali activators preparation	Default from inhouse recipe	-25%	50%	
Route A				
Sodium silicate [kg]	50.07	37.6	75.1	

Sodium hydroxide [kg]	10.06	7.5	15.1	
Lithium chloride [kg]	10.02	7.5	15.0	
Route B				
Sodium silicate [kg]	41.81	31.4	62.7	
Sodium hydroxide [kg]	8.40	6.3	12.6	
Lithium chloride [kg]	8.37	6.3	12.6	
Upgrade strategies at ceramic plant	Base case	-10%	25%	
Route A				
Natural gas as heat [MJ]	23030.9	20727.8	28788.6	
Electricity [kWh]	1371.1	1234.0	1713.9	
PM emitted (kg PM/ t ceramic)	8.71	1.03	8.71	
Route B				
Natural gas as heat [MJ]	19230.8	17307.7	24038.54	
Electricity [kWh]	1079.4	971.4	1349.2	
PM emitted (kg PM/ t ceramic)	8.71	1.03	8.71	

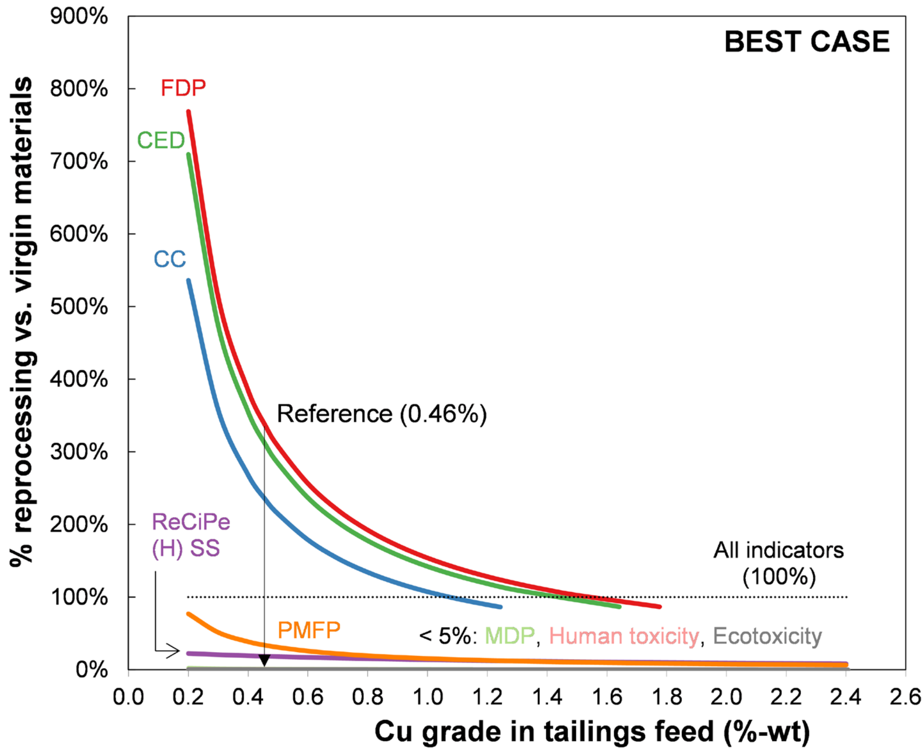


Figure B10. Cu in tailings vs. ratio of reprocessing to metal-only credits (route B without valorization—best case). MDP, human toxicity, and freshwater ecotoxicity are hardly visible due to low values below 5%. FDP = fossil depletion potential, MDP = metal depletion potential, ReCiPe (H) SS = single score using ReCiPe method at endpoint level.

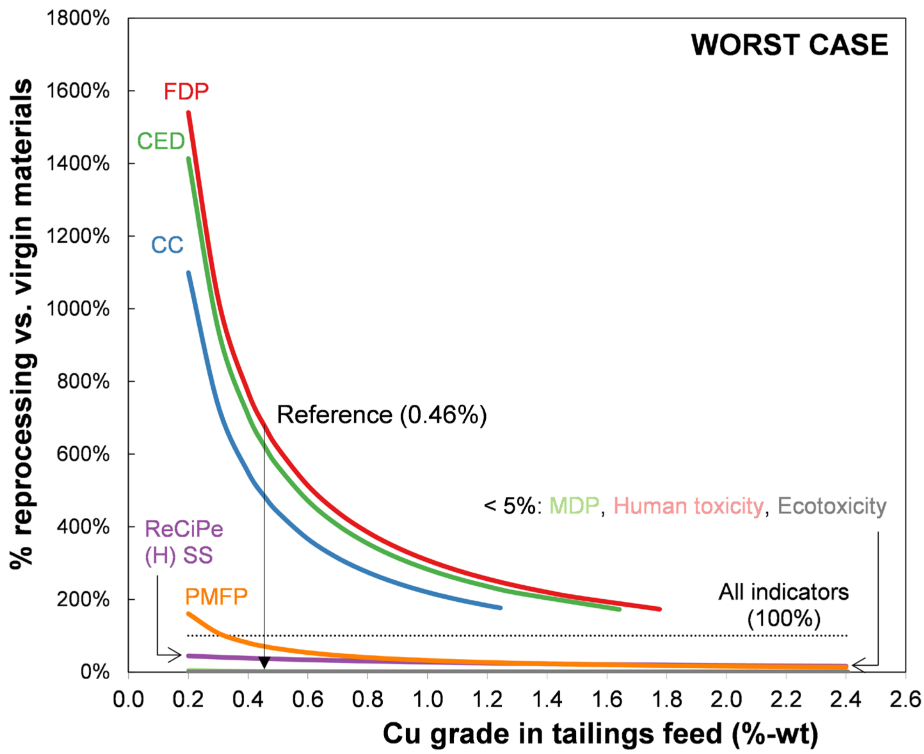


Figure B11. Cu in tailings vs. ratio of reprocessing to metal-only credits (route B without valorization—worst case). MDP, human toxicity, and freshwater ecotoxicity are hardly visible due to low values below 5%. FDP = fossil depletion potential, MDP = metal depletion potential, ReCiPe (H) SS = single score using ReCiPe method at endpoint level.

2) Transport distance

Distance 1, that is around 50 km on the map, connects the location of mine tailings site to the city of Loulé. Distance 2, meanwhile, is the average distance from the same origin to several cities up north (~300 km, from the shortest Setubal to Coimbra). Results are presented in Figure B13.

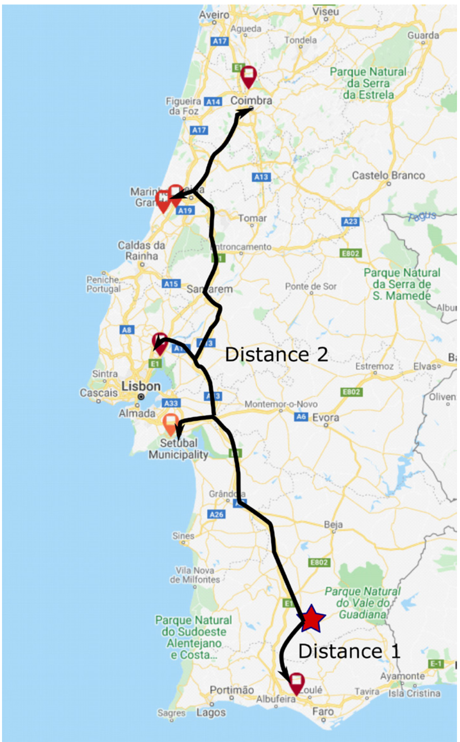


Figure B12. Transport distances to assess the impacts of transport from source to reprocessing sites. The red star indicates the waste production site, while the other pin symbols indicate the location of cement/ ceramic industries in Portugal. Map data from Google© (2021) and cement plants data from Cemnet(Cemnet, 2020).

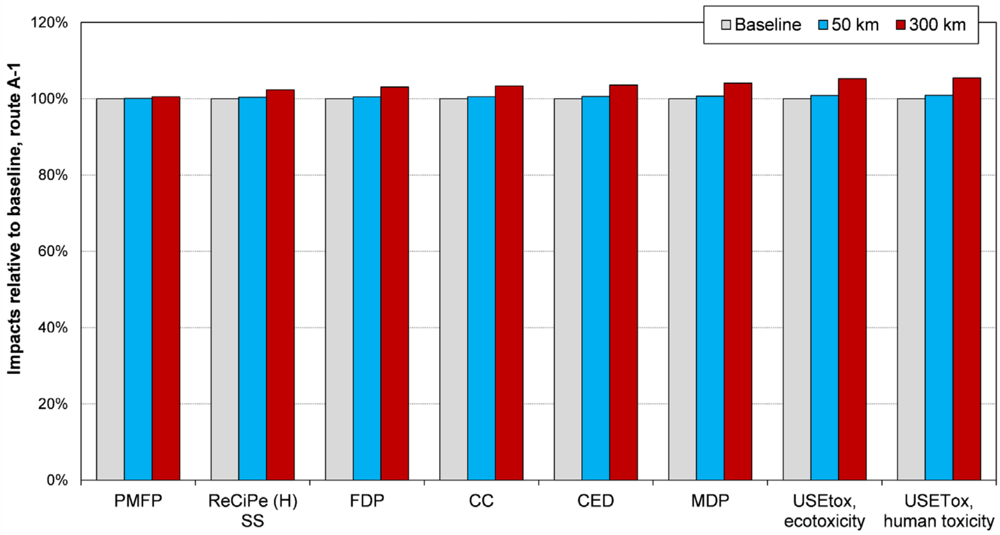


Figure B13. Effect of transport to point of reprocessing vs. base case. Grey bars are set to 100%, representing the base case reprocessing burdens of route A-1 without credits

3) Substitution ratio (full and value-corrected)

Table B29. The substitution ratios used in the LCA models are reported. The numbers behind colons indicate the amount of equivalent primary product that can be substituted for 1 unit of produced secondary materials.

Secondary products	Displaced primary product	Base case	Plausible case	Reference
Secondary copper	Cathodes of primary copper	1:1	1:0.95	(Koffler & Florin, 2013)
Secondary zinc	Primary zinc	1:1	1:0.95	(Koffler & Florin, 2013)
Secondary CSA cement	Primary CSA cement	1:1	1:0.85	(Rigamonti et al., 2020)
Secondary ceramic tile	Primary ceramic tile	1:1	1:0.85	(Rigamonti et al., 2020)
Geopolymer binder	Primary ordinary Portland cement	1:1	1:0.75	(Hassan et al., 2019)
Sulfuric acid	Primary sulfuric acid from elemental sulfur	1:1	1:1	Assumed

4) Future electricity mixes in Portugal

We used the data from Ecoinvent 3.7.1 for baseline year in all electricity input flows, which is equivalent to the data for 2015 electricity data from IEA 2017 energy statistics for Portugal as originally described in the Ecoinvent dataset (Ecoinvent, 2021). To model the electricity production in year 2030, we collected the energy mixes data from EU national energy and climate plan 2030(European Commission, 2019). A more extended version can be found in the country-specific modeling approach as explained in the national long-term strategies of Portugal power sector(Environment Portugal, 2019). It puts emphasis on the increased capacity of renewable energy share (up to 80%), with top three sources are coming from hydropower (22%), wind (31%), and solar (27%). With this information, we then adapted the background data used for LCA modeling in the sensitivity analysis (also see Appendix-3 “EL mixes” tab for more information). The compiled data is shown in Table B30.

Table B30. Portugal electricity mix in year 2015 and 2030 according to the national energy and climate plan (NECP 2030)(European Commission, 2019)

Energy source	Proportion in the 2015 electricity mix*	Proportion in the 2030 electricity mix
Cogeneration	10%	9%
Waves	0%	0%
Geothermal	0%	1%

Biomass	1%	4%
Solar	2%	27%
Wind	22%	31%
Hydro	14%	22%
Oil	0%	0%
Natural gas	24%	7%
Coal	26%	0%

*Electricity mix data taken from Ecoinvent 3.7.1 as is

Next to changing electricity to future mix in 2030, we also test the environmental performances when assuming 100% solar PV consumption on site. This assumption is based on a scenario for independent electricity supply without relying from national grid, exemplifying independent off-grid electricity production near the area. Although it does not represent reality nor sound implementation, this hypothetical scenario gives speculation of how much gains or loss can be obtained from maximum renewables capacity, i.e., solar PV as shown in Figure B14. Many impact categories are decreased when electricity from solar PV is used. Three impacts associated with metal production in the upstream are nevertheless worsened, namely MDP, USETox ecotoxicity, and human toxicity. Overall, owing to the endpoint methods with distinct weighting for all impact assessment categories in ReCiPe (H) V1.13, the hypothetical PV scenario can achieve slightly better environmental profiles than 2030 energy plan. Indeed, one should consider this as theoretical case as the precise results may depend on the country's actual mix, which may well be situated somewhere between these explorative future projections. The effects on other reprocessing routes (A2, B1, and B2) would also show similar outcomes as motivated in the Figure 3.5 of the thesis.

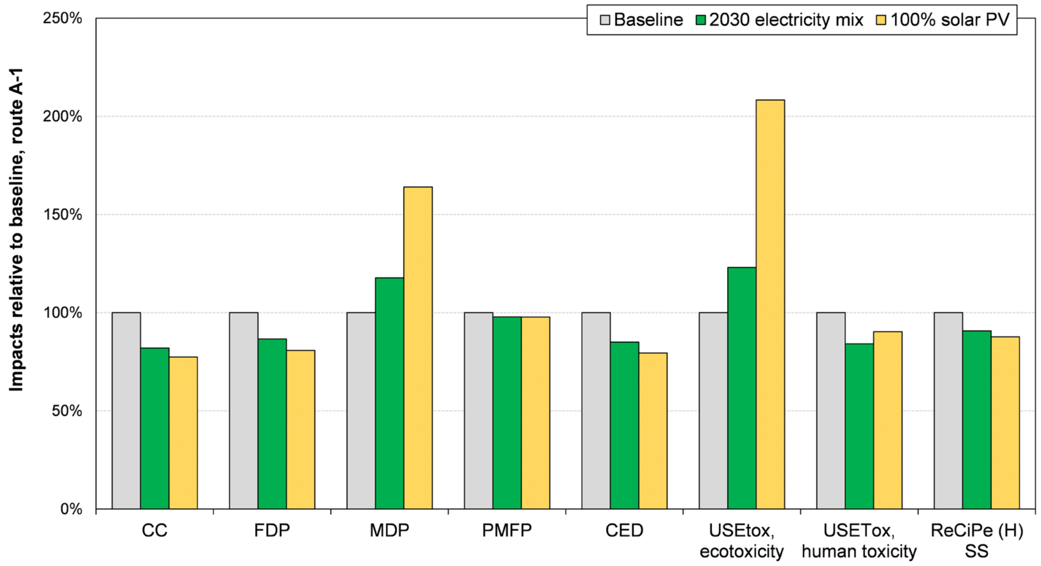


Figure B14. Effect of electricity mix vs. base case. Grey bars are set to 100%, representing the base case reprocessing burdens of route A-1 without credits

B.11 Instructions to run or modify the LCI and LCA models

We use the Activity Browser (Steubing et al., 2020) for the LCA modeling, which is a graphical user interface of the previously developed open-source Brightway2 (Mutel, 2017) in Python. The following steps explain how users can reuse, modify the datasets, and reproduce the results generated in this study.

1. Download the Activity Browser from the official Github page https://github.com/LCA-ActivityBrowser/activity-browser
2. Update through command prompt if necessary and activate the ab_dev environment
3. Download the SI spreadsheet from ACS website, containing the life cycle inventory data of this study. For this, refer to sheet 2 – 8 in Appendix-3 online file
4. Import the Excel sheets into your system by clicking import database on Activity Browser "File" menu on top left corner
5. Relink the standard Ecoinvent database as your used background database. Prior to that, make sure that you have the official license to use Ecoinvent 3.7.1 database https://www.ecoinvent.org/database/ecoinvent-371/ecoinvent-371.html
6. Select relevant impact categories from the second tab, should you wish to run LCA models for other LCIA methods
7. Define process parameters with new results or user-defined statistical distributions, simply by entering new values in the "parameters" tab on the right side
8. Once previous steps are done, users can either perform standard LCA, scenario analysis, or uncertainty simulations on their own system. The results of these computations are also provided in the Appendix-3 online file.

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Appendix C

Toward sustainable reprocessing and valorization of sulfidic copper tailings: scenarios and prospective LCA

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Supplementary information for the following paper:

Toward sustainable reprocessing and valorization of sulfidic copper tailings: scenarios and prospective LCA in ***Science of the Total Environment*** **(2023)**. Link.

C. Supplementary information for Chapter 4

C.1 Methods overview and additional description

The following section describes the detailed methods and calculation frameworks used in this study. Figure C1 depicts the framework used in this study with the steps.

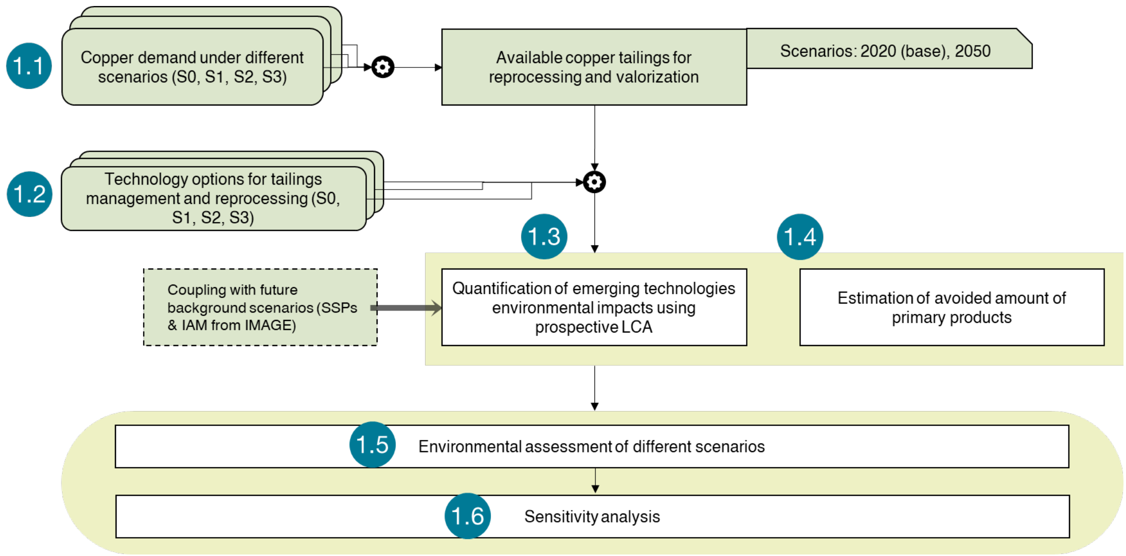

Figure C1. Overview of the framework with steps, adapted from Figure 4.1.

Details regarding resources, assumptions, and rationale taken for each step are explained as follows in the subsection.

C.1.1 Copper demand and scenario narratives

Copper demand scenarios were generally retrieved from the past studies of Ciacci et al. (2017, 2020), with some updates for the copper transition demand from the study of Gregoir and van Acker (2022). They both simulated mass flow data reflecting the historical copper flows, including inflow/ outflow trades of the European copper sector. The demand projection is inherently added on top, assuming that future forecasts follow UNEP Geo outlook scenarios (UNEP, 2019). We specifically picked a sustainability-focused scenario from the original studies: TE (Towards Equitability). This scenario represents a storyline that fits with the objectives of the present study to align with the 1.5-degree climate goals. For more details about other future exploratory scenarios, readers can access other studies in which they applied similar projection methods for the metals (Ciacci et al., 2020; Elshkaki et al., 2018; Kuipers et al., 2018; UNEP, 2019; Van der Voet et al., 2019).

Table C1. Domestic copper demand, and primary + secondary supply at current recycling in 2020.

Year 2020 - Flow in kt copper	BAU Scenario 0
Cu demand	3833
Domestic extraction	750
Mining loss (to tailings)	83
Cu old scrap generated	2940
Secondary Cu (direct melting)	118
Secondary Cu (cathodes)	1646
Import to cover total demand	1319
% From recycling	46%

Table C2. Projected domestic copper demand without transition demand and primary + secondary supply in 2050.

Year 2050 - Flow in kt copper	Scenario 1	Scenario 2	Scenario 3
Cu demand	3146	3146	3146
Domestic extraction	750	750	750
Mining loss	83	83	83
Cu old scrap generated	2858	2858	2858
Secondary Cu (direct melting)	114	114	114
Secondary Cu (cathodes)	1600	1600	1600
Import to cover total demand	682	682	682
% From recycling	54%	54%	54%

Table C3. Projected domestic copper demand with transition and primary + secondary supply, with standard recycling rates of old Cu scrap in 2050.

Year 2050 - Flow in kt copper	Scenario 1	Scenario 2	Scenario 3
Cu demand (standard + transition) [a]	4646	4646	4646
Domestic extraction	750	750	750
Mining loss	83	83	83
Cu old scrap generated	4221	4221	4221
Secondary Cu (direct melting)	168	168	168
Secondary Cu (cathodes)	2363	2363	2363
Import to cover total demand	1365	1365	1265
% From recycling	54%	54%	54%
Additions from tailings reprocessing	-	-	100
% From recycling (inc. tailings)	-	-	57%

Note : [a] = total demand for copper is estimated by summing up copper's projected standard demand in other applications (construction, electronics, industrial machinery, transportation, and consumer goods) with the copper's transition demand (Ciacci et al., 2020). Future scenarios require an additional 1500 kT copper for systems transition, respectively (Gregoir & Van Acker, 2022), which are based on TE = towards equitability scenario.

C.1.2 Treatment options for mine tailings

There are several options to manage copper tailings from the copper ore beneficiation/ metallurgical processing stage. Traditionally, copper tailings are sent to the disposal facility and contained for a long period (Reid et al., 2007, 2009). Alternative approaches are emerging these days, leveraging novel metallurgical techniques and advances to minimize environmental liability while maximizing resource value. In our study, we extended the results of the EU mine waste valorization project (Machiels et al., 2021) by applying novel resource recovery techniques as a simulation for large-scale, alternative copper tailings management. Past studies have successfully screened relevant technologies and reprocessing/ valorization routes for managing copper tailings (Adrianto & Pfister, 2022; Niu et al., 2021; Veiga Simão et al., 2022). It is important to note that we apply prospective LCA through scaling of respective technologies as described in detail in the study of Adrianto and Pfister (2022). No technology learning effects were considered in this assessment. We summarize here the key technology descriptions for each scenario (Table C4).

Table C4. Overview of treatment options for copper tailings, adapted from Adrianto and Pfister (2022).

Process steps	Treatments in scenario 2	Treatments in scenario 3
Beneficiation	Flocculation-flotation	-
Extraction	-	MW-roasting and leaching
Recovery	-	Ion flotation and precipitation
Residue valorization	Sulfur rich fraction: CSA cement production Aluminosilicate fraction: Ceramic production	Aluminosilicate fraction: Geopolymer production
Displaced metals	-	Primary metals (copper and zinc)
Displaced building materials	▪ Primary CSA cement ▪ Primary ceramic roof tile	Primary Portland cement
Other by-products	-	Primary sulfuric acid, heat

- Beneficiation, scenario 2

In the beneficiation stage, sulfur rich fraction (pyrite) is separated from the tailings feedstock using flocculation techniques (Broadhurst et al., 2015; Norgate & Haque, 2010). Reagents such as polyacrylamide and xanthate are injected to help improve pyrite separation. At the outlet, pyrite residues fraction and gangue materials rich in aluminosilicate fractions are generated for further processing.

- Extraction and recovery, scenario 3

A combined microwave-roasting and chemical leaching developed in the project is employed to extract copper and zinc from tailings (Kamariah et al., 2022; Xanthopoulos et al., 2021). The feedstock must first be dried and then roasted at a temperature of up to 700C. Afterwards, leaching processes take place with the aid of lixiviants, a mixture of ammonia and ammonium carbonate—each assumed to reach 95% yield rates. Finally, copper metals are recovered using ion flotation techniques while zinc is precipitated. All solvents in the system are assumed to be recuperated at 95% recovery rates (Amelio et al., 2014; Xanthopoulos & Binnemans, 2021).

- Residue valorization, scenarios 2 and 3
    - Scenario 2

    Calcium sulfoaluminate (CSA) production is chosen as the valorization technique to treat pyrite rich fraction from the beneficiation stage (Martins et al., 2021). In general, the manufacturing step of CSA cement mimics what ordinary Portland cement has: the raw material acquisition, calcination, and gypsum additions. The main changes are related to input materials (bauxite consumption) and process emissions due to modifications in the cement formulation (Ren et al., 2017).

    As for the aluminosilicate fraction, previous studies have concluded that such residues can be used as raw materials for ceramic tile manufacturing (Veiga Simão et al., 2021). This provides a basis that 10%-wt of the ceramic tiles can be substituted with the cleaned fraction from the beneficiation stage.

    - Scenario 3

    Contrary to scenario 2, the aluminosilicate tailings fraction can also be used as raw materials for alternative cement, that is, inorganic polymer or geopolymer (Niu et al., 2020, 2021). Drawing on these findings, we distinguished the valorization step in scenario 3 by hypothetically producing geopolymer that can substitute ordinary Portland cement. The processing steps include chemical activation via alkali-activator agents, namely sodium silicate and sodium hydroxide, to promote polymer-like structures.

- Byproduct utilization, scenario 3

Sulfuric acid plants are installed next to the pyrite roasting plants to eliminate sulfur dioxide off-gas direct releases to the environment (Runkel & Sturm, 2009). Wet cleaning steps and catalytic reactions are employed to convert SO_2 off gas into SO_3. Follow-up hydration, exothermic reactions take place in the absorption columns, which generate sulfuric acid in gaseous form for later condensation as liquid, according to the best available technology handbook (European Commission, 2007).

Overall, all these reprocessing routes constitute the basis of life cycle inventory and analysis for the prospective assessment in step 3. The life cycle inventory modelling is described in detail in the previous study (Adrianto & Pfister, 2022).

C.1.3 Prospective assessment

We constructed prospective life cycle assessment models by considering different treatment options for tailings, metal scenarios, and background system changes. This best practice of prospective LCA has been recommended elsewhere (Arvidsson et al., 2018; Tsoy et al., 2020; van der Giesen et al., 2020) and is still a subject of evolving improvement. In our study, for the background system changes we relied on the general narratives defined by Shared Socioeconomic Pathways or SSPs (Riahi et al., 2017). This set of scenarios stems from large scholarly efforts, which attempt to create robust and credible future storylines for integrated forward-looking analysis. To facilitate systematic integration of future background datasets in the simulation of each scenario developed, we used the framework ‘premise’ (Sacchi et al., 2022), which is a specifically built tool for running futurized LCA models. Future background datasets allowed us to assess the environmental performance at defined time horizons in this study (i.e., 2020 and 2050). 2020 is chosen as the base year, while 2050 is assumed to be the year when technology scale up and changes are expected to materialize. In addition, 2050 is typically the defining target year for many decarbonization visions and climate change mitigation in many industries, including copper (Chan et al., 2019; IEA, 2020, 2021; Wesseling et al., 2017; World Economic Forum, 2015).

C.1.4 Secondary production from reprocessing

For each secondary product generated in each scenario, we calculated the realistic volume by 1) comparing the production output to the primary demand or 2) adjusting the volume in view of the availability of raw materials/ other critical ingredients in the supply chains. Accordingly, it is assumed that the secondary production of all materials must be less than the primary supply such that all secondary materials can be consumed in the region without market disruptions. The primary demand for materials in 2050 is derived from a combination of the consumption growth (projection of past data with the same trends), market share of new materials, experts’ predictions (as part of research studies/ analyses), and industry roadmaps (noted exhaustively in Table 4.2 of the manuscript). The calculation steps are summarized in Table C5.

Table C5. Methods and assumptions to estimate product supply in 2050.

Product	Approach to estimate primary supply in 2050	Product supply in 2050 [million tonne]
Ceramic tile	According to the market outlook report, future production follows a compound annual growth rate of 4.1% from 2020. (Inkwood Research, 2022). Total 2020 production data in the EU was obtained from the Ceramic World report (2021), by converting total production in surface areas to mass. Total surface area for ceramic tiles is 1035 million m^2. Area to mass conversion for ceramic $(1m^2) =$ 22.7 kg (Ros-Dosdá et al., 2018).	72 (+50)
CSA cement	According to the prediction of industry roadmaps (ECRA and CSI, 2017; IEA, 2018), cement production in the EU would remain stagnant. Thus, using a market penetration rate of 15% from alternative cement market diffusion (Favier et al., 2018; Habert et al., 2020), we derived the future supply of CSA cement. $Supply\ in\ 2050 = 15\% \times OPC\ supply\ in\ 2050$	25 (+25)
Geopolymer	It is assumed that geopolymer will substitute ordinary Portland cement in the future. Based on the report published by the EU cement association (Cembureau, 2022), the region contributes 4% to the total world's share. In 2020, totally there was 4170 million tonnes of OPC manufactured. The future supply in 2050, assuming that the cement market will stabilize in Europe, is calculated by multiplying EU's share with the global cement production.	167 (-)
Copper	Copper supply in 2050 is derived from other studies in the context of European production as secondary adapted data (Gregoir & Van Acker, 2022).	4.6 (+1)
Zinc	Zinc supply in 2050 is derived from other studies in the context of European production as secondary adapted data (Gregoir & Van Acker, 2022).	2.9 (+0.1)

Sulfuric acid	Linear forecast based on historical and projected data from the year 2015 – 2029. (ChemIntel360, 2022). The production data is presented in Figure C2. From the linear regression modelling, we used the following correlation to estimate future production: $y = 4.37x - 8562.1$ x = production year Europe's market share is assumed to be stable at 6.4% of global production (y). (King et al., 2013).	25 (+9)

Note: For product supply in 2020, no forecast is needed since the market supply is already recorded in the baseline year (see Table C8). Numbers in brackets represent the volume increases from the 2020 levels.

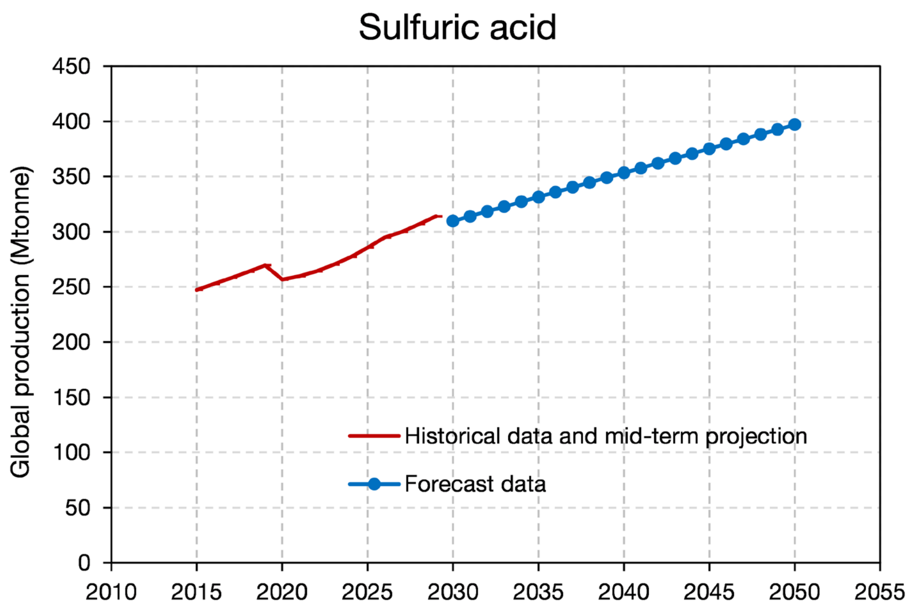

Figure C2. Global sulfuric acid production and future estimates. Data built on (ChemIntel360, 2022).

C.1.5 Environmental assessment

We relied on the life cycle assessment (LCA) to evaluate the environmental impacts of every scenario in this study. The LCA is based on the standardized ISO 14040 method (ISO, 2006), which was then complemented with the prospective or ex-ante LCA approaches mentioned in section 4.1.3. All emissions caused by implementing scenarios were counted as positive (+) values, while all avoided emissions or substitution credits were defined as negative (-) values. For the latter elements, we assumed that in the base case (full substitution rates applied) an equivalent amount of primary production was substituted by the secondary products generated in the associated scenario, i.e., x amount of CSA cement in scenario 2 displaces x amount of primary CSA cement (see section 5.2).

The rest of products follows the same approach, and the substituted products are listed in Table C7.

C.1.6 Recycling potential

There are many ways to measure circular economy in socio-economic and ecological cycling (Mayer et al., 2019). We defined a mass-based indicator by calculating the share of secondary materials in the total supply. To illustrate this method, we take the example of copper. In 2050, 0.1 million tonne of copper can be recovered from tailings if scenario 3 were to be applied. Meanwhile, we estimated that Europe would need approximately 4.6 million tonnes of copper to support its economy. Thus,

$$Recycling\ rates\ for\ copper\ from\ tailings\ 2050\ (\%) = \frac{0.1\ mT\ copper}{4.6\ mT\ copper} = 2\%$$

C.2 Description of impact assessment categories

The selected life cycle impact assessment (LCIA) methods are ReCiPe 2016 (Huijbregts et al., 2017), plus additional impact assessment indicators such as CED for fossil energy consumption, abiotic resource depletion for resource availability, and USEtox for toxicity metrics. All used LCIAs are listed in Table C6.

Table C6. The list of LCIA methods in this study.

Environmental impact indicator	Unit	Description
IPCC 2013, climate change impact	CO_2-eq	IPCC 2013 method is used to characterize different gaseous emissions according to their global warming potential and the aggregation of different emissions in the impact category climate change (IPCC, 2014).
USETox, freshwater ecotoxicity	CTUe	USEtox model is an environmental model for characterisation of human and ecotoxicological impacts in LCIA. It has been developed by a team of researchers from the Task Force on Toxic Impacts under the UNEP-SETAC Life Cycle Initiative. USEtox is designed to describe the fate, exposure and effects of chemicals (Rosenbaum et al., 2008).

<table>
<tr><td>Cumulative energy demand (CED), fossil</td><td>MJ-eq</td><td>Cumulative Energy Demand analysis aims to investigate the energy use throughout the life cycle of a good or a service. This includes the direct uses as well as the indirect or grey consumption of energy due to the use of, e.g. construction materials or raw materials (Frischknecht et al., 2015). In this study, we specifically selected non-renewable sources, fossil in the CED subcategory.</td></tr>
<tr><td>Abiotic depletion potential</td><td>Sb-eq.</td><td>Abiotic depletion refers to the depletion of nonliving (abiotic) resources such as fossil fuels, minerals, clay, and peat. Abiotic depletion is measured in kilograms of Antimony (Sb) equivalents (van Oers & Guinée, 2016).</td></tr>
<tr><td>ReCiPe 2016, damage to ecosystems quality impacts</td><td>Species.year</td><td rowspan="4">ReCiPe 2016 provides a harmonised implementation of cause-effect pathways for the calculation of both midpoint and endpoint characterisation factors. Ecosystems quality human health, and resource scarcity/availability are selected in the LCIA as the three areas of protection (Huijbregts et al., 2017).
The unit for ecosystem quality is the local species loss integrated over time (species year).
DALYs (disability adjusted life years), relevant for human health, represent the years that are lost or that a person is disabled due to a disease or accident.
The unit for resource scarcity is the dollar (USD), which represents the extra costs involved for future mineral and fossil resourceextraction.</td></tr>
<tr><td>ReCiPe 2016, damage to human health impacts</td><td>DALY</td></tr>
<tr><td>ReCiPe 2016, damage to resource availability impacts</td><td>USD</td></tr>
<tr><td>ReCiPe 2016, Endpoint (H), total aggregated single score</td><td>Points</td></tr>
</table>

C.3 Secondary material production

For the error bars in Figure 4.4 of the manuscript, we calculate the net impacts of each scenario by varying the possible secondary material production as a function of market penetration/ availability (a modified calculation for Table 4.2 of the manuscript). The penetration rates are adapted only for building materials. The values are listed in

Table C7. Variation in market penetration rates for secondary material (2050 cases). Volume unit in million tonnes.

Scenario	Secondary material	Maximum possible secondary production	Realistic secondary production (base case)	Best case – higher penetration rate for secondary materials	Worst case – lower penetration rate for secondary materials
2	Ceramic tile	539	61	61	27
	CSA cement	127	19	25	6
3	Geopolymer	64	6	32	3
	Copper	0.1	0.1	0.1	0.1
	Zinc	0.08	0.08	0.08	0.08
	Sulfuric acid	12	12	12	6

Note: Best cases assume penetration rates are set to maximum or capped at total primary supply for respective materials. Worst cases assume 5% and 50% penetration rates for building materials and sulfuric acid, respectively, equivalent to half of the realistic values (Habert et al., 2020).

Table C8. Secondary production potential vs. primary material demand in EU. Volume unit in million tonnes for the year 2020.

Scenario	Secondary Material	Maximum possible secondary supply, million tonne	Product penetration/ availability	Adjusted secondary production, million tonne	Primary material substituted	Primary demand in 2020, million tonne	Data and calculation source
2	Ceramic tile	372	Assumed 3%	11	Ceramic tile	21	(Cerame-Unie, 2021); (Ceramic World Web, 2021)
	CSA cement	88	Penetration 0.15%[i]	0.1	CSA cement	0.2	(Favier et al., 2018; Habert et al., 2020)
3	Geopolymer	44	Penetration 0.05%[i]	0.02	OPC cement	167	(Cembureau, 2022; Favier et al., 2018; Habert et al., 2020)
	Copper	0.09	Could be 100%	0.09	Primary copper	3.6	(Ciacci et al., 2020; Gregoir & Van Acker, 2022)
	Zinc	0.08	Could be 100%	0.08	Primary zinc	2.8	(Gregoir & Van Acker, 2022)
	Sulfuric acid	8	Could be 100%	8	Sulfuric acid	16	(ChemIntel360, 2022; King et al., 2013)

Note: the same calculation procedure and results as in Table 4.2 but with the year 2020.

C.4 Additional LCA results

Environmental performances based on (1) two additional midpoint impact categories and (2) three endpoint impact categories of ReCiPe 2016 are presented below.

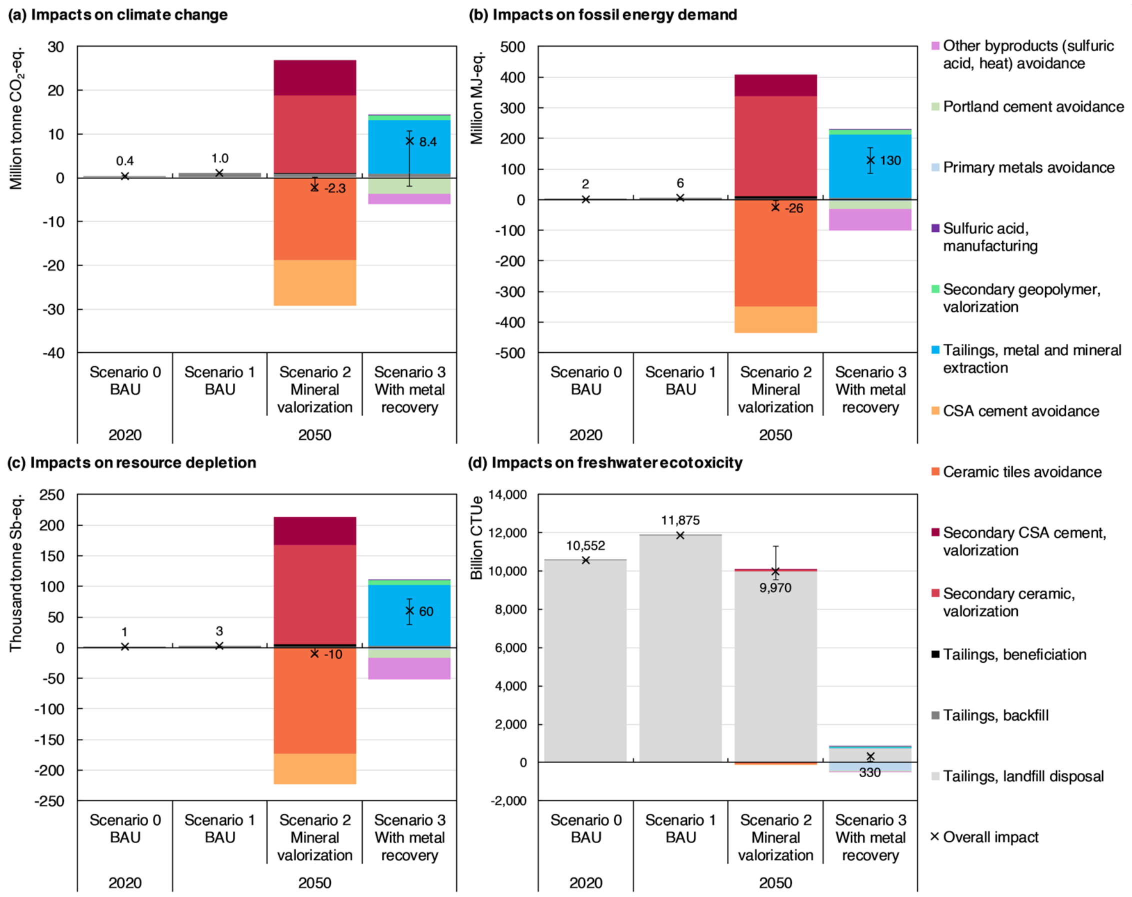

Figure C3. Comparison of the different scenarios for copper tailings management on a system-wide analysis: a) climate change impacts, b) cumulative energy demand, c) abiotic resource depletion, and d) USEtox freshwater ecotoxicity.

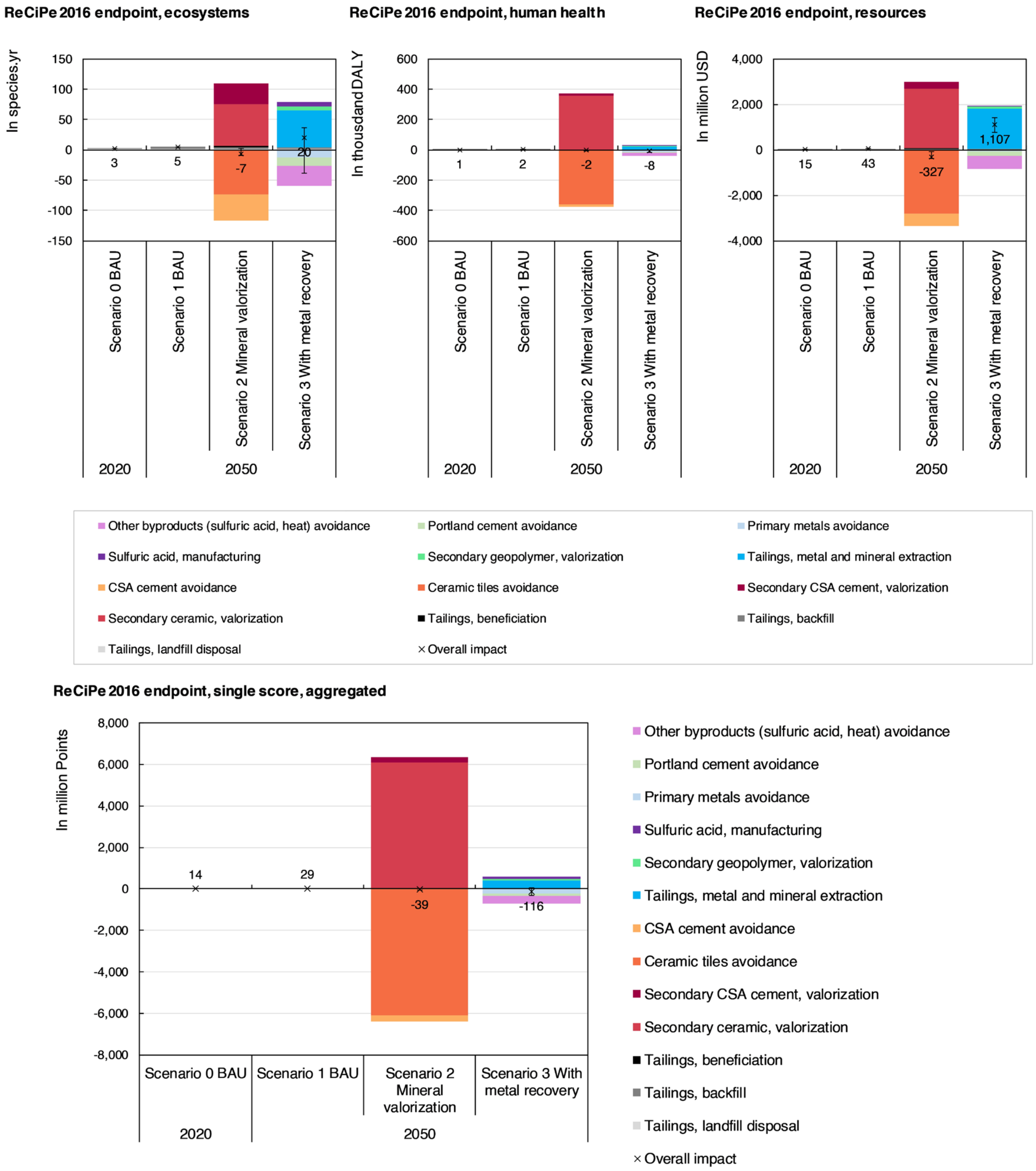

Figure C4. Comparison of the different scenarios for copper tailings management on a system-wide analysis: ReCiPe 2016 damage to ecosystems, human health, and resource availability impacts (top) and aggregated, single score (bottom).

C.5 Sensitivity analysis

We performed sensitivity analyses to the following parameters in the models:

C.5.1 Selection of marginal production technologies

In this sensitivity test, we identify different market suppliers for sourcing the primary products. A summary of marginal technologies in the sensitivity analyses is presented in Table C9.

Table C9. Choice of marginal production technologies in the sensitivity analyses.

Material	Base case	Alternative marginal 1	Alternative marginal 2	Alternative marginal 3
Ceramic tiles	Europe 100% (electricity and natural gas)	Europe 80%, China 20% (electricity, natural gas, and coal)	China 40%, India 60% (natural gas and coal)	China 100% (coal only)
CSA cement	Europe 100%	China 50%, Middle East 50%	Middle East 100%	China 100%
OPC cement	Production in Europe, with MEA-based CCS	Production mix in India and China, with MEA-based CCS	Production in India without CCS	-
Metals (only copper)	Production in Europe	Production mix in LatAm (Chile and Peru) as dominant suppliers	Production mix global via hydrometallurgical technologies	-

Note: In the default case, we assume all European-centric production, implying that displacement of respective materials only occurs domestically, so emission mitigation effects can only be claimed within the continent. This is performed to separate environmental impacts generated in the domestic region from those abroad. For demonstration, see Section C.9 ‘Contribution of tailings management to GHG reduction targets’ of the Supplementary Materials.

The assumptions and description for each material are explained as follows:

- Ceramic

China could become the next major exporters of ceramic products when the local domestic manufacturers cease production (European Commission, 2022). A mix of ceramic production in Europe and China is defined as the marginal choice in the first test. In the second test, we assume that China would become the single ceramic tiles supplier to fulfill European ceramic demand (100% ceramic from China, with energy supplied by coal). However, a growth decline in China would mean that other countries would need to ramp up production to make up for the world's demand (Sangwan et al., 2017; Wang et al., 2020). In the third test, we assume that 40% and 60% of a ceramic marginal mix are manufactured in China and India, respectively. Energy and fuel efficiency gains are expected in the future production according to European Best Available Technology Document for ceramic (European Comission, 2007), which are assumed to be 20% reductions by 2050 (Alig et al., 2021; Ros-Dosdá et al., 2018).

- CSA cement

Instead of sourcing most CSA cement from China in the base case, Middle East, due to its growing economic development and construction activity, may become a prominent player in low-impact cement production. Moreover, sulfur — one of the key ingredients for the production of CSA cement as an alternative to traditional sulfates – can be closely sourced near the oil and gas industry in the region (Al Horr et al., 2017). Thus, the Middle East would emerge as one of the marginal suppliers in the future if this market trend and technological innovation continue (Gálvez-Martos et al., 2021).

- Ordinary Portland cement

Cement consumption in China is expected to peak between 2015 and 2030, as per capita cement consumption declines towards more developed nation levels. After 2030, global cement production will be enabled by strong production growth in India and other developing Asian countries, Africa, and the Middle East. We assumed that alternative marginal suppliers are cement produced solely in India, the second largest producer in 2050, without CCS technologies installed. Additionally, cement can also be produced domestically in Europe, as observed by the stagnant yet steady growth in the continent. Internal cement players in Europe can also take the role as the marginal supplier in the market (IEA, 2018). Meanwhile, feasible technology improvements are taken from the decarbonization of cement industry reviews (Habert et al., 2020; Watari et al., 2022).

- Metals, namely copper and zinc

In the alternative marginal scenarios, we assumed Chile would supply most of the copper worldwide, as production in China and other regions is declining. Production of copper via the hydrometallurgical route appears as the third marginal scenario, assuming that the share of copper metals from oxide deposits are increasing in the future (Rötzer &

Schmidt, 2020). On the contrary, no modifications to the marginal data for zinc are exercised due to the dominant position of China as the marginal supplier.

C.5.2 Substitution factor of construction materials (minimum, average, maximum)

The allocation problem in LCA was avoided using the system expansion methodology. The adopted procedure from Vadenbo et al. (2017) is applied, quantifying the substitution potential of market products with adapted values of secondary resources. This enables a better representation of substitution in the simulation than the standard approach, as it avoids the standard 1:1 displacement.

Table C10. Substitution ratios varied in the sensitivity analyses.

Substitution ratio	Range of value
Ceramic	0.5 – 1.0
CSA cement	
OPC cement	
Metals (copper, zinc)	

Varying substitution factors for all the materials result in the following Figure C5. Net GHG emissions for each future scenario are presented, with horizontal axes indicating the secondary products substitution rates while the vertical axis indicates the net environmental impacts.

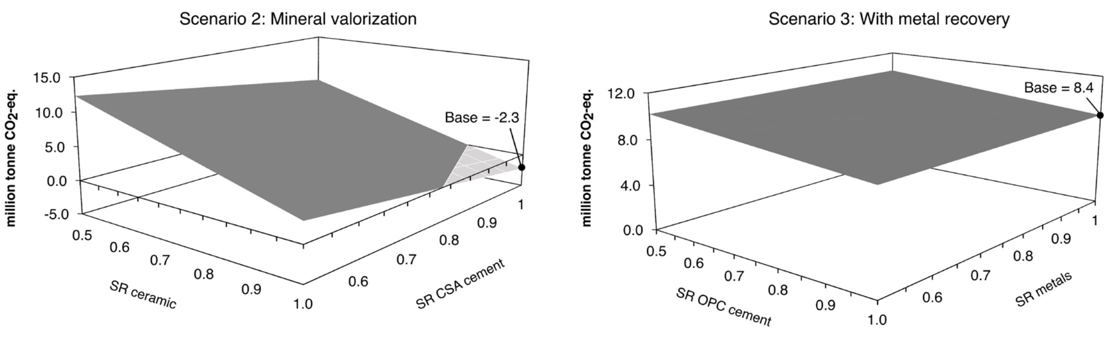

Figure C5. Influences of substitution ratios for secondary products recycling on climate change potential. Net climate change impacts for scenarios 2 (left) and 3 (right). SR = Substitution ratio.

C.6 Life cycle inventories data used for substituted primary products/ avoided services

The main sources of background data were derived from Ecoinvent 3.8 life cycle database (Ecoinvent, 2021). The data for 2020 was obtained as is, while the data for 2050 was generated using the tool 'premise'.

Key procedures to run the 'premise' tool are listed below. More detailed information can be accessed on the official website (https://premise.readthedocs.io/en/latest/index.html).

1. IMAGE model was selected for the IAM. The SSP2-RCP 1.9 W/m^2 was defined as the scenario that aligns with the study's objective (i.e., limit the global temperature increase to 1.5C by 2100, compared to pre-industrial levels).
2. Extract the ecoinvent 3.8 databases and make them useable on the brightway2 project or Activity Browser.
3. Open premise documentation to install the package on the system
4. Ask for a decryption key from the maintainers. Choose SSP2-RCP19, which is readily available when installing premise
5. Transform the selected ecoinvent 3.8 databases using premise by running a Jupyter notebook similar to this example. Codes were adapted according to the desired scenario output and original parent database.
6. Load the futurized database back into the brightway2 / Activity browser project
7. Perform LCA modeling using the systematically modified database

To illustrate, Figure C6 depicts the coupling workflow and the selected components to create life cycle databases with IAM scenarios.

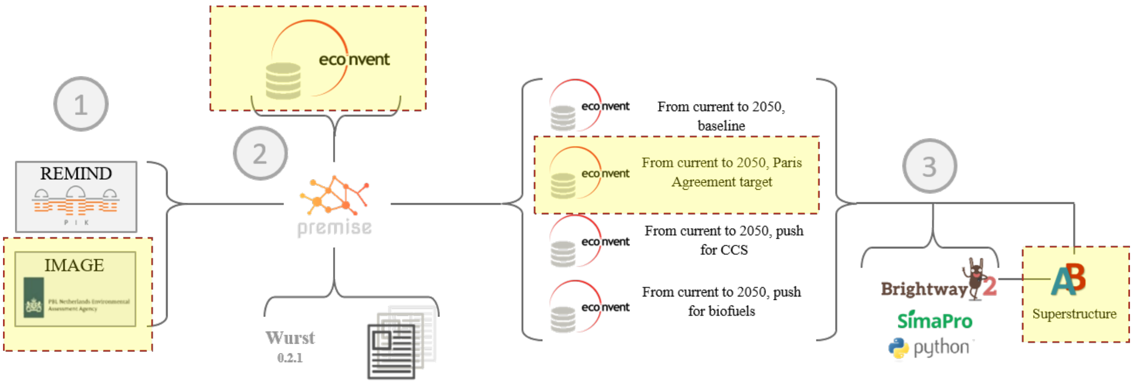

Figure C6. General IAM-LCA coupling workflow, adapted from Sacchi et al (2022). Yellow boxes indicate elements that were selected in this study.

We provided the list of key datasets used and updated for the purpose of this study (see Table C11).

Table C11. Ecoinvent 3.8 processes for the substituted primary materials production/ avoided services (including building materials, energy, and other credits).

Primary material production	Substituting secondary material	Ecoinvent 3.8 process	Unit	Remark
CSA cement	CSA cement from tailings	NA (Built upon secondary LCA data of past studies)	kg	Adapted primary production inventory based on secondary data
Ceramic tile	Ceramic tile with tailings incorporation	Ceramic tile production	kg	Adapted primary production inventory based on own calculation data
Ordinary Portland cement	Geopolymer made with tailings aggregates	Cement production, Portland	kg	Estimated market data
Sulfuric acid	Sulfuric acid from tailings off-gas	Market for sulfuric acid	kg	Prospective market data, based on future Ecoinvent database.
Heat	Surplus heat from sulfuric acid plant	Market group for heat, district or industrial, natural gas	MJ	Prospective market data, based on future Ecoinvent database.
Copper	Secondary copper recovered from tailings	Market for copper	kg	Estimated market data
Zinc	Secondary zinc recovered from tailings	Market for zinc	kg	Estimated market data
Treatment of sulfidic tailings via landfill	-	NA (Built upon emissions data of (Doka, 2017) in Ecoinvent)	kg	Adapted waste treatment inventory based on recent studies on regionalized mine tailings model (Adrianto et al., 2022).

Note: For future datasets, they were all generated using 'premise' based on Ecoinvent 3.8. Datasets for building products were taken from the life cycle inventory of tailings reprocessing and valorization in the previous study (Adrianto & Pfister, 2022). For the sulfidic tailings via landfill, results from the study of Adrianto et al. (2022) were collected and aggregated to represent total impacts of tailings disposal in the EU.

C.7 Comparison with other studies

It should be noted that it is difficult to compare the results of this study with the previous studies because 1) other studies presented their LCA results in relative values instead of absolute values, 2) LCA studies of past research were based on retrospective data collection method, not forward-looking ones as applied in the current study, and 3) the system boundary, technical processes, and materials involved frequently carry implicit differences between studies. Nevertheless, we compare our findings to the literature in Table C12.

Table C12. Comparison of environmental performance per product with previous studies.

Product	Reference	Country/ region	Processing technology	Primary energy demand [GJ/ t]	GHG [t CO2-eq./ t]	Ecotoxicity potential [Million CTUe/ t]
Copper	This study	Europe	Tailings reprocessing	1269	73.9	-115
	(Northey et al., 2013)	Global	Various (Pyro, hydro)	10 – 70	1 – 9	NA
	Ecoinvent 3.8	Global	Combined (Primary, secondary, recycling, and others)	23	1.3	4.1
Zinc	This study	Europe	Tailings reprocessing	1506	88.0	-132
	(Van Genderen et al., 2016)	Global	Electrometallurgy	37.5	2.6	NA
	Ecoinvent 3.8	Global	Combined (Primary, secondary, recycling, and others)	27	1.2	0.32
Geopolymer	This study	Europe	Tailings valorization	23.5	1.7	-1.8E-03
	(Salas et al., 2018)	Europe	Alkali activation	NA	0.13	NA
Ceramic tile	This study	Europe	Tailings valorization	5.6	0.28	-2.5E-05
	(Almeida et al., 2016)	Europe (Portugal)	Conventional	7.2	0.52	6.0E-06
	Ecoinvent 3.8	Global	Combined	6.0	0.33	1.4E-06
CSA cement	This study	Europe	Tailings valorization	3.0	0.37	-9.8E-05
	(Ren et al., 2017)	China	Conventional	4.9	0.68	NA

Note: LCA results for materials, except Ecoinvent 3.8 with IAMs, were taken based on the publication year. Therefore, the results of our study are not directly comparable to those of other studies. They are presented as a reference point for the respective product family (i.e., products made using different raw materials and processes).

For both secondary metals and building products, there are noticeable differences in GHG and primary energy demand of our study and others in the literature. This is due to i) energy/resource intensive extraction processes for recovering trace metals in tailings and ii) limited credits from low volume valorized materials. However, the toxicity impacts for the secondary metals are substantially lower than the primary counterparts (or even negative values), which means that the valorization/reprocessing has mitigating effects for such toxicity impact categories. The toxicity impacts have particularly high relevance and implications for the copper sector, since it is globally responsible for a large share of toxicity impacts compared to other metal resources (IRP, 2019).

C.8 GHG accounting for the EU copper sector

The calculation and data used to visualize results in Figure 4.6 of the thesis are summarized in Table C13. We carried out this analysis to quantify how much impact is generated per material flow category as done in the study of Ciacci et al. (2020) and to quantify how much GHG emissions are generated in the continent/ outside of the regional boundary as done in the study of Muller et al. (2020). The MFA data from Ciacci et al. (2020) was used to update the GHG results in this work (see Table C3).

Table C13. Consumption-based GHG accounting for copper sector in the EU. Values in $MtCO_2$-eq. Derived from Ciacci et. al (2020).

Year 2050	Scenario 1 /	Scenario 2 /	Scenario 3
Primary Cu production (occurs in the EU)	0.6	0.6	0.6
Import from other countries (embedded GHG) without copper recovery from tailings	3.8	3.8	3.8
Secondary supply (direct melting)	0.0	0.0	0.0
Secondary supply (secondary cathode)	2.2	2.2	2.2
Total emissions without tailings management	6.7	6.7	6.7
Domestic emissions (% of emissions in the EU territory incl. secondary production)	43%	43%	43%

Note: for scenario 3, we calculated the imported emissions equal to scenarios 1 and 2 to avoid double crediting in the table. Tailings management for scenario 3 has GHG reduction effects from metal/copper avoidance, aggregated in the total environmental performance as shown in Figure C3. Data was derived from (Ciacci et al., 2020).

We used year 2000 as the reference year to determine the GHG emission targets to achieve decarbonization goals of the copper sector. In brief, the methods to estimate GHG emissions at 2000 levels were taken from the study of Ciacci et al. (2020). Applying the model developed to account for Cu demand and the associated energy requirements and GHG emissions, we estimated that about 4.8 million tonne copper entered the use phase in the EU-28 in 2000 supplied from primary and secondary forms. Table C14 lists the data for copper flow and estimated GHG emissions.

Table C14. Mass flow, carbon intensity, and GHG emissions estimation for copper production in the EU in 2000. Data extracted from other study (Ciacci et al., 2020).

Copper flow	Volume (Mtonne copper)	Carbon intensity (Mtonne CO_2-eq/Mtonne copper)	GHG emissions (Mtonne CO_2-eq.)
Pimary copper			
Domestic	0.8	1.5	1.2
Import	3.1	3.0	9.3
Secondary copper			
Direct melting	0	0.2	0.0
Secondary cathodes	0.9	1.4	1.26
Total	4.8		11.8
80% reduction – reference line			2.4

C.9 Contribution of tailings management to GHG reduction targets

From the GHG performance we obtained, specifically from alternative tailings management, we estimated GHG reduction/ addition due to switching from traditional copper tailings management to the improved ones in scenarios 2 and 3. The results of this analysis are shown in Table 4.3 of the manuscript. Herewith we provide additional assumptions, methods, and interpretations to supplement what was discussed in the paper.

Historical GHG inventory and emission reduction targets. Past data for EU's GHG inventory was taken from the recent analysis of the European Environment Agency (2022). Under the assumption that every sector has similar GHG reduction portion, we linearly scaled the reduction amount to each major sector defined in the original study, i.e., in line with the 1.5-degree pathway, the industry category overall needs to mitigate 95% of GHG emissions from its 1990 levels by 2050. The total annual CO_2 emissions in 1990 were 4.6 billion tonne CO_2-eq. It is notable to acknowledge that this way of allocating emissions budget is only one way of many methods available, which are still under continuous discussion (Williges et al., 2022).

Tailings management impacts. The net tailings management impacts for copper were quantified for every scenario. For future scenarios, one result shows values for the base case scenario with standard/ realistic market penetration, while those in brackets (Table 4.3) show values for the best case with higher secondary market penetration. In addition, we also calculated the 'what-if' scenarios if the displacement of primary products may occur outside of the regional (EU) borders. However, this needs to be interpreted with caution: the GHG and emission reporting should be carefully reported to avoid double-counting issues, i.e., not claiming avoided emissions twice or more in the separate reporting guidelines.

Share of contribution. We calculated the contribution of improved tailings management by dividing the net negative impacts of copper tailings management by emission reduction targets, both for only the industry category and all categories. This was performed to measure and contextualize the large-scale reprocessing strategies in the overall systems. We found that emissions from tailings management would grow due to more waste volume being generated in the future. Nevertheless, we also found that specific future scenarios (Scenario 3) could lead to higher impacts compared to the business-as-usual approaches, if substitution credits of secondary production were smaller than impacts from its processing/ recycling. For these cases, either minimizing energy and resources consumption for such energy-intensive secondary processes or increasing product penetration, or a combination of both would help make scenario 3 more favorable from an environmental standpoint. We found that rising secondary market penetration—acknowledging how optimistic it is—in effect counterbalances the reprocessing burdens, making it compatible with emission reduction goals (Table 4.3 of the manuscript).

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Curriculum vitae

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Education

Jan 2019 – Dec 2022	PhD in Environmental Engineering ETH Zurich, Switzerland
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Sep 2017 – Sep 2018	MSc in Energy Environment: Sci., Tech. & Management L'X École Polytechnique, France
Feb 2017 – Jul 2017	Certificate in Management and Entrepreneurship ESADE Business School, Spain
Aug 2016 – Jun 2017	MSc in Mechanical Engineering, focus: Renewable Energy KTH Royal Institute of Technology, Sweden
Aug 2010 – Jul 2014	BSc in Chemical Engineering Bandung Institute of Technology, Indonesia

Work experiences

Jul 2021 – Dec 2021	Sustainability specialist and visiting researcher Wienerberger and KU Leuven, Belgium
Mar 2018 – Sep 2018	Research engineer / process innovation intern Électricité de France (EDF), France
Sep 2016 – Sep 2017	Technology consultant International Student Consulting Network, Sweden
Apr 2015 – Aug 2016	Account manager Dow Chemical, Indonesia
Jul 2014 – Dec 2014	Process engineer consultant LAPI ITB Consulting and World Bank, Indonesia

Languages

Indonesian/Malay	Native or mother tongue
English	Advanced
German	Good
French	Good

Publication list

This section comprises records of scientific outputs and related activities.

Journal publications

1. Scientific article: **Adrianto, L. R.***, Pfister, S., & Hellweg, S. (2022). Regionalized Life Cycle Inventories of Global Sulfidic Copper Tailings. *Environmental Science and Technology*, *56*(7), 4553–4564. https://doi.org/10.1021/acs.est.1c01786

2. Scientific article: **Adrianto, L. R.***, & Pfister, S. (2022). Prospective environmental assessment of reprocessing and valorization alternatives for sulfidic copper tailings. *Resources, Conservation and Recycling*, *186*, 106567. https://doi.org/10.1016/j.resconrec.2022.106567

3. Scientific article: **Adrianto, L. R.***, Ciacci, L., Pfister, S., & Hellweg, S. (2023). Toward sustainable reprocessing and valorization of sulfidic copper tailings: Scenarios and prospective LCA. *Science of The Total Environment, 871*, 162038. https://doi.org/10.1016/j.scitotenv.2023.162038

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Related scientific activities

Reviewers for scientific journals:

- Journal of Industrial Ecology
- Resources, Conservation & Recycling
- Minerals Engineering
- Integrated Environmental Assessment and Management
- Journal of Cleaner Production
- Indonesian Journal of Energy

Conference organization:

- As organizer: Life Cycle Assessment Discussion Forum 76 "The use of LCA for forecasting and steering new technologies" on 19 November 2020 in Zurich, Switzerland. Link to program and recordings