Environmental impacts of processing secondary cobalt raw materials

Riina Aromaa-Stubb

Aalto University

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Environmental impacts of processing secondary cobalt raw materials

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ABSTRACT

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The demand for cobalt is predicted to continue growing in the next decades and is currently being supplied practically entirely by primary production, which is highly concentrated geographically and raises environmental concerns. Increasing the supply of cobalt from secondary raw materials is necessary to meet the growing demand and can help diversify the supply and lower the environmental impacts of cobalt production. The goal of this thesis was to assess the environmental impacts of cobalt recycling from hardmetal scrap and spent cobalt catalysts.

The methodology used in this study was simulation-based life cycle assessment (LCA). The studied processes were simulated with HSC Sim software based on information and values from the literature to gather an inventory of direct emissions and consumed chemicals and utilities. This allows for investigation of the environmental impacts of processes still in the development stage. The environmental impacts were calculated in GaBi or openLCA software using the ecoinvent database for the background data, such as the production of the chemicals and electricity consumed. Contribution, scenario, sensitivity and uncertainty analyses were used in the interpretation phase to formulate the conclusions and recommendations.

In total nine scenarios were assessed in this work. Five recycling scenarios examined hardmetal recycling with three focusing on tungsten and cobalt recycling and two on tantalum and niobium recycling. In addition, four catalyst recycling scenarios were investigated. Three were focused on cobalt recycling from a Fischer-Tropsch catalyst, and one on molybdenum, vanadium, nickel and cobalt recycling from a Mo/Co/γ-Al_2O_3 catalyst. All scenarios apart from the tantalum and niobium recycling scenarios showed that the environmental impacts of the recycling systems were lower than those of primary production of the same products.

Data produced by process simulation is considered secondary data, which has a higher uncertainty than primary data measured directly from a plant. This study used several methods of sensitivity and uncertainty analysis to estimate the effect of data uncertainty on the conclusions and recommendations formulated based on process simulation data. The analyses were performed both directly on the inventory items of the LCA model, such as the consumption of chemicals, and the process simulation parameters that affected the formation of the inventory. For the process simulation parameters related to the consumption of chemicals and utilities the results were similar to the results of the analyses focused on the inventory, while the parameters related to direct emissions showed a larger difference between the analyses.

Aalto-yliopisto

TIIVISTELMÄ

Tekijä Riina Aromaa-Stubb

Väitöskirjan nimi Sekundääristen kobolttiraaka-aineiden prosessoinnin ympäristövaikutukset

Artikkeliväitöskirja

Sivumäärä 84

Avainsanat prosessimallinnus, ympäristövaikutukset, kovametallin kierrätys, katalyyttien kierrätys

Koboltin kysynnän on ennustettu kasvavan tulevina vuosikymmeninä. Nykyisellään lähes kaikki koboltti tuotetaan primääriraaka-aineista ja sen tuotanto on keskittynyt maantieteellisesti harvoille alueille. Tuotantoon liittyy myös ympäristövaikutuksia sekä huolta toiminnan kestävyydestä. Kierrätysraaka-aineiden käytön lisääminen koboltin tuotannossa on käytännössä välttämätöntä ja sen avulla voidaan myös parantaa raaka-ainesaatavuutta sekä laskea tuotannon ympäristövaikutuksia. Tämän väitöskirjan tutkimuksen tavoitteena on arvioida koboltin kierrätyksen ympäristövaikutuksia, kun raaka-aineina käytetään kovametallia ja kobolttikatalyyttejä.

Tutkimuksessa käytettiin tutkimusmenetelmänä prosessimallinukseen perustuvaa elinkaariarviointia (LCA). Tutkittavat prosessit mallinnettiin HSC Sim -ohjelmalla kirjallisuudesta saatujen prosessitietojen ja -arvojen avulla. Kierrätysprosessimallin perusteella voitiin rakentaa inventaario, joka sisältää numeraalisina arvoina sekä prosessin suorat päästöt että käytettyjen kemikaalien ja hyödykkeiden kulutuksen. Tämänkaltainen inventaario mahdollistaa kehitteillä olevien prosessien ennakoivan ympäristövaikutusten arvioinnin. Ympäristövaikutukset laskettiin GaBi tai openLCA -ohjelmalla ja taustaprosessien (kuten käytettyjen kemikaalien ja sähkön tuotanto) arvoina käytettiin ecoinvent-tietokannan arvoja. Tulosten tulkinnassa sekä johtopäätösten ja suositusten määrityksessä käytettiin erilaisia kontribuutio-, skenaario-, herkkyys-, ja epävarmuusanalyysejä.

Tutkituissa kierrätysprosesseissa arvioitiin yhteensä yhdeksän prosessiskenaariota. Viisi kierrätysprosessia keskittyi kovametallin kierrätykseen ja näistä kolme volframin ja koboltin kierrätykseen, kun taas kaksi tantaalin ja niobiumin kierrätykseen. Lisäksi tutkittiin neljää katalyyttien kierrätysprosessia. Kolme tutkittua prosessia keskittyi koboltin kierrätykseen Fischer-Tropsch-katalyytistä ja yksi molybdeenin, vanadiinin, nikkelin ja koboltin kierrätykseen Mo/Co/γ-Al_2O_3-katalyytistä. Kaikissa skenaarioissa - tantaalin ja niobiumin kierrätystä lukuun ottamatta - kierrätysprosessien ympäristövaikutukset olivat pienemmät kuin samojen tuotteiden ympäristövaikutukset olisivat valmistettaessa primääriraaka-aineista.

Tutkimuksessa käytettävällä prosessimallinnuksella kerätyt lähtötiedot ovat toissijaista, ja niillä on korkeampi epävarmuus kuin ensisijaisesti suoraan laitokselta kerätyillä massa- ja energiatase tiedoilla ja arvoilla. Tässä tutkimuksessa käytettiin useita herkkyys- ja epävarmuusanalyysimetodeja arvioimaan, miten mallinnettuun dataan liittyvä epävarmuus vaikuttaa tulosten perusteella määritettyihin johtopäätöksiin ja suosituksiin. Nämä analyysit tehtiin sekä suoraan elinkaariarvion mallin inventaariolle, kuten kemikaalien kulutukselle, että prosessimallinnuksen parametreille, jotka vaikuttavat inventaarion muodostumiseen. Prosessimallinnuksen parametreilla, jotka liittyivät kemikaalien ja hyödykkeiden kulutukseen tulokset olivat samankaltaisia inventaarion analyysien tulosten kanssa, kun taas suoriin päästöihin liittyvillä parametreilla erot olivat suurempia.

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My time at Aalto University spans almost a decade, all the way from starting as a bachelor student in the autumn of 2014 to this doctoral thesis in 2025, although there was a year-long break after graduating in 2019. During my studies I was mostly interested in the environmental impacts of industrial production and was adamantly against both studying more hydrometallurgy than was mandatory and pursuing doctoral studies. However, life has a way of changing a young person's mind so when it became possible to do a doctorate on a life cycle assessment topic, I jumped at the chance. The decision might have been spontaneous, but it led to a great journey that was made that way by many wonderful people.

I want to start by thanking my supervisor, Professor Mari Lundström, for giving me this opportunity to pursue doctoral studies in her research group and all her support and academic advice throughout these years. This has allowed me to take on duties and projects that have pushed me out of my comfort zone and helped me develop a skillset that I value highly.

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Espoo, 9 July 2025
Riina Maria Aromaa-Stubb

Contents

List of Abbreviations and Symbols

Abbreviations

AP	Acidification potential
APT	Ammonium paratungstate
DQI	Data quality indicator
DRC	Democratic Republic of the Congo
EIA	Environmental impact assessment
EoL	End-of-life
EP	Eutrophication potential
EU	European Union
EV	Electric vehicle
FT	Fischer-Tropsch
GWP	Global warming potential
HDS	Hydrodesulphurisation
HPAL	High-pressure acid leaching
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
LCO	Lithium cobalt oxide
LIB	Lithium ion battery
NMC	Nickel-manganese-cobalt
ODP	Ozone depletion potential
POCP	Photochemical oxidant creation potential

Symbols

A	Technology matrix
B	Intervention matrix
f_n	Final demand vector
g_n	Inventory vector
h_n	Impact vector
P	Process matrix
p_n	Vector to describe unit process
Q	Characterisation matrix
q_n	System vector
s_n	Scaling vector

List of Publications

This doctoral dissertation consists of a summary and of the following publications which are referred to in the text by their Roman numerals.

I. **Aromaa, Riina**; Rinne, Marja; Lundström, Mari. 2022. Comparative Life Cycle Assessment of Hardmetal Chemical Recycling Routes. American Chemical Society. ACS Sustainable Chemistry & Engineering, volume 10, issue 31, pages 10234-10242. DOI:10.1021/acssuschemeng.2c01969.

II. **Aromaa, Riina**; Rinne, Marja; Lundström, Mari. 2023. Life Cycle Assessment of Tantalum and Niobium Recycling from Hard Metal Scrap. American Chemical Society. ACS Sustainable Chemistry & Engineering, volume 11, issue 41, pages 14997-15005. DOI:10.1021/acssuschemeng.3c03540.

III. **Aromaa-Stubb, Riina**; Rinne, Marja; Lundström, Mari. 2024. Life Cycle Assessment of Cobalt Catalyst Production and Recycling. Journal of Sustainable Metallurgy, volume 10, pages 1795-1806. DOI: 10.1007/s40831-024-00897-0.

IV. **Aromaa-Stubb, Riina**; Rinne, Marja; Lundström, Mari. 2025. Life Cycle Assessment of Mo, V, Ni, and Co Recovery from Spent Catalyst. ACS Sustainable Resource Management. DOI: 10.1021/acssusresmgt.5c00243.

Authors’ Contribution

Publication I: Comparative Life Cycle Assessment of Hardmetal Chemical Recycling Routes

Conceptualisation and formulation of scenarios was done by RA with the guidance of MR and ML. RA performed the process simulations and life cycle assessment along with data analysis and visualisation with the guidance of MR. The manuscript was written by RA and reviewed by MR and ML. Supervision and funding acquisition was by ML.

Publication II: Life Cycle Assessment of Tantalum and Niobium Recycling from Hard Metal Scrap

Conceptualisation and formulation of scenarios was done by RA with the guidance of MR and ML. RA performed the process simulations and life cycle assessment along with data analysis and visualisation. The manuscript was written by RA and reviewed by MR and ML. Supervision and funding acquisition was by ML.

Publication III: Life Cycle Assessment of Cobalt Catalyst Production and Recycling

Conceptualisation and formulation of scenarios was done by RA. RA performed the process simulations and life cycle assessment along with data analysis and visualisation. The manuscript was written by RA and reviewed by MR and ML. Supervision and funding acquisition was by ML.

Publication IV: Life Cycle Assessment of Mo, V, Ni, and Co Recovery from Spent Catalyst

Conceptualisation and formulation of scenarios was done by RA. RA performed the process simulations. Life cycle assessment was conducted by RA and MR. RA performed the data analysis and visualisation. The manuscript was written by RA and reviewed by MR and ML. Funding acquisition was by RA and ML. Supervision was by ML.

1. Introduction

The consumption of cobalt is growing at a fast pace, with the demand being almost entirely supplied by primary production in the Democratic Republic of the Congo (DRC) and Indonesia (Cobalt Institute, 2023a). The recent growing demand can mostly be attributed to the consumption of cobalt in the battery sector, but cobalt is also an essential material for the manufacturing and tooling industries and the production of certain alloys and catalysts (European Commission, 2020). Substantial stocks of cobalt become available each year in end-of-life (EoL) products from which it could be recovered. Currently, however, only a minor proportion of new cobalt is produced from secondary raw materials (Cobalt Institute, 2023a). The drive towards the circular economy raises the question of how the impact of cobalt production would be affected by increased utilisation of secondary resources.

Primary production of cobalt is associated with concerns regarding water and air pollution and effects on biodiversity (Cobalt Institute, 2023a). A commonly held assumption is that the environmental impacts of recycling are lower than those of primary production. In reality, however, environmental benefit from recycling is not a given and depends on multiple factors such as raw material composition and the unit processes chosen.

1.1 Background

The use of cobalt dates back to the Bronze Age, when it was first used to give a blue colour to glass, glazes and ceramics (Roberts and Gunn, 2014). Cobalt oxide, although black, when added to molten glass gives it a blue colour (Habashi, 2016). Blue pigments produced from cobalt ore along with cobalt-aluminium blues have been traded since ancient times (Kaczmarcyk, 1986), but after the first uses of cobalt for aesthetic purposes it took until the 20th century for it to find a use in technology applications (Roberts and Gunn, 2014). As a pure metal, cobalt was first isolated in 1735 but the first real demand for it came at the turn of the 20th century with the development of cobalt-chromium alloys (Roberts and Gunn, 2014).

The non-oxidizing properties of cobalt-chromium alloys and their resistance to changes under heat were discovered during World War I and in the following decades, the production of cobalt multiplied at a fast pace, from 400 tons in

1920 to 5 500 tons in 1940, driven in part by its use in heat-resistant alloys (Bilbrey, 1962). A further increased demand for cobalt came from the aircraft industry, which required high-temperature alloys as the invention of the jet engine and stockpiling of cobalt during the Korean war drove production up to 17 300 tons in 1959 (Bilbrey, 1962).

The early 20th century was also an important period of development in industrial catalysis. One of the most well-known applications of cobalt in catalysis has been in Fischer-Tropsch (FT) synthesis. An early standard FT catalyst with cobalt was developed in the 1930s, although during World War II, research shifted to finding cheaper catalysts due to the limited availability and higher price of cobalt (Gholami *et al.*, 2020). By the 1980s, cobalt catalysts in the form of cobalt-molybdena on alumina support had become some of the most common catalysts used in hydrogenation units to remove sulphur and nitrogen from crude oil (Heinemann, 1982).

By the 1960s, use in pigments had become a minor application and cobalt had established a lasting foothold in the manufacturing and tooling industries. Major applications of cobalt included permanent-magnet alloys, high-temperature alloys, hardmetals and tool steels (Bilbrey, 1962). The early 20th century, which saw the development of high-temperature alloys, also witnessed the invention of hardmetals, also known as cemented carbides. Tungsten carbide cemented in a cobalt matrix drastically reduced the costs of manufacturing and increased the output of the American metalworking industry by a factor of three between 1939 and 1945 (Ettmayer *et al.*, 2014).

In the current market, the traditional applications of cobalt have been surpassed by the battery industry, with electric vehicle (EV) batteries and portable batteries becoming the largest consumers of cobalt with a 40% and 30% share, respectively, in 2022 (Cobalt Institute, 2023a). Superalloys, hardmetals, and catalysts account for 9%, 5% and 3% of the demand, respectively (Cobalt Institute, 2023a).

The demand for cobalt is currently mostly supplied by primary production. The largest cobalt producer is the DRC, which in 2022 produced 73% of mined cobalt (Cobalt Institute, 2023a). China is heavily involved in cobalt mining in the DRC with the majority of operations either partially or fully Chinese-owned and it also produced 76% of refined cobalt globally in 2022 with a large proportion of the intermediates originating in the DRC (Cobalt Institute, 2023a). In the DRC cobalt is produced as a by-product of copper. The minerals are divided into sulphide and oxide minerals, with the oxides in reality being mostly carbonates and hydroxides. The main minerals in sulphide ore are chalcopyrite ($CuFeS_2$), bornite (Cu_5FeS_4) and carrollite ($CuCo_2S_4$) and in oxide ore the main minerals are malachite ($Cu_2CO_3(OH)_2$) and heterogenite ($CoO(OH)$) (Crundwell *et al.*, 2020). The extraction technologies depend on the mineralogy of the ores, with flotation used for sulphide ores and leaching and solvent extraction for oxide ores (Crundwell *et al.*, 2020).

Cobalt is also produced as a by-product of nickel from nickel sulphide ores and limonite in nickel laterite ores. Nickel sulphide ores have traditionally been the larger source of cobalt with cobalt contained in minerals such as pentlandite

((Ni, Fe, Co)$_9$S$_8$) and they are typically processed using flotation and matte smelting (Crundwell *et al.*, 2011). The main mineral in limonite is goethite (α-FeO(OH)) in which nickel and cobalt are present alongside iron and substitute for iron in the structure (Crundwell *et al.*, 2011). Limonite ores can be treated by various methods such as reduction roasting and leaching (the Caron process) or high-pressure acid leaching (HPAL) (Crundwell *et al.*, 2011). Indonesia, which became the second largest cobalt producer in 2022 accounting for 5% (Cobalt Institute, 2023a), represents cobalt mining from laterites (Dehaine *et al.*, 2021). China is also heavily involved in cobalt and nickel production in Indonesia, where all of the HPAL plants currently in operation are owned by Chinese-Indonesian companies (Cobalt Institute, 2023a).

The market size of cobalt is expected to double by 2030 relative to 2022, with most of the growth attributed to the battery industry (Cobalt Institute, 2023a). Currently, 86% of the growth in annual demand of cobalt is due to its use in EVs while the traditional non-battery applications account for only 6% of the growth (Cobalt Institute, 2023a).

The largest primary producers of cobalt, the DRC and Indonesia, are expected to contribute most of the necessary growth in cobalt production to meet the increasing demand (Cobalt Institute, 2023a). The DRC is expected to supply 44% of the growth in primary production by 2030 and Indonesia could contribute 37% of the growth (Cobalt Institute, 2023a).

The fact that the production of cobalt is geographically concentrated in only a limited number of regions, along with other factors, makes its supply vulnerable to disruption. This is further exacerbated by the significant involvement of China in the DRC and Indonesia that together represent almost 80% of cobalt mining, which poses a risk to the wider market (Cobalt Institute, 2023a). The production of cobalt as a by-product is also highly dependent on continued demand for its host metals. At present, 70% of cobalt is produced as a by-product of copper, 20% as a by-product of nickel, 8% in operations extracting both copper and nickel, and only a very minor amount of cobalt (2%) is produced in operations where it is the main commodity (van den Brink *et al.*, 2020). Due to the supply risks and demand for cobalt in essential applications where it cannot be substituted practically, cobalt has been included in the European Union's (EU) list of critical raw materials in every iteration since the first one was released in 2011 (European Commission, 2023).

There is a need for regionalised supply chains of cobalt in Europe. Finland is the only European country to contribute to the mined supply of cobalt, but the output is small compared to production in the DRC and Indonesia (Cobalt Institute, 2023a). Finland produces 10% of refined cobalt globally, making it the second largest cobalt refiner after China (Cobalt Institute, 2023a). Finland supplies 54% of the refined cobalt consumed in the EU, and Belgium and Norway supply an additional 7% each (European Commission, 2020). However, Europe is still reliant on imports from the DRC as they constitute 68% of EU sourcing of cobalt ores, concentrates and intermediates while domestic production in Finland contributes 14% (European Commission, 2020).

Environmental damage can add to the supply risk when the production causes a level of environmental damage that is unacceptable to the surrounding society. The level of risk for current cobalt deposits is relatively low but stronger environmental regulations could hinder new operations or expansion of existing operations (van den Brink *et al.*, 2020). The cobalt mining industry in the DRC has faced issues with water and air pollution, waste management and biodiversity risks due to lax mining standards and concerns about biodiversity impacts and adherence to environmental standards are also associated with the rapidly growing cobalt production in Indonesia (Cobalt Institute, 2023a). One way of reducing the pressure on local environments and finite resources is by increasing the supply from secondary sources of cobalt (Cobalt Institute, 2023a).

With the estimated increase in cobalt demand, the sufficiency of the supply has been called into question. Although not enough to fully bridge the gap on its own, one source of cobalt that can provide a substantial share is EoL products (Fu *et al.*, 2020). So far recycling has played only a minor role in the supply of cobalt and currently the share of cobalt demand being supplied from secondary resources is only 5%, however, it has been estimated that this will increase to 15% by 2030 and exceed 40% by 2040 (Cobalt Institute, 2023a). The share is expected to grow as larger volumes of EoL batteries become available and the recycling capacity to process it increases (Cobalt Institute, 2023a).

While some cobalt uses such as in pigments are dissipative, cobalt can be collected and recycled from its largest applications in batteries, superalloys, hardmetals and catalysts (European Commission, 2020). The majority (65%) of the secondary cobalt currently recycled is from spent batteries and the rest is from hardmetal scrap (24%), and alloy scrap and spent catalysts (11%) (Cobalt Institute, 2023b).

The recycling rates of the different applications vary by a significant degree. While an estimated 46% of hardmetal scrap is recycled (Zeiler *et al.*, 2021), it is estimated that only 5% of spent catalysts are recovered (Sun *et al.*, 2019). The collection of alloys and hardmetal scrap as well as their recycling is generally operated by the superalloy and hardmetal industries while the recycling of spent catalysts and battery waste is done within the cobalt industry or dedicated plants for battery waste (European Commission, 2020). Some recycling efforts, such as the remelting of alloy scrap into new alloys, aim at the direct recycling of the secondary raw material as-is. Other methods target the separation and individual recovery of valuable elements, such as in the case of the chemical recycling of hardmetal scrap or hydrometallurgical treatment of battery waste.

The environmental impacts of recycling are often lower than those of primary production, although this is not true in all instances. The environmental impacts of metals refining depend particularly on the raw material composition and the process used. Secondary raw materials from EoL products with cobalt typically contain not only cobalt but other valuable materials as well. Complex compositions may increase the complexity of the process necessary for the separation and recovery of all the valuable materials. An increase in the number of unit processes that make up the process can lead to increased consumption of chemicals and energy and generate more wastes of varying quality. As the environmental impacts cannot be determined directly, a methodology such as life cycle assessment (LCA) is required to assess them.

Since the environmental impacts are so dependent on the raw material and process used, the accuracy of the LCA results is reliant on the availability of representative data. This poses a challenge as the aggregated and average data available in LCA databases and literature is rarely specific enough to describe any particular case well. In the absence of readily available data, alternative methods are necessary to produce it. After primary data measured and collected at a facility, process simulation is the most accurate way of producing the necessary life cycle inventory (LCI) data (Parvatker and Eckelmann, 2019). The material and energy balances from process simulation can be used to assist in the collection of the input and output streams that form the LCI of the system. The methodology has been used to evaluate the environmental impacts of both primary metals production (Elomaa *et al.* 2020a; Elomaa *et al.*, 2020b; Rinne *et al.*, 2021a; Rinne *et al.*, 2022; Rinne *et al.*, 2023; Lappalainen *et al.*, 2024; Nan *et al.*, 2024a) and secondary metals production (Rinne *et al.*, 2021b; Rinne *et al.*, 2024; Arellano-Sanchez *et al.*, 2024; Nan *et al.*, 2024b). The method requires detailed knowledge and data gathering of the process conditions and chemical reactions for example but enables a high resolution of the different unit processes of the system.

Process simulations for LCA are often based on up-scaled laboratory data and estimations in the absence of more accurate industrial-scale data. This affects the uncertainty associated with the life cycle impact assessment (LCIA) results. Uncertainty is ever-present in LCA and may originate from several different sources. Sources of uncertainty that relate to process simulation-based LCA include the uncertainty related to modelling choices and parameter uncertainty (Bojarski *et al.*, 2008). The inclusion of uncertainty analysis in simulation-based LCAs of metal refining has been limited but it provides vital information on the robustness of the conclusions drawn from the results.

1.2 Objectives and scope

In the interests of the circular economy and due to its criticality, cobalt should be recovered from EoL products. The environmental benefit of this, however, is not a given and the environmental impacts of recycling cannot be estimated based on the environmental impacts of primary production alone, due to different raw material composition and process configurations, for example. This thesis investigates the industrial recycling of hardmetal scrap and spent catalysts in chosen process routes by means of LCA. As primary LCI data for the studied processes is unavailable due to their development-stage status or for some other reason, process simulation has been used to generate the LCI, which affects the uncertainty related to the data.

Thus, the objectives of this thesis were as follows:

i. to simulate hardmetal and catalyst recycling processes to produce LCI data,
ii. to perform LCA of hardmetal and catalyst recycling processes to compare their environmental impacts,
iii. to utilise different uncertainty propagation methods to determine their effect on conclusions drawn from impact assessment results based on the LCI generated by process simulation.

The results obtained in this research are included in the publications (I-IV) and the compendium section of this thesis. **Figure 1** presents how the topics of the publications (I-IV) are related and their links to Objectives i-iii as described above.

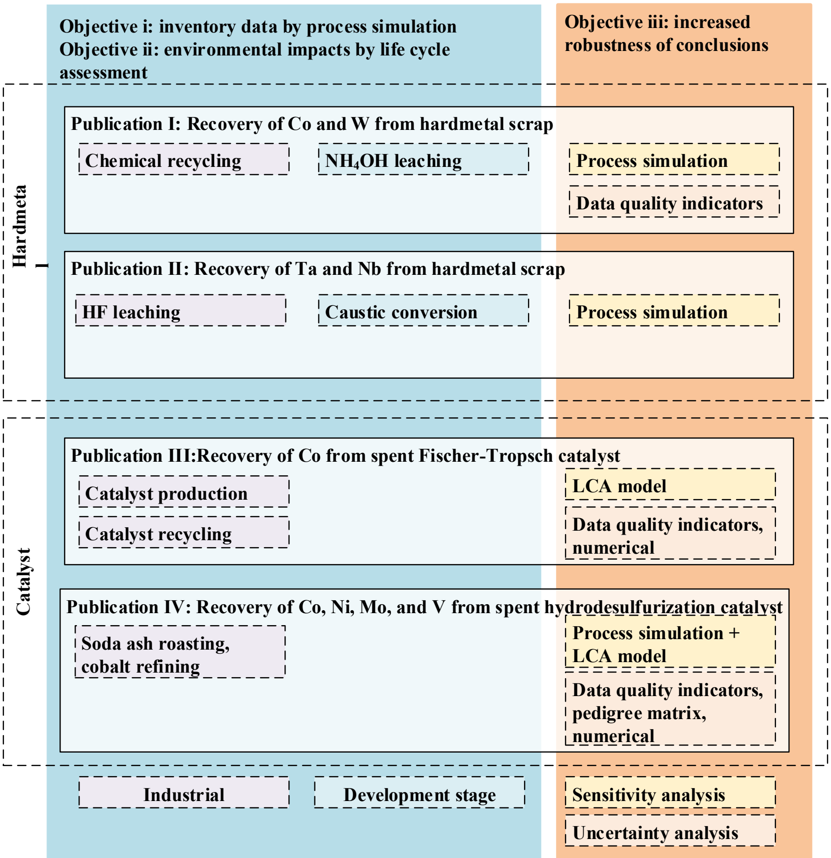

Figure 1. Schematic of thesis publications and their link to the objectives.

1.3 New scientific contribution

This study contributes to the existing scientific knowledge regarding the environmental impacts of cobalt recycling concerning both industrial operation and proposed development-stage processes. The following case studies and findings presented in the publications are proposed as original:

Publication I: Chemical recycling of cobalt and tungsten from hardmetal scrap was studied by process simulation and LCA. A process based on the industrial processes for tungsten refining was compared to a development-stage process.

i. A fairly large proportion of the total impacts of chemical recycling was caused by a few inputs to the process: heating for oxidation of scrap, alkali consumption in leaching and stripping, electricity consumption, and ammonia emissions from ammonium paratungstate (APT) crystallisation.
ii. The environmental impacts calculated for the development-stage process were 7.7-25.7% larger than those calculated for industrial chemical recycling. This was due to longer multi-step oxidation, longer leaching time, higher concentration of alkali and larger volumes. However, considering the uncertainty, the difference cannot be considered significant in all impact categories.
iii. The environmental impacts of industrial chemical recycling were small compared to the other stages in the production of tungsten carbide cemented in a cobalt matrix. When using 50/50 primary and recycled APT and cobalt hydroxide, the impact of recycling was highest in global warming potential (GWP) but only accounted for 14% of the total impacts.

Publication II: The possible recovery of tantalum and niobium from a leach residue from the chemical recycling of hardmetal scrap was investigated by process simulation and LCA. A process based on tantalum and niobium refining from coltan ore was compared to a development-stage process. The effect of the tantalum and niobium concentration in the hardmetal scrap on the recycling benefit was investigated.

i. The impacts of the HF-based leaching process were found to be mostly due to the leaching and effluent treatment process steps as a result of the consumption of hydrofluoric and sulphuric acids as well as lime.
ii. A large share of the impacts of the development-stage HF-free process were found to be due to the caustic conversion and solvent extraction steps. This was mostly due to the consumption of sodium hydroxide and oxalic acid.
iii. The HF-free process showed some promise of being less environmentally harmful than the traditional HF-based process. However, the very high impacts due to the consumption of oxalic acid make it unrealistic unless the oxalic acid is produced from non-fossil sources or replaced in the process.
iv. The recycling of tantalum and niobium from the leach residue of hardmetal chemical recycling has the potential to be environmentally beneficial when the content of the metals in the hardmetal scrap is high enough. With a combined 5 wt.% of metals in the scrap, the calculated impacts are lower than those of primary production, although the conclusion cannot be drawn directly considering the uncertainty and more refined data would be necessary. With higher tantalum and niobium contents in the scrap, the environmental benefit is clear.

Publication III: Recycling of cobalt from spent FT catalyst was studied by process simulation and LCA with three intermediate products: cobalt hydroxide, cobalt sulphate and cobalt carbonate. The impact of using primary and secondary cobalt hydroxide for catalyst production was investigated.

i. Production of cobalt hydroxide used to prepare the cobalt precursor and titanium dioxide which was used as the support contribute the majority of the impacts originating from the catalyst production
ii. Using approximations of cobalt catalyst may substantially underestimate the true impacts when the catalyst production process is omitted. The impacts of a cobalt catalyst production process cannot be accurately described by using generic cobalt production impact data.
iii. In the recycling of the spent catalyst in the cobalt refining process, the majority of the impacts are due to the consumption of sodium hydroxide and sulphuric acid.
iv. Cobalt chemicals produced from the spent catalyst raw material had substantially lower environmental impacts when compared to production of cobalt chemicals from primary raw materials. The environmental impacts resulting from catalyst production could be substantially reduced by using secondary cobalt to produce the cobalt precursor.

Publication IV: Recycling of cobalt, molybdenum, vanadium and nickel from spent MoCo/ɣ-Al2O3 catalyst was investigated by process simulation and LCA. The effect of the concentration of the valuable metals in the spent catalyst on the recycling benefit was studied.

i. The largest contributors to the environmental impacts of the recycling process varied by impact category but included the consumption of electricity in the process, ammonia and hydrochloric acid in molybdenum recovery, sodium hydroxide in nickel and cobalt recovery, and sulphuric acid in leaching and waste treatment as well as the emissions of ammonia and carbon dioxide.
ii. Recycling of the spent catalyst to recover the metals is only environmentally beneficial if the metal content in the catalyst is high enough. When the recoverable metal content is low (6 wt.%), the global warming potential of the recycling process is 1.5 times higher than that of primary production, although in other impact categories the recycling is still beneficial.
iii. The recovery of cobalt from spent catalyst with high enough metal content is also environmentally beneficial. The allocated results showed that the environmental impact of the recycled cobalt is lower than production from primary raw materials in all impact categories.
iv. The sensitivity and uncertainty analyses showed that performing such analysis on the LCA model alone is insufficient to capture the parameter uncertainty originating from the simulation when the value of large contributors is highly dependent on uncertain simulation parameters.

1.4 Structure of this thesis

This thesis consists of three scientific peer-reviewed journal publications [I - III], one manuscript submitted to a peer-reviewed journal [IV] and a compendium section. The publications are attached to the thesis as appendices.

Chapter 2 in the compendium section discusses the theoretical background of the research by presenting the status of cobalt recycling along with the framework of LCA for recycling processes. A review of uncertainty and sensitivity analysis methods in LCA is also presented. Chapter 3 presents the framework for the process simulation and LCA used in Publications I-IV. Chapter 4 outlines the environmental assessment results which are discussed in Chapter 5 along with recommendations for future research. Finally, the conclusions are presented in Chapter 6.

2. Theoretical background

2.1 Cobalt recycling

In the production of new cobalt products, currently only 5% of the raw materials used are secondary (Cobalt Institute, 2023a). The majority of cobalt recycling takes place in China, where almost half of recycled cobalt is produced while Europe and North America account for a quarter each (Cobalt Institute, 2024). Large stocks of cobalt would be available for recovery from secondary resources in the form of manufacturing scrap and EoL products.

The leading application of cobalt is currently in battery applications which in total accounted for 72% of cobalt use in 2022 and 40% of cobalt is used for EVs while another 30% is used for portable electronics (Cobalt Institute, 2023a). The demand for cobalt for battery applications was driven by lithium cobalt oxide (LCO) chemistries until 2022 when nickel-manganese-cobalt (NMC) chemistries became the largest driver (Cobalt Institute, 2023a). The different cathode chemistries such as LCO and NMC contain different amounts of cobalt with the content in LCOs being much higher (60%) than in NMC (6-20%) (Cobalt Institute, 2025).

In line with lithium-ion batteries (LIB) being the largest application of cobalt, the majority of recycled cobalt on the market also comes from batteries. Most of this (74%) comes from battery manufacturing scrap which accounted for 9.3 t of recycled cobalt in 2022, whereas cobalt in EoL products is largely wasted (Cobalt Institute, 2025). Currently, the largest wasted recovery potential is in portable electronic batteries of which the majority end up mostly abandoned or disposed of in landfills or incinerators as recycling is limited by low collection rates and availability of recycling capacity (Cobalt Institute, 2025). The amount of cobalt recycled from EoL LIBs was 2.4 t in 2022 (Cobalt Institute, 2025) while the total of possible cobalt resources in electronic waste is more than 34 000 t annually (Baldé *et al.*, 2024). Only 22.3% of e-waste is formally collected or recycled (Baldé *et al.*, 2024). As EVs become the largest source of potential secondary cobalt, the collection and recycling systems will be equally important, and the total amount of recycled cobalt will be affected by whether EVs are scrapped in developed economies or in low-income countries with a lack of LIB recycling capacity (Cobalt Institute, 2025).

Most LIB recyclers use a combination of mechanical, pyrometallurgical and hydrometallurgical processes to recover the valuable metals contained in batteries. Most of the large-scale commercial recyclers focus on the recovery of copper, nickel and cobalt and the recycling flowsheets vary significantly between recyclers (Sommerville *et al.*, 2021). Spent batteries can be processed by deactivation and mechanical pretreatment to produce black mass or they can be fed directly into a pyrometallurgical process to produce a metal alloy with the investigated recycling processes for black mass including direct recycling to produce a cathode active material and pyro- or hydrometallurgical recycling (Sommerville *et al.*, 2021). Black mass can be fed into a pyrometallurgical process followed by hydrometallurgical treatment of the alloy or fed directly into a hydrometallurgical process which can recover individual metals, typically as metal salts (Sommerville *et al.*, 2021).

In addition to LIBs, cobalt can be recycled from superalloy scrap, hardmetal scrap and spent catalysts (European Commission, 2020). Superalloys, which are the second largest application of cobalt, are difficult to recycle due to their physical and mechanical properties (Srivastava *et al.*, 2014). The recycling processes are based on either pyrometallurgy or hydrometallurgy or a combination of the two and for the most part, the recycling of superalloys has been limited to only the re-melting of scrap in the manufacturing industries due to the difficulty of recycling and large variation in composition (Srivastava *et al.*, 2014). However, re-melting by mixing with fresh material can lose up to 20% of the alloying elements, many of which are considered critical (Srivastava *et al.*, 2014). Cobalt can therefore be recycled from superalloys in closed-loop recycling but is not recovered as a chemical product available for further use in other applications as it is from batteries. Applications besides batteries from which cobalt can be recycled as cobalt chemicals in open-loop recycling include hardmetal scrap and spent cobalt catalysts.

The intended product, raw material composition as well as the recycling flowsheet and technology used have a significant effect on the environmental impacts of a metal product produced from secondary raw materials. The research into the environmental impacts of the recycling of cobalt-containing raw materials has been focused on LIBs with quantification of the impacts of different battery chemistries in pyrometallurgical processes (Hendrickson *et al.*, 2015; Boyden *et al.*, 2016; Cusenza *et al.*, 2019; Ciez and Whitacre, 2019; Rajaeifar *et al.*, 2021, Kallitsis *et al.*, 2022), hydrometallurgical processes (Dunn *et al.*, 2012; Hendrickson *et al.*, 2015; Boyden *et al.*, 2016; Cusenza *et al.*, 2019; Ciez and Whitacre, 2019; Raugei and Winfield, 2019; Rinne *et al.*, 2021b; Duarte Castro *et al.*, 2022; Kallitsis *et al.*, 2022; Rinne *et al.*, 2024) and direct recycling processes (Dunn *et al.*, 2012; Ciez and Whitacre, 2019) . In addition to cobalt recycling from batteries, recycling from hardmetal scrap has been quantified for zinc process recycling and chemical recycling by electrolysis (Furberg *et al.*, 2019).

2.1.1 Recycling of hardmetal scrap

Hardmetals are widely used in the manufacturing industry in applications such as drilling, cutting tools and wear parts (Konyashin, 2014). They consist of a tungsten carbide (WC) matrix and a metal binder, for which cobalt is often used and most industrial grades for different applications are formed by varying the WC grain size and the binder content (Konyashin, 2014). The grain size is typically 0.2-10 μm and the cobalt content is typically 6-16% (Fang *et al.*, 2014). In addition, hardmetal can include coatings or grain growth inhibitors to achieve the desired properties. The coatings can be Ti, Cr, Al, Hf or Ta as carbides, nitrides or carbonitrides (Fang *et al.*, 2014) while grain growth inhibitors include VC, Cr_3C_2, TaC, NbC, TiC, ZrC, HfC and Mo_2C (Konyashin *et al.*, 2014).

Recycling processes for hardmetal can be classified as direct, indirect or semi-direct. Direct recycling transforms the scrap material into a powder that has the same composition as the original material and therefore, high purity powder can only be achieved with high purity scrap (Shemi *et al.*, 2018). Indirect recycling methods do not suffer from limitations of scrap purity and can be used to treat most tungsten-containing products and they are based on chemical treatment of the scrap material to produce intermediate products such as APT (Shemi *et al.*, 2018). Semi-direct recycling refers to selective dissolution of either the binder or the WC phase, while leaving the other phase intact and most methods involve selective leaching of the binder, which leaves the integrity of the WC phase weakened, allowing easy attrition (Shemi *et al.*, 2018). Different methods for hardmetal recycling are presented in **Figure 2**.

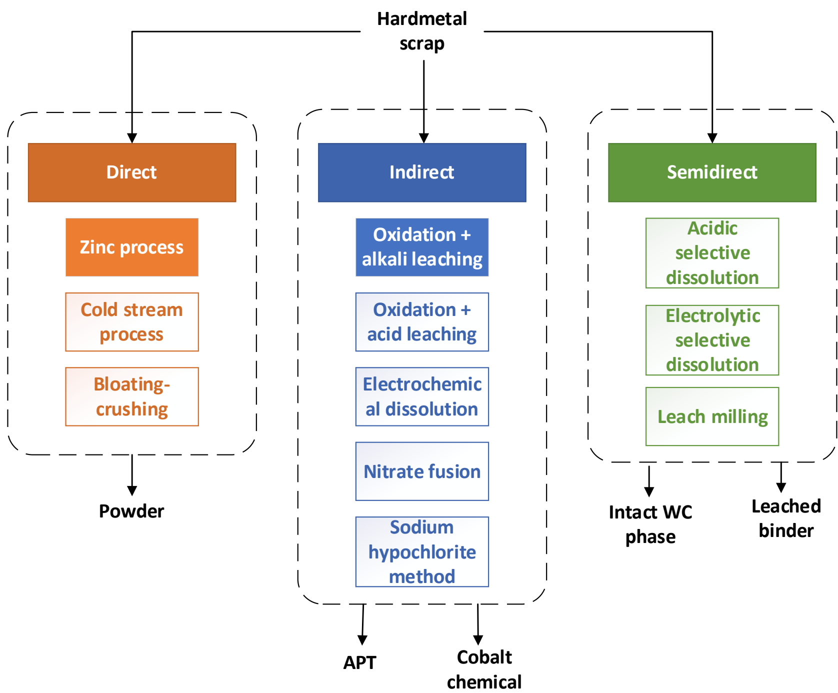

Figure 2 Hardmetal recycling processes and their products, industrially significant processes on darker background.

Direct recycling methods are either pyrometallurgical or mechanical processes (Srivastava *et al.*, 2019). Examples of direct recycling processes studied for recycling of hardmetal scrap include the zinc process (Trent, 1946; Barnard, 1971), the cold stream process (Hartline III *et al.*, 1976) and the bloating-crushing method (Lassner and Schubert, 1999). Of the direct recycling methods, the zinc process is the most widely used and in it, the binder cobalt is separated from the WC phase by molten zinc, which is then evaporated (Shemi *et al.*, 2018). The cold stream process is a mechanical process in which the material is broken into small fractions by forced air and adiabatic expansion cools the air which protects the material from oxidation (Srivastava *et al.*, 2019). In the bloating-crushing method, the raw material is heated to a high temperature and rapidly quenched, which embrittles the hardmetal (Lassner and Schubert, 1999).

Indirect methods include oxidizing pretreatment in combination with alkali (MacInnis *et al.*, 1975; Martin *et al.*, 1981) or acid (Seegopaul and Wu, 1996; Seegopaul and Gao, 2003) leaching, electrochemical dissolution (Latha and Venkatachalam, 1989; Zaichenko *et al.*, 2010), the nitrate fusion method (Bhosale *et al.*, 1990) and the sodium hypochlorite method (Bhosale *et al.*, 1990). Oxidation and leaching methods are based on an oxidizing pretreatment, which transforms tungsten into a soluble form and either alkali or acid leaching can be used to dissolve tungsten or cobalt, respectively (Srivastava *et al.*, 2019). Electrochemical methods involve the oxidation of the hardmetal scrap at the anode to produce tungstic acid and cobalt ions in solution (Latha and Venkatachalam, 1989; Zaichenko *et al.*, 2010). The nitrate fusion process functions by using nitrate salts in a roasting pretreatment that converts WC into sodium tungstate, which is easily dissolved in water (Bhosale *et al.*, 1990). The sodium hypochlorite process treats powdered scrap by leaching in a sodium hypochlorite solution, which transforms tungsten into sodium tungstate (Bhosale *et al.*, 1990).

Semi-direct recycling methods include acidic selective dissolution (Edtmaier *et al.*, 2005; Freemantle and Sacks, 2015), electrolytic selective dissolution (Ghandehari *et al.*, 1982; Malyshev and Hab, 2004) and leach milling (Seo and Kim, 2016). Both acidic selective dissolution and electrolytic selective dissolution solubilise the cobalt binder while leaving the WC phase as a solid dissolution (Ghandehari *et al.*, 1982; Malyshev and Hab, 2004; Edtmaier *et al.*, 2005; Freemantle and Sacks, 2015). Organic acids have been utilised in the selective leaching of the binder, but the reaction kinetics are slow, with leaching taking place over a period of several days (Edtmaier *et al.*, 2005; Freemantle and Sacks, 2015). In leach milling, the hardmetal material is first oxidised followed by simultaneous pulverisation of the material and dissolution of cobalt in acid (Seo and Kim, 2016).

The main industrial processes for recycling hardmetal are direct recycling using the zinc process and chemical recycling (Leal-Ayala *et al.*, 2015). The zinc process is a direct recycling method, whereas chemical recycling is an indirect method (Shemi *et al.*, 2018). Although no reports exist on how much tungsten is recycled by each process, it has been assumed that at least 50% is recycled by the zinc process and the remaining tungsten by the chemical recycling process (Leal-Ayala *et al.*, 2015).

In the zinc process, the hardmetal scrap is brought into contact with molten zinc and the binder metal is separated from the WC as the zinc penetrates the structure (Srivastava *et al.*, 2019). This is followed by evaporation of zinc, which leaves the binder metal cobalt on the surface of the WC phase (Srivastava *et al.*, 2019). The material produced is friable and, once it has cooled down, it can be ground into a powder which has the same composition as the original scrap material and can be used in place of primary material (Shemi *et al.*, 2018). The zinc process can have good recoveries of up to 95% although it does consume considerable amounts of power (Leal-Ayala *et al.*, 2015). It is also focused mostly on the recovery of tungsten, whereas the recovery of other elements such as cobalt is not considered as important even though they do end up in the new material (Shemi *et al.*, 2018).

Where the zinc process and other direct recycling methods are limited by the purity of the scrap material, chemical recycling can be used to treat hardmetal scrap regardless of purity and composition (Shemi *et al.*, 2018). Chemical recycling follows largely the same unit processes as those used in tungsten refining from primary resources but unlike concentrates, hardmetal scrap must first be oxidised in order to achieve a soluble form of tungsten (Zeiler *et al.*, 2021). Following oxidation, however, the material can be treated the same in the primary refining process (Zeiler *et al.*, 2021). Dissolution of tungsten is performed by alkaline pressure digestion using either sodium hydroxide or soda ash, producing a sodium tungstate solution (Shemi *et al.*, 2018). The solution is then purified by means of ion exchange or solvent extraction and the solution is converted into ammonium tungstate solution which can be crystallised as ammonium paratungstate (APT) (Shemi *et al.*, 2018).

Cobalt is left in the leach residue following the alkaline dissolution step in the chemical recycling process (Zeiler *et al.*, 2021). Hydrometallurgical unit processes can be used to extract the cobalt from the leach residue and recover it as a cobalt chemical product (Chandra *et al.*, 2021). Various acids such as HCl, H_2SO_4 and HNO_3 can be used for cobalt dissolution (Chandra *et al.*, 2021). Organic acids have also been investigated as a result of their assumed environmental benefit due to biodegradability and the fact that their use does not release toxic or harmful gas emissions (Srivastava, 2019). However, so far, they have not been used in commercial practice with one issue being the slower dissolution kinetics (Srivastava, 2019). Precipitation and solvent extraction are often utilised to recover the cobalt from the solution (Chandra *et al.*, 2021). Industrially, cobalt can be recovered by the same unit processes used to refine cobalt concentrates and crude cobalt hydroxide (Jervois, 2024). The process is based on leaching in sulphuric acid, precipitation of impurities, solvent extraction and final product recovery by precipitation or crystallisation (Jervois, 2024).

The recycling rate of hardmetal scrap is relatively high, at 46% on average, although it varies somewhat depending on the application with cutting tools having the highest recycling rate, i.e. 55% (Zeiler *et al.*, 2021). For hardmetal used in mining, construction and energy as well as in wear parts and chipless forming, the recycling rate is estimated to be 40% (Zeiler *et al.*, 2021).

2.1.2 Recycling of spent catalysts

Catalysts containing cobalt are used in two primary applications: in the hydrodesulphurisation (HDS) process to remove sulphur from oil and in FT synthesis to turn natural gas into synthetic fuels (Chandra *et al.*, 2022). FT catalysts contain cobalt as the active component along with a support and potentially some promoters but the cobalt content in FT catalysts varies and can be anything from a few percent of cobalt to more than 20% (Chandra *et al.*, 2022). Typical supports in FT catalysts include titanium dioxide, silicon dioxide, aluminium dioxide and carbonaceous material while promoters used can include noble metals, transition metal oxides or alkali and alkaline earth metals (Gholami *et al.*, 2020). HDS catalysts contain a number of valuable metals, often including molybdenum, vanadium, nickel and cobalt along with an alumina support (Chandra *et al.*, 2022). The cobalt content in HDS catalysts is typically lower than in FT catalysts, at only 0-6% (Chandra *et al.*, 2022).

When the activity of a catalyst decreases below an acceptable level it can no longer perform its function in the system and is either regenerated or replaced. Typically, regeneration is possible to a certain extent and for a number of times but eventually the catalyst must be disposed of and replaced, at which point it may become available for recycling (Sun *et al.*, 2015). Both FT and HDS catalysts can be recycled industrially, although the processes and means of cobalt recovery vary.

Spent cobalt FT catalyst can be recycled in the cobalt refining process also used to treat primary raw materials and other secondary raw materials such as cobalt from hardmetal scrap (Jervois, 2024). This is a hydrometallurgical process based on leaching to solubilise the valuable metals (Jervois, 2024). After leaching the solution is purified in iron and copper removal steps using precipitation and solvent extraction is used to remove the remaining impurities (Jervois, 2024). Different cobalt chemical products can be produced from the cobalt-containing solution following solvent extraction, for example by crystallisation or precipitation (Jervois, 2024).

Most investigated processes for cobalt recovery from spent HDS catalysts are based on hydrometallurgy, although several studied methods also follow pyro-hydrometallurgical routes (Chandra *et al.*, 2022). A purely pyrometallurgical process based on calcination and smelting has also been studied (Howard and Barnes, 1991). The different methods are presented in **Figure 3**.

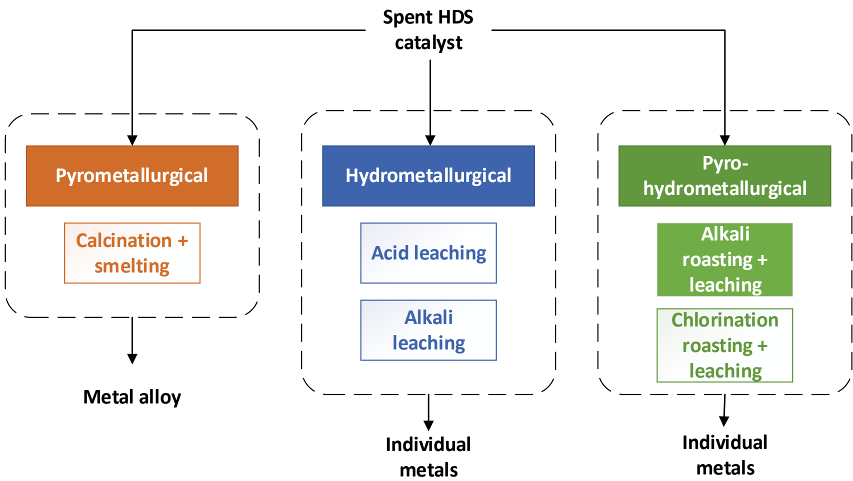

Figure 3 HDS catalyst recycling processes and their products, industrially significant processes on a darker background.

The pyro-hydrometallurgical recycling routes include alkali roasting and chlorination roasting in combination with leaching (Chandra *et al.*, 2022). The solubility of molybdenum and vanadium in water is enhanced by roasting in the presence of an alkali compound (Llanos *et al.*, 1997; Chen *et al.*, 2006). In contrast, chlorination roasting volatilises molybdenum and vanadium, which can be separated by selective condensation while nickel and cobalt are leached in water (Gaballah *et al.*, 1994).

Hydrometallurgical processes may begin with a pretreatment step to clean the catalyst surface and remove volatile species (Valverde *et al.*, 2008). The possible pretreatment can then be followed by either acid leaching (Valverde *et al.*, 2008; Banda *et al.*, 2012; Hamza *et al.*, 2019; Wiecka *et al.*, 2020) or alkali leaching (Hamza *et al.*, 2019; Yaraş and Arslanoğlu, 2020). Unit processes such as solvent extraction and precipitation can then be utilised to further separate and recover the individual metals (Valverde *et al.*, 2008; Banda *et al.*, 2012; Hamza *et al.*, 2019; Wiecka *et al.*, 2020).

Most industrial systems separate molybdenum and vanadium from nickel and cobalt early in the process, after which individual metals can be recovered as pure products, e.g. by precipitation (Llanos and Deering, 1998; Wang *et al.*, 2015). Cobalt, along with nickel, is typically left in the filter cake following the leaching of molybdenum and vanadium and the filter cake can be used in cement manufacturing or nickel refineries or treated in an electric arc furnace (Llanos and Deering, 1998; Wang *et al.*, 2015). Another option could be to leach the nickel- and cobalt-containing leach residue in sulphuric acid (Chen *et al.*, 2006).

The recycling rate of cobalt-bearing catalysts has been considered to be low, at 5%, although this is only an estimate (Sun *et al.*, 2015). In 2020, the total recovery of cobalt from secondary resources was 10 600 t, of which 11% came from catalysts and alloy scrap, with alloy scrap accounting for the majority (Cobalt Institute, 2021).

2.2 Life cycle assessment

LCA is a standardised method that can be used to estimate the environmental impacts of product systems (ISO 14040, 2006; ISO 14044, 2006). It addresses the environmental aspects of a product system, and the potential environmental impacts related to it throughout its life cycle. The life cycle of a product includes everything from raw material acquisition to production and manufacturing, use, recycling and final disposal, and LCA determines both the direct environmental impacts, such as the carbon dioxide emissions associated with them, as well as the indirect impacts that relate to the production of the chemicals used during the production stage, for example.

LCA consists of four phases: goal and scope definition, inventory analysis, impact assessment and interpretation, as depicted in **Figure 4** (ISO 14040, 2006). The workflow is not straightforward moving from one phase to the next but rather iterative, both within the interpretation phase and between the other phases. Initial results provide information for the following iteration to improve data collection as necessary to fulfil the goal of the LCA.

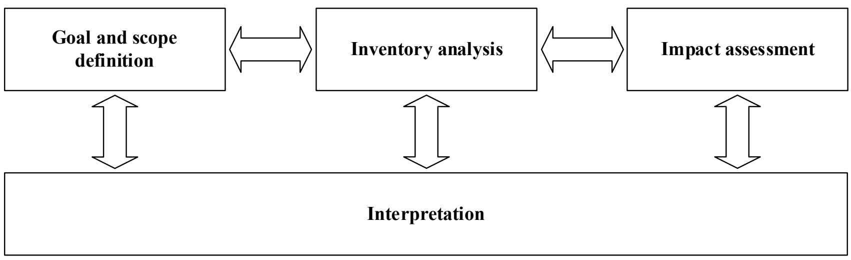

Figure 4. Phases of LCA and their interconnections.

2.2.1 Goal and scope definition

Goal definition involves defining the intended application and audience of the study along with the reasons for carrying out the study. Scope definition addresses the items that define the focus and detail of the study so that they meet the stated goal. The described items include the product system and its functions, the functional unit, the system boundary, allocation methods, impact assessment methodology, interpretation used, data and data quality requirements, assumptions, value choices, limitations, critical review requirements and reporting (ISO 14044, 2006).

The function describes the performance characteristics of the system. The functional unit, on the other hand, provides a reference to which the inputs and outputs of the system are normalised. Any comparisons between products or processes should use an identical functional unit. A reference flow is used to describe the amount of a product necessary to provide the functional unit. The system boundary determines what processes are included in the LCA and needs to be consistent with the stated goal. Any life cycle stage, process, input or output may only be excluded if it does not significantly affect the conclusions that can be drawn from the results. In general, all flows and processes that contribute to the system should be included but in practise it is not possible or efficient use of resources to do so. For less relevant ones, lower quality data or data estimates may be deemed enough, and irrelevant ones may be cut-off entirely if justified to be insignificant to the outcome of the study. The determination of which flows and processes can be cut-off is informed by the iterative nature of LCA and expert judgement.

The data used in the LCA must be of suitable quality to be able to meet the stated goal and scope. The coverage and representativeness of the data related to time, geography and technology should be considered. The uncertainty and variability of the data should also be addressed, and qualitative and quantitative aspects of the data quality should be characterised. For the unit processes that contribute most of the mass and energy flows as well as most of the environmental impact, data concerning the representativeness and specificity is especially important.

2.2.2 Inventory analysis

The initial plan for conducting the inventory analysis phase of the LCA is provided by the goal and scope phase. Inventory data on the inputs and outputs for each of the unit processes included in the system boundary is collected. Collection of the data may involve measurements, calculations and estimations, and details on the data quality should be provided, especially for the unit processes that contribute the most. Computationally, the inventory analysis can be represented by matrices, although in practice it is performed using LCA software.

The basic model of inventory analysis uses matrices to contain the economic and environmental information and vectors to scale it to match the system under study. A unit process can be described by a vector, p, which contains the inventory information. The order of the elements such as kWh of electricity or kg carbon dioxide is fixed. A system consists of multiple unit processes and can be described by process matrix P in Equation 1 (Heijungs and Suh, 2002a):

$$P = (p_1|p_2|\dots p_n) \tag{1}$$

The process matrix along with the unit processes can be divided into two parts. The first represents flows within the economic system and the second environmental flows. The partitioned process matrix can be described by Equation 2 (Heijungs and Suh, 2002a):

$$P = \left(\frac{A}{B}\right), \tag{2}$$

in which matrix A represents economic flows and is called the technology matrix and matrix B represents the environmental flows or environmental interventions of the unit processes and is called the intervention matrix (Heijungs and Suh, 2002a).

In the goal and scope phase of LCA, a reference flow is chosen that fulfils the determined functional unit. A final demand vector, f, can be used to represent the economic flows that correspond to the reference flow. The environmental flows corresponding to the reference flow can be represented by the inventory vector, g. Using the final demand and inventory vectors, the system can be represented by the system vector q, according to Equation 3 (Heijungs and Suh, 2002a):

$$q = \begin{pmatrix} f \\ g \end{pmatrix} \tag{3}$$

A vector, s, with scaling factors, is used to scale the unit processes in the technology matrix to match the final demand vector, according to Equation 4 (Heijungs and Suh, 2002a):

$$s = A^{-1}f \tag{4}$$

As both the technology matrix and the final demand vector are known, Equation 4 can be solved to provide the values of the scaling factors in the scaling vector. Scaling of a unit process affects not only the economic flows but also the environmental flows. Thereby, inventory vector g can be solved from Equation 5 (Heijungs and Suh, 2002a):

$$g = Bs, \tag{5}$$

in which B is the intervention matrix, s is the scaling vector and g is the inventory vector (Heijungs and Suh, 2002a).

2.2.3 Impact assessment

LCIA utilises the information from the inventory analysis to determine the potential environmental impacts of the system. Impact assessment consists of three mandatory elements: selection of impact categories, classification, and characterisation.

The selection of impact categories and the category indicators alongside them should be done so that they comprehensively reflect the environmental issues related to the system. For LCAs involving metals the recommended impact categories include global warming potential (GWP), acidification potential (AP), eutrophication potential (EP), photochemical oxidant creation potential (POCP) and ozone depletion potential (ODP) (Santero and Hendry, 2016). The models behind the characterisation of impacts are specific for each impact category and are of varying maturity. For the recommended impact categories, the maturity of the impact assessment methods is high and the LCI data that contributes to them is typically well available and of decent quality (Santero and Hendry, 2016). Resource depletion categories such as abiotic depletion of elements and abiotic depletion of fossil resources, as well as toxicity categories such as human toxicity have been considered less robust (Santero and Hendry, 2016). However, their inclusion may in some cases be beneficial to avoid bias (Peters *et al.*, 2017).

Classification assigns all the LCI items to the impact categories selected. Different items and emissions affect one or multiple impact categories through different mechanisms. Characterisation involves calculation of the impact category indicator results. It is done by converting the classified LCI results into the units used by each of the impact categories and aggregating the converted results.

Classification and characterisation are both done in practice using LCA software utilising different impact assessment methods such as ReCiPe (Huijbregts *et al.*, 2017), CML (CML, 2016) or TRACI (Bare, 2012). The environmental models in the methods are detailed and complex but the computational structure of impact assessment in LCA is more straightforward as it only utilises the characterisation factors which are the results of these models (Heijungs and Suh, 2002b).

The indicator results can be represented by an impact vector, h, which has as many elements as there are chosen indicator categories. In addition to the impact vector, a characterisation vector, q, is used for each impact category. The characterisation vector contains the information of the characterisation factors provided by the impact assessment methods. A characterisation matrix, Q, can be formed with the characterisation vectors for each of the impact categories, according to Equation 6 (Heijungs and Suh, 2002b):

$$Q = (q_1|q_2|\dots q_n) \tag{6}$$

When characterisation matrix Q and inventory vector g are known, impact vector h can be solved according to Equation 7 (Heijungs and Suh, 2002b):

$$h = Qg \tag{7}$$

2.2.4 Interpretation

The results of the inventory analysis and impact assessment phases are interpreted according to the set goal and scope of the study. The interpretation consists of several elements. Firstly, it should identify the significant issues based on the inventory analysis and impact assessment results. Significant issues may include inventory data, impact categories, or significant contributors to LCI or LCIA results. Secondly, it should evaluate the completeness of the data needed, sensitivity of the results and conclusions to uncertainties or methodological choices, and consistency of the assumptions, methods and data with the goal and scope. Finally, it provides the conclusions, limitations and recommendations of the study. (ISO 14044, 2006)

Contribution analysis is an item that is commonly included in the interpretation phase to investigate which unit processes or inventory items contribute significantly to the total environmental impact of the system. The inventory results can be summarised by Equation 8 (Heijungs and Suh, 2002b):

$$g = BA^{-1}f, \tag{8}$$

in which g is the inventory vector, B is the intervention matrix, A is the technology matrix and f is the final demand vector (Heijungs and Suh, 2002b). The same can also be written as Equation 9 (Heijungs and Suh, 2002b):

$$\forall k:\ g_k = \sum_{\forall j} \sum_{\forall i} b_{kj}\,(A^{-1})_{ji} f_i, \tag{9}$$

where the sum over j relates to aggregation over unit processes, the sum over i to the aggregation over economic flows and k is an environmental flow. For contribution analysis, partial aggregation of the inventories is also possible and the set of unit processes in the system may be partitioned into smaller sets with the most detailed partition being one where each subset only contains one unit process. Thereby, the contribution of processes can be investigated on the desired level. (Heijungs and Suh, 2002b)

The interpretation phase of the LCA should also provide information and try to quantify the uncertainty and variability related to the data and results. The items that most affect the uncertainty may also be identified and, together with information on the largest contributors, used for key issue analysis. Key issue analysis identifies which data to focus on to improve the LCI quality in the following iteration.

2.3 Uncertainty and sensitivity analysis

Numerous sources of uncertainty contribute to the total uncertainty related to the results produced by an LCA model. One classification includes uncertainty in the form of parameter uncertainty, model uncertainty and uncertainty due to choices as well as temporal variability, spatial variability, and variability between objects and sources (Huijbregts, 1998a). Different aspects of parameter uncertainty can include data inaccuracy, data gaps and unrepresentative data (Björklund, 2002). In addition, epistemological uncertainty or lack of knowledge about the system, sheer mistakes and estimation of uncertainty itself can be considered to contribute to the total uncertainty (Björklund, 2002).

The term uncertainty is used in LCA with some variation in the definition, sometimes including and sometimes excluding the concepts of variability and sensitivity (Rosenbaum *et al.*, 2018). They are, however, distinct concepts and should be distinguished from each other. Uncertainty relates to the lack of knowledge of the real value of a quantity while variability is due to inherent variation in the real world and the natural spread of values in a system (Björklund, 2002). Sensitivity refers to how much a change in an input parameter value will change the LCA model result (Rosenbaum *et al.*, 2018).

An LCA model only produces a single impact value output, a point estimate, when variations in inputs are not considered. When uncertainty is considered, the impact values can be represented by a distribution, which allows for a better evaluation of the results in terms of drawing conclusions and recommendations (Rosenbaum *et al.*, 2018). For example, when comparing two options, if the overlap of the distributions is high, it is unlikely that one option is better than the other even though the point estimates would suggest so (Rosenbaum *et al.*, 2018).

Many methods have been used and proposed to address the issue of uncertainty in LCA (Huijbregts, 1998a; Björklund, 2002; Lloyd and Ries, 2007). Different methods are needed to address the different types of uncertainty. Some, such as the variability of measurable parameters, are more straightforward to quantify while others, such as uncertainty related to mistakes or epistemological uncertainty, are more difficult to characterize (Rosenbaum *et al.*, 2018). The treatment of uncertainty can involve both uncertainty analysis, which is the propagation of uncertainty to the outputs, as well as sensitivity analysis, which describes how the input uncertainty influences the output uncertainty (Igos *et al.*, 2018).

Sensitivity refers to how variation in an input parameter or a choice changes the results of the model. The sensitivity of the model to a parameter can be considered to be high when a small change in the parameters leads to a large change in the model result (Rosenbaum *et al.*, 2018). Sensitivity analysis can be performed for both continuous input parameters and discrete choices and it is performed by changing the parameters individually and recalculating the model results (Rosenbaum *et al.*, 2018).

The LCA method requires the practitioner to make a number of choices and assumptions, which introduces some degree of uncertainty into the model results (Heijungs and Lenzen, 2014). These may include choices on how to deal with co-product or EoL multifunctionality, the choice of functional unit, and the chosen time horizon of certain impact indicators, such as global warming (Huijbregts, 1998b). The influence of choices on the model results can be examined using scenario analysis. (Huijbregts, 1998b) Scenario analysis is a type of sensitivity analysis in that it involves making a change in the model and recalculating the model results. It shows how much the model results differ from the baseline according to each choice tested (Igos *et al.*, 2018).

Numerical or sampling methods are methods that utilise computers for repeat calculations. It requires that each input parameter can be described by a distribution and when the model results are calculated, they will vary from run to run (Heijungs and Huijbregts, 2004). With enough samples, the statistical properties can be investigated, including the mean, the standard deviation, the median and 95% confidence intervals (Heijungs and Lenzen, 2014). Monte Carlo simulation is the most commonly used sampling method, although other sampling methods such as the Latin hypercube method have been proposed (Heijungs and Lenzen, 2014). Numerical methods in the form of Monte Carlo simulation are the most commonly applied approach to estimate uncertainty in LCA studies as it has been built into many commercial LCA software tools, making it an easily accessible choice for LCA practitioners (Guo and Murphy, 2012; Heijungs and Lenzen, 2014; Rosenbaum *et al.*, 2018; Igos *et al.*, 2018).

It is also possible to use analytical methods to estimate the effect of uncertainty on the results of an LCA, but their use has not been widespread as they have not been implemented in LCA software (Heijungs and Huijbregts, 2004; Heijungs and Lenzen, 2014). Analytical approaches require less computing time compared to numerical methods but they require more mathematical understanding as they are not generally implemented in commercial software (Heijungs and Lenzen, 2014). They are based on a first order approximation of the Taylor expansion of the model (Heijungs and Huijbregts, 2004). Where numerical methods provide an output probability distribution, analytical methods estimate the mean and standard deviation or variance (Heijungs and Lenzen, 2014).

Semi-quantitative methods can be used to describe the impact of uncertainty associated with the elementary flows that make up the LCI when the information is not available, although they only describe parameter uncertainty (Rosenbaum *et al.*, 2018). This combines the basic uncertainty due to variation and stochastic error of the elementary flow values with an additional uncertainty based on data quality (Rosenbaum *et al.*, 2018). A semi-quantitative pedigree matrix approach has been applied in the LCA database ecoinvent (Ciroth *et al.*, 2016). It evaluates the data using five data quality indicators of reliability, completeness, temporal correlation, geographical correlation and further technological correlation. The data is scored with an indicator score from 1 to 5 for each of the indicators in the matrix which in turn are translated into uncertainty factors which represent a geometric standard deviation (Ciroth *et al.*, 2016).

3. Materials and methods

3.1 Raw materials and process description

Hardmetal scrap (**Publications I** and **II**) and spent catalyst (**Publications III** and **IV**) were selected as the raw materials to be studied in this thesis. The feed material compositions were defined based on the literature. Representative process flowsheets for the recycling of these materials were then built and simulated in HSC Sim 10, using industrially proven hydrometallurgical unit processes and with simulation parameters gathered from the literature.

3.1.1 Hardmetal

The simulated processes in **Publications I** and **II** describe the recycling of cobalt, tungsten, tantalum and niobium from hardmetal scrap. The raw material in **Publication I** is hardmetal scrap, H1, which consists mostly of tungsten carbide cemented in a cobalt matrix. In addition, it contains carbides of chromium, vanadium, tantalum, titanium and niobium in the form of coatings and grain growth inhibitors. The raw material in **Publication II** is the leach residue, H2, which is left once tungsten and cobalt have been leached out in two consecutive leaching steps. The leach residue contains mostly of tantalum and niobium, which are difficult to leach, in addition to the remaining titanium, tungsten and cobalt that were not solubilised in previous leaching steps. The compositions of the two hardmetal raw materials investigated (H1, H2) are provided in **Table 1**.

Table 1. Composition of the hardmetal raw materials (H1, H2) in **Publications I** and **II**

	H1, wt.%		H2, wt.%
Co	11	$Co(OH)_2$	2.3
WC	80.8	$CoWO_4$	0.1
TaC	2.5	WC	26.5
NbC	2.5	WO_3	3.1
TiC	2.5	$WO_3 \cdot H_2O$	2.9
Cr_3O_2	0.5	$NaTaO_3$	21.4
VC	0.2	$NaNbO_3$	19.8
		TiO_2	19.8
		Cr_3C_2	4.2

A block diagram of the two investigated recycling systems for hardmetal raw materials (H1, H2) is presented in **Figure 5**. For industrial chemical recycling of hardmetal scrap the same processes can be utilised as those used for primary tungsten and cobalt refining. Direct leaching of the hardmetal scrap is difficult and therefore the scrap is first oxidised at a high temperature to make the tungsten soluble (Zeiler *et al.*, 2021). The high-temperature treatment oxidises the tungsten and cobalt to form tungsten trioxide, WO_3, and cobalt tungstate, $CoWO_4$ (Yang *et al.*, 2016). The oxidised material is ground to increase the surface area for leaching and after the treatment, tungsten is easily dissolved with sodium hydroxide and forms sodium tungstate, $NaWO_4$ (Yang *et al.*, 2016). Solvent extraction is used to extract tungsten from the solution and stripping with ammonium hydroxide produces ammonium tungstate, $(NH_4)_2WO_4$, which can be crystallised as APT (Wolfe *et al.*, 2014).

An overview of the hardmetal recycling options in addition to chemical recycling is provided in the theoretical background section of the thesis in Section 2.1.1. One of the investigated options is a modification on the chemical recycling process in which leaching with sodium hydroxide is replaced by leaching with ammonium hydroxide (Luidold *et al.*, 2012). The form of WO_3 produced in the traditional high-temperature oxidation step is not soluble in ammonium hydroxide and the oxidation is therefore performed in three stages: a high-temperature oxidation similar to the one in the traditional flowsheet, a reduction step with hydrogen and another oxidation at a lower temperature (Seegopaul and Wu, 1996). After the three-step oxidation, the calcine is subjected to leaching in ammonium hydroxide which leaches tungsten as ammonium tungstate directly, without the need for solvent extraction and tungsten can be crystallised from the solution as APT (Seegopaul and Wu, 1996).

Cobalt is not leached in the alkaline leaching in either sodium hydroxide or ammonium hydroxide and is therefore left in the leach residue (Yang *et al.*, 2016). The cobalt in the leach residue is in the form of cobalt hydroxide (Yang *et al.,* 2016) but must be purified to produce a refined cobalt hydroxide product. The process simulated in **Publication I** achieved this by first leaching the residue in sulphuric acid to solubilise cobalt as cobalt sulphate. The leaching was then followed by removal of titanium from the solution, after which cobalt was precipitated as cobalt hydroxide. This flowsheet shares some unit processes with existing industrial cobalt refining but is not representative. In the compendium the recycling of cobalt from the leach residue by industrial cobalt refining process was simulated. The industrial process starts with leaching in sulphuric acid followed by solution purification steps in the form of precipitation and solvent extraction, after which cobalt can again be precipitated as cobalt hydroxide (Jervois, 2024).

Tantalum and niobium cannot be leached in sulphuric acid, and they therefore remain in the leach residue after cobalt leaching (Xie *et al.*, 2022). The recovery of tantalum and niobium from hardmetal scrap is not industrial practice but flowsheets for their recovery can be modelled after the treatment of columbo-tantalite “coltan” ore, in which both metals appear together. The typical flowsheet for the refining of tantalum and niobium from coltan ore starts with leaching in hydrofluoric and sulphuric acids (Zhu *et al.*, 2011). Tantalum and niobium in the leach residue are in the form of sodium tantalate, $NaTaO_3$, and sodium niobate, $NaNbO_3$, which are not directly soluble in the commonly used acids and must first be roasted to make them soluble (Yang *et al.*, 2013; Ghambi *et al.*, 2021). It has been suggested that acid leaching could be substituted by caustic conversion in sodium hydroxide followed by water leaching, where tantalum and niobium dissolve (Deblonde *et al.*, 2016; Shikika *et al.*, 2020). In both flowsheets the solubilisation of tantalum and niobium is followed by solvent extraction to separate the two metals by co-extracting and selectively stripping them, although different chemicals are necessary depending on whether the leaching is done in acid or water (Shikika *et al.*, 2020; Deblonde *et al.* 2019). Following the separation of the metals by solvent extraction they can be selectively precipitated as hydroxides with ammonia and the hydroxides can be calcined as tantalum pentoxide, Ta_2O_5, and niobium pentoxide, Nb_2O_5 (Shikika *et al.*, 2020).

For this thesis, the processes were simulated using industrially proven unit processes for the raw materials whenever possible and with laboratory-scale experimental data on the behaviour of similar materials in similar conditions. More detailed process descriptions and chemistries, along with the process simulation parameters used, are available in the supporting information of **Publication I** for cobalt and tungsten recovery and the supporting information of **Publication II** for tantalum and niobium recovery.

3.1.2 Catalysts

The raw material, i.e. feed compositions of cobalt catalysts are provided in **Table 2**. These materials (C1, C2) were used in **Publications III** and **IV** to study the recycling of cobalt catalysts. The raw material in **Publication III**, C1, is a FT catalyst and has an active phase of cobalt on a titania, alumina or silica support with manganese and calcium added as promoters. The active phase in the catalyst while in use is metallic cobalt, which turns into oxide or mixed metal oxide upon deactivation. The raw material in **Publication IV**, C2, is a $Mo/Co/\gamma\text{-}Al_2O_3$ catalyst used in petroleum refining with an active phase of molybdenum with an alumina support and cobalt added as a promoter. Upon deactivation the catalyst also contains vanadium and sulphur for example, along with other minor impurities.

Table 2. Composition of the catalyst raw materials (C1, C2) in **Publications III** and **IV**

	C1, wt.%		C2, wt.%
Co_3O_4	20.0	Al_2O_3	55.5
TiO_2	16.0	C	15
Al_2O_3	47.6	S	10
SiO_2	16.0	Mo	7.5
Mn	0.2	V	6
Ca	0.2	Ni	2
		Co	1

		P	1
		SiO_2	2

A block diagram of the two recycling systems investigated for cobalt catalysts is presented in **Figure 6**. Cobalt catalysts used in FT synthesis, C1, can be industrially recycled in the same refining process used to process primary cobalt raw materials and cobalt from hardmetal scrap. The spent catalyst is first solubilised as cobalt sulphate by leaching in sulphuric acid (Jervois, 2024). Precipitation of impurities and solvent extraction are then used to purify the solution and different cobalt products can be produced from the purified cobalt sulphate solution (Jervois, 2024). **Publication III** focused on three different cobalt products: cobalt hydroxide, produced by precipitation with sodium hydroxide, cobalt sulphate, produced by crystallisation from the cobalt sulphate solution, and cobalt carbonate, produced by precipitation with sodium carbonate.

A typical recycling process for the Mo/Co/γ-Al_2O_3 catalyst in **Publication IV** starts with soda ash roasting during which molybdenum forms sodium molybdate, Na_2MoO_4, and vanadium forms sodium orthovanadate, Na_2VO_3 (Llanos and Deering, 1998; Wang *et al.*, 2015). Both sodium molybdate and sodium orthovanadate are highly soluble in water and can therefore be selectively leached, while other elements such as aluminium, cobalt and nickel remain in the leach residue (Wang *et al.*, 2015). Vanadium is first precipitated from the solution as ammonium metavanadate, NH_4VO_3, using ammonium sulphate, $(NH_4)_2SO_4$ (Llanos and Deering, 1998). Ammonium metavanadate is then calcined into vanadium pentoxide, V_2O_5 (Llanos and Deering, 1998). After vanadium precipitation from the solution, molybdenum can be recovered by first converting sodium molybdate with hydrochloric acid to molybdenum chloride, $MoCl_6$ (Park *et al.*, 2006). Neutralisation with ammonium hydroxide converts molybdenum chloride to ammonium molybdate $(NH_4)_2MoO_4$, which can be calcined to molybdenum trioxide, MoO_3 (Park *et al.*, 2006). The cobalt and nickel contained in the leach residue are typically not recovered as pure products on-site but rather fed as secondary raw material to nickel refineries or for alloy production (Llanos and Deering, 1998; Wang *et al.*, 2015). Based on the metals present in the leach residue it is technically possible to recover both cobalt and nickel in the same industrial cobalt refining process used to treat other cobalt-containing raw materials, which was also used for leach residue in hardmetal recycling. Cobalt and nickel along with aluminium are solubilised from the leach residue by leaching in sulphuric acid. Aluminium is removed from the solution by precipitation after which cobalt and nickel are separated by solvent extraction (Sole *et al.*, 2019). Both cobalt and nickel can then be precipitated as hydroxide products using sodium hydroxide.

The processes were simulated using information on existing industrial processes that treat raw materials similar to the ones studied. These included the GCMC soda ash roasting process for Mo/Co/γ-Al_2O_3 catalyst (Llanos and Deering, 1998; Wang *et al.*, 2015) and a hydrometallurgical cobalt refining process (Jervois, 2024). The information was then supplemented by the laboratory-scale experimental data available in the literature. More detailed process descriptions and chemistries along with the process simulation parameters used are available in **Publication III** and its supporting information regarding FT catalyst recycling and in **Publication IV** and its supporting information for the recycling of Mo/Co/γ-Al_2O_3 catalyst.

3.2 Goal and scope definition

3.2.1 Study goals

The reasons for carrying out the study were firstly to provide detailed process-level inventory data on the hydrometallurgical recycling of hardmetal scrap and spent cobalt catalysts and secondly to determine the environmental hotspots of the processes and how the environmental impacts of cobalt production are affected by the chosen process and simulation parameters. The recycling impacts were also compared to the impacts of primary production to determine the potential environmental benefit gained by recycling. The stated goals in each publication were as follows:

- **I.** Study the environmental impacts of recycling cobalt and tungsten from hardmetal scrap by both industrial and proposed chemical recycling processes.
- **II.** Investigate the potential environmental benefit from recovery of tantalum and niobium from hardmetal scrap by methods used to treat columbo-tantalite ore both industrially and on pilot scale.
- **III.** Study the environmental impacts of recycling cobalt from spent FT catalyst by means of industrial cobalt refining.
- **IV.** Investigate the potential environmental benefit from recycling not only molybdenum and vanadium but also cobalt and nickel as pure intermediate products from spent Mo/Co/γ-Al_2O_3 catalyst.

The intention is for the results of the study to be used by LCA researchers and practitioners as well as metallurgical experts and stakeholders within the related industries. All the information in the thesis is available publicly with the studies published in open access journals that have been reviewed externally in the journals' peer review process.

3.2.2 System boundaries and scenarios

The technical boundary of the studied recycling processes was gate-to-gate. The boundary included the solubilisation of the recovered metals, solution purification, separation of the recovered metals if necessary and recovery of the metals as intermediate products. High-temperature pretreatment was included in the recycling systems studied in **Publications I, II** and **IV,** while in **Publication III** pretreatment of the spent catalyst was left outside the technical boundary as it was assumed to take place at the use site. The technical boundary for the hardmetal material (H1, H2) recycling systems is shown in **Figure 5** and the boundary for catalyst material (C1, C2) recycling systems in **Figure 6**.

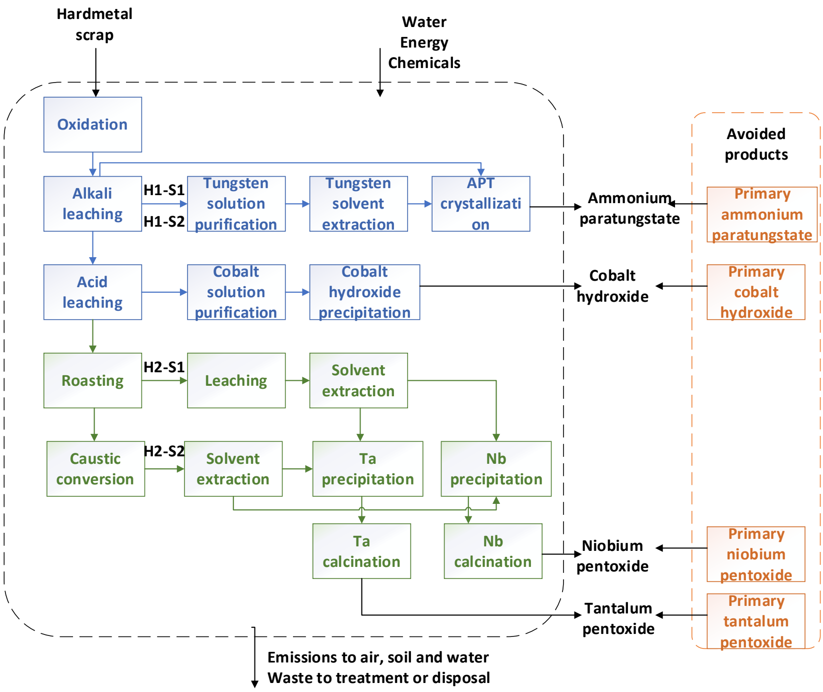

Figure 5. System boundary of the hardmetal recycling systems with recycling of cobalt and tungsten in blue and recycling of tantalum and niobium in green. H1-S1 refers to typical chemical recycling, and H1-S2 to the development-stage scenario. H2-S1 is based on industrial HF leaching and H2-S2 is a proposed HF-free process.

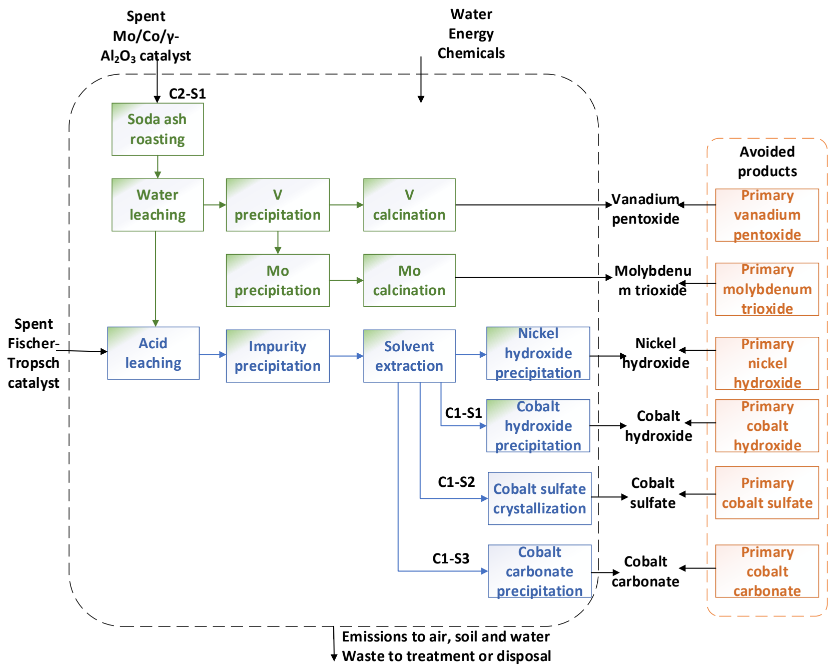

Figure 6. System boundary of the catalyst recycling systems with the cobalt refining process in blue and additional units for Mo/Co/γ-Al_2O_3 catalyst recycling in green. Blue-green units are common for both raw materials. C2-S1 refers to the Mo/Co/γ-Al_2O_3 catalyst recycling process. C1-S1, C1-S2 and C1-S3 refer to cobalt recycling from FT catalyst as hydroxide sulphate and carbonate, respectively.

The geographical boundary used in the studies was Europe and European activities were used for the background processes whenever possible. In the absence of activities modelled for the European region, global values were used. Of the studied processes, chemical recycling of hardmetal, H1, and spent FT catalyst, C1, are currently recycled industrially to produce cobalt products. Chemical recycling in Europe is performed in the tungsten refining process in Germany and cobalt refining is assumed to be conducted in Finland. The cobalt refining industry in Finland also uses spent catalyst as raw material.

Only selected metals contained in the studied secondary raw materials were recovered in the recycling processes and considered as products in the LCAs. The recovered metal products in **Publications I-IV** and in the compendium are provided in **Table 3**. The other metals contained in the hardmetal scrap and spent catalyst compositions (detailed in **Tables 1** and **2)** were simulated to end up in solid residue streams and considered as waste in the LCAs.

Table 3. Summary of the recycling scenarios for different raw materials (hardmetal scrap and spent catalyst) and products in **Publications I-IV** and in the compendium

Process	Feed	Products	Abbr.	Publ.
Hardmetal scrap				
Chemical recycling	H1	APT, $Co(OH)_2$	H1-S1	I
NH_4OH leaching	H1	APT, $Co(OH)_2$	H1-S2	I

Chemical recycling, industrial cobalt refining	H1	APT, $Co(OH)_2$	H1-S3	Compendium
HF leaching	H2	Ta_2O_5, Nb_2O_5	H2-S1	II
Caustic conversion	H2	Ta_2O_5, Nb_2O_5	H2-S2	II
Spent catalyst				
Co refining	C1	$Co(OH)_2$	C1-S1	III
Co refining	C1	$CoSO_4$	C1-S2	III
Co refining	C1	$CoCO_3$	C1-S3	III
Soda ash roasting, Co refining	C2	MoO_3, $Co(OH)_2$, $Ni(OH)_2$, , V_2O_5	C2-S1	IV

The definition of scenarios by nature in LCA introduces uncertainty and bias into the study when it is not only focused on an existing industrial process with primary data. In other cases, uncertainty is present and related to e.g. technological advancements and industry operation. Choices in how the scenarios are defined may therefore affect the results of the study. Multiple different scenarios can in these cases be explored to incorporate the uncertainty.

In this thesis, several different production processes and technologies were studied for the production of cobalt chemicals from secondary raw materials. Different raw materials and products were also explored. The simulated recycling systems were based on both current industrially operated processes as well as some proposed processes still in development phase. The simulated industrial systems are representative of industrial operation while the development stage processes are either relatively small changes to industrial flowsheets or based on pilot scale data of continuous operation and therefore also representative of plausible operation.

The studies assume a current time frame and the largest difference in the environmental impacts in the future apart from technological advancement is likely to be due to the changing electricity mix. The studies use the current Finnish electricity mix which is already relatively clean and therefore the effect of decarbonization of the electricity mix in the coming decades is assumed to be relatively small for the direct electricity consumption.

3.2.3 Functional units and multifunctionality

The functional unit provides a reference for the normalisation of the system inputs and outputs. The function of the systems studied in **Publications I-IV** was considered to be the recycling of scrap material and therefore the functional units were formulated on the input side rather than based on metal output. As the functional units were formulated on a mass basis, they were also considered as the reference flows. In **Publication I** the functional unit was defined as the treatment of 1 t hardmetal scrap and in **Publication II** it was defined as the treatment of 1 t of leach residue. In **Publications III** and **IV** the functional unit was defined as the treatment of 1 t spent catalyst.

Publications I, II and **IV** dealt with systems that produced multiple products. **Publication III** only recycled cobalt and therefore did not deal with co-product multifunctionality. Scenario analysis was used to assess the effect on the interpretation of the LCIA results caused by different methods of dealing with co-product multifunctionality when necessary. Several allocation keys were included in the analysis when it was unclear whether the produced products were by- or co-products. Economic value was included in the analysis when the price difference between two or more the co-products was considered substantial.

The impacts described in **Publication I** were allocated using the mass of the product, the mass of the metal content in the product and the economic value of the product as the allocation keys. In addition, the allocation was performed on both the subdivided and the unsubdivided systems. Based on the results, subdivision was performed in the later publications when allocation was necessary. In **Publication II**, the allocation was made based on the economic value of the co-products as it was assumed that interest in the recovery of the elements would be driven by demand for the more valuable tantalum. In **Publication IV**, the allocation was made using the mass of the metal content in the products and the economic value of the products as allocation keys.

The environmental impacts of the recycling processes were compared to the impacts of the primary production of equivalent products. This was done in **Publications I, II** and **IV** both on an individual metals basis after allocation and on a system level using system expansion. A simple cut-off was used as the EoL allocation method. Since metals are infinitely recyclable and the hydrometallurgical processes studied produce products of a quality equal to primary production, the recycled products were considered to substitute primary products with a 1:1 ratio.

The functional units and modelling choices related to multifunctionality are presented in **Table 4** for all the publications.

Table 4. Functional units and choices regarding multifunctionality in **Publications I-IV**

	Functional unit	**Subdivision**	**Allocation**	**EoL allocation**
I	1 t hardmetal scrap	Subdivided and unsubdivided system	Product mass, metal mass, economic value	Simple cut-off
II	1 t leach residue	Subdivided system	Economic value	Simple cut-off
III	1 t spent FT catalyst	n/a	n/a	Simple cut-off
IV	1 t Mo/Co/γ-Al_2O_3 catalyst	Subdivided system	Metal mass, economic value	Simple cut-off

3.3 Inventory analysis

In the absence of primary data on the inputs and outputs in the studied processes, process simulation was used to assist in the inventory analysis phase.

Flowsheets and scenarios were formulated based on a literature analysis after which the processes were simulated in HSC Sim. The modelling of hydrometallurgical processes in HSC Sim is based on chemical reactions. In addition to the reactions, parameters such as reaction extent, process conditions and distribution between phases are defined for each of the unit processes in the simulation. Values for the process parameters were defined based on literature information and details for each simulated process are available in each of **Publications I-IV**.

The electricity consumption by equipment in the simulation was not calculated by HSC Sim. Estimated electricity consumptions of the simulated processes were calculated based on the mass balances in the simulations with the methodology detailed by Elomaa *et al.* (2020a), Rinne *et al.* (2021a) and Rinne *et al.* (2021b). The throughput was scaled to industrial volumes and dimensions calculated for the main equipment: reactors, thickeners and filters. The methodology was used to provide an estimate in **Publications I-IV,** although **Publications III** and **IV** also utilised information from two environmental impact assessments (EIA) by cobalt producers (Umicore, 2022; Jervois, 2023) to refine the estimate.

Background data describing the production of the chemicals and utilities consumed by the studied processes was mostly obtained from ecoinvent LCA database versions 3.7 (**Publication I**), 3.8 (**Publications II and III**) and 3.10 (**Publication IV and compendium**) as well as GaBi Professional 2020 database (**Publication I**). The ecoinvent databases were also used as the source for the environmental impacts of primary production with a few exceptions. In **Publication II** the production of tantalum and niobium from coltan ore was simulated using the recycling model based on HF leaching. In **Publication III** the environmental impacts of primary cobalt sulphate production were gathered from the results of an LCA by the Cobalt Institute (2023c). In **Publication IV** the production of V2O5 was modelled based on the LCI by Weber *et al.* (2018).

3.3.1 Hardmetal recycling inventories

The inventories for hydrometallurgical processing of hardmetal scrap in the systems studied are provided in **Table 5**. The process consists of oxidation of the hardmetal scrap (H1), alkali leaching of tungsten, solvent extraction (not in the H1-S2 scenario) and crystallisation of APT to recover tungsten. Cobalt is recovered from the alkali leach residue by leaching in sulphuric acid and is recovered as cobalt hydroxide by precipitation. Tantalum and niobium are extracted from the cobalt leach residue (H2) by roasting followed by either leaching (H2-S1) or caustic conversion (H2-S2) and recovered as oxides by precipitation and calcination. The parameters used in the process simulations were mainly based on publicly available laboratory-scale studies, pilot-scale studies and patents. Process simulations and LCIs on unit process level are presented in **Publication I** for cobalt and tungsten recovery and in **Publication II** for tantalum and niobium recovery. Simulation of the H1-S3 scenario has not been published earlier and is presented for the first time in this compendium.

Table 5. Compiled LCI for hardmetal recycling scenarios, per functional unit. * refers to new scenario investigated in this compendium.

Input	**Unit**	**H1-S1**	**H1-S2**	**H1-S3***	**H2-S1**	**H2-S2**
Scrap	t/t	1	1	1	-	-
Leach residue	t/t	-	-	-	1	1
Air	t/t	2.87	3.10	2.87	-	-
Hydrogen	t/t	-	0.006	-	-	-
Ammonium hydroxide	t/t	0.60	1.05	0.58	-	-
Hydrogen peroxide	t/t	0.04	0.04	0.04	-	-
Magnesium chloride	t/t	0.07	-	0.07	-	-
Ferric chloride	t/t	0.04	0.04	0.04	-	-
Heat	MJ/t	18 200	19 900	19 300	7 800	1 300
Electricity	MJ/t	13 400	19 600	16 800	32 500	26 300
Organic	t/t	1.13	-	1.26	30.6	54.0
Water	t/t	21.1	19.0	23.2	145	108
Sodium hydroxide	t/t	0.97	0.69	0.75	0.40	10.0
Sulphuric acid	t/t	1.93	2.75	1.84	11.8	-
Lime	t/t	0.14	0.40	0.18	19.5	-
Carbon dioxide	t/t	-	-	0.07	-	-
Bisulphate	t/t	-	-	-	0.74	0.74
Ammonia	t/t	-	-	-	2.76	0.31
Carbon dioxide	t/t	-	-	0.070	5.1	5.4
Hydrogen fluoride	t/t	-	-	-	5.69	-
Oxalic acid	t/t	-	-	-	-	0.49
Nitric acid	t/t	-	-	-	-	0.55
Ammonium nitrate	t/t	-	-	-	-	0.57
Output						
Carbon dioxide	t/t	0.21	0.21	0.21	-	-
Ammonia	t/t	0.03	0.03	0.03	-	-
Vapour	t/t	3.09	6.29	2.89	7.77	20.6
Solid waste	t/t	2.44	1.75	3.83	50.0	3.1
Wastewater	t/t	17.0	12.5	21.1	140	121
APT	t/t	0.97	0.94	0.97	-	-
$Co(OH)_2$	t/t	0.16	0.16	0.16	-	-
Ta_2O_5	t/t	-	-	-	0.16	0.085
Nb_2O_5	t/t	-	-	-	0.099	0.10

3.3.2 Catalyst recycling inventories

The inventories for the systems studied for cobalt catalyst recycling are provided in **Table 6**. Spent FT catalyst (C1) is recycled in a cobalt refining process consisting of sulphuric acid leaching, solution purification steps and recovery by precipitation or crystallisation, depending on the desired cobalt product. Spent Mo/Co/γ-Al_2O_3 catalyst (C2) is first treated by soda ash roasting, followed by water leaching and recovery of molybdenum and vanadium by precipitation and calcination. Leach residue from the water leaching containing cobalt and nickel is then treated in the same cobalt refining process as the spent FT catalyst. The parameters used in the process simulations were mainly based on publicly available laboratory- and pilot-scale studies supplemented by EIA information on the cobalt refining units. The process simulations for the raw materials are presented in detail in **Publication III** for spent FT catalyst and in **Publication IV** for spent Mo/Co/γ-Al_2O_3 catalyst.

Table 6. Compiled LCI for catalyst recycling scenarios, per functional unit

Input	**Unit**	**C1-S1**	**C1-S2**	**C1-S3**	**C2-S1**
Spent catalyst	t/t	1	1	1	1
Sulphur dioxide	t/t	0.0016	0.0016	0.0016	-
Sodium carbonate	t/t	-	-	0.23	-
Flocculant	t/t	0.031	0.031	0.031	-
Extractant	t/t	0.0038	0.0038	0.0038	-
Kerosene	t/t	0.015	0.015	0.015	0.075
Lime	t/t	0.15	0.55	0.14	0.098
Sodium hydroxide	t/t	0.81	0.49	0.66	0.19
Carbon dioxide	t/t	0.032	0.028	0.025	0.075
Water	t/t	7.9	14	8.0	25
Sulphuric acid	t/t	1.7	2.2	1.7	0.78
Hydrogen peroxide	t/t	0.0096	0.0096	0.0096	0.0062
Electricity	MJ/t	3200	3400	3200	13 000
Heat	MJ/t	2900	2900	2800	12 600
Air	t/t	0.019	0.019	0.019	3.1
Soda ash	t/t	-	-	-	0.17
Ammonium hydroxide	t/t	-	-	-	0.22
Hydrochloric acid	t/t	-	-	-	0.23
Output					
Vapour	t/t	0.032	6.6	0.032	-
Solid waste	t/t	4.9	5.8	4.9	0.60
Wastewater	t/t	6.6	4.8	6.7	25
Carbon dioxide	t/t	-	-	-	0.61
Ammonia	t/t	-	-	-	0.0044
Cobalt product	t/t	0.25	0.71	0.32	0.012
Molybdenum trioxide	t/t	-	-	-	0.11
Vanadium pentoxide	t/t	-	-	-	0.10
Nickel hydroxide	t/t	-	-	-	0.022

3.4 Impact assessment

ReCiPe 2016 was used as the impact assessment method for the classification and characterisation of the impacts in all **Publications I-IV**. The optional steps of weighing and normalisation were excluded.

The impact categories evaluated in the publications were global warming potential (GWP, kg CO_2-eq.), acidification potential (AP, kg SO_2-eq.), eutrophication potential (EP, kg P-eq.), ozone depletion potential (ODP, kg CFC-11-eq.), photochemical oxidant creation potential (POCP, kg NO_x-eq.) and freshwater consumption (FC, m^3). The choice of impact categories was guided by the recommendation of Santero and Hendry (2016) for metal and mining industry LCA studies. The chosen impact categories were deemed to sufficiently describe the different impacts on air, water and soil.

ReCipe 2016 groups certain sources of uncertainty and choices into three perspectives: individualistic, hierarchist and egalitarian and these value choices impact the derivation of characterisation factors as the perspectives take different views on time horizon, impact pathways and adaptation potential (Huijbregts *et al.*, 2016). Of the considered impact categories the choice is relevant to GWP and ODP, and the hierarchist perspective, which is based on the scientific consensus of the time horizon and impact mechanisms, was used in the assessment.

3.5 Interpretation

The interpretation phase in LCA is important for deriving robust and reliable conclusions and recommendations from the results of the LCI and LCIA phases. The results as a whole are analysed and considered in relation to the data quality in terms of accuracy, completeness and reliability, and any assumptions made. Interpretation is performed in relation to the intended application to answer the questions posed in the goal and scope definition phase.

Significant issues in the form of key unit processes, process parameters and assumptions were identified and assessed in the publications by means of contribution analysis, sensitivity analysis and uncertainty analysis. Contribution analysis was used to identify the main contributors to the LCIA results, including the most relevant unit processes within the simulated recycling processes and the most relevant life cycle stages in the systems expanded to include primary production. The choices with potential to influence the LCIA results were evaluated using scenario analysis.

The sensitivity of the LCIA results to individual process simulation parameters as well as individual chemical, utility and emission streams in the LCA model was assessed in the publications by different methods. In **Publications I** and **II** the sensitivity of the results to process simulation parameters was assessed, while the LCA model was the focus of **Publication III** and the results of both analyses were compared in **Publication IV**. The sensitivity analyses involved changing the values of the process simulation parameters or the streams in the LCA model individually by ±20% and recalculating the LCIA results.

The influence of parameter uncertainty on the conclusions and recommendations formulated based on the LCIA results was investigated by allowing the parameter values to vary within limits determined by uncertainty estimates. Different methods of estimating the level of uncertainty were studied in **Publications I, III** and **IV**. All three publications utilised data quality indicators (DQI) with expected levels of accuracy that were assigned to the values of the chemical, utility and emission streams in the system. The expected levels of accuracy were based on the cost estimate classification system of the American Association of Cost Estimation and modified for LCA by Schreiber *et al.* (2016). In addition to the DQIs, the pedigree matrix used by Ciroth *et al.* (2016) for the ecoinvent 3 LCA database was utilised in **Publication IV** to estimate the effect of uncertainty associated with the LCA model on the results and conclusions of the study. The uncertainties estimated in **Publications III** and **IV** for the LCA model were also used to perform a Monte Carlo simulation. In addition to estimating the impact of the uncertainty originating from the LCA model, **Publication IV** estimated the influence of the uncertainty from the process simulation parameters. DQIs were assigned to the process simulation parameters and a Monte Carlo simulation was performed. The minimum and maximum values found were used in the LCA model to perform a Monte Carlo simulation to determine the influence of the process simulation parameter uncertainty on the LCIA results.

4. Results and discussion

4.1 Impact assessment

The environmental impacts of recycling hardmetal scrap (H1, H2) and cobalt-containing catalysts (C1, C2) are presented in **Figure 7**. Three scenarios are shown for hardmetal recycling. In the first (H1-S1+H2-S1), tungsten and cobalt are recycled by an industrial chemical recycling process, and tantalum and niobium are recycled by an HF leaching process based on industrial practices. In the second (H1-S2+H2-S2), tungsten and cobalt are recycled by a development-stage chemical recycling process and tantalum and niobium are recycled by a development-stage process based on caustic conversion. In the third (H1-S3+H2-S1), tungsten is recycled by an industrial chemical recycling process, cobalt by an industrial cobalt refining process (new unpublished results presented in this compendium), and tantalum and niobium by an industrial HF leaching process. Three scenarios are shown for cobalt recycling from FT catalyst in a cobalt refining process: (i) C1-S1, in which cobalt is recovered as cobalt hydroxide, (ii) C1-S2, in which cobalt is recovered as cobalt sulphate and (iii) C1-S3, in which cobalt is recovered as cobalt carbonate. Finally, one scenario (C2-S1) is shown for molybdenum, vanadium, nickel and cobalt recycling from a Mo/Co/γ-Al_2O_3 catalyst. The contribution to the environmental impacts of the recovery processes for the different metals, pretreatment steps such as oxidation and first leaching to separate metals, and abatement and effluent treatment steps are presented below for each of the scenarios.

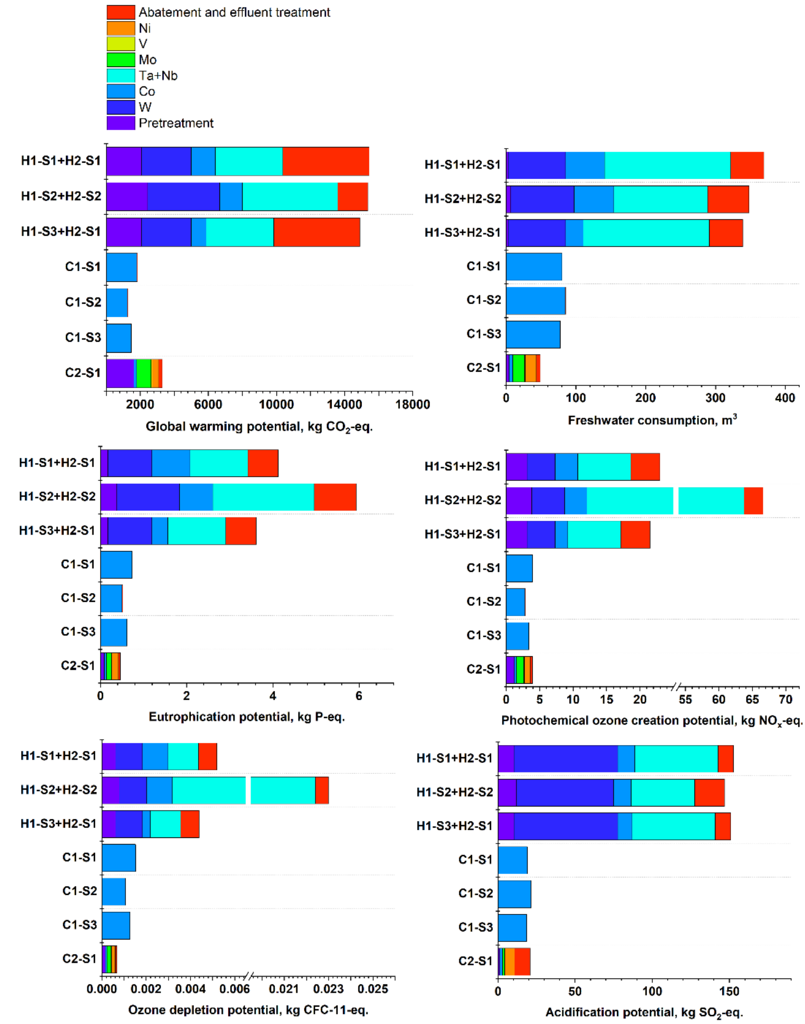

Figure 7 Environmental impacts of hardmetal scrap and spent cobalt catalyst recycling. Functional units: 1 t of hardmetal scrap (H1-S1+H2-S1, H1-S2+H2-S2 and H1-S3+H2-S1), 1 t of cobalt FT catalyst (C1-S1, C1-S2 and C1-S3) and 1 t Mo/Co/γ-Al_2O_3 catalyst (C2-S1).

In the hardmetal scrap chemical recycling processes, the contribution of the different process steps to the environmental impacts varies, depending on the scenario and impact category. The environmental impacts of tungsten recycling are larger than those of cobalt recycling. Tungsten recycling contributes 5.4-44.6% of the total environmental impact of the system depending on the scenario and impact category, whereas the contribution of cobalt recycling is 5.0-22.3%. For primary production of hardmetal as well, previous research by Furberg *et al.* (2019) and Ma *et al.* (2017) have found the hydrometallurgical production of APT to be a major contributor. For APT, the primary and secondary production processes are equivalent. Tungsten recycling only contributes less than 10% of the impacts in the H1-S2+H2-S2 development-stage scenario for POCP and ODP, which are dominated by tantalum and niobium recycling. The recycling of tantalum and niobium from the leach residue following cobalt recovery is a large contributor in the other impact categories as well. All in all, it contributes 26.3-83.6% of the total environmental impact. Pretreatment contributes 0.8-15.8% of the impacts and is most pronounced in GWP (13.3-15.8%) and POCP (5.7-14.6%). Abatement and effluent treatment steps contribute 2.6-34.0%, mostly to GWP (11.6-34.0%) in the two industrial scenarios. This is mostly due to the abatement and effluent treatment necessary in the recycling of tantalum and niobium by HF leaching.

The calculated environmental impacts of the different hardmetal recycling processes vary somewhat from each other. Regarding tungsten and cobalt recovery, the industrial process with the updated cobalt refining flowsheet in the H1-S3 scenario shows the smallest impacts in most impact categories. However, in some impact categories, such as POCP, the difference is negligible particularly between the two industrial scenarios H1-S1 and H1-S3. The development-stage chemical recycling process modelled in scenario H1-S2 has larger impacts than the other scenarios, particularly for GWP and EP. Concerning the recycling of tantalum and niobium also, the difference between all scenarios is quite small for GWP, FC and AP. The combined development-stage scenario H1-S2+H2-S2 shows higher impacts than the industrial scenarios for EP, POCP and ODP. The difference is especially large for POCP and ODP due to the oxalic acid consumed in the solvent extraction stage to separate tantalum and niobium in the development-stage recycling process.

The environmental impacts of tantalum and niobium recycling are dependent on the content of the metals in the hardmetal scrap. This is illustrated in **Figure 8**, which shows the environmental impacts of tantalum and niobium recycling by the H2-S1 scenario based on industrial HF leaching compared to the environmental impacts of the primary production of equivalent products with varying tantalum and niobium content in the hardmetal scrap. The results in **Figure 7** were calculated according to the tantalum and niobium content given in **Table 2**, which was 2.5 wt.% each, 5 wt.% total. As the content of the two metals in the hardmetal scrap increases, the environmental impacts are substantially decreased.

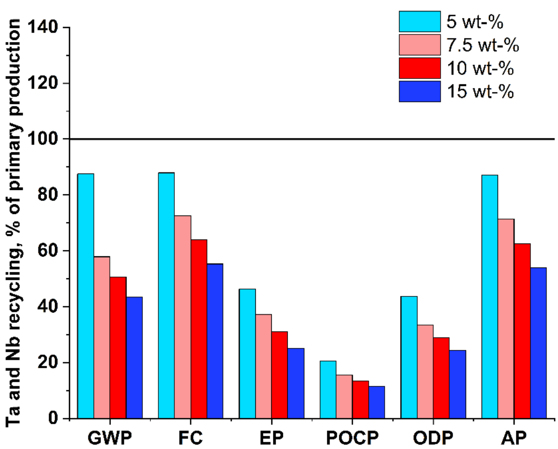

Figure 8 Environmental impacts of tantalum and niobium recycling by HF leaching (H2-S1) with different tantalum and niobium total content (wt.%) in hardmetal scrap compared to environmental impacts of primary production of equivalent products. 100% represents the level of environmental impacts from primary production. (Adapted from **Publication II** © 2023 The authors, reproduced from ACS Sustainable Chemistry & Engineering under CC BY 4.0).

In the recycling of FT catalyst in scenarios C1-S1, C1-S2 and C1-S3, only cobalt is recycled from the spent catalyst. Therefore, the environmental impacts presented in Figure 7 for the scenarios are only divided between cobalt recycling and abatement and effluent treatment. The majority of the impacts originate in the cobalt recycling process itself, whereas the contribution of the abatement and effluent treatment steps is low, i.e. 0.2-3.7% of the total impacts. The total impacts are in a similar range for all scenarios. The largest difference can be found in GWP, EP, POCP and ODP, where the impacts of recycling as cobalt hydroxide are 27-30% and 13-19% larger than those of recycling as cobalt sulphate and cobalt carbonate, respectively. The difference in FC and AP is smaller, with recycling as cobalt sulphate having 7-13% higher impacts than recycling as cobalt hydroxide or cobalt carbonate.

In the recycling of Mo/Co/γ-Al_2O_3 catalyst in the C2-S1 scenario, molybdenum, vanadium, nickel and cobalt were recycled. The largest contributing process step varied somewhat depending on the impact category. Molybdenum contributed only 6.6% to AP but 25.3-32.9% to all other impact categories. Similarly, nickel contributed only 11.8% to GWP but 21.4-32.6% to the other impact categories. Pretreatment contributed heavily to GWP, EP, POCP and ODP with contributions ranging from 18.3% to 49.2%. The abatement and effluent treatment steps only have a large contribution to AP with 48.6%. Cobalt and vanadium contributions are relatively small, with cobalt contributing 4.9-12.2% of the total environmental impact depending on the impact category and vanadium contributing 0.6-3.6%. The recycling of Mo/Co/γ-Al_2O_3 catalysts has also been studied by Benavides *et al.* (2017), who reported the GWP of petrochemical and biorefinery catalyst recycling as 3.8 kg and 1.0 kg CO_2-eq./kg spent catalyst, respectively. Considering the differences in the studied processes, the results presented in this thesis are in line with the previous work, with GWP ranging from 2.7 to 4.0 kg CO_2-eq./kg spent catalyst.

4.2 Contribution analysis

The contribution to the environmental impacts of direct emissions as well as the upstream impacts of the production of the chemicals and utilities consumed by the processes is shown in **Figure 9** for each of the scenarios described in **Table 3**. The largest contributors varied depending on the system, scenario and impact category. Apart from a few exceptions, the contribution of direct emissions was small or non-existent and the majority of the impacts originated upstream in the production of the chemicals and utilities consumed by the processes.

Direct emissions were large contributors in the hardmetal recycling scenarios (H1-S1, H1-S2 and H1-S3) and the Mo/Co/γ-Al_2O_3 catalyst recycling scenario (C2-S1). In the hardmetal recycling scenarios the contribution was due to ammonia emissions from APT production which contributed substantially to AP. In the Mo/Co/γ-Al_2O_3 catalyst recycling scenario the contribution was from carbon dioxide emissions from soda ash roasting to GWP and from ammonia emissions from the scrubbing of calcination offgas to AP. The simulation parameters controlling the gaseous emissions were in the sensitivity and uncertainty analyses treated as more uncertain than the parameters controlling e.g. reagent consumption due to the lack of representative data.

Electricity was a large contributor to many of the scenarios, particularly the hardmetal recycling scenarios for tungsten and cobalt (H1-S1, H1-S2 and H1-S3) and the Mo/Co/γ-Al_2O_3 catalyst recycling scenario (C2-S1). The used electricity production mix represents electricity consumption in Finland, which is the largest cobalt producer in Europe. The impacts originating from the production of the electricity consumed in the processes is dependent on the production mix, which are highly regional. Some regional production mixes such as in Norway and Sweden are have mostly lower impacts compared to the Finnish one while the impacts of the average European production mix are higher. For the scenarios where electricity was a major contributor (H1-S1, H1-S2, H1-S3 and C2-S1), changing the production mix from Finnish to average European does increase the impacts of the studied processes but does not affect the conclusions drawn from the results regarding recycling benefit.

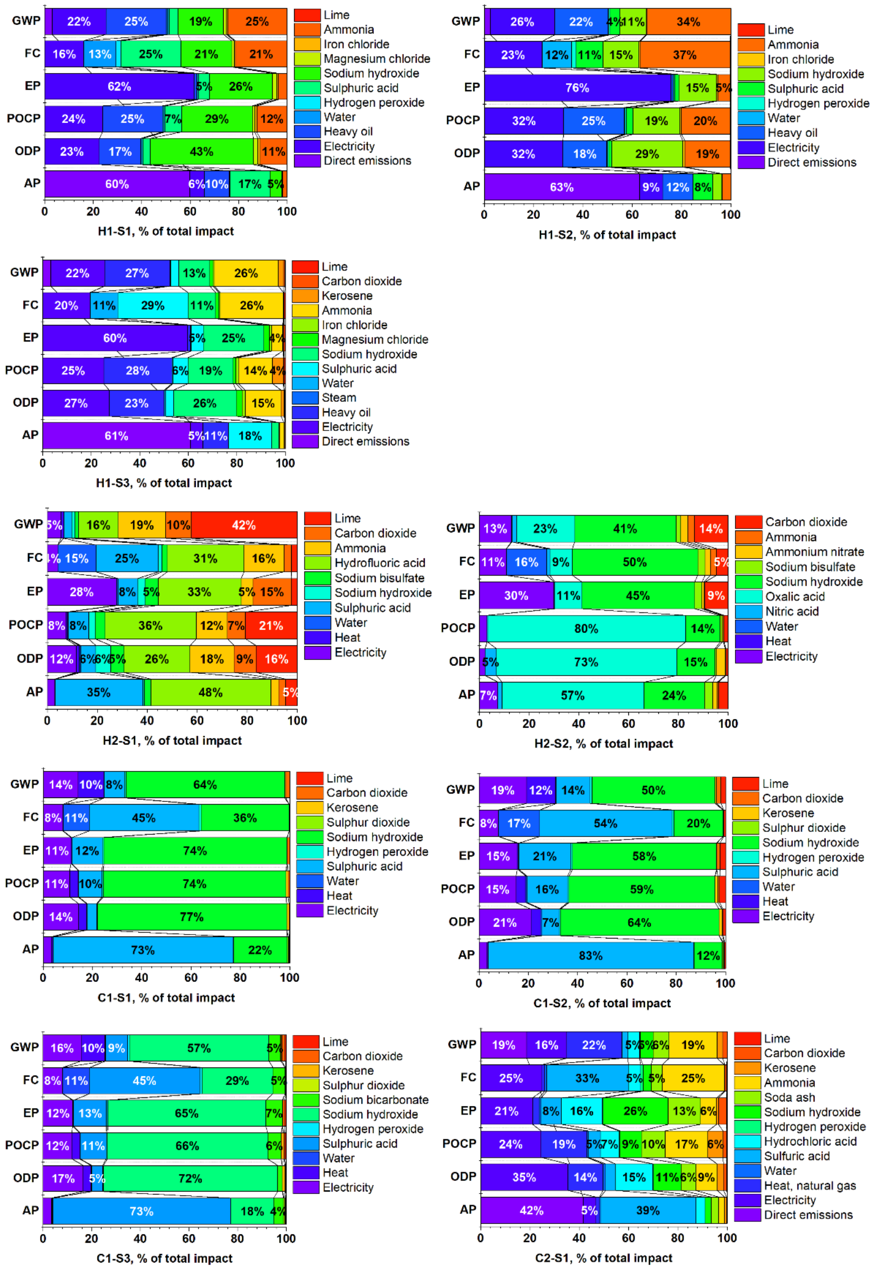

Figure 9 Contribution of direct emissions as well as chemical and utilities consumption to the environmental impact of different hardmetal scrap and spent cobalt catalyst recycling processes.

The results of the hardmetal recycling scenarios for tungsten and cobalt (H1-S1, H1-S2 and H1-S3) are all quite similar. Direct emissions only contribute substantially to AP, where they are the dominant contributor (60-63%). The direct emissions in this case are ammonia emissions from APT production, which were also identified by Furberg *et al.* (2019) as a major contributor to AP in the non-Chinese primary production of hardmetal. By far the largest contributor to EP

(60-76%) is electricity, which also contributes substantially to GWP, FC, POCP and ODP (16-32%). Ma *et al.* (2017) also identified electricity as the major contributor in the Chinese primary production of hardmetal. Another notable contributor is heating, which is mainly required in the oxidation of hardmetal scrap and contributes 17-28% to GWP, POCP and ODP. Ammonia is used in the H1-S1 and H1-S3 industrial scenarios in tungsten solution purification and tungsten solvent extraction as well as in the H1-S2 development-stage scenario in leaching of the oxidised scrap. It contributes substantially to GWP, FC, POCP and ODP. The contribution to industrial scenarios H1-S1 and H1-S3 is between 11% and 26% and the contribution to the development-stage scenario H1-S2 between 19% and 37%. The sulphuric acid used in tungsten solvent extraction and in cobalt leaching is a substantial contributor to FC (25-29%) and AP (17-18%) in industrial scenarios H1-S1 and H1-S3, while in the development-stage scenario H1-S2 its contribution is smaller, 11% to FC and 8% to AP. The largest contribution from cobalt recycling is in the form of sodium hydroxide consumption, which contributes substantially to GWP, FC, EP, POCP and ODP in all scenarios, although it is highest in H1-S1 (19-43%) and somewhat lower in H1-S2 and H1-S3 (11-29%).

In the hardmetal recycling scenarios for tantalum and niobium (H2-S1 and H2-S2), there are substantial differences in the contribution results due to the larger difference in the chemicals used in the processes. The H2-S1 process based on industrial HF leaching is mainly dominated by the acids used in the leaching step: hydrofluoric acid and sulphuric acid. Hydrofluoric acid contributes substantially to all impact categories. The lower contributions are to GWP and ODP with 16% and 26%, respectively, while the contribution to other categories is between 31% and 48%. Sulphuric acid contributes substantially to FC and AP (25% and 35%, respectively). Other large contributors include lime (16-42% to GWP, POCP and ODP), ammonia (12-19% to GWP, FC, POCP and ODP) and electricity (28% to EP). The H2-S2 development-stage process based on caustic conversion is dominated by the sodium hydroxide used for caustic conversion and the oxalic acid used in solvent extraction. The sodium hydroxide contribution is larger to GWP, FC and EP, where it contributes 41-50% and smaller to POCP, ODP and AP (14-24%). In contrast, the contribution of oxalic acid is the reverse, i.e. it contributes 9-23% to GWP, FC and EP and 57-80% to POCP, ODP and AP. Similarly to the industrial H2-S1 scenario, electricity is another large contributor with 30% to EP.

In the recycling of FT catalyst in scenarios C1-S1, C1-S2 and C1-S3, the environmental impacts are dominated by the consumption of sodium hydroxide and sulphuric acid in all scenarios. Sodium hydroxide is the largest contributor to GWP, EP, POCP and ODP with 50-77%, depending on the impact category and scenario, while the contribution to FC and AP is 20-36% and 12-22%, respectively. Sulphuric acid is the largest contributor to FC and AP with 45-54% and 73-83%, respectively. Electricity is another notable contributor with 11-21% to GWP, EP, POCP and ODP.

In the recycling of Mo/Co/γ-Al_2O_3 catalyst in the C2-S1 scenario, the largest contributor varies depending on the impact category. Direct impact contributes substantially to GWP (19%) and AP (42%) as a result of carbon dioxide and ammonia emissions. Of the consumed utilities, electricity contributes substantially to all impact categories apart from AP with 16-35%, and heat contributes 14-22% to GWP, POCP and ODP. Of the consumed acids, sulphuric acid contributes 33% and 39% to FC and AP, respectively, and hydrochloric acid contributes 16% and 15% to EP and ODP, respectively. Of the other chemicals consumed, ammonia contributes 17-25% to GWP, FC and POCP, sodium hydroxide contributes 26% to EP, and soda ash contributes 13% to EP.

4.3 Recycling impact

The environmental impacts of the recycling scenarios described in **Table 3** are shown in **Figure 10** compared to the environmental impacts of producing equivalent products in equivalent amounts from primary resources. In the figure, 100% represents the environmental impacts of primary production and the impacts of the recycling processes have been scaled accordingly. The information is missing for FC and ODP in the C1-S2 scenario producing cobalt sulphate as they were not included in the reference for the primary impacts in the results of the Cobalt Institute LCA (2023c). In addition, the values for cobalt sulphate in the ecoinvent database are likely to be unrealistically high as the activity uses product grade cobalt hydroxide as the feed material rather than crude cobalt hydroxide.

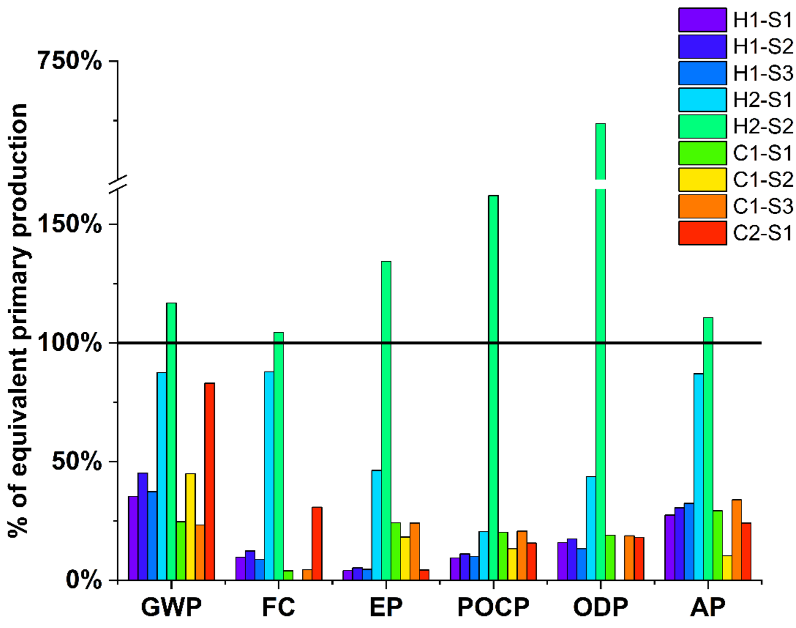

Figure 10 Environmental impacts of the different hardmetal scrap and cobalt catalyst recycling scenarios compared to the environmental impacts of equivalent primary production. 100% represents the impacts of primary production.

In the hardmetal recycling scenarios focusing on tungsten and cobalt refining (H1-S1, H1-S2 and H1-S3), the environmental impacts of the recycling processes are all clearly below the level of impacts from primary production. There are some differences between the three scenarios due to different process flow-sheets and amounts of products produced. The environmental benefit of tungsten and cobalt recycling from hardmetal is evident as the recycling impacts are less than 50% of those of primary production. Of the different impact categories, GWP and AP are the closest to primary production impacts at 35-45% and 27-32%, respectively.

In the hardmetal recycling scenarios focusing on tantalum and niobium recycling (H2-S1 and H2-S2), the situation is not as clear. For scenario H2-S1 based on industrial HF leaching, the recycling impacts are below those of primary production. However, in several of the impact categories the difference to primary production is less than 15% and considering the possible sources of uncertainty a clear environmental benefit cannot be determined directly. However, as shown in **Figure 8**, as the tantalum and niobium content increases, the impacts decrease, and recycling becomes more clearly environmentally beneficial. When the total content of tantalum and niobium in the hardmetal scrap is 7.5 wt.% or above, the environmental benefit is clear even considering the uncertainty. The content of cubic carbides in hardmetal commonly ranges from 0 to 15% but can be as high as 50% in specific applications (Mehrotra, 2014). The impacts of the development-stage process in scenario H2-S2 are substantially higher than those of H2-S1 and are also higher than those of primary production in all impact categories. This is especially the case for ODP, which is dominated by oxalic acid and is more than seven times the ODP of primary production. In the current form the process does not seem to provide any environmental benefit as a recycling method.

The impacts of the HF-free process are heavily dominated by two chemicals: oxalic acid and sodium hydroxide. Therefore, less environmentally intense production of the two chemicals would also decrease the impacts of the process. Sodium hydroxide production, in contrast, is electricity-intense and a move towards wider utilisation of green energy sources could decrease the impacts originating in its production (ecoinvent, 2025). Currently, organic chemicals such as oxalic acid are often produced from non-renewable fossil resources and coal but production from other feedstocks, such as CO_2, could decrease its impacts (Schuler *et al.*, 2021). The dataset used in the thesis is based on oxalic acid production from e.g. naphtha via propylene, which has high carbon dioxide emissions and high consumption of nitric acid. Data is not available for alternative production routes such as production from biomass or by direct electrochemical CO_2 reduction. In some impact categories such as AP, the impact is both dominated by oxalic acid and close to the level of primary production and the traditional HF-based process. There a substantial decrease in the environmental impacts of oxalic acid production might lead to environmental benefit large enough to change the relative ranking of the processes. However, it is difficult to determine without representative data on alternative oxalic acid production and even more so for the impact categories such as ODP and POCP for which almost all impact is due to oxalic acid consumption while the impact of primary production and the HF-based process is substantially lower.

In the recycling of FT catalyst in scenarios C1-S1, C1-S2 and C1-S3, the environmental benefit gained by recycling is also clear. Although the process to recover the different products only differs in the final step, there are some small differences between the products in how much they decrease the impacts compared to primary production. The decrease in impacts from cobalt hydroxide production in C1-S1 and cobalt carbonate production in C1-S3 are very similar. Cobalt sulphate production in C1-S2, on the other hand, shows less of a decrease in GWP compared to the two other scenarios and a larger decrease in AP. The GWP of cobalt sulphate is 45% of primary production, whereas the GWP of cobalt hydroxide and cobalt carbonate is 25% and 23%, respectively. The AP of cobalt sulphate is 10% of primary production while the AP of cobalt hydroxide and cobalt carbonate is 29% and 34%, respectively. The impacts in the other impact categories are less than 25% of those of primary production for all scenarios.

The source of cobalt used in the production of cobalt FT catalyst has a substantial effect on the environmental impacts of that catalyst. The environmental impacts of cobalt FT catalyst production from cobalt hydroxide via cobalt nitrate and with a titania support are shown in **Figure 11** using three different amounts of secondary and primary cobalt hydroxide. First is a closed loop recycling scenario in which 2/3 of the cobalt hydroxide is secondary and 1/3 is primary, the second uses only secondary cobalt hydroxide and the third uses only primary cobalt hydroxide. It is clear that the use of cobalt produced from secondary resources substantially decreases the impacts of the catalyst production. The need for case-by-case assessment of the environmental impacts of a product or process is highlighted by the fact that the GWP when using only secondary cobalt (8.8 kg CO_2-eq./kg catalyst) is close to the value used by Iribarren *et al.* (2013) in their LCA of production of FT products from biosyngas (approximately 8 kg CO_2-eq./kg catalyst), which was based on an approximation. It is unlikely that only secondary cobalt would be used in the production of the catalyst and therefore possible that the use of approximations not including the full catalyst production process and materials can underestimate the true catalyst impacts. When using the mix of secondary and primary cobalt and only primary cobalt, the GWP of catalyst production was markedly higher, 11.5 kg CO_2-eq./kg catalyst and 17.1 kg CO_2-eq./kg catalyst, respectively.

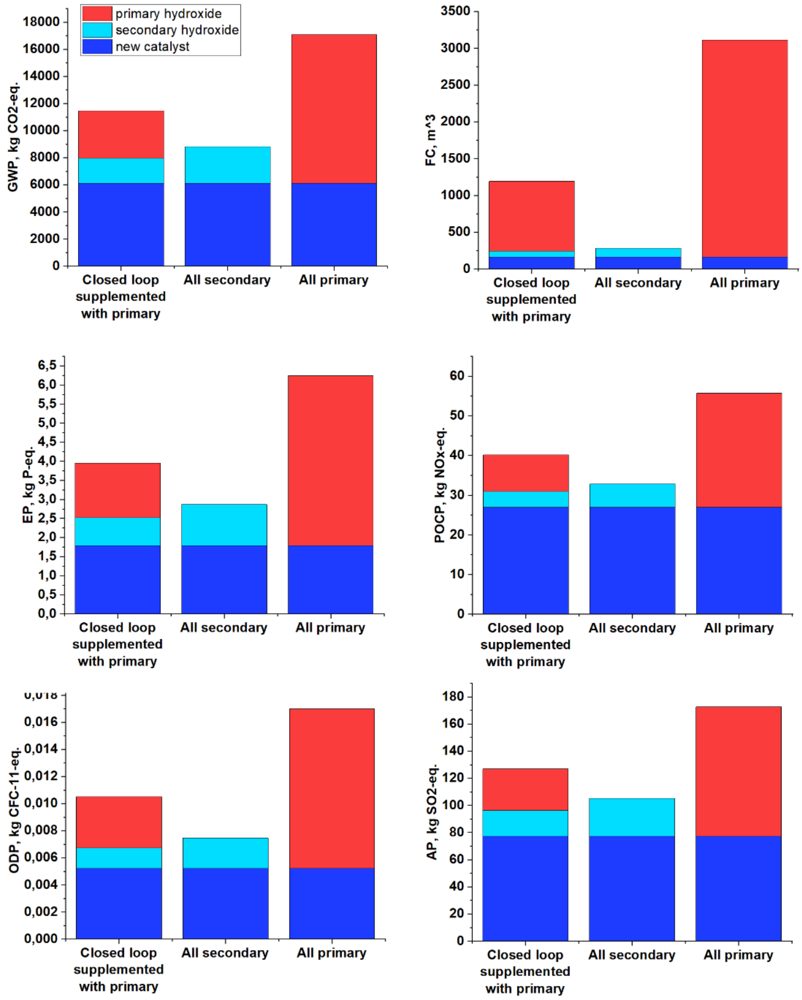

Figure 11 Environmental impact of cobalt FT catalyst production with different amount of primary and secondary cobalt: 2/3 secondary and 1/3 primary cobalt hydroxide, all secondary cobalt hydroxide and all primary cobalt hydroxide. New catalyst impacts are catalyst production impacts other than cobalt production.

In the recycling of Mo/Co/γ-Al_2O_3 catalyst in the C2-S1 scenario, the recycling impacts in all impact categories besides GWP are well below those of primary production. The GWP is 83% of that of equivalent primary production and considering the uncertainty the possible environmental benefit is not directly evident. It can be seen in **Figure 12** that, although vanadium production is the largest contributor in primary production, it is the recovery of molybdenum and to a lesser extent nickel from the catalyst that are the cause of the relatively high GWP. The benefit is much clearer in the other impact categories where FC and AP are 31% and 24% of primary production figures while the other impacts are less than 20%.

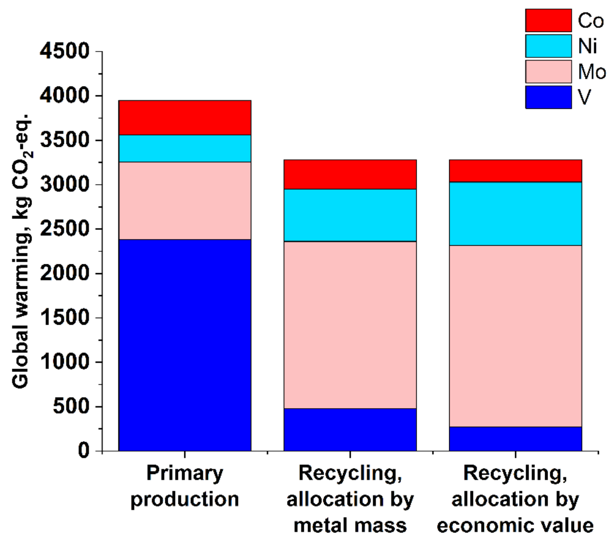

Figure 12 GWP of recycling of 1 t Mo/Co/γ-Al_2O_3 catalyst compared to the GWP of equivalent primary production.

A closer view of the possible environmental benefit gained by recycling cobalt from the different raw materials is presented in **Figure 13**. It shows the environmental impacts of cobalt recycling from hardmetal scrap, cobalt FT catalyst and Mo/Co/γ-Al_2O_3 catalyst scaled to 1 kg cobalt along with the impacts of equivalent primary production. The cobalt recycling processes in scenarios H1-S3, C1-S1, C1-S2, C1-S3 and C2-S1 are all based on the same flowsheet of leaching in sulphuric acid to solubilise cobalt, precipitation and solvent extraction for impurity removal, and precipitation or crystallisation to produce the desired cobalt chemical product. As in **Figure 10**, the information for primary cobalt sulphate is missing regarding FC and ODP.

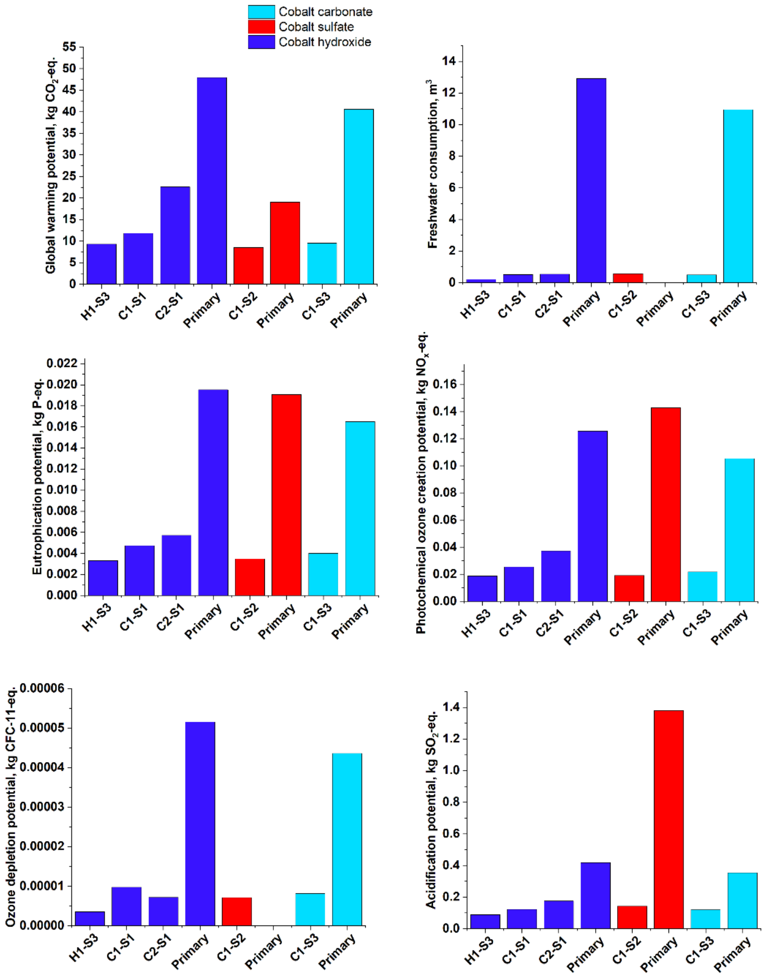

Figure 13 Environmental impacts of producing 1 kg Co as different cobalt products in scenarios H1-S3, C1-S1, C1-S2, C1-S3 and C2-S1 as well as primary production.

In all scenarios the recycling of cobalt is environmentally beneficial as the recycling impacts are substantially below those of equivalent primary production. For cobalt hydroxide, the recycling impacts are 53-98% lower than those of primary production depending on the raw material and impact category. Cobalt hydroxide is recycled from all the studied raw materials and the trend in most impact categories is that the impact of recycling from hardmetal in the H1-S3 scenario is the lowest while the impact of recycling from Mo/Co/γ-Al_2O_3 catalyst in the C2-S1 scenario is the highest. Impacts from hardmetal recycling are 42-60% lower, while impacts from FT catalyst recycling are 5-48% lower. ODP forms the only exception, with recycling from cobalt FT catalyst having the highest impacts by 64% and 27% compared to hardmetal and Mo/Co/γ-Al_2O_3 catalyst, respectively. For cobalt sulphate the impacts were 55-90% lower than those of equivalent primary production and for cobalt carbonate they were 66-95% lower.

The EoL allocation method used was the cut-off method which considers the recycled products responsible only for the environmental impacts of the recycling process to which the material enters burden-free. Similarly, primary products are only responsible for the primary production impacts. Other EoL allocation methods assign burdens differently. For example, the 50/50 method assigns primary production and waste treatment burdens equally between the first and last product life cycles and the circular footprint formula assigns recycling impacts to both the user of recycled material and the producer of the product that was recycled. Naturally, the application of different EoL allocation methods may therefore give different impact values for the same material. The cut-off method mainly benefits the user of recycled material as the material is not burdened with impacts from primary production or final disposal. However, when the difference between the environmental impacts of primary and secondary cobalt is large, as is the case for all scenarios examined in this thesis, the conclusion of environmental benefit is likely to remain the same regardless of chosen allocation method.

Currently, the amount of secondary cobalt on the market is low compared to cobalt produced from primary resources. In 2022 only 5% of cobalt was supplied from secondary resources with 9.3 t of it being produced from manufacturing scrap and 2.4 t from EoL LIBs (Cobalt Institute, 2023a; Cobalt Institute, 2025). The cobalt market is expected to grow substantially with most of the growth expected to be mostly supplied by primary production, however, the role of recycling will also be substantial with 15% of the total cobalt production expected to come from recycling in 2030 and over 40% in 2040. (Cobalt Institute, 2023a). The results of this thesis show that the increasing utilization of secondary cobalt resources has the potential to also substantially decrease the environmental impacts related to the growing cobalt market.

4.4 Uncertainty

4.4.1 Sensitivity analysis

Sensitivity analysis was utilised in **Publications I-IV** to study the effect of uncertainty in individual items on the LCIA results. **Publications I** and **II** examined the effect of uncertainty in process simulation parameter values while **Publication III** examined the effect of uncertainty in the items that form the inventory used for the LCA model. **Publication IV** considered both and contrasted the results of sensitivity analysis on process simulation and LCA model levels.

The sensitivity of the LCIA results to uncertainty in process simulation parameters was investigated in **Publications I** and **II** by varying selected parameter values by ±20% and recalculating the inventory and LCIA results. A 20% change in the values was considered substantial enough to reveal possible sensitivity in the LCA model to the process simulation parameters. The choice of parameters examined was guided by the results of the contribution analysis and the quality of the data on which separate parts of the process simulation were based. **Publication I** examined the cobalt content in hardmetal scrap, leaching chemical concentrations, ammonia circulation and parameters related to the thickeners and filters. **Publication II** examined the solid-liquid ratio, leaching chemical concentration, aqueous-to-organic ratios in stripping, tantalum extraction in niobium stripping and precipitation pH.

The sensitivity analyses showed that the LCA models in **Publications I** and **II** were sensitive to certain process simulation parameters. The tungsten and cobalt recycling processes simulated in **Publication I** in scenarios H1-S1 and H1-S2 were both sensitive to ammonia circulation and acid concentration in cobalt leaching. The results for H1-S2 were also sensitive to the concentration of alkali in the leaching of oxidised scrap. For leaching chemicals, the change in the results was between 3% and 11% in various impact categories, while the change caused by the ammonia circulation affecting the ammonia emissions was between 16% and 20% in AP. However, the level of sensitivity was not considered to be high enough to affect the major conclusions drawn from the results regarding environmental benefit. The tantalum and niobium recycling processes simulated in **Publication II** in scenarios H2-S1 and H2-S2 were both sensitive to the solid-liquid ratio, aqueous-to-organic ratio in niobium stripping and the concentration of chemicals used in the solubilisation stages. The changes in H2-S1 were between 0% and 23% depending on the parameter and impact category. The changes in H2-S2 were between 16% and 30%. Thereby, even though the environmental impacts of tantalum and niobium recycling in H2-S1 scenario are lower than in primary production, it cannot be directly determined to be environmentally beneficial when the concentrations are low considering the level of sensitivity of the results to the simulation parameters. However, the situation changes as the concentration in the hardmetal scrap increases and the difference with regard to primary production is larger.

The results from the sensitivity analysis of process simulation parameters shows whether the LCA model is substantially sensitive to parameters that are known to vary (e.g. cobalt content in hardmetal), those based on uncertain literature data (e.g. tantalum extraction in niobium stripping) or parameters that have been estimated based on similar processes (e.g. solid-liquid ratio). This can be useful information to guide further data collection to refine the process simulation and thereby the LCA model. However, the process is quite time- and labour-intensive and requires some manual modification of the process simulation.

LCA software typically has the capability of performing sensitivity analysis on the LCA model, which is more straightforward than sensitivity analysis of the process simulation parameters. The sensitivity of the LCIA results to uncertainty in the inventory items was investigated in **Publication III** by varying the values of the inventory items in the LCA software and recalculating the LCIA results. A ±20% variation was again used. Included in the analysis were all chemicals and utilities consumed by the process. The recycling models in scenarios C1-S1, C1-S2 and C1-S3 were sensitive to variation in the consumption of sodium hydroxide and sulphuric acid. The same chemicals were also the largest contributors to the environmental impacts. The change was between 2% and 17% depending on the chemical and impact category. The environmental benefit from recycling compared to primary production was great enough that the level of sensitivity did not affect the conclusions.

Sensitivity analysis of the process simulation parameters and of the inventory items provides similar but not exactly the same information. In **Publication IV**, the sensitivity of LCIA results to both was investigated by varying the inventory items and chosen process simulation parameters by ±20% and recalculating the LCIA results. Simulation parameters assumed to be uncertain or to affect the consumption of large contributors were included in the analysis. Sensitivity analysis of the inventory items found the largest changes, between 5% and 8%, to come from variation in the values of ammonia emissions and consumption, electricity consumption, sulphuric acid consumption and sodium hydroxide consumption, depending on the impact category. Sensitivity analysis of the process simulation parameters found that the largest changes, between 2% and 74%, resulted from variation in parameters affecting ammonia emissions, heat consumption, electricity consumption, sodium hydroxide consumption, sulphuric acid consumption and soda ash consumption. The changes were generally between 2% and 5%, apart from changes to AP from ammonia emissions which were +74% and -42%. Mostly, the sensitivity analyses identified the system as being sensitive to the same items (ammonia emissions, consumption of sodium hydroxide, sulphuric acid and electricity). Regarding the sensitivity to consumed chemicals and utilities, the results were quite well aligned, although for the direct emissions of ammonia the analysis of process simulation parameters found the system to be much more sensitive than the analysis of inventory items.

Sensitivity analysis provides useful information on the effect of the process simulation parameters or inventory items of the LCIA results and can be used to target data collection effectively. However, even though the analysis is, for example, focused on process simulation parameters considered uncertain in one way or another, a fairly rigid ±20% change in the parameter values does not describe the level of uncertainty in the data. Sensitivity analysis results should therefore be combined with information provided by uncertainty analysis methods.

4.4.2 Uncertainty analysis

Different methods of uncertainty analysis were used in **Publications I, III** and **IV** to study the robustness of the conclusions drawn from the LCIA results. **Publication I** assigned DQIs, each with an expected level of accuracy to all chemicals, utilities and direct emissions making up the inventory. **Publication III** also utilised DQIs but also used them as a basis for Monte Carlo simulation. **Publication IV** used both DQIs and a pedigree matrix to estimate the uncertainty levels of inventory items, which were then used in the Monte Carlo simulations. DQIs were also assigned to chosen process simulation parameters to perform Monte Carlo simulation.

The impact of uncertainty related to the inventory items was described in **Publication I** based on DQIs. The method has previously been used in LCA studies focusing on metals refining (Schreiber *et al.*, 2016; Marx *et al.*, 2018; Pell *et al.*, 2019) and is based on the cost estimate classification system of the American Association of Cost Estimation (Bredehoeft *et al.*, 2020). The DQIs range from DQ1, with an expected accuracy of -10% to +15%, to DQ5, with an expected accuracy of -50% to +100%. DQ1 and DQ2 (-15% to +25%) are assigned to measured data and literature data of industrial operations, respectively, and neither was assigned to the flows in the inventory. Most of the inventory items were considered DQ3 (-20% to +30%), which is assigned to modelled, calculated and upscaled data. Some flows related to electricity consumption and oxidation were considered DQ4 (-30% to +50%), referring to data estimated based on similar processes and DQ5, referring to estimated data. Assigning DQIs gives a qualitative assessment of the uncertainty but does not show how the uncertainty affects the LCIA results.

The impact of uncertainty related to the inventory items was also analysed based on DQIs in **Publication III** and their effect on the LCIA results was studied using Monte Carlo simulation. The majority of the inventory items were again classified as DQ3. The consumption of electricity, hydrogen peroxide in leaching, flocculant in thickeners, and diluent and extractant in solvent extraction were considered DQ2 as the data was acquired from an EIA of industrial production (Jervois, 2024). Normal distributions were used in the Monte Carlo simulation with the minimum and maximum values determined by the DQIs. The standard deviations varied between 6% and 11% and were generally the highest for AP. It was considered that the level of uncertainty in the model was not high enough to affect the conclusions drawn regarding the environmental benefit.

In **Publication IV** the impact of uncertainty related to the inventory items was analysed based on both DQIs and a pedigree matrix used in the ecoinvent database. Based on the DQIs, most of the inventory items were classified as DQ3 apart from heat and electricity which were classified as DQ4 and hydrogen peroxide and kerosene which were considered DQ2. Based on the pedigree matrix, the reliability of the flows related to the consumption of chemicals and utilities was mostly considered to be 4, except for hydrogen peroxide and kerosene which were considered to be 2. Completeness, temporal correlation and geographical correlation were mostly considered to be 1, apart from electricity and heat which were considered to be 3. Further technological correlation was considered to be 2 for all, except for hydrogen peroxide and kerosene for which it was considered to be 1. For the direct emissions of ammonia and carbon dioxide, reliability, completeness and further technological correlation were considered to be 5 while temporal and geographical correlation were 1. The uncertainties estimated by DQIs and the pedigree matrix were used to perform Monte Carlo simulations. The median values from both simulations were 4-8% apart, with values from the simulation based on the pedigree matrix being smaller. Both simulations also produced mostly similar standard deviations, which were 2-12% apart, except for GWP and AP. For GWP and AP the results from the simulation based on the pedigree matrix were 208% and 231% higher, respectively. GWP and AP are highly impacted by the direct emissions, which were considered quite uncertain. Estimation of uncertainty by both DQIs and the pedigree matrix focused on the uncertainty in the level of inventory items but did not consider the effect of the process simulation parameters.

The impact of uncertainty related to the process simulation parameters was analysed in **Publication IV** by assigning DQIs to chosen parameters, performing a Monte Carlo simulation and using the achieved new minimum and maximum values for the inventory items in the LCA model to perform another Monte Carlo simulation. Of the chosen parameters, DQ2 was assigned to hydrogen peroxide concentration, DQ3 was assigned to soda ash input, solid-liquid ratios in leaching steps, concentration of acid in leaching and stripping acid consumption, and DQ4 was assigned to ammonia circulation, filter cake moisture, filter wash water efficiency and thickener underflow solids. **Figure 14** shows a comparison of the results of this analysis with the results of the two analyses focusing on inventory items based on DQIs and the pedigree matrix. The median was mostly similar to the other two analyses, and the difference was 1-7% for impact categories other than AP for which the difference was higher, i.e. 50% and 38% than the analyses based on the pedigree matrix and DQIs, respectively. Substantially larger differences were found in the standard deviations. The difference to the analysis based on the pedigree matrix was between -91% and +10% and the difference to the analysis based on DQIs was between -51% and +263% depending on the impact category.

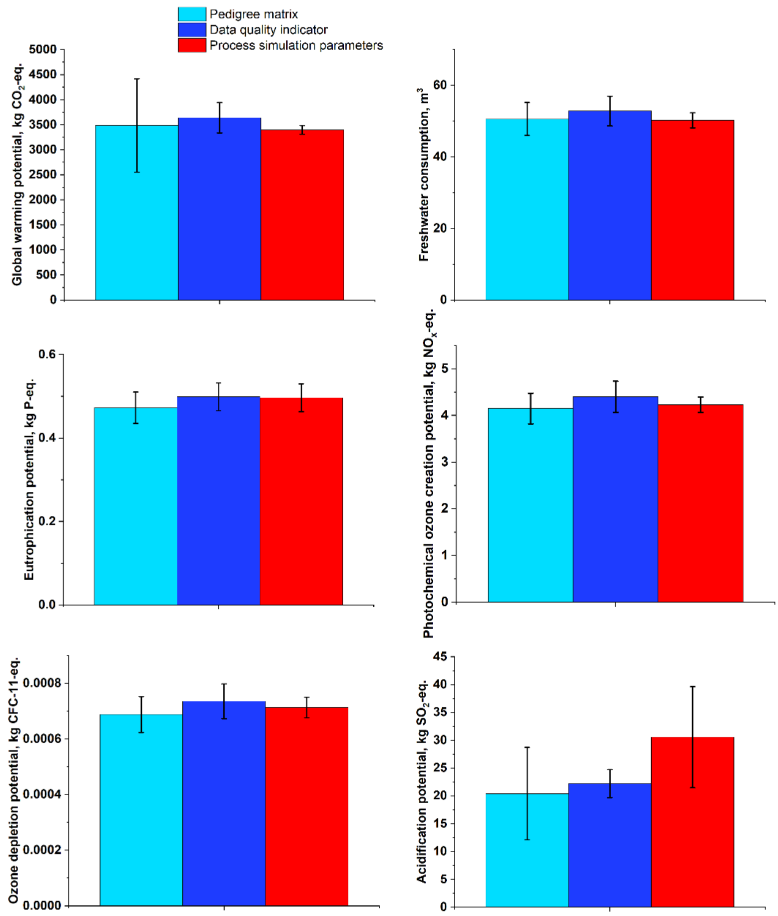

Figure 14 Environmental impacts of recycling 1 t $Mo/Co/\gamma\text{-}Al_2O_3$ catalyst based on different uncertainty analysis methods. (**Publication IV** © 2025 The authors, reproduced from ACS Sustainable Resource Management under CC BY 4.0).

The largest differences between the methods are found for AP, to which direct ammonia emissions from the catalyst recycling process are the largest contributor. The emissions are determined in the process simulation by the parameter of ammonia circulation, which was considered relatively uncertain, i.e. DQ4. For impact categories which are dominated by the consumption of chemicals and utilities, the results of the different uncertainty analyses are more in line with each other. The uncertainty analysis involving process simulation parameters is more time- and labour-intensive than the analyses focusing purely on inventory items and the LCA model. However, when uncertain parameters that directly control large contributors are included in the process simulation, their impact should also be considered.

4.5 Applicability of the results

Publications I-IV provided gate-to-gate process inventory data and impact assessment results on the recycling of hardmetal and cobalt catalysts in the geographical area of the EU. The results on the environmental impacts of the recycling were interpreted taking into consideration the uncertainty from various sources. The data can be used to estimate the environmental impacts of tungsten, cobalt, tantalum and niobium recovery from hardmetal via chemical recycling, cobalt recovery from FT catalyst via cobalt refining, and molybdenum, vanadium, nickel and cobalt recovery from Mo/Co/γ-Al_2O_3 catalyst via soda ash roasting and cobalt refining processes. The impacts were assessed for specific raw material compositions in specific processes. The models and inventories would need to be updated if the product, raw material or process of interest differed substantially from those presented in the current work. Particularly the concentration of the recovered metals such as cobalt is likely to substantially impact the possible environmental benefit gained by recycling.

The geographical area of **Publications I-IV** was considered as the EU and the majority of the used background processes used were for production within the EU or Europe. Differences to other production regions are most likely to arise due to differences in either the production processes and technology or the electricity mix. The used electricity mix for the direct consumption of electricity in the simulated processes was the current Finnish one which is a relatively clean one. The production methods and electricity mix should be carefully considered in any reuse of the generated inventories.

Only certain metals were modelled to be recycled as products while others, such as titanium and aluminium, were removed from the solutions and treated as solid waste. In **Publication I**, tungsten and cobalt were recycled from hardmetal scrap while other metals in the material ended up in leaching residues and other solid wastes. This system was expanded in **Publication II** to consider the recycling of tantalum and niobium. In **Publication III** only cobalt was recycled from spent FT catalyst. The catalyst contains also substantial amounts of titanium, which could possibly be recovered although data to model such a process is not available in public literature. In **Publication IV**, molybdenum, vanadium, nickel and cobalt were recycled from Mo/Co/γ-Al_2O_3 catalyst while e.g. aluminium was removed from the solution in solution purification steps. Inclusion of other metals contained in the raw materials as products if recycled at sufficient purity could affect the environmental impacts of the system and should be considered if using the inventories as proxy data for treatment of similar raw materials.

The largest limitation to the reliability of the results is the quality and availability of data. As primary data is not publicly available for the studied processes, the inventories have been generated with the aid of process simulation. The process simulations in the presented work are based mostly on laboratory- and pilot-scale experiments which have been scaled up to represent industrial scale production. However, process parameters acquired from batch experiments on laboratory-scale may not directly describe continuous industrial operation. Continuous operation and higher efficiency in industrial processes could affect e.g. the concentration of compounds in the solution and retention times which in turn may affect the electricity consumption. Uncertainty also arises from the databases used in the simulation which do not cover all the species in the simulation making the prediction of solution properties difficult. This mainly affects the calculations of solution density and heat balance and the effect on the inventory is likely to be small.

Regardless of its shortcomings, in the absence of measured data from a plant, process simulation can be considered a good alternative. In some cases, it can result in underestimation of the impacts but is more likely to overestimate them (Parvatker and Eckelmann, 2019). Because information on the exact chemical behaviour of the studied materials under different process conditions is limited in the literature, the uncertainty of the process simulation parameters has been considered using various methods and mostly did not affect the conclusions drawn. Nevertheless, the process simulations and LCA models should be updated if more representative information becomes available, particularly for the streams considered more uncertain in the uncertainty and sensitivity analyses.

These studies provide only an estimate of the environmental impacts of the recycling processes and do not take a more holistic view of sustainability. Inclusion of the social and economic pillars of sustainability would allow for a better understanding of the recycling systems. For instance, tantalum is considered a conflict mineral and the social impacts of its recycling compared to primary production might contrast with the environmental impacts of recycling. The implementation of methods for sustainability assessment such as social LCA and life cycle costing in addition to LCA should be considered for future work.

5. Conclusions

The growing demand for cobalt is almost exclusively being met by highly geographically concentrated primary production, with mining in the DRC and Indonesia and refining in China. Environmental concerns have also been associated with the primary production of cobalt, particularly regarding effects on biodiversity and pollution to water and air. An increased proportion of secondary cobalt in the production of new cobalt products could help diversify the supply and has also been assumed to lower the environmental impacts of cobalt production.

The environmental impacts of a recycling process are dependent on the raw materials, products and unit processes involved. Estimating impacts even based on similar processes or raw materials is therefore difficult and information is lacking for many recycling systems. Information is not available publicly for the industrial processes investigated in this thesis on a detailed enough level for assessment and information on the investigated development-stage processes does not exist at all. Therefore, in this thesis, process simulation was used in combination with LCA to gather the inventory of process data employed as the basis for the calculation of environmental impacts. Process simulation allows for prediction of mass and energy balances of industrial-scale processes based on information from laboratory or pilot-scale studies.

In this thesis, nine scenarios were simulated with different products, and flowsheets were created to study the recovery of metals from hardmetal scrap and two spent cobalt catalysts. **Publications I** and **II** focused on metal recovery from hardmetal scrap with **Publication I** estimating the environmental impacts of tungsten and cobalt recycling in chemical recycling and **Publication II** focusing on tantalum and niobium recycling from a chemical recycling leach residue. An additional scenario based on the chemical recycling process in **Publication I** but featuring an updated industrial cobalt refining process was presented for the first time in this compendium. **Publications III** and **IV** focused on metal recovery from spent cobalt catalysts. **Publication III** estimated the impacts of cobalt recycling from a cobalt FT catalyst in an industrial cobalt refining process, while **Publication IV** examined molybdenum, vanadium, nickel and cobalt recycling from a $Mo/Co/\gamma\text{-}Al_2O_3$ catalyst by soda ash roasting and an industrial cobalt recycling process.

Almost all recycling systems investigated in the thesis were shown to have lower environmental impacts than the production of equivalent products from primary raw materials. Tantalum and niobium recycling by an industrial HF leaching process was shown to provide a clear environmental benefit only when the tantalum and niobium content in the hardmetal scrap was high. The development-stage scenario with an HF-free process suffered from high environmental impacts due to oxalic acid. The potential environmental impacts were shown to be much higher when compared to primary production. The calculated impacts of cobalt recycling in particular were lower than those of primary production for all raw materials. Cobalt was modelled as being recycled as cobalt hydroxide from all investigated raw materials. When comparing the different cobalt hydroxide results, the environmental benefit was greatest for recycling from hardmetal scrap. The least environmental benefit, on the other hand, was found for most impact categories in recycling from Mo/Co/γ-Al_2O_3 catalyst, although the benefit was still clear and the impacts considerably lower than in primary production. The differences between the scenarios are mostly due to different process configurations and cobalt content in the raw materials.

In the EU, the majority of cobalt ores, concentrates and intermediates are sourced through imports from the DRC while the majority of refined cobalt comes from domestic production in Finland (European Commission, 2020). A large share of the refined cobalt from Finland is produced from imported concentrates but a substantial amount is also produced from spent cobalt-bearing materials. A large cobalt refiner in Finland reported that in 2024 15-20% of the production came from secondary sources (Jervois, 2025). Production from domestic secondary sources improves the resilience of European cobalt supply while decreasing the environmental impacts of cobalt production. Of the secondary raw materials studied in this thesis the largest environmental benefit was found in the recycling of hardmetal scrap for which the process is already well established. Environmental impacts of cobalt production could therefore be further decreased by even relatively simple means of increased collection at hardmetal use sites in the manufacturing industry. In addition to filter cakes from hardmetal recycling, also the recycling of spent FT catalyst in the cobalt refining process is performed industrially in Finland for which the results from this thesis showed clear environmental benefit compared to primary production. Closed-loop recycling systems with cobalt users in hardmetal and catalyst industries also promote the collection and thereby recycling of valuable secondary cobalt raw materials.

The reliability of the conclusions and recommendations of an LCA study is determined by the quality of the data used in it. Uncertainty analysis can provide information on how the results should be interpreted but is not always straightforward to perform. In this thesis, several methods for sensitivity and uncertainty analysis were used to estimate the uncertainty related to the data in LCA based on process simulation. On a case study basis, it was found that, for the parameters related to the consumption of utilities, the results were similar regardless of whether the analysis was done on the LCA model inventory items or the process simulation parameters. For these it might be possible to avoid the more time- and labour-intensive analysis of the process simulation parameters. However, for uncertain parameters related to direct emissions, omitting the analysis of the process simulation parameters would have skewed the estimation of uncertainty contained in the LCIA results.

The use of secondary data, such as that produced by process simulation, increases the uncertainty when compared to the use of primary data measured directly from operations. However, it is also associated with some benefits. Process simulation produces data that is highly specific to the studied system. It also allows for division of a multifunctional process, which metals recycling processes often are, into the smallest unit processes possible, allowing accurate division of the inventory between multiple products. When the limitations of the method are sufficiently taken into consideration, process simulation can be used with reasonable reliability and accuracy to estimate the environmental impacts of an existing or development-stage process.

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Appendix

Key terms of the doctoral thesis

English
Key term: life cycle assessment
Definition: A method to calculate how a product impacts the environment over its entire life cycle.
Explanation: Life cycle assessment calculates the direct and indirect environmental impacts of a product's entire lifecycle including resource extraction, production processes, transportation and end-of-life. In practice a life cycle assessment consists of four stages: goals and scope definition, inventory analysis, impact assessment and interpretation. To compare different products, they must all deliver the same function, which is described by a functional unit and reference flow.

Finnish
Käsite: elinkaariarviointi
Määritelmä: Menetelmä, jolla voidaan laskea, kuinka tuote vaikuttaa ympäristöön koko elinkaarensa aikana.
Selite: Elinkaariarvioinnilla määritetään tuotteen koko elinkaaren suorat ja epäsuorat päästöt, joita aiheutuu raaka-aineiden hankinnasta, tuotantoprosesseista, kuljetuksesta ja tuotteen hävittämisestä. Käytännössä elinkaariarviointi koostuu neljästä vaiheesta: tavoitteiden ja soveltamisalan määrittely, inventaarioanalyysi, vaikutusarviointi ja tulosten tulkinta. Eri vaihtoehtojen vertailemiseksi täytyy niiden kaikkien täyttää sama toiminto, jota kuvataan toiminnallisella yksiköllä ja vertailuvirralla.

English
Key term: secondary raw material
Definition: Raw material that is produced when waste or end-of-life products are recycled.

Explanation: New products can be produced from primary raw materials which are entering the life cycle for the first time after resource extraction or secondary raw materials which have been recycled from waste or end-of-life products. Secondary raw materials can be used to replace all or some of the primary raw material in a production process.

Finnish
Käsite: sekundäärinen raaka-aine
Määritelmä: Raaka-aine, joka saadaan jätteitä tai käyttöikänsä päähän tulleita tuotteita kierrättämällä.
Selite: Uusia tuotteita voidaan valmistaa joko primäärisistä raaka-aineista, jotka on saatu suoraan luonnosta ja ovat elinkaarensa alussa tai niitä voidaan valmistaa sekundäärisistä raaka-aineista, jotka on kierrätetty jätteistä tai käyttöikänsä päähän tulleista tuotteista. Sekundäärisiä raaka-aineita käyttämällä voidaan korvata kaikki tai osa valmistuksen kuluttamista primäärisistä raaka-aineista.

Business, Economy

Art, Design, Architecture

Science, Technology

Crossover

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