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Water and carbon footprints of mining and producing Cu, Mg and Zn: A comparative study of primary and secondary sources

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Vatten- och kolavtryck av brytning och produktion av Cu, Mg och Zn:
En komparativ studie av primära och sekundära källor

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Abstract
Finding sustainable alternatives to the current degrading activities associated with metal mining and production is essential in order to meet increased resource demand and stricter environmental regulations. One option that recently has gained interest is to recover metals from fly ash, which is produced when incinerating municipal solid waste. As of today, metal resources in fly ash are not utilized in Sweden or in most other parts of the world, but are put on landfills where they are made unavailable for future use. Incentives in terms of economic and environmental gains of recovering metals from fly ashes are needed in order to realize the required installments for such recovery. A few studies have been dedicated to examine the economic (and to a smaller extent environmental) benefits of up-scaling successful laboratory attempts of these practices in the past. However, these studies are often compared to the current practices at a certain waste-to-energy plant, or for a specific metal. In general data on environmental parameters, especially water use, for implementing techniques for metal recovery from fly ash is scarce. This study compared the environmental burden in terms of water and carbon footprint of recovering Cu, Mg and Zn from fly ash, compared to conventional mining and production of these metals. To enable such a comparison, a literature study of water use and greenhouse gas emissions for the respective metal recovery methods has been made. Both the current practices in use today, as well as a few potential future alternatives are discussed. The overall findings are that both the carbon- and the water footprint is much larger for mining and producing the studied metals using present practices, i.e. primary metal recovery, compared to recovering them from fly ash. The investigated sources indicate that greenhouse gas emissions for magnesium and zinc recovery could be reduced by a factor of 2 and 15 respectively, if secondary sources are used instead. With currently available techniques, the water footprint can be considered negligible when recovering metals from fly ash. A general lack of data on water use and large inconsistencies in ways of reporting water footprint were affecting the results of this study markedly. Still, it could firmly be concluded that if Cu, Mg and Zn were recovered from fly ash, large savings of CO_2e emissions and water could be made compared to only producing these metals from primary sources.

Key words: metal production, fly ash, water footprint, carbon footprint, mining.

Sammanfattning
I Sverige förbränns årligen ca 4.5 miljoner ton avfall. Från förbränningen bildas en askrest, som delvis består av så kallad flygaska. I dagsläget används inte denna aska i Sverige, utan läggs på deponi i Norge eller Tyskland, trots att den innehåller värdefulla metaller. Deponering av användbart material kan inte anses som ett hållbart alternativ i en värld med snabbt växande befolkning och ökande belastning på naturens resurser. Då återvinning av material och minskad miljöpåverkan blivit alltmer aktuellt har potentialen av metallutvinning ur flygaska under senare år undersökts allt mer runt om i världen. Att hitta alternativ till dagens tillvägagångssätt att framställa rena metaller är extra intressant då brytning och produktion av metaller tyvärr ofta innebär kraftig negativ miljöpåverkan av olika slag. Den här studien jämför miljöpåverkan mellan traditionell brytning och produktion av metallerna koppar, magnesium och zink med framställning av dessa metaller från flygaskor. De parametrar som undersöks är vattenanvändning och växthusgasutsläpp. Baserat på de källor som undersökts kan man dra slutsatsen att det är betydligt mer miljövänligt att återvinna metaller ur flygaska än att bryta dem från jordskorpan och producera dem på traditionellt vis. Då metallutvinning ur flygaska idag främst sker i liten skala, ofta i form av laboratorie-försök, bör potentialen i implementering av fullskaliga anläggningar undersökas mer, då stora besparingar på miljöbelastande aktiviteter kan undvikas vid denna typ av metallproduktion. Förutom miljömässig prestanda bör även tekniska och ekonomiska aspekter vid denna typ av utvinning undersökas för att indikera om metallproduktion från flygaska i framtiden ska kunna agera komplement till nuvarande metoder.

Nyckelord: metallproduktion, flygaska, vattenavtryck, kolavtryck, gruvindustri.

List of abbreviations

BES – Bio-Electrochemical Systems

CO_2e – Carbon dioxide equivalents

Cu - Copper

CF – Carbon Footprint

GHG – Greenhouse Gas

GWP- Global Warming Potential

LCA – Life Cycle Assessment

LCI – Life Cycle Inventory

LMG – Latrobe Magnesium

MI – Material Intensity

Mg – Magnesium

MSW – Municipal Solid Waste

MSWI – Municipal Solid Waste Incinerator

ISO – International Organization of Standardization

SHG – Special High Grade

SRB – Sulfur Reducing Bacteria

SX-EW – Solvent Extraction - Electrowinning

WF – Water Footprint

Zn - Zinc

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1. Introduction

Metals are present all around us and within us, acting as vital compounds for the functionality of nature, humans and the modern society. Over millennia, humans have developed a strong dependency of metals as construction material and for technological purposes. In recent years this has allowed us to efficiently store and use renewable energy, which is a vital ingredient to a sustainable future. To obtain the metals needed to supply the current demand, extensive mining of the Earth is required. The mining industry is unfortunately strongly associated with significant negative impact on the environment in terms of land degradation, toxic waste, airborne emissions and water pollution, to name a few (Widerlund and Öhlander 2014). As with fossil fuels and groundwater, the metals of the Earth belong to a closed system and are not an endless resource. The availability of ore that is profitable to mine is for certain metals predicted to reach its peak within a few decades, as ore grades are declining and demand is rising from a growing human population and technological advancement (Northey et al. 2014a; Brown 2006). Altogether, this have given rise to a strong interest of investigating the potential in recycling and recovering metals from waste, in order to reduce the dependency of mining virginal material and to meet requirements from stricter environmental regulations. As of today, some metals are directly recycled from households and industries, but there are still vast metal stocks in waste that are not utilized (UNEP 2011).

In recent years, researchers have been devoted to find different ways of efficiently extract and upgrade metals from ashes from municipal solid waste incinerators (MSWI) (Pettersson et al. 2013; Tang and Steenari 2015; Karlfeldt Fedje 2010; Karlsson et al. 2010; Schlumberger et al. 2007). Metals in fly ash have previously been recovered successfully through different methods, including leaching, which for certain metals have proven to be both resource-economic and sustainable options compared to conventional metal production (Schlumberger et al. 2007; Karlsson et al. 2010; Schlumberger and Beuhler 2012). Most of these studies have yet mainly been carried out in laboratory scale, but efforts towards large-scale implementations are ongoing.

The economic feasibility of installing industrial plants dedicated to recover metals from waste is naturally of high interest and has therefore been reported to some extent previously (e.g. Pettersson et al. 2013; Schlumberger et al. 2007, Johansson et al. 2013; Karlsson et al. 2010). However, literature on the *environmental* gains of recovering metals from waste, and specifically fly ash, compared to mining and producing metals conventionally, is very sparse.

Aim: This study sets out to fill parts of this knowledge gap by giving the reader an overview of the environmental burden of current practices of mining and production of copper, zinc and magnesium. It explicitly aims to find whether there is an environmental advantage in terms of water and carbon footprint to recover these metals from fly ash, i.e. secondary sources, instead of mining them from the crust of the Earth.

The results for conventional metal mining and production are discussed and compared with corresponding data for different existing methods of metal recovery from fly ash,

and a few potential options for future secondary metal recovery are introduced as well. The report is carried out as a literature study, where data on water and carbon footprints for the different ways of metal production is collected from scientific articles, databases and personal communication. The reported material will give an indication on how much greenhouse gas emission and water use that can be avoided if implementing the presented recovery techniques, compared to the present mining and production activities in place.

2. Background

2.1. Metals, mining and metal production

Metals are necessary to the well-being of humans and our society in uncountable ways. They are crucial for the functioning of all living organisms on Earth as nutrients, parts of proteins or catalyzers in biochemical reactions (Kessissoglou 1995). Humans started to master metals around 4000 B.C. (Reardon 2011), and they have acted an integral part of the development of the human species ever since. Metals have numerous applications and have played a major role in modernizing our society in terms of industrialization and technological development and are crucial ingredients for a sustainable future as they allow us to move away from fossil fuels. For instance, some metals act as essential compounds in new technologies needed for capturing and storing energy from renewable resources, such as solar panels and batteries (UNEP 2013). Lightweight metals, such as magnesium, are continuously becoming more attractive in many everyday products such as laptops, cellphones and other products that profit from being light (Johansson et al. 2013). With a growing middle-class, the demand for these kind of products are likely to continue rising. Another important market for light metals that is likely to see an increase in the near future is electric vehicles, especially as technology improves and prices continue to drop (Pollet et al. 2012). This increased demand of modern technology and vehicles means a greater need of material and more mining.

Unfortunately, the extraction of these valuable materials comes at a price. Mining industry is known to cause extensive damage on the natural environment and at times even on the workers and the surrounding society in terms of health (UNEP 2013). Furthermore mining and the production of metals use significant amounts of water and degrade surrounding nature in different ways. This might for instance include occasional contamination of the environment through different kinds of spills, increased nitrogen and salinity levels in receiving waters, emissions of particulate matter and greenhouse gases (GHGs), reduction of biodiversity in the area, or other environmentally degrading activities (Tibbett 2015; Widerlund and Öhlander 2014; Mudd 2010a)

The steadily increasing human population keeps the mining industry flourishing, as more material is needed to supply demand. Over time this naturally leads to raw materials becoming less abundant in the crust of the Earth (Vieira et al. 2012). Most of the major metals used for industrial purposes are present in immense amounts in the lithosphere. In many cases however, only a small fraction of the material is economically profitable to mine due to low concentration in the ore, which means lower yields and greater costs per unit produced metal (Northey et al. 2014a). Profitability will become an increasingly critical issue as ore grades are likely to

decline over time since ores of higher metal concentrations are principally mined (Mudd 2010b). Lower ore grade also means higher environmental impact per unit produced metal, as less metal can be extracted after processing the ore (Northey et al. 2013).

As negative effects of climate change are steadily increasing both in frequency and intensity on a global scale (IPCC 2014), the need to minimize GHG emissions and move towards a resource efficient future is imperative. The need to enable lucrative recycling of metals is especially urgent as metal reserves (profitable for mining) are limited in nature, and that mining is an environmentally degrading activity that cannot be considered sustainable. By recovering metals from waste, substantial amounts of emissions, land and water degrading activities and other negative environmental impacts associated with mining and production can be avoided.

With respect to the Swedish environmental targets adopted by the Government, especially '*Reduced Climate Impact', 'Good Quality Groundwater' and 'Flourishing Lakes and Streams'* (Naturvårdsverket 2016), alternatives to current ways of obtaining metals, and their individual performance on environmental impact, are scrutinized in this report.

2.1.1. Choice of metals

The metals copper, magnesium and zinc were chosen for this study due to their individual importance for the global economy, for the society and for the environment (UNEP 2013). Data availability was also a necessary requirement when choosing objects of investigation, in order to generate higher quality of the results. Further reasons for this pick is their relatively high concentration and monetary value in Swedish fly ashes as well as their promising potential to be recovered from fly ashes (Pettersson et al. 2013; Karlfeldt Fedje 2010; Tang and Steenari 2015; Johansson et al. 2013).

2.1.2. Characteristics and production of Cu, Zn and Mg

Mining is in essence extraction of resources from the crust of the Earth, and can be carried out very differently depending on what resource (e.g. natural gas, oil, water or ore) is mined, what operation is in place and where. In the context of metal mining, one normally divides mining activities into either open-pit or underground mines. Copper, magnesium and zinc are mined and processed using different techniques over the world. The most common practices are presented below.

Copper (Cu):

Copper is a widely used metal globally, and is considered the third most important metal for industry after iron and aluminum (Johansson et al. 2013). It is mined both from open pit and underground mines, however most major production countries mine Cu from open pit mines (Dudka and Adriano 1997). Most of the global Cu reserves are present in Latin America, from where the European Union imports around half of the copper used within the EU (Johansson et al. 2013). Sweden holds seven copper mines, and makes up about 10% of the production within the EU as of 2013 (SGU 2016).

The global reserves of copper in the crust of the Earth are extensive, to say the least. However, only a fraction of the total reserves are considered economically profitable to extract with current techniques. With the rapid and vast extraction of copper that

has been taken place over the past century, some estimates indicate that copper will reach its peak as an available resource for mining within only a few decades (Brown 2006; Vieira et al. 2012; Northey et al. 2014a).

In broad terms, copper ore normally occurs as either oxide ore or sulfide ore. The sulfide copper ore chalcopyrite ($CuFeS_2$), is the most common one making up roughly 50% of the mined copper globally (CDAA 2016). Copper is mainly recovered through two different processes – pyrometallurgy (thermal treatment) and hydrometallurgy (wet treatment) (Figure 1). In general, iron-containing sulfide ores are treated pyrometallurgically. After mining the ore, this process includes concentration, smelting and refining stages to produce high-grade copper (Northey et al. 2013). Oxide ores and sulfide ores with lower iron content are instead processed by hydrometallurgical techniques, which incorporates leaching followed by so called solvent extraction and electrowinning (SX-EW) steps, after the ore has been mined (Ayres et al. 2003, Norgate et al. 2007). Solvent extraction is a purification step often involving metal ions transfer from aqueous to organic phase (Tang and Steenari 2015) and electrowinning involves extraction of pure Cu by passing the copper concentrate through electrolytic cells after it has been heated and filtered (BGS 2007). Hydrometallurgical processes are advantageous in terms of the ability to extract lower ore grades as well as emitting less gaseous toxins since the waste is liquid, thus easier to neutralize and handle (BGS 2007). However, hydrometallurgy is in general more energy demanding than pyrometallurgy (UNEP 2013).

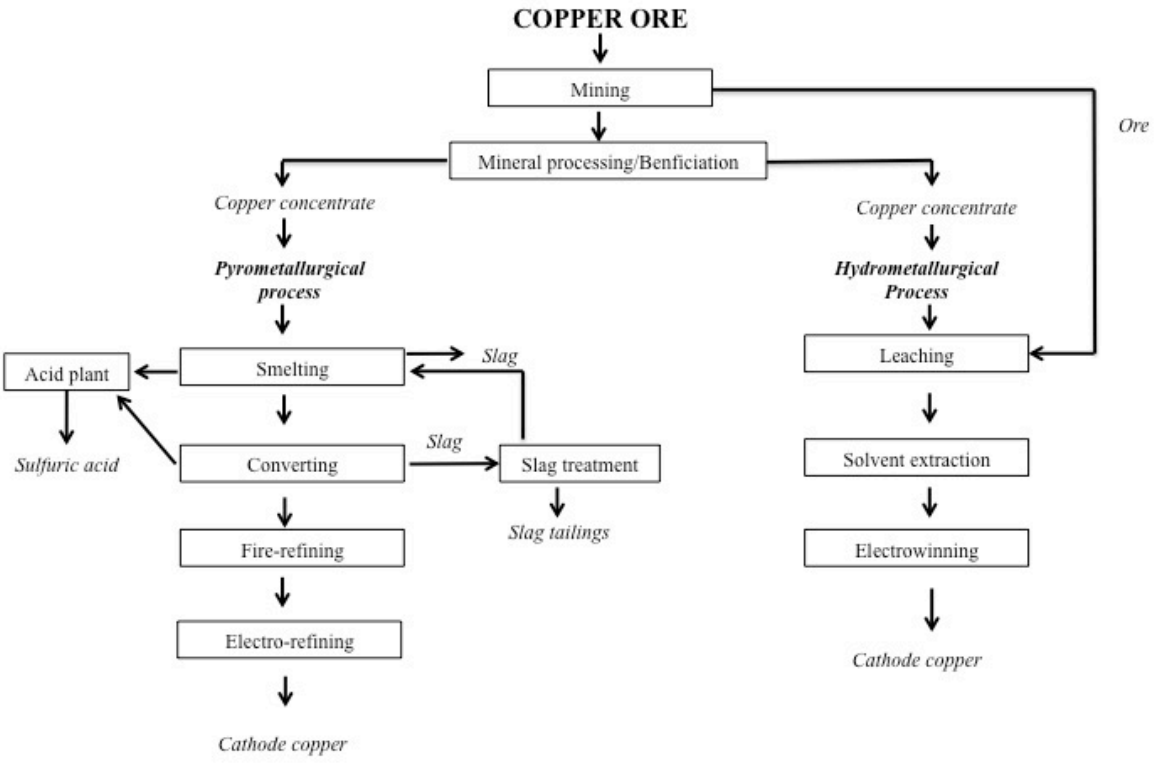

Figure 1: A flow scheme of the dominate ways of producing Cu from copper ore followed by either pyrometallurgical or hydrometallurgical processes. Figure adapted from Norgate et al. 2007.

Today, copper is recycled to a fairly large extent, where approximately 35% of the total copper on the market originates from recycled material. This recycled material consists of scrap, either from end-of-life material or scrap that is contained from

downstream manufacturing (International Copper Association 2014). In Europe the corresponding recycling rates of Cu are 48% on average (Bertram et al. 2002).

Magnesium (Mg):

Magnesium is the eighth most abundant and widespread metal on Earth, occurring in great amounts both as minerals and as dissolved in the ocean or salt lakes. Mineral sources of Mg are nowadays dominating the market, where dolomite makes up roughly half of the world Mg output, followed by magnesite and carnallite (BGS 2004). Mg has a density of 1.74 g/cm^3, making it roughly 35% lighter that aluminum and a very attractive metal (Johansson et al. 2013). It has many applications, however its light characteristics make it especially suitable for products such as batteries, cameras, laptops, electric cars etc. (Cherubini et al. 2008).

When it comes to magnesium mining, China is by far the largest producer, generating roughly 85% of the world's magnesium (Johansson et al. 2013). To produce high purity magnesium, either thermal or electrolytic processes are used. The thermal method is the most cost-effective and common option, but is also very energy demanding. The raw material treated thermally is primarily dolomite, while magnesite, brines and seawater is treated through electrolytic processes. Dolomite does not require that many stages of complex purification as the inputs of electrolytic processes do, which is part of why it is more cost-efficient (BGS 2004). The two major ways of producing Mg, from dolomite and magnesite ore, is illustrated in Figure 2 below. Thermal (i.e. pyrometallurgical) processes for Mg production typically include crushing, calcination, briquetting and reduction before melting and refining the concentrate to pure magnesium. In electrolytic (i.e. hydrometallurgical) processes the ore is normally leached with hydrochloric acid, followed by purification, dehydration and extraction through electrolysis (Norgate et al. 2007).

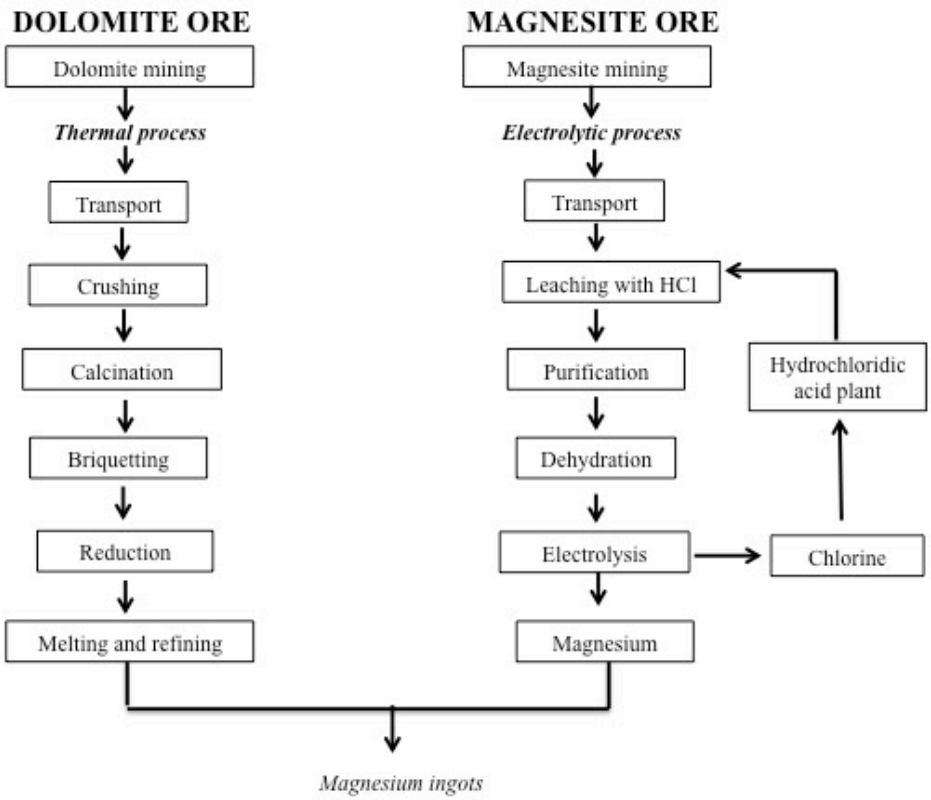

Figure 2: Depiction of the two major Mg mining and production processes, adapted from Cherubini et al. 2008.

Zinc (Zn):

Australia, China and Peru make up the large majority of the world's Zn production (U.S. Geological Survey 2015), while Europe only holds a tiny fraction of the global Zn reserves (about 1%)(European Commission 2015a). From that fraction, Sweden, having seven zinc mines, makes up roughly a fourth of the production within the EU (SGU 2016). Zinc is considered a "strategic substance" in the EU as it holds a high economic importance, is very scarce in Europe and has many applications for industry (European Commission 2015a).

Most zinc mines are present underground (80%) and the rest is mined from open pits or from a combination of the two types of mine (IZA 2016). Zinc never occurs as pure metal in nature, but often appear as a zinc-lead ore, or as other combinations with copper, gold or silver. The greatest source of zinc is from the mineral sphalerite, ZnS, or zinc blende, which is present in most zinc deposits over the world (The U.S. Department of Energy 2002). There are many uses for zinc, but due to its both rust-preventive and anti-oxidizing properties, it is especially popular as rust and corrosion prevention on other metals, and as the anti-oxidizing element in medical creams and products (SGU 2016).

There are two major ways of producing Zn; through electrolytic (hydrometallurgical) or imperial smelting (pyrometallurgical) processes. Some zinc may however also be extracted from lead concentrate through blast furnace processes (Norgate et al. 2007), which is illustrated in Figure 3. Today about 90% of the global zinc is produced through hydrometallurgy involving electrolysis. The process involves roasting, leaching, purification and electrolysis stages (Norgate et al. 2007, IZA 2016).

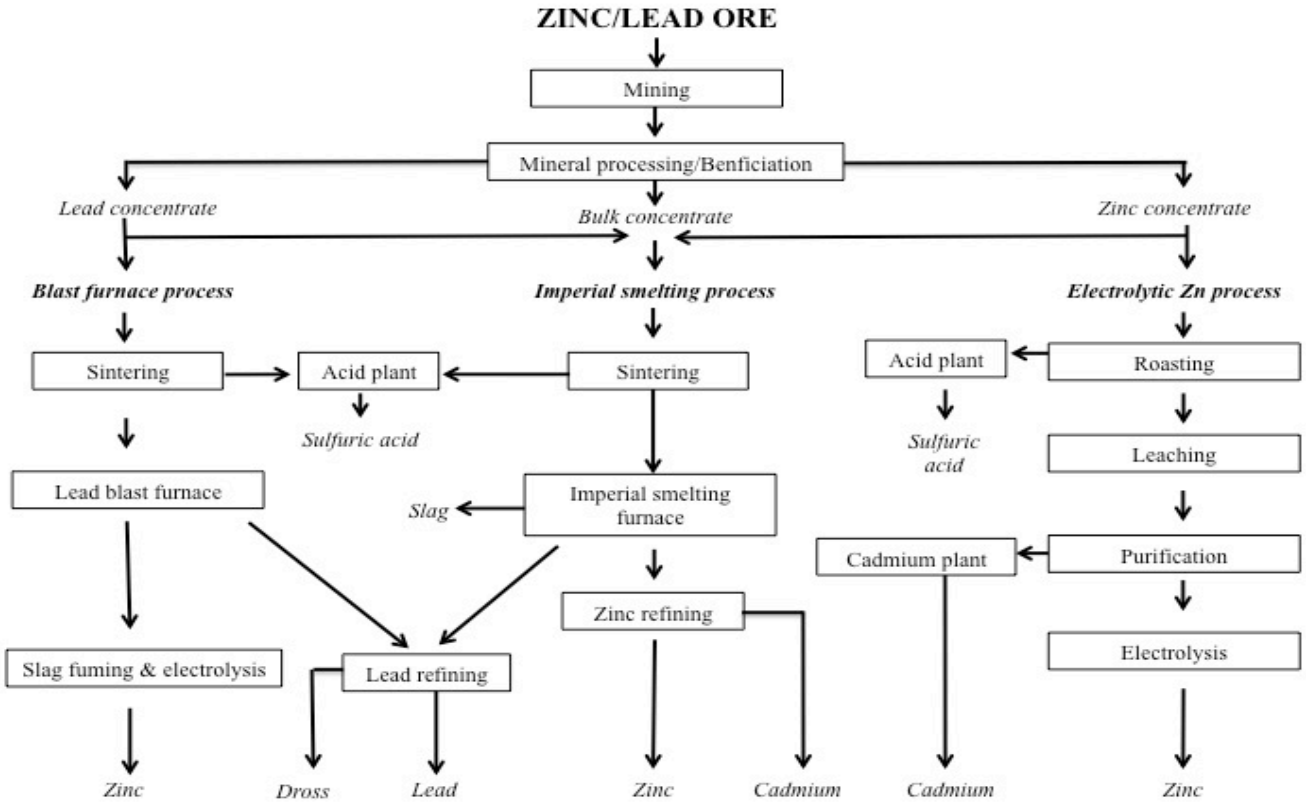

Figure 3: Illustration of the major Zn production processes fron Zn/Pb ore. Figure adapted from Norgate et al. 2007.

2.2 MSW, fly ash and metal recovery

2.2.1. Waste and fly ash in Sweden

Every year about 4.5 million tons of municipal solid waste (MSW) is treated in Sweden, which corresponds to roughly half a ton per person annually (Avfall Sverige 2015). The definition of MSW varies between countries, but within the EU it normally incorporates waste that is collected by or on behalf of municipalities, i.e. from households, commerce, offices and public institutions (European Environment Agency 2013).

In Sweden, nearly 50% of the produced waste is used for energy supply, making it the top producer of energy per kilo waste in Europe. This energy is mainly used as district heating (88%) and the remaining 12% becomes electricity. The rest of the waste is recycled, either for material or biological purposes and about 1% is put on landfill. Nowadays, due to high recycling rates, Sweden needs to import parts of the MSW that goes to incineration to supply energy demand. In 2014, 1.5 million tons of waste was imported, mainly from the UK, Norway and Ireland. (Avfall Sverige 2015).

When incinerating MSW, a rest product in form of ash is produced, which makes up about 20-25% of the total weight of the waste (Johansson et al. 2013). This ash is divided into two different categories - bottom ash and fly ash. Bottom ash generally contains minerals of high melting points, ceramics and larger metal objects, while fly

ash is made up of finer particles, containing a lot of metals, salts and organic compounds (Tang and Steenari 2016). Depending on which process the fly ash is generated from (i.e. fluidised bed boiler or grate fired boiler), the fraction of the total ash varies. Together, waste-to-energy (WTE) plants in Sweden produces about 200,000 tons of fly ash every year (Pettersson et al. 2013; Johansson et al. 2013).
In these ashes a substantial amount of metals are present, sometimes comparable to the metal content in the Earth's crust, or occasionally even higher (Johansson et al. 2013). Table 1 indicates the average content of the selected metals Cu, Mg and Zn in Swedish fly ashes.

Table 1: Average fractions of Cu, Mg and Zn in Swedish fly ashes.
Source: Johansson et al. 2013.

Fraction in fly ash	Amount (%)
Cu	0.22
Mg	0.14
Zn	1.7

Still, as of today fly ash is not utilized as its contents are considered toxic and contains potentially soluble chlorides and heavy metals (Mangialardi 2003; Pettersson et al. 2013). It is currently unaffordable to process fly ash to a non-toxic state in Sweden, so it is instead treated and solidified so that no leaching will occur, and is either put on landfill or shipped to quarries in Langoya, Norway or in Germany, where it is used as filling material (Pettersson et al. 2013). This cannot be considered sustainable in the long run as the quarries hold limited space and the metals present in the buried fly ash becomes unavailable for future use, which means less resources and a loss of economic value (Tang and Steenari 2016). Moreover, the immobilization of the fly ash content cannot be guaranteed to remain in a very distant future. Strict laws on limiting the amount of landfilling was implemented in the EU in 2008 and landfilling of organic waste got prohibited in Sweden in 2005 (Avfall Sverige 2009). This cut-down on landfills is another argument to investigate the potential in sustainable alternatives.

The potentially toxic leaching issues with fly ash has led to development of different techniques aiming to reduce the metal content or immobilize the metal compounds in ashes (Mangialardi 2003; Lam et al. 2010). There are several ways of achieving this, including e.g. magnetic recovery of iron alloys and thermal treatment. Treatments involving magnetism are not relevant for this study as it is primarily used for bottom ashes. Thermal treatment has proven to be very efficient in immobilizing metals as well as reducing the amount of metals in fly ash (to less than <5%) as some metals evaporate around temperatures of 1000 °C (Karlfeldt Fedje 2010). These techniques are however very costly and involves high energy consumption during the heating processes and is not particularly common within the EU (Sabbas et al. 2003).

2.2.2. Metal recovery from fly ash – current practices and findings

Instead of solely removing metals or solidifying fly ash to a non-leaching substance, which has been in focus in the past, the potential of upgrading waste and ash to high-grade metals has been investigated over recent years (Schlumberger et al. 2007; Karlfeldt Fedje 2010; Lam et al. 2010). For fly ash, the probably most common

method for this purpose is wet treatment, i.e. leaching, which is the method in main focus for this report. When fly ash is treated with an acidic liquid (at times alkaline substrates are used, e.g. Karlfeldt Fedje 2010), the low pH allows the metals to be released from their, in most cases, oxide or chloride form (Pettersson et al. 2013). This makes them available for further processing and extraction that lastly results in pure metal compounds (Karlfeldt Fedje 2010; Tang and Steenari 2015). In general, parameters such as type of acid, reagent, leaching time and pH affect the total recovery rates significantly. Recovery rates are further affected by ash chemical speciation of metals, liquid-to-solid (L/S) ratio, particle size and mineralogy. (Karlfeldt Fedje 2010).

There are numerous positive aspects of extracting metals and making them available for reuse. Not only are the degrading activities of mining avoided and the amounts sent to landfills reduced, but as the ashes become less toxic they can be mixed up with cement and also be used as construction material (Lam et al. 2010, Pettersson et al. 2013).

As of today, there is however only one single full-scale facility devoted to upgrading metals from fly ash to pure metallic compounds. This plant is situated in Zuchwil, Switzerland, and has a capacity of producing 1 ton of special high-grade (SHG) Zn (99.995%) daily, using a highly successful method called FLUREC, which has been running since 2013 (BSH Umweltservice AG 2016). In the FLUREC process, acid water produced from the waste incineration process is used for leaching. Through a number of steps, including solvent extraction and electrowinning, pure Zn is extracted. A rest product of hydroxide sludge is produced in the process, but this can be reintroduced in the incoming waste and incinerated again (Schlumberger and Beuhler 2012). If this process would be used for all MSWI ashes in Switzerland, it would make up roughly a third of the total usage of Zn in Switzerland (Pettersson et al. 2013).

In Sweden, similar attempts have been made on fly ashes from Swedish WTE plants. A report by Pettersson et al. (2013) aimed to find the efficiency of Zn recovery by also using acid process water produced from incineration of Swedish MSW from ten different facilities, to investigate whether it might be profitable to install a similar metal recovery plant in Sweden. The study found that 85% of the Zn could successfully be recovered from the investigated fly ashes, while simultaneously making great savings on GHG emissions from avoided mining activities and landfill transport. In Sweden, it might however be more profitable to produce lower grade Zn and send it for further refining elsewhere according to the study.

Studies on Cu recovery from fly ash using different extraction methods have also been carried out (e.g. Yang et al. 2012; Karlfeldt Fedje 2010; Karlsson et al. 2010). Karlfeldt Fedje (2010) investigated the potential of recovering Cu using different reagents and managed to selectively recover about 90% of Cu leachates using leaching with ammonium nitrate or acid followed by solvent extraction. The same study estimated that with an 80% recovery rate of Cu from fly ash, an annual Cu production in Sweden of 1000 tons could be achieved, assuming a Cu concentration of 1500mg/kg in fly ashes. Tang and Steenari (2015) used hydrochloric acid (HCl) at a pH of 2 to leach and recover Cu and Zn from fly ash using two different methods of solvent extraction, which gave Cu yields of 69-87% and 75-80% of Zn respectively.

Magnesium recovery from fly ash using wet treatment has not been studied to the same extent. However, the Australian company Latrobe Magnesium will start running by the end of 2016, aiming to initially produce 5000 tons of pure Mg per year using combined hydrometallurgical and thermal treatment of fly ashes originating from industrial waste. The company estimates Mg recovery rates of up to 95%, based on laboratory experiments (LMG 2014).

Many of these studies and techniques are however still at a pilot stage and have only been carried out in laboratory scale. Still, most of the presented studies hold great potential for being implemented in full scale and are the best alternatives to date in terms of secondary metal production from fly ash. Therefore the above-mentioned findings/projects have been chosen as subject for comparison to conventional mining and production of metals originating from virginal material. Their individual performance in terms of water- and carbon footprint will be presented and compared to primary metal production in more detail in the discussion. A few other interesting topics in the context of metal mining are also presented in the discussion, namely biometallurgy (using sulfur reducing bacteria), bio-electrochemical systems (BES) and landfill mining.

2.3. Terminology and definitions

This study uses the environmental parameters *water footprint* and *carbon footprint* to give an indication on the respective environmental impact of producing Mg, Cu and Zn from primary sources (i.e. mining virginal material and using contemporary producing techniques and from secondary sources (i.e. recovering them from fly ash). There are several concepts and terms associated to these parameters that need to be explained in order to get an understanding of the content reported in this study and the analysis connected to it, which are presented below.

2.3.1. Carbon footprint

The carbon footprint (from now on termed CF) is a widely used and well known parameter that has been in use in different kind of environmental inventories and Life Cycle Assessments (LCAs) since more than a decade back (Ertug Ercin and Hoekstra 2012; Santero and Hendry 2016). The term is thought to be originating from the so-called "ecological footprint" which was developed by Wackernagel and Rees in 1996, aiming to estimate the environmental burden of a certain activity.

Despite the wide use of the CF in LCAs and its general acceptance in society, no universal definition have been made up until very recent years (Ertug Ercin and Hoekstra 2012). Pandey et al. described the term in 2010 as the amounts of GHGs, expressed in terms of CO_2e, that are emitted into the atmosphere by an individual, organization, process or product from within a specified boundary. Based on the conducted literature research, this gives a representative picture of the previous attempts of defining it in literature. In 2013 the ISO-standard ISO14067:2013 defined the CF as the:

"Sum of greenhouse gas emissions and removals in a product system, expressed as CO_2-equivalent and based on a life cycle assessment using the single impact category of climate change".

This description include some terms that require further definitions (taken from the ISO14067:2013 and ISO12064-1:2006);

Greenhouse gases (GHGs): "As of the Kyoto Protocol, the six main types of GHGs are considered carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), hydrofluorocarbons (HCFs), perfluorocarbons (PFCs) and sulfur hexaflouride (SF_6)."

CO_2-equivalents (CO_2e.): "Unit for comparing the radiative forcing of a GHG to that of carbon dioxide. The mass of a greenhouse gas is converted into CO_2 equivalents using *global warming potentials"*

Global Warming Potential (GWP): "Factor describing the radiative forcing impact of one mass-based unit of a given GHG relative to an equivalent unit of carbon dioxide over a given period of time."

Impact Category: "Class representing environmental issues of concern to which life cycle inventory analysis (LCI) results may be assigned." … "[LCI is a] phase of LCAs involving the compilation and quantification of inputs and outputs for a product throughout its life cycle."

As increased levels of atmospheric carbon dioxide and other GHGs are well known to increase the temperature of our planet (IPCC 2014), many incentives to reduce these emissions have been made on all different levels across the planet. To the maybe the most important one belongs the formation of the UNFCCC (United Nation Framework Convention on Climate Change) in 1992, from where environmental treaties such as the Kyoto Protocol and the Paris Agreement were born, which legally binds ratifying countries to actively reduce their GHG emissions (UNFCCC 2016). Due to these international agreements, carbon footprints are nowadays readily reported in large corporate businesses, industries and organizations. Carbon footprint has for the same reasons also been a popular subject of investigation in many scientific papers. This makes collection of data on GHG emissions relatively accessible, even for specific cases such as mining and producing of Zn, Cu and Mg to pure metals from primary sources. Databases such as Ecoinvent v.3 (Weidema et al. 2013) and GaBi (Thinkstep 2015) facilitate access to carbon footprints and other environmental parameters as they hold environmental data on a vast range of commodities, production processes and means of transport from all over the world, in one place.

2.3.2. Water footprint

Water footprint (from now on termed WF) on the other hand has not been used and reported as extensively in Life Cycle Assessments, despite its high importance in mining industry (Santero and Hendry 2016; Peña and Huijbregts 2013). A universal standard for reporting WF was not developed until late 2014 by the International Organization of Standardization (ISO14046:2014). Berger and Finkbeiner (2010) mean that this might be explained by the fact that the LCA method was created and has mainly been used in developed countries, were water normally is not in scarcity. Another reason also lay in the difficulty of reporting water use, as the water cycle is a closed system were water circulates, rather than gets consumed (Mudd 2008).

In general, studies regarding mining and production of metals often report water use

in different ways and units, and often data is lacking completely for certain processes (UNEP 2013; Molinare 2014). In a comprehensive review on WF reporting in LCAs (Berger and Finkbeiner 2010) it was more specifically found that WFs reported from mining companies or in scientific reports vary in terms of including water origin, discharge location, water balance of the area, water quality as well as temporal and spatial scope. Even the terminology of water use was found to be inconsistent. The same review however found that the great majority of reported WFs refer to "consumptive use of off-stream freshwater" that is taken from fresh surface or ground water. Here "consumptive use" means loss of freshwater due to water being incorporated into a product, is discharged to another watershed than it was taken from, evaporated, or other withdrawals of water from the original catchment area. "Off-stream water use" refers to any prior removal of freshwater from the water body, in contrast to in-steam water use, which is freshwater used on site for e.g. hydropower generation or marine traffic (Berger and Finkbeiner 2010). This report considers a water footprint as the definition formulated by Hoekstra (2011) for the Water Footprint Network, which is a widely recognized definition and has been developed in cooperation with a vast number of other environmental organizations and institutions worldwide (Hoekstra 2011);

"*The water footprint of a product is defined as the total volume of fresh water that is used directly or indirectly to produce the product. It is estimated by considering water consumption and pollution in all steps of the production chain."*

The water footprint is further divided into blue, green and grey water footprint, according to the methodology of Hoekstra (2011), which has been applied in many other studies (e.g. Peña and Huijbregts 2013; Ranchod et al. 2015). According to the assessment manual the *blue water footprint* refers to the consumptive use of fresh surface and groundwater; the *green water footprint* to the consumptive use of rain water stored in, or temporarily on top of, soil or vegetation (i.e. not run-off or groundwater); while the *grey water footprint* refers to the amount of water required to dilute polluted water to acceptable water quality levels and natural background standards (Hoekstra 2011). These footprints will however not be treated separately in the report, as there is inconsistency in reporting, and as stated above, most WFs refers to the consumption of blue water.

Water directives in Europe and Sweden:

A brief understanding of how the quality of water is graded and regulated is appropriate in the context of water reporting. The apprehension of "good" water quality might vary between different directives and regions, but according to the European Water Framework Directive, "good status" of a water body is reached when it meets its Environmental Quality Standards (EQS), stipulated in the EQS Directive (2008/105/EC)(European Commission 2015b). For surface water, this involves meeting certain ecological and chemical criterions and for groundwater there are quantitative and chemical requirements. Chemical parameters for water quality standards could for instance involve parameters such as turbidity, pH, biochemical oxygen demand, total dissolved solids, organic carbon content etc. (EPA 2001). These requirements should, according to the directive, be further specified on a national level. In Sweden, surface water quality standards are regulated by the Swedish Agency for Marine and Water Management, and groundwater standards by The Geological Survey of Sweden (SGU)(SwAM 2016). The European Framework

directive was adopted into Swedish law in 2004 and obligated the member states of the EU to reach "good status" in all water bodies by 2015 (Willquist et al. 2015). In the context of mining industry, these regulations are relevant in terms of the quality of wastewater from mining operations. If the discharged water quality from a certain operation is considered acceptable or not, is both based on directions of relevant regulations, but also on the water quality status of the water body where the water is discharged (e.g. Länsstyrelsen Västerbotten 2012).

2.3.3. Life Cycle Assessment

Life Cycle Assessment has already been mentioned briefly in this text. It commonly appears in the context of environmental issues and product analysis, and the term itself gives the reader a general idea of its application. However, it needs to be clarified.

In essence, an LCA produces a kind of environmental inventory of relevant inputs, outputs or "exchanges" during the life cycle of a product, activity or process. It further incorporates evaluation of the inventory data and estimates potential environmental impact based on the reported values (Norgate et al. 2007). It therefore acts as a great tool to depict the environmental burden of a certain object or process of interest, as all relevant components of its lifecycle are identified, quantified and weighed (Werder and Steinfeld 2000).

The ISO has devoted a standard of how LCAs should be implemented (ISO14040 series). Here they chose to define LCA as [a tool] for *"addressing the environmental aspects and potential environmental impacts (e.g. use of resources and the environmental consequences of releases) throughout a product's life cycle from raw material acquisition through production, use, end-of-life treatment, recycling and final disposal (i.e. cradle-to-grave)"* (ISO14040:2006).

The definition includes the term "cradle-to-grave", which is the general approach in LCA's. The life cycle of a product or a system is however often divided into the following three different stages: cradle to entry gate; entry gate to exit gate and exit gate to grave. In the context of metal mining, the first stage refers to the extraction of raw material and refining, the second stage to the manufacturing of the product and the "exit gate to grave"-stage corresponds to the use of the product as well as its recycling and disposal (Norgate et al. 2007). When WF or CF is reported in literature for primary metal mining and production, the two first stages are normally the single focus of the LCA. The last stage of the life cycle is often not reported, as this requires detailed data on recycling rates which is in general more difficult to access (Nuss and Eckelman 2014). This means that most studies report environmental data in a fused "cradle-to-gate" manner (Northey et al. 2013), which consequently has been the adapted approach in this report.

2.4. Restrictions

This study exclusively focuses on the fly ash fraction from ash residues of incinerated MSW, and completely excludes bottom ash. This choice was made partly based on the fact that fly ashes typically are enriched with Zn and Mg, compared to bottom ashes, and the value of zinc alone in Swedish fly ashes is estimated to a yearly value of 75 million SEK (Pettersson et al. 2013; Johansson et al. 2013). Fly ashes often

include toxic compounds such as heavy metals and chlorides to a greater extent than of bottom ashes (Tang and Steenari 2015), which is another reason to study the potential in purification and metal recovery from these ashes. Bottom ashes have been studied in the past and are already, to some extent, used as construction material in some countries (Allegrini et al. 2014; Lam et al. 2010). Fly ashes have great potential for similar purposes and deserve more attention.

Data has been collected without limitations to specific regions or countries, but when comparing and discussing potentials in metal recovery from fly ash the report holds a focus on current practices in Sweden. In general, special attention to the major production countries of the respective metals is given as this gives the best representation of the current environmental impact of mining and production activities globally.

In order to enable a fair comparison of water and carbon footprint for metals produced from mining respectively from fly ash, it is crucial to define the start and the end of the different production processes carefully so they are as conforming as possible. As stated above, the reported WF and CF of producing a pure metal is normally expressed using a "cradle-to-gate" approach. In this context, the "cradle" is the initiation of mining the ore, and the "gate" is considered the finished, pure product after it has been processed and treated through different processes. This logic is kept in this report, following the same cradle-to-gate approach. If data is reported in other manners it is discarded in order to avoid misleading results.

For the fly ash scenario, the boundaries of start and end of the different footprints are not as straightforward. As incineration of waste already is taking place, the fly ash itself is here considered the corresponding "cradle", i.e. incineration processes are not accounted for when estimating a carbon or water footprint. The end, or the "gate", is considered the final pure metal left from leaching, and following stages for metal extraction and purification (normally solvent extraction, stripping and electrowinning processes). The transportation of the remaining fly ash to landfills, after metal extraction has taken place, is not included either, as transport of fly ash to landfills is already a running procedure (Pettersson et al. 2013).

According to the ISO standard 14067:2013, a carbon footprint should be reported in carbon dioxide equivalents (CO_2e) (ISO 2013). This directive is followed in the report, thus data on only CO_2 or separate GHG's are discarded for the analysis.

For the WF analysis, no such stringent restrictions on data collection could be made due to the late standardization and varying ways of reporting WF up until recently. Therefore, all types of reported water footprints are collected for this study, and where background on methodology for WF reporting is given, this is noted in the tables of gathered data (Appendix II). This naturally means that the reported values for WF used in this report are not always harmonized. This issue is further discussed in the sources of error section.

Other environmental parameters such as toxicity, total land use, emissions of NO_x gases and particulate matter and such have not been included in this report due to limits in terms of time and data availability. Water use and GHG emissions were considered the most apt parameters to study, as CF is a good indicator of

environmental burden that people in general are familiar with, while WF is highly relevant as water is heavily consumed within mining industry and is a central issue that needs to be taken into consideration (Northey et al. 2013). Water footprint reporting is underrepresented in the metal mining and production context compared to CF, despite the fact that mining activities often occur in dry areas where water balance is especially critical (Peña and Huijbregts 2013). The increasing problems around groundwater depletion and water scarcity around the world (Hamdy et al. 2003) further enhanced the choice to study WF.

3. Method and data for literature review

The general methodology of this study has been to gather and harmonize data from the sources stated below (3.1.). Comparison and evaluation has been enabled by first defining reciprocal start and end stages for the different ways of producing metals (production from primary vs. secondary sources), followed by a small statistical analysis consisting of finding averages and standard deviations for the different metals and their CF's and WF's for respective production method (based on data from Appendix I-IV). Details on methodology and background when reporting these parameters have been noted where available. Only data that are reported based on a "cradle-to-gate" approach has been used in the report, both for CF and WF. Individual presentation and comparison of a selected number of current or potential future practices were made with respect to conventional mining of virginal material and current means of production, in order to get a better picture of the current research and practices in use for fly ash recovery.

The template that was used when collecting data on resource use for metal recovery, both from primary and secondary sources (see Appendix I-IV), included the following parameters:

- Author
- Metal
- Amount (WF = L/kg metal and CF = kg CO_2e/kg metal)
- Processes
- Comments
- Country

3.1. Data

Data on water use and CO_2e emissions for primary metal mining and production of Mg, Cu and Zn has primarily been collected through published papers in scientific journals that were accessed through Lund University's search tool LUBsearch and through Google Scholar.

The SimaPro8 software (PRé Sustainability, Amersfoort, Netherlands), which is a tool for LCA purposes and automatically gives access to the database Ecoinvent (version 3), has also been used extensively for the reporting of resource use in mining and production of the selected metals. The methodology used for CF reporting in SimaPro8 was the International Reference Life Cycle Data System (ILCD) created by the European Commission (European Commission 2010).

For data collection on water use and CO_2e emissions from secondary resources, i.e. when recovering metals from fly ash, scientific papers were used as a resource. However, the most valuable information was collected through personal contact with key persons within the field, who have published a number of papers in the topic and are experts on metal recovery from fly ash. The two major sources have been Karin Karlfeldt Fedje (Associate professor at Division of Water Environment Technology at the Department of Civil and Environmental Engineering at Chalmers University, Sweden and Researcher at Renova, Sweden) and Dr. Stefan Schlumberger (Centre of Excellence Hydrometallurgy, ZAR, Switzerland). Contact with these sources has been conducted through email.

The flow scheme of the bioprocess, depicted and described in the discussion (Fig. 7), is based on a preliminary concept-design of metal recovery at SP and created in consultation with Karin Willquist (Energy and Bioeconomy department at the Technical Research Institute of Sweden). This bioprocess is further supported by the sources given in the report of Willquist et al (2015).

Calculations:
The averages for CF and WF of primary metal production are based on all data given in Appendix I and II respectively. For secondary metal production, data given in Appendix III and IV act as the foundation for the results.

The average WF from secondary metal production for Zn was based on estimations by Schlumberger (2016) that in Switzerland about 30L water/kg Zn is needed, and that up to 5% of this water is lost through evaporation and landfilling of residues. Here the upper limit of 5% was chosen, resulting in the WF of 1.5 L/kg Zn given in Table 2. Water use for waste treatment at Swiss MSWI plants was given in the supplementary material of Boesch et al. (2014). Here estimations on water requirements per kg waste were made, and that 1ton of waste generates 20kg fly ash. The value 0.0013 (Table 2) is generated by using this data together with data from Schlumberger (2016), that from 1ton fly ash, 52 kg Zn can be recovered using the FLUREC process (assuming a Zn content of 6.5% and a recovery rate of 80%).

Table 2: Data on WF estimates and connected data to it needed for calculations of WF for different processes for secondary Zn production.

Author	Metal	Amount	Unit	Processes	Comments	Country
Schlumberger (2016)	Zn	1.5	L/kg Zn	MSWI plants	Assuming loss of 5% on evap. + landfill of residues	Switzerland
Schlumberger (2016)	Zn	52	Kg/ton fly ash	FLUREC	From 1 ton fly ash → 52 kg SHG Zn can be recovered	Switzerland
Boesch et al. 2014, suppl. Material	-	1	L water/kg waste	Incineration to FLUREC	From waste to end product. 20kg fly ash/ton waste	Switzerland
Boesch et al. 2014, suppl. Material	Zn	0.0013	L/kg Zn	FLUREC	Based on estimations from Schlumberger (2016)	Switzerland

The numbers given by Schlumberger (2016) regarding amount of Zn recovery per ton fly ash were also used when calculating CF in one case (Boesch et al. 2014 (suppl. material), Table 7). The estimation of CF for the study of Karlfeldt Fedje et al. (2014), given in the result section (Table 7) is based on a yearly production of 800ton Zn and 120MWh/year for Zn recovery process, which was data provided in the report. A Swedish electricity mix is applied for conversion into CO_2e.

Where conversion from GHG data to CO_2e has been necessary, GWP for the respective GHGs has been collected from IPCC's Annual Report 5 (Myhre et al. 2013).

4. Results

4.1. Comparison of CF and WF between primary and secondary sources

Figure 4 shows the results of the comparative analysis of carbon footprints between primary and secondary resources. The results show that in the case of Mg and Zn the footprint can be reduced by a factor 2 and 15 respectively. There is no data on Cu extraction from ashes. The results for the different metals and processes are presented in more detail below and data on averages and standard deviations are given in Table 3-6.

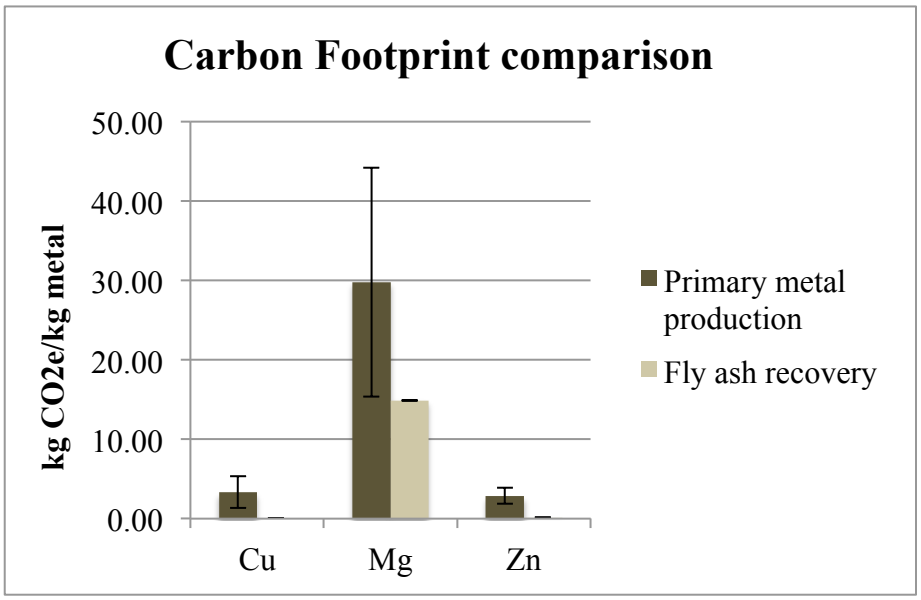

Figure 4: The graph illustrates the respective carbon footprints of Cu, Mg and Zn, both for metal production from primary (dark brown) and secondary (beige) sources and their standard deviations.

The corresponding comparison for water footprint is presented in Figure 5. It can be read from this graph that the WF of Zn recovery from primary resources is about 3.7 times larger than of Cu and Mg. For all the investigated metals, the WF is either negligible or not reported when it comes to metal recovery from fly ash, which is why no bars can be seen in the graph. Zn was the only metal that had some estimates on water use reported in literature, resulting in an average of 0.5L/kg Zn. For Cu and Mg only theoretical estimates of a net 0 WF were available. These estimates have not been examined in practice or published, but are accounted for as results and are presented further in the discussion section.

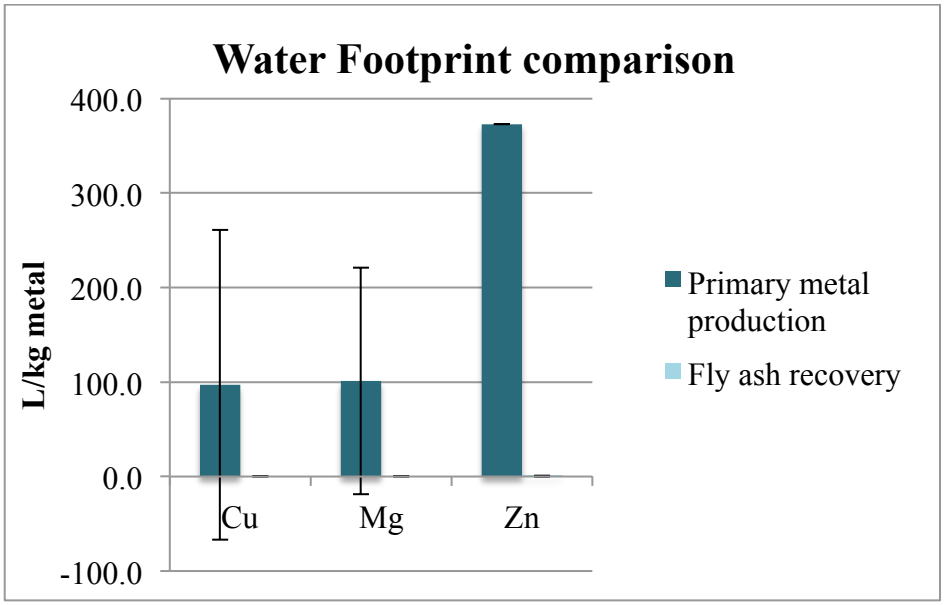

Figure 5: The graph illustrates the respective water footprints of Cu, Mg and Zn, both for metal production from primary (dark blue) and secondary (light blue) sources and their standard deviations.

4.2. CF and WF for primary metal production

In this section the published data on primary metal production, i.e. conventional mining from the Earth's crust and current means of production of metals, is reviewed. All the collected data relevant for this section are presented in Appendix I and II. These tables act as the foundation of the results and should be consulted for more detailed data.

Based on the averages of the CF for Cu and Zn presented in literature, seen in Table 3, there is no significant difference between the two metals. The CF for magnesium was found to be roughly ten times higher than for Cu and Zn, with a value of 29.8 kg CO_2e/kg Mg. The standard deviations for the investigated metals followed the relative size of the respective CFs quite well (Table 3). Carbon footprints for Cu production ranged between 0.4 kg CO_2e/kg Cu in Finland, up to 8.5 kg CO_2e/kg Cu for the mining company Palabora in South Africa (Northey et al. 2013). GHG emissions for Mg ranged between 5.4 kg CO_2e/kg Mg (Nuss and Eckelman 2014) up to 47 kg CO_2e/kg Mg in China (Ehrenberger et al. 2008), while for Zn values ranged between 1.0 kg CO_2e/kg Zn in Sweden up to 4.6 kg CO_2e/kg Zn in Australia (Norgate et al. 2007).

Table 3: The average carbon footprints and their respective standard deviations from primary metal production of the investigated metals

Average CF mining & production		
Metal	**Amount (kg CO2-eq./kg metal)**	**S.D.**
Cu	3.4	2.0
Mg	29.8	14.4
Zn	2.9	1.0

The number of sources that each of these averages are based on are presented in Figure 6, which clearly indicates that Cu reporting is far more common both in terms of water and carbon footprint compared to the other metals.

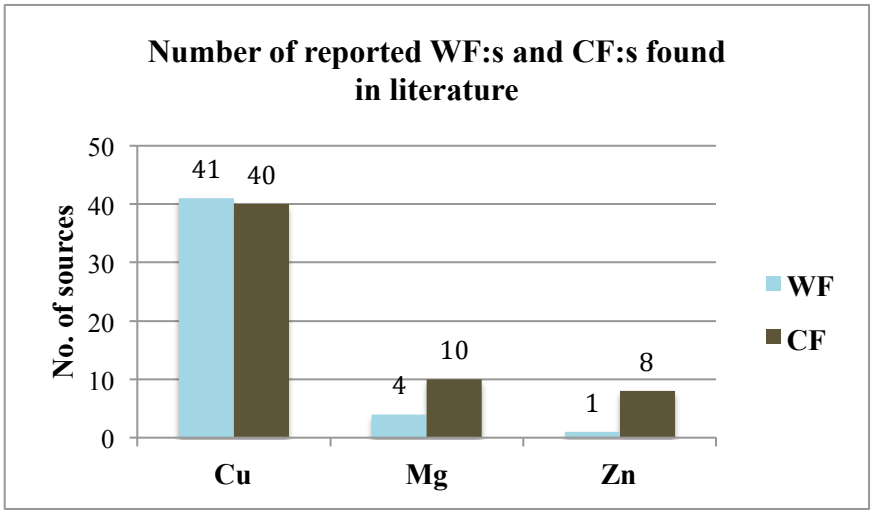

Figure 6: The amount of sources for CF (brown) and WF (blue) found in literature for primary metal production using a cradle-to-gate approach.

The water footprints for mining and producing metals from primary sources are given in Table 4. As only one WF was reported for Zn, no standard deviation could be found. The variation of WF between data sources for Cu, Mg and Zn are large, with standard deviations over 100L/kg metal (Table 4).

Table 4: The average water footprints and their respective standard deviations from primary metal production of the investigated metals.

Average WF mining & production		
Metal	**Amount (L/kg metal)**	**S.D.**
Cu	97.2	164.0
Mg	101.3	120.0
Zn	372.9	-

Looking only at Zn production (excluding mining) in China, pyrometallurgical (pyromet) processes used about eight times more water and emitted nearly five times more CO_2e than hydrometallurgical (hydromet) processes did (Xiao et al. 2003). Peña and Huijsbregts (2013) found similar trends, with pyromet practices having 2.4 times larger WF than hydromet practices for primary Cu production in Chile. In the case of Cu production, the opposite trend can be seen for GHG emissions, with hydromet processes having roughly twice as large CF compared to pyromet processes (Norgate et al. 2007, see Appendix I). No comparative statistical analysis of the variation of CF and WF between different processes were made due to lack of process-specific data for most reported CF's and WF's.

Northey et al. (2013) did a thorough analysis of environmental aspects of mining and production of Cu globally. The authors covered both CO_2e emissions from the different companies as well as WF, and had a strong focus on Australia. For the WF they chose to follow the method of Ridoutt and Pfister (2013) and found an average

WF of 74 L/kg Cu, which can be compared to the average WF for Cu in this study of 97 L/kg Cu.

The WF for Mg was found from Cherubini et al. (2008), where the authors compared two different ways of reporting water use: Material Intensity (MI) and Life Cycle Inventory (LCI), for four different mining methods. This resulted in one method generating much lower WFs (LCI), which might explain the high S.D. If the S.D. where calculated separately for the different methods, an S.D. of 120.2 for the MI and 14.4 for the LCI method.

4.3. CF and WF for secondary metal production

Data on CO_2e emissions from fly ash originated metal production is very limited. However, some results were obtained, presented in Table 5. For Zn, the CF averaged on 0.19 kg CO_2e/kg Zn, with values ranging from 0.0062 (Karlfeldt Fedje et al. 2014) to 0.33 kg CO_2e/kg Zn (Bjurström and Steenari 2003). For Mg a CF of about 15 kg CO_2e/kg Mg was estimated based on data gathered for this study. No data on CO_2e emissions per kg produced Cu from fly ash was found. In Table 6 the data found for water footprints of secondary metal production is given. Water footprint values for Cu and Mg are theoretical values given by Karlfeldt Fedje (2016), assuming the same WF's as for current practices for Zn recovery. The WF for Zn is based on three reported cases (Schlumberger 2016; Boesch et al. 2014; Karlfeldt Fedje 2016), see Appendix IV. The results for the different metals and processes are presented in more detail below.

Table 5: Averages of reported CFs and their respective standard deviations for recovering the investigated metals from fly ash.

Average CF fly ash recovery		
Metal	**Amount (kg CO2-eq./kg metal)**	**S.D.**
Cu	N.D.	N.D.
Mg	14.89	-
Zn	0.19	0.1

Table 6: Averages of reported WFs and their respective standard deviations for recovering the investigated metals from fly ash.

Average WF fly ash recovery		
Metal	**Amount (L/kg metal)**	**S.D.**
Cu	0	-
Mg	0	-
Zn	0.5	0.7

4.3.1. FLUREC process, Zn

The process of producing SHG Zn from fly ash at the plant in Zuchwil, Switzerland (using FLUREC) has a WF of close to 0 L/kg Zn (Schlumberger 2016). Minor losses (<5%) of water occur due to evaporation and landfilling of residues. Water required at Swiss MSWI plants is taken from rivers and is mainly used for wet flue gas treatment systems, while some water is needed for rinsing purposes and make-up of chemicals.

Over all, to produce SHG Zn using the FLUREC methodology, about 30L water/kg Zn is required. All water is however purified until the water quality is well in accordance with the legal limits and then fed back into the same river of origin Schlumberger 2016), which is why the consumptive use of water can be considered negligible and a net 0 WF can be assumed. The CF for Zn production in Switzerland has been reported slightly differently among the studied sources. According to Schlumberger (2016) the energy required for producing 1kg Zn using the FLUREC process is 3.6 kWh. This corresponds to 0.15 kg CO_2e/kg Zn if a Swedish electricity mix is assumed. In the supplementary material of Boesch et al. (2014) it is reported that the energy requirements for fly ash treatment is 387 kWh, when recovering metals using the FLUREC process. This would correspond to a value of 0.31 kg CO_2e/kg Zn (Table 7), i.e. about twice as much emissions, assuming a Swedish electricity mix and an overall Zn yield of 52kg Zn per ton fly ash (see method), which represent the current situation at the plant. The latter reported value (from Boesch et al. 2014), is however based on the maximal energy consumption of the process, while the value of Schlumberger (2016) represents the normal, average value, which is why these numbers differ (Schlumberger 2016).

Table 7: Data on which sources and values the CF of secondary Zn production is based on.

Author	Metal	Amount (kg CO2-e/kg metal)	Comments	Country
Karlfeldt Fedje et al. 2014	Zn	0.0062	Acidic scrubber water + local deposit	Sweden
Schlumberger and Beuhler 2012	Zn	0.15	FLUREC	Switzerland
Bjurström and Steenari 2003	Zn	0.33	Appendix K – acid process water + metal upgrade	Sweden
Boesch et al. 2014, suppl. mat.	Zn	0.31	FLUREC	Switzerland
Schlumberger 2016	Zn	0.15	FLUREC	Switzerland

4.3.2. Swedish research, Cu & Zn

Götaverken AB and Renova are currently underway performing a pilot project in Sweden where they try out a process similar to that of FLUREC, where leaching is done using acidic process water (mainly containing HCl) - with the exception that the final product is a lower grade Zn that is instead sent to metal-upgrading facilities that are already in place (Karlfeldt Fedje 2016). The WF for Zn production using this methodology would result in net 0, as no extra water is added and the wastewater is purified and fed back to the recipient, according to Karlfeldt Fedje (2016). No data on CF of this process has been estimated yet.

In Karlsson et al. (2010), a plant for Cu recovery from MSW fly ash was suggested using ammonium nitrate for leaching, followed by an SX-EW process. This method was compared in terms of CO_2 emissions and total cost to the current practice where Cu is obtained from Cu ore, and fly ash is sent directly to a landfill in Norway. The study found that approximately 450ton CO_2 emissions could be saved every year only by recovering Cu from waste instead of mining it, compared to current practices. Another 80 tons CO_2 emissions would be saved on reduced transport to landfills, if the rest product was put on a local deposit. The study did not report water use and did not take other GHGs into account, but only reported estimated CO_2.

As briefly described previously in the report, Pettersson et al (2013) did a similar study on Zn recovery, also using acid leaching from process water, were they introduced a few new scenarios of fly ash deposit and recovery methods as an alternative to conventional Zn management in Sweden. Two different MSWI plants were used for the experiments and calculations of environmental and economic gains with conventional and alternative methods. It was found that total CO_2e emissions could be reduced with up to roughly 1600 ton CO_2e annually for both plants, if Zn was recovered as zinc hydroxide, $Zn(OH)_2$ and upgraded in Odda, Norway, while the rest of the ash was deposited on a local landfill. This study included transports as well when estimating avoided CO_2e emissions, which does not follow the methodology of CF reporting chosen for this report (see Restrictions section).

4.3.3. Latrobe Magnesium, Mg

Latrobe Magnesium (LMG) claims that the WF for Mg recovery at their facility will be more or less negligible, as the water used in the process will be cleaned and fed back to the power station and re-used for the ash slurry (LMG 2014). LMG reported that the CF for Mg production would be approximately 50% of the CF of conventional way of mining and producing Mg today, which would correspond to a value of 14.89 kg CO_2e/kg Mg, based on the collected data. However, this Mg recovery method is not solely done through leaching, but on a combination of hydromet and pyromet treatment. The fly ash is also originating from brown coal industrial waste and not MSWI fly ash.

5. Discussion

5.1. Primary vs. secondary sources

From the results presented above, it is evident that both greenhouse gas emissions and use of water resources can be greatly reduced if metals are recovered from fly ash, instead of mined and produced from primary sources. This is valid for all studied metals. However, this report is limited by the fact that only two environmental parameters (GHGs emissions and water use) are considered. To acquire a full and fair comparison of the environmental burden between the two different ways of obtaining metals, a thorough LCA of all investigated cases would be preferable, where far more elements that impact on the environment are taken into account. For instance, in terms of metals extraction from fly ash, type and amount of chemicals, neutralizing agents, energy demand for electrowinning, toxic waste management and much more can have significant impact on the total environmental burden.

Furthermore, as the error bars in Figure 4 and 5 indicate, large insecurities of the reported values prevail; especially for the WFs of primary sources and for the CF of Mg (primary sources). Estimations of GHGs emissions and water use from secondary metal production are based on very few sources, which is further discussed in chapter six. Drastic assumptions of the environmental benefits from metal production from secondary sources should therefore be avoided until a more thorough analysis (such as an LCA) has been undertaken and analyses of statistical significance have been conducted.

5.2. CF & WF for primary metal production

The tenfold larger carbon footprint for magnesium production from primary mining, in relation to the other metals (Table 3), can partly be explained by the process that it is extracted using thermal processes, which is very energy demanding *per se* (Cherubini et al. (2008). Furthermore, the large majority of world Mg is mined in China, which mainly generate their electricity from coal (75% of total electricity in 2006-2010, The World Bank 2016). Ehrenberger et al. (2008) showed that by changing their main source of electricity from coal to gas, a reduction in GHG emissions could be reduced to as much as 45% only in China.

Given that WF for Zn production and mining in a cradle-to-gate approach was only reported in one database, doubt of representativeness may arise. However, this study covered 44% of the global Zn mining and 32% of global Zn production (EPLCA 2016), thus giving a fairly representative picture of the current situation. Compared to Mg and Cu, Zn had almost four times as high water use per unit produced metal (Table 4). This is another strong incentive on finding sustainable alternatives to the current practices of Zn production. No proper explanation of the notably higher WF for Zn production compared to Mg and Cu could be found in the studied sources.

Data on CF for different mining activities as well as for production of metals is more available than data on WF of corresponding activities. This is probably due to that GHG emissions are well known to have an intricate connection with global warming, thus CF acts as a good indicator of the environmental impact of an activity, making this parameter interesting to study. Furthermore, as previously stated, difficulties in correctly estimating water use from different sources (groundwater, surface water, evaporation etc.) prevail due to lacking standards of reporting (Berger and Finkbeiner 2010), which might explain the underrepresentation of WF reporting.

An evident need for a standardized way of reporting water footprints and what specific components to include in the term “water footprint” has been seen in the reported literature. The ways of reporting WF is varying significantly between the studied mining companies and databases as well as between techniques and mining plants (see Appendix II). However some trends can be seen such as pyrometallurgical practices tend to generate higher WFs compared to hydrometallurgical processes, based on the few sources giving data on this.

As there is now one existing standard of how to report WF in LCAs, developed by the ISO, future comparison of efficiency in water use and water management will hopefully be less troublesome. It would however be desirable to have industry-specific instructions on water footprint reporting, as interpretation and ways of applying the standard might differ between companies and geographical regions. Stakeholders with an interest in comparing environmental performance between companies would also benefit from knowing that resource management data is reported similarly (Cote et al. 2009). Naturally, detailed data on water management requires a range of measurements and thorough additional research, which might be very time consuming and costly. If however, it would be a legal requirement for all mining companies to follow the same standard of reporting LCAs, this would be a fair, universal “issue”, that all stakeholders, end users and decision makers would benefit from in the end.

The importance of selecting the right methodology for WF analysis was illustrated by Cherubini et al. (2008), where their comparison between Material Intensity (MI) and Life Cycle Inventory (LCI) for four different primary mining methods for Mg resulted in a S.D. an order of magnitude higher for the MI than for the LCI method. This effect is central when relating to the high standard deviation in general and when interpreting results assessed with different methodology.

In general there is more published literature on environmental impact of copper mining and copper production compared to magnesium and zinc mining/production (Figure 6). This might be partly explained by that copper is used to a wider extent in metal industry compared to magnesium and zinc (Johansson et al. 2013). For Cu, there was one statistical outlier among the reported WFs (1047 L/kg Cu compared to average of 97L/kg Cu). If this one was removed, an S.D. of 70.3 was generated instead of 164.0.

As was carefully explained by Northey et al. 2013 there is a variation of carbon and water footprint of primary Cu mining around the world. The authors chose to follow the method of Ridoutt and Pfister (2013) and found an average WF 25% lower than the reported average in this study (Table 4). As there was a strong focus on Australia in Northey et al. (2013), this might indicate that Australia holds stricter environmental laws for water management within the mining industry, maybe due to large parts of the country being classified as hot arid desert, according to the Köppen-Geiger climate classification (Peel et al. 2007), where efficient water management becomes critical.

5.3. CF & WF for secondary metal production

Over all, data access is extremely limited. Most data on CF and WF was found for Zn, possible due to the fact that fly ashes are normally enriched in Zn compared to other metals and bottom ashes (Johansson et al. 2013), thus a greater potential for recovery has been seen here. For Mg, only one reported case of CF and WF was found and in the case of Cu recovery from secondary sources, no CF was reported in literature. However, theoretical estimations on WF for Cu and Mg have been made by Karlfeldt Fedje (2016).

5.3.1. FLUREC process

The practices at the Swiss Zn recovery plant act as an excellent example of a business where environmental friendly practices can be economically profitable and natural resource depletion is avoided (Schlumberger and Beuhler 2012; Schlumberger et al. 2007). What needs to be kept in mind is that a successful concept in one country might not be equally successful elsewhere. Pettersson et al. (2013) raised differences between Sweden and Switzerland in terms of environmental laws, landfilling costs, fraction Zn in fly ashes as well as required amounts of import. Switzerland is exclusively dependent on import of Zn and does not produce any Zn except through recycling, while Sweden holds large reserves of zinc-containing ore (SGU 2008). As Sweden is more self-sufficient in terms of Zn supply, import rates are avoided to a greater extent and economic savings of recycling metals becomes less significant (Pettersson et al. 2013). The same authors state that it might be more profitable to purify low-grade Zn in Sweden rather than implementing the FLUREC technology, as there is already a well-developed mining industry with metal upgrade facilities in

place. These factors and many others need to be carefully considered before assumptions on economic and environmental gains can be made.

5.3.2. Swedish research

According to Karlfeldt Fedje (2016), the WF of Zn recovery would be negligible in the pilot project as acid process water from MSW incineration is used (following a similar method to FLUREC). In theory, the same techniques could be applied to Cu and Mg recovery from fly ash, resulting in a 0 WF for these metals as well (Karlfeldt Fedje 2016). This has however not yet been tested and confirmed in practice, and recovery rates might differ due to ash mineralogy, acid requirements (for desired pH) and chemical bond of the metal, etc., which might make this process unprofitable and not relevant for larger scale/production. An assumption of a WF of 0 for Mg and Cu should therefore be taken with care.

In the study of Karlsson et al. (2010) where yearly savings of 450 tons CO_2e were estimated from secondary Cu production, other parameters required for the method, such as process chemicals, contributed to an increased CF. This was estimated to account for up to 200 tons higher CO_2e emissions per year compared to the reference case. Still, a net saving of roughly 300 ton CO_2e/year could be gained using the suggested method, if the reduced transport is included. In order to make this method economically preferable to current practices, the recycling rates of the chemicals need to be >99%, and the recovery rates of Cu around 95%.

In the study of Pettersson et al. (2013) where an estimated 1600tons CO_2e could be saved annually using the suggested methods compared to current practices of attaining Cu in Sweden, the main savings of emissions were from avoided mining activities, and some from transport. Although these studies (Karlsson et al. 2010 and Pettersson et al 2013) gives a clear indication that large amounts of CO_2e emissions could be avoided if fly ash metals were recycled and the landfilling practices were changed, it still do not enable the desired comparison of this study, i.e. exactly how much emissions that could be avoided per unit produced metal, as the data is reported in CO_2e savings per year. The studies also include transport to landfills and use two specific WTE plants as reference cases, which make a conversion to the desired unit extremely complex.

5.3.3. Latrobe Magnesium

The reason that CO_2e emissions are thought to be reduced by roughly 50% compared to normal Mg production at The Latrobe Magnesium facility, Australia, is partly due to the feedstock of Mg being from magnesium oxide instead of magnesium carbonate, as many magnesium plants have, according to the company. However, it is not specified how this estimate is generated, and if it follows the same cradle-to-gate scenario as of this report. Still, this data was used as it refers to savings in terms of CO_2e emissions from mining and production, which seems to be in accordance with the restrictions of the report, however uncertainties remains. Furthermore, it was the only source that could be found covering environmental data on Mg recovery from fly ash, thus a selective pick of high-quality data was not possible.

As this is still not a running facility, even though several test studies has been made on efficiency and related costs, emissions and environmental burdens (LMG 2014), the reported numbers should be interpreted with great care until objectively validated

in full-scale. Furthermore, no published papers on the findings reported on the homepage of LMG could be found which further hampers the credibility of the reported data.

The potential of recovering magnesium from MSWI fly ash would be interesting to look at, as Mg demand is likely to rise due to its apt characteristics for lightweight products. However, the average Mg content in Swedish fly ashes is quite low (0.14%), and given that SHG Zn recovery probably is not profitable in Sweden (Pettersson et al. 2013), it might be an indication that so will be the case for high-purity Mg recovery.

5.4. Alternative methods for secondary metal production

5.4.1. Biometallurgy

When metals are leached from fly ash, a liquid product is created, which contains a range of toxic, organic and inorganic compounds. As sulfuric acid often is the acid used for metal leaching purposes (Tang and Steenari 2016), the liquid is typically enriched in sulfates. Today solvent extraction, stripping and electrowinning are common procedures that follow after leaching. The potential of other methods to replace certain steps in this standard procedure is readily being investigated in order to reach greater efficiency, generate less waste and have less negative environmental impact. One alternative that recently has caught much attention is the potential in using sulfur-reducing bacteria (SRB) for metal upgrade after leaching the fly ash. These anaerobic bacteria are fast growing, harmless and naturally occurring microorganisms that reduce sulfates into sulfides (bioreduction), which lead to metal precipitation (Muyzer and Stams 2008). The process uses bioethanol or other substrates (such as sludge, lactate, H_2/CO_2) to reduce the sulfate in the process water. This substrate however increases the chemical oxygen demand (COD) of the water and care must be made for high substrate conversion yields. After the biological treatment about 30% of the sulfates remain and 20% of the substrate, given that bioethanol is used (Liamleam and Annacchatre 2007). The metals are selectively precipitated using different pH, and the remaining water is purified until water quality standards are met. Thus, by using SRB, the contaminated water can be upgraded while valuable metals are made available for recovery (Huisman et al. 2006). This bioprocess is illustrated and explained in more detail in Figure 7.

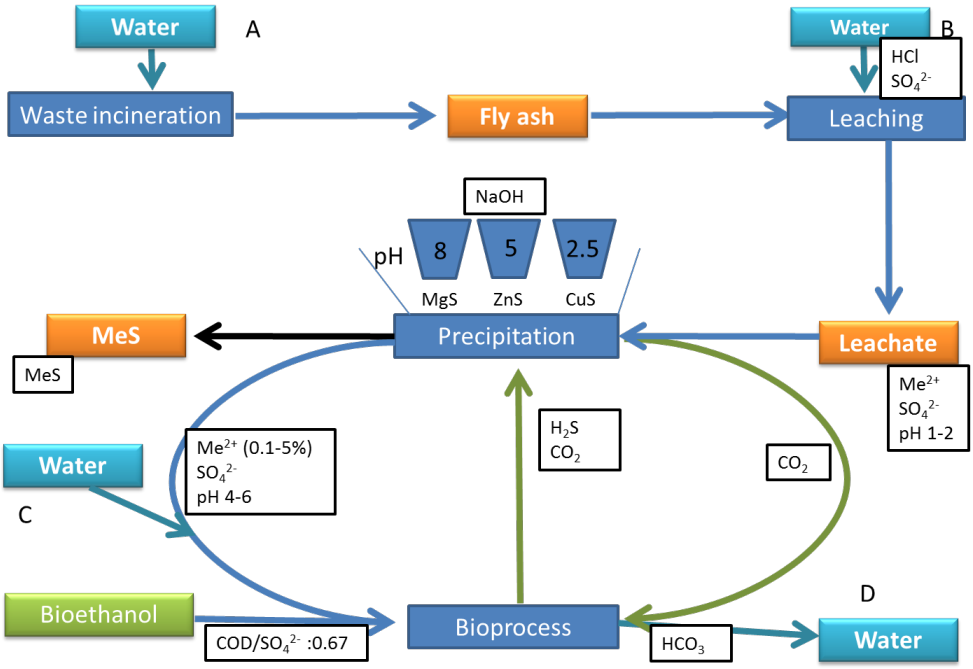

Figure 7: *Illustration of biometallurgical process using a sulfate reducing anaerobic bioreactor, based on a preliminary concept-design discussed at SP.* The fly ash from MSW incineration is leached with process water from incineration with possible addition of sulfuric acid (pH 1-2). The resulting metal solution (leachate) is then precipitated with biogenic hydrogen sulfide gas (H_2S; green line shows gas flow). The addition of the reduced H_2S results in a selective precipitation of Cu at pH 2.5, ZnS at pH 5 and MgS, complexed with other metals at pH 8 (black line) at a high yield (99.9-99.5%; Willquist et al. 2015). The remaining metal solution and sulfuric acid is pumped into the bioreactor which convert the oxidized sulfuric acid to the reduced gas H_2S and CO_2 by using bioethanol as carbon and energy source. CO_2 is formed during biological oxidation of ethanol. The water balance should be close to zero as the water output (D) will be recycled to give the water input (A-C). Possibly an additional membrane filtration step (not shown) step might be needed to reduce the salt concentration in the effluent water (D). Addition of water in A comes from the water scrubber system during gas upgrading in the incineration process. This water contains hydrochloric and sulfuric acid and can be used for leaching the metals so that the addition of water in B can be assumed to be zero (Karlfeldt Fedje 2016). Addition of water before the bioprocess can be necessary to dilute the acid and metal concentrations to tolerable levels for the SRB (Hao et al. 2014). Limestone or NaOH is used to increase the pH in the precipitation step. The H_2S and CO_2 produced in the bioreactor that is not consumed during precipitation can be re-circulated in the process and the produced CO_2 can be used as a buffer to increase the pH in the bioreactor, according to the suggested concept.

Biometallurgy using SRB is an attractive alternative to chemical reduction methods for several reasons including:

- High metal recovery rates (ranging from about 88%-99%) (Muyzer and Stams 2008);
- Higher metal selectivity (Huisman et al. 2006), which means higher market value;
- Bioethanol can be used as a substrate (electron and carbon source) needed for the microorganism to survive and reduce sulfate to hydrogen sulfide, where the CO_2 released during growth corresponds to the same CO_2 taken up by the plants used for bioethanol production, i.e. low CF. Ethanol has shown to have a high conversion yield (80%, Liamleam and Annacchatre 2007) and the carbon dioxide produced can be used to increase the pH during the precipitation step;

- Precipitation at lower pH reduces the need for chemical addition (limestone), which reduces climate impact as chemicals themselves have a carbon and water footprint (e.g. Karlsson et al. 2010);
- The bioprocess itself is a bioremediation and water management process that upgrades the process water from both inorganic and organic substances in the wastewater stream (Willquist et al. 2015).

However, there are some drawbacks with the bacteria, including sensitivity to parameters such as pH, temperature, liquid flow rate, sulfate concentration ratio and metal concentration (Hao et al. 2014; Willquist et al. 2015). The bioprocess itself takes up to 6-90 hours to be completed (Willquist et al. 2015), making it a relatively slow method. Moreover, when sulfates have been reduced to sulfides there is still a need of smelting or other treatment of the metal sulfides in order to get pure metallic compounds, which requires high temperatures and is energy demanding, which naturally would affect the CF and should be taken into consideration when estimating the environmental impact of the bioprocess.

As the bacteria are sensitive to metal and sulfate concentrations, the liquid product after leaching might need to be diluted to reach tolerable concentrations before biological treatment is possible (Figure 7, water C). However, if this water is purified to acceptable levels and fed back to the same water source or recycled, no water is consumed *per se*, thus a WF of 0 L/kg ash can be assumed.

5.4.2. BES – Bioelectrochemical systems

Electrowinning, which normally is used for metal recovery after leaching, is rather energy demanding (Pettersson et al. 2013). Therefore, Pettersson et al. (2013), among others, have investigated the potential in using bioelectrochemical systems (BES) as an alternative to this process for Zn recovery. These BES' are based on microbes converting the chemical energy of organic compounds into electricity (Pant et al. 2012). In the study it was found that by using BES, energy savings of about 40% compared to conventional electrolysis could be gained, with a Zn purity of 96%. Higher purities could probably be reached by increasing the time of the BES process and more energy could likely be saved by optimizing the design of the BES reactor (Pettersson et al. 2013). This would reduce the carbon footprint of secondary metal recovery further, thus BES' act as an interesting option for future recovery techniques.

5.4.3. Landfill mining

Landfill mining has in the past been used as a common term for many different operations. It has for instance been described as preservation or reduction of landfill space, energy or material recovery from landfills and elimination of pollution sources, to name a few (Krook et al. 2012). Lately landfill mining has however been more associated with material recovery from these deposits (e.g. Van der Zee et al. 2004). Seeing landfills as a source for material instead of an end-station of products is becoming increasingly relevant as environmental problems and raw material prices are increasing while natural reserves are declining (Krook et al. 2012). Krook and co-authors (2012) show that considering landfills as potential sources is valid by referring to a number of studies that have shown that vast amounts of metals are present in landfills today, where for instance the landfill stocks of Cu has been estimated up to nearly 400 million metric tons globally. Barriers in terms of

technology, high process costs, unknown metal concentration and distribution and unfavorable characteristics of the waste has however been prevalent in the context of landfill mining (Wagner and Raymond 2015). Still, as of today, some potential in landfill mining, or more specifically ashfill mining, exists using current techniques. Wagner and Raymond (2015) showed in their study that metals could be profitably mined and recovered from ashfills in the U.S., without any funding from the government. These ashfills however only make up a small fraction of landfill masses globally and contain a higher metal content than other landfill waste. A great challenge remains in making it beneficial to mine other fractions of landfills, i.e. raw waste with lower metal content, as this requires more processing thus higher costs (Wagner and Raymond 2015). However, great savings on the environment could possibly make up for the excessive economic costs of (raw waste) landfill mining and act as sufficient incentives for governments to fund these kind of mining projects in the future.

In sum, not only using future incoming waste as metal recovery sources, but also taking advantage of already existing metal deposits on landfills, is an interesting subject for investigation. This could in a best-case scenario both lead to avoiding degrading mining practices of virginal material, while saving natural resources and decreasing amount of occupied land by landfills.

6. Sources of error

The aim of the report was to give a comparable overview of the different ways of producing metals – from ore or from fly ash. Due to time and resource limits, no thorough analysis of the different ways of producing metals could be made, but an average of all practices of recovering the metals from ore were instead taken. As there can be large differences between e.g. pyrometallurgical and hydrometallurgical processes in terms of energy demand (Xiao et al. 2003; Norgate et al. 2007), taking an average can generate misleading result when estimating the WF and CF gains of a specific country. For instance, if Sweden principally imports Cu from a country where pyrometallurgical practices are used, the CF might be smaller, compared to the average value given in this study, thus less relative savings of greenhouse gas emissions could be made if implementing secondary metal recovery. It would therefore be highly desirable to know exactly where and what processes the metals used in Sweden are originating from. These details were however not obtained, despite many attempts to access this kind of data.

As previously mentioned in the text, inconsistencies in way of reporting water use in mining industry and production of all metals pervade. Variations occur to some extent for carbon footprint reporting too. The majority of the studied sources report emissions in CO_2 equivalents, but some articles only give data on CO_2 emissions. Where only CO_2 was reported, the values were exuded in the analysis, as they were not considered representative. The general scarcity of data on water footprint in this study can partly be explained by the fact that not all companies have a cradle-to-gate approach. Some companies/papers only give data of water use in terms of mining, others only consider the production.

In resource inventories, water use is at times only reported as ‘water’, and specifies that it includes all components of mining and production (e.g. EPLCA 2016), without

giving any further details on water use which makes it hard to evaluate. According to Cote et al. (2009), there is a general lack within the mining industry of reporting amounts of polluted water, source of water extraction, recycling rates as well as local water balance. These shortcomings might originate from the fact that many water-reporting methods do not require data on where water is taken from and where it is released, nor if or how much the water quality differs between input and output water (Berger and Finkbeiner 2010). These are severe shortages in methodology as water balance is of critical importance for the ecosystem of a watershed, and will be altered differently depending on what source it is extracted from. Likewise, water quality data is vital for assuring no human or environmental harm is imposed due to the activities in place. All these differences in data collection, along with differing mining and production practices, contribute to the very large standard deviation values seen in the results for WF in mining and production. The observed inconsistencies in WF reporting, the lack of data and the high standard deviations most definitely impact on the reliability of results, which should be taken into consideration by the reader.

In some reports (e.g. Karlfeldt Fedje et al. 2014) total CO_2e emissions from an activity has been based on gross energy requirement data and multiplied with a factor for national electricity mixes given by IPCC (IPCC/TEAP 2005) in order to convert MWh into CO_2e. As Sweden has a nearly six times higher factor compared to Switzerland (0.041 vs. 0.07), this might give misleading results. Where possible, conversion to Swedish electricity mixes has been made in order to make CFs from different studies and countries more comparable.

The database Ecoinvent v.3 was very helpful in terms of reporting CF for the different metals and provided several thorough datasets. There was however no suitable data for WF in any available methodology within the program. When using ILCD, the category ‘water resource depletion’ could be chosen. This category was however rejected as it generated highly unlikely values very different to those reported in literature. The ILCD handbook (European Commission 2010) was consulted, but no clear instructions on what or how data in this category was reported could be found.

The need to decide on a “cradle” and “gate” for the fly ash scenario that corresponded to the cradle-and-gate restrictions present in conventional mining and production was somewhat subjective. If incineration processes of the fly ash would have been taken into account, much higher CF for secondary metal recovery would most likely have been seen. This is however processes that are already in place, which is why this was excluded and has been motivated previously in the text. Transports to landfills were also excluded, which made comparison with environmental impact data from Karlsson et al. (2010) and Pettersson et al. (2013) difficult. This choice is also motivated in the text.

Lastly, for almost all described cases that currently or potentially could recover metals from fly ash, only one, or very few sources were found. Much information for the specific methods was based on personal communication with key persons or researchers behind a certain technique or experiment. In some aspects this can be preferable as it gives clear, relevant and updated information on the requested data. However, what is said has not been published and as in all cases, more sources increase the credibility of stated information.

7. Conclusions

Primary metal recovery, i.e. mining virginal material and producing Cu, Mg and Zn using current practices, exercise a greater environmental burden than of metal recovery from fly ash, both in terms of water use and GHG emissions. The carbon footprint of Mg and Zn production can be reduced by a factor 2 and 15 respectively if the metals are recovered from secondary sources instead of primary sources. No such comparison could be made for Cu, as no CF value was found for secondary production of this metal.

Regarding primary metal production, Cu and Zn had similar CFs with values of around 3 kg CO_2e/kg metal, while the CF for Mg was an order of magnitude larger than this. The noticeably high CF for Mg is likely a result of that around 85% of global Mg mining and production takes place in China where a large fraction of the electricity is coal powered, which contributes to large quantities of CO_2e emissions. Considering the substantial carbon footprint of primary Mg production, finding alternatives to current mining and production practices of this metal is particularly important. Incentives for more research on the potential of recovering Mg from secondary sources are strong. The average water footprints for the same processes and metals from primary sources were 97.15 L/kg Cu, 101.30 L/kg Mg and 372.9 L/kg Zn respectively. Uncertainties around these numbers however prevail, as standard deviations were notably high with values of more than 100L/kg metal. No suitable explanation could be found for the much higher WF for Zn.

Corresponding data on CF and WF for metal recovery from fly ash is scarce. This is partly explained by this being a relatively new field of research that incorporates new techniques which are not implemented in full-scale yet, with the Zn recovery facility in Switzerland being an exception. The few studies that did investigate large-scale implementation of successful laboratory attempts focused on economic aspects to a greater extent than environmental performance. In the case of Switzerland, the water footprint for Zn recovery using the FLUREC process can be considered approximately net 0 L/kg Zn, with small losses (<5%) of water through evaporation and landfilling of residues. From laboratory estimates carried out in Sweden, the WF for Zn recovery can also be considered negligible as the water used for leaching is purified to acceptable water quality standards and fed back to recipient, which in theory could apply for Mg and Cu too (Karlfeldt Fedje 2016).

With recovery rates up to 90% (Karlfeldt Fedje 2010) and 85% (Pettersson et al. 2013) for Cu and Zn respectively, large potential exists for secondary metal recovery in Sweden, using different leaching methods. By recovering metals from fly ash, savings in terms of GHG emissions, water and expenses could be made under certain circumstances. CO_2e emissions are mainly reduced by avoided mining activities and transport to landfills, if local deposits are used instead. Still, the current amounts of the investigated metals have yet not been proven profitable to refine to pure metals from fly ashes due to factors such as available techniques and facilities for metal upgrade and landfill taxes in Sweden.

In addition to comparing environmental performance of recovering metals from primary and secondary sources, technical and economic aspects are vital to study as

well, in order to indicate whether secondary metal production might act as a suitable complement to current metal production or not.

In a greater context, even though recycling of material to a large extent is vital for the well being of the planet and society, it cannot be seen as a definite solution to sustainability. The society as a whole needs to consume less and terminate the dependency of fossil fuels and limited natural resources in order to reach a more sustainable world. To achieve this state, world leaders must strive towards making "environmentally friendly" living effortless and profitable. This can for instance mean assuring better prices for products made with care for the environment and for the society. Moving towards a cyclic economy, where recycling of material and increased lifetimes of all kinds of products is beneficial both for end-users and manufacturers, is another central action in order to create a more sustainable world.

8. Future recommendations

As much data has been collected for this study, it would have been desirable to dedicate more time and resources for a more thorough analysis of the studied material, such as more statistical analyses and comparison between different countries and recovery processes. It would also be of great interest to include other environmental parameters such as land use, NO_x gas emissions, toxic waste etc., and most preferably economic aspects should be taken into account in the analysis as well. A thorough cost-benefit-analysis including both environmental, social and economic aspects would be an apt option.

For future studies in the field, it would be appropriate to make a similar comparison but for different metals. Metals listed as critical raw material would be especially interesting, as these are often less abundant in the Earth's crust, or considered exceptionally important for the economy or industry (European Commission 2015).

As magnesium had about a tenfold higher carbon footprint than of Cu and Zn, the need to find alternative ways of Mg extraction and production is urgent. More research on the potential of recovering Mg from secondary sources is strongly recommended, including analysis of both environmental and economic aspects. Investigation of which parameters contribute to the markedly larger water footprint for primary Zn production compared to Cu and Mg production would be of further interest, as this WF might be reduced if the responsible factors are identified.

The environmental performance when recovering ashes from bottom ashes would also be interesting to scrutinize, as these typically contain a greater fraction of metals than fly ashes do (with the exception of Zn, Mg and a few other metals) (Johansson et al. 2013), which possibly could result in less environmental impact per unit produced metal.

Better reporting and investigation of water use, GHG emissions and other environmental parameters when examining the potential in up-scaling laboratory findings of secondary metal recovery would also be desirable. Preferably it should be reported in a cradle-to-gate approach, as this is the most common way when reporting

primary metal production, which would facilitate comparison between primary and secondary metal production significantly.

More research on alternative ways of recovering metals from secondary sources and how to minimize energy use and environmental impact when doing so is encouraged. Biometallurgical processes using SRB, bioelectrochemical systems and landfill mining are all good examples and interesting topics for future research within this field.

Measuring and comparing environmental performance of a mining company would be a lot easier if all environmental parameters would be reported in the exact same way. Developing industry-specific directives on how to interpret and apply the ISO standard on LCA reporting would therefore be advantageous for all stakeholders.

Lastly, under all circumstances, it is vital to carefully evaluate the profits from mining activities and compare this to potentially lost ecosystem services and other negative impacts on the environment or the society. As opinions might be strongly contradicting among stakeholders, it is crucial to take all voices into account. Thorough analyses and research on potential effects on the environment and society from mining activities are strongly encouraged before mining operations are initiated, in order to make sound decisions for our future.

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Appendices

I. CF for metal mining and production

Copper:

Author	Emissions (kg CO2-eq./kg metal)	S.D.	Processes/mining company	Comments	Country
Norgate et al. 2007	3.3	0.006	Pyro (Smelting/converting and electro-refining)	LCA	Australia
	6.2	7.971	Hydro (Heap leaching and SX-EW)	LCA	Australia
Nuss and Eckelman 2014	2.8	0.333	Average of processes	LCA	Global
Ecoinvent v.3	3.6	0.049	Various processes		Australia
	5.8	6.043			Asia Pacific
	3.5	0.028			Latin Am.
	5.5	4.615			North Am.
	8.0	21.329			Global
	5.4	4.224	Electrolytic		Global
	6.6	10.662	SX-EW		Global
	2.1	1.702			Sweden
	6.6	10.539	Various	Various. 99.95% Cu. Electrolytic refining	Global
Northey et al. 2013	5.2	3.325	Cadia-Ridgeway		Australia
	2.3	1.159	Ernest Henry		Australia
	1.7	2.811	Golden Grove		Australia
	2.1	1.630	Mount Isa		Australia
	4.1	0.523	Northparkes		Australia
	5.0	2.635	Olympic Dam		Australia
	2.2	1.385	Prominent Hill		Australia
	1.2	4.738	Rosebery		Australia
	4.7	1.751	Telfer		Australia
	0.9	6.134	Highland Valley		Canada
	2.3	1.159	Kidd Creek		Canada
	1.0	5.649	Andina		Chile
	3.1	0.077	Codelco Norte		Chile
	2.5	0.769	Collahuasi		Chile
	2.0	1.895	El Soldado		Chile
	1.4	3.907	El Teniente		Chile

	1.7	2.811	Escondida		Chile
	1.1	5.183	Los Bronces		Chile
	3.1	0.077	Mantos Blancos		Chile
	2.3	1.159	Mantoverde		Chile
	3.5	0.015	Salvador		Chile
	3.9	0.274	Quebrada Blanca		Chile
	0.4	8.861	Pyhäsalmi		Finland
	2.8	0.333	Sepon		Laos
	8.5	26.248	Palabora		South Africa
	1.0	5.649	Cayeli		Turkey
	1.2	4.738	Ok Tedi		PNG
	4.3	0.853	Kennecott Utah		USA

Magnesium:

Author	Emissions (kg CO2-eq./kg metal)	S.D.	Processes	Comments	Country
Ehrenberger et al. 2008	47.00	296.87	Pidgeon	Conventional process, coal powered	China
Gao et al. 2009	36.60	46.65	Pidgeon	Coal fueled	China
	41.90	147.14	Pidgeon	Producer gas + coal	China
	34.10	18.75	Pidgeon	Coke oven gas, short transport.	China
Nuss and Eckelman 2014	5.4	593.90	Average of processes		Global
Cherubini et al. 2008	24.5	27.77	AM, electrolyctic		Australia
	10.4	375.20	Bolzano, thermal		Brazil
	13.8	255.04	Magnetherm, thermal		France
	42	149.57	Pidgeon, thermal		China
Ramakrishnan and Koltun 2004	42.00	149.57	Pidgeon		China

Zinc:

Author	Emissions (kg CO2-eq./kg metal)	S.D	Processes	Comments	Country
Norgate et al. 2007	4.6	3.2	Electrolytic		Australia
	3.3	0.2	Imperial smelting		Australia
Nuss and Eckelman 2014	3.1	0.1	Average of processes		Various countries
Ecoinvent v.3	2.8	0.0	Various processes		Global
Ecoinvent v.3	1.0	3.4	Several mines		Sweden
Werder and Steinfeld 2000.	3.1	0.1	Leaching + roasting + electrolysis		Central Europe
	1.5	1.7	Solar thermal (experimental)		Central Europe
EPLCA 2016	3.2	0.1	mining + pyro smelt (10%) & electro smelt (90%)	Represents 32% of world production, and 44% of global Zn mining (2005).	Various countries

II. WF for metal mining and production

Copper:

Author	Water use (L/kg metal)	S.D	Processes/mining company	Comments	Country
Peña and Huijsbregts 2013	96.0	1.3	Pyrometallurgy	Only blue WF	Chile
	40.0	3266.7	Hydrometallurgy	Only blue WF	Chile
EPLCA 2016	15.3	6700.2	Mining + hydro & pyrometallurgy	LCI. Ca 90% coverage of mining and production of copper sheet (2011)	Europe
	11.2	7388.2	Mining + hydro & pyrometallurgy	LCI. Ca 90% coverage of mining and production of copper tube (2011)	Europe
	12.9	7098.9	Mining + hydro & pyrometallurgy	LCI. Ca 90% coverage of mining	Europe

				and production of copper wire (2011)	
Northey et al. 2013	74.0	536.1	Various processes	Average value, LCI	Various countries
Northey et al. 2014b	1.4	9165.2	Ernest Henry	Method of Ridoutt and Pfister. 2013.	
	27.0	4923.1	Mount Lyell	Method of Ridoutt and Pfister. 2013.	
	1.5	9142.2	Golden Grove	Method of Ridoutt and Pfister. 2013.	
Mudd 2008	173.3	5798.1	Various techniques, lit. study	1.27 (ore throughput) + 172 (ore grade). Diff. ways of water reporting from companies	Various countries
Northey et al. 2013	49.0	2318.9	Cadia-Ridgeway	LCI	Australia
	42.3	3009.1	Ernest Henry	LCI	Australia
	33.0	4115.8	Golden Grove	LCI	Australia
	19.5	6030.3	Mount Isa	LCI	Australia
	74.0	536.1	Northparkes	LCI	Australia
	46.6	2555.8	Olympic Dam	LCI	Australia
	39.8	3289.6	Prominent Hill	LCI	Australia
	1046.9	902015.8	Rosebery	LCI	Australia
	161.1	4089.0	Telfer	LCI	Australia
	91.3	34.3	Alumbrera	LCI	Argentina
	135.4	1462.7	Highland Valley	LCI	Canada
	76.7	418.4	Kidd Creek	LCI	Canada
	99.4	5.0	Andina	LCI	Chile
	53.3	1923.3	Codelco Norte	LCI	Chile
	31.9	4258.2	Collahuasi	LCI	Chile
	48.2	2396.6	El Soldado	LCI	Chile
	139.7	1810.1	El Teniente	LCI	Chile
	52.5	1994.1	Escondida	LCI	Chile
	75.3	477.6	Lomas Bayas	LCI	Chile
	80.7	270.8	Los Bronces	LCI	Chile
	226.5	16730.2	Mantos Blancos	LCI	Chile
	46.6	2555.8	Mantoverde	LCI	Chile
	321.3	50241.0	Salvador	LCI	Chile
	21.9	5663.3	Quebrada Blanca	LCI	Chile
	211.0	12960.7	Pyhasalmi	LCI	Finland
	34.0	3988.5	Sepon	LCI	Laos
	94.4	7.6	Palabora	LCI	South

					Africa
	87.3	97.1	Cayeli	LCI	Turkey
	42.5	2987.2	Tintaya	LCI	Peru
	38.8	3405.3	Ok Tedi	LCI	PNG
	9.8	7630.9	Kennecott Utah	LCI	USA

Magnesium:

Author	Water use (L/kg metal)	S.D. (whole sample)	S.D. (MI)/(LCI)	Processes	Comments	Country
Cherubini et al. 2008	28.21	5342.51	24685.91	Bolzano, thermal	Material Intensity	Brazil
	156.3	3024.73	842.60	AM, electrolytic	Material Intensity	Australia
	191.9	8207.91	43.20	Magnetherm, thermal	Material Intensity	France
	364.9	69483.64	32246.28	Pidgeon, thermal	Material Intensity	China
Cherubini et al. 2008	5.84	9113.09	130.82	Bolzano, thermal	LCI	Brazil
	17.63	7001.09	0.12	AM, electrolytic	LCI	Australia
	4.92	9289.59	152.71	Magnetherm, thermal	LCI	France
	40.72	3670.24	549.55	Pidgeon, thermal	LCI	China

Zinc:

Author	Water use (L/kg metal)	S.D.	Processes	Comments	Country
EPLCA 2016	372.90	-	Mining + pyro & electrolytical	LCI. Represents 32% of world production and 44% of global Zn mining (2005)	Global
Xiao et al. 2003	16.24		Hydro	Only processing, mining not included	China
	130		Pyro	Only processing, mining not included	China

III. CF for metal recovery from fly ash

Author	Metal	Amount (kg CO2-e/kg metal)	Process	Comments	Country
Karlfeldt Fedje et al. 2014	Zn	0.0062	Acidic wastewater	800ton Zn/year, 120MWh/year	Sweden
Schlumberger and Beuhler 2012	Zn	0.15	FLUREC	Average of processes	Switzerland
Bjurström and Steenari 2003	Zn	0.33	Acid leaching	Appendix K	Sweden
Boesch et al. 2014 (suppl. material)	Zn	387 kWh	FLUREC	7.4 kWh/kg Zn (assuming 52 kg Zn/ton fly ash)	Switzerland
Boesch et al. 2014 (suppl. material)	Zn	0.31	FLUREC	Conversion based on data from Schlumberger	Switzerland
Schlumberger 2016	Zn	0.15	FLUREC		Switzerland
LMG 2014	Mg	50%	Thermal + hydro	Estimating a CF of 50% comp. to current mining and production of Mg	Australia

IV. WF for metal recovery from fly ash

Author	Metal	Amount	Unit	Processes	Comments	Country
Karlfeldt Fedje (2016)	Zn	0	L/kg Zn	Acid leaching + wash	Acid process water from incineration + purification	Sweden
Schlumberger (2016)	Zn	1.5	L/kg Zn	MSWI plants	Minor losses (<5%) on rinsing + chemical treatment	Switzerland
Boesch et al. 2014, supp. Material	Fly ash	1	L/kg waste	Incineration to FLUREC	From waste to end product	Switzerland
	Zn	0.0013	L/kg Zn	FLUREC	Based on estimations from Schlumberger (2016)	Switzerland
LMG 2014	Mg	0	L/kg Mg	Thermal + hydro	Not in practice yet	Australia

V. Fly ash data - recovery rates and concentrations

Recovery rates:

Source	Metal	Amount recovered (%)	Comments
Karlfeldt Fedje 2010.	Cu	90	Cu2+ ions, 3M NH4NO3 extraction (in conclusion)
Karlfeldt Fedje 2010.	Cu	95	Cu2+ ions, 2M H2SO4 stripping (paper 4)
Karlfeldt Fedje et al. 2012.	Cu	90	NH4NO3 and HNO3
Boesch et al. 2014	Cu	25	Acidic scrubbing
Tang and Steenari 2015.	Cu	50.9	Sulfuric acid
Tang and Steenari 2015.	Cu	68.2	Acidic leaching, hydrochloric acid
Tang and Steenari 2016	Cu	78.2	Acidic leaching, extraction, stripping, further processing (Av. Of ash A & B)
Yang et al. 2012	Cu	59.8	Acid leaching + electrodeposition
Tang and Steenari 2015.	Zn	80	Sulfuric acid
Tang and Steenari 2015.	Zn	80.8	Acidic leaching, hydrochloric acid
Tang and Steenari 2016.	Zn	78.1	Acidic leaching, extraction, stripping, further processing (Av. Of ash A & B)
Nagib and Inoue 2000.	Zn	35.3	Acid leaching
Johansson et al. 2013	Zn	85	Acid leaching from process water from incineration
Boesch et al. 2014	Zn	70	Acidic scrubbing

LMG 2014	Mg	95	Combined hydromet and thermal treatment

Concentration in fly ashes:

Author	Metal	Amount	Unit	Processes	Comments	Country
Johansson et al. 2013	Cu	0.22 +/- 0.09	%		Swedish ash (Average of fluidbädd and rosterpannor)	Sweden
Brunner and Rechberger 2015	Cu	3250	mg/kg (DS)	Swiss WTE facilities, from (Schlumberger and Bühler 2012).	1500-5000	Switzerland
Funari et al. 2014	Cu	952	mg/kg		330-5530 mg/kg in litt. (9)	Italy
Funari et al. 2014	Cu	2400	kg/a	Thermo recycling	Two incinerator plants. Flow FA. ± 50	Italy
Karlfeldt Fedje 2010 (ref to Chandler et al. 1997)	Cu	1900	mg/kg	Various techniques	600-3200	Various countries
Average	Cu	**2034**	mg/kg			
Johansson et al. 2013	Mg	0.14 +/- 0.02	%		Swedish ash (Average of fluidbädd and rosterpannor)	Sweden
Johansson et al. 2013	Mg	12000	mg/kg		11 MSWI stations, min 3900-max 22000	Various countries
Funari et al. 2014	Mg	78500	kg/a	Thermo recycling	Two incinerator plants. Flow FA ± 350	Italy
Brunner and Rechberger 2015	Mg	12000	mg/kg (DS)	Swiss WTE facilities, from (Schlumberger and Bühler 2012).	6000-18000	Switzerland
Avfall Sverige 2015:10.	Mg	14000	mg/kg			
Johansson et al. 2013	Mg	2655	ton/year		2012	Sweden
Average		**12667**	mg/kg			
Fedje 2010 (Chandler et al. 1997)	Zn	39500	mg/kg	Various techniques	9000-70000	Various countries
Brunner and Rechberger 2015	Zn	70000	mg/kg (DS)	Swiss WTE facilities, from (Schlumberger and Bühler 2012).	20 000-120 000	Switzerland

Funari et al. 2014	Zn	13.417	mg/kg		10-20,000 mg/kg in litt. (11)	Italy
Johansson et al. 2013	Zn	1.7 +/- 0.65	%		Swedish ash (Average of fluidbädd and rosterpannor)	Sweden
Schlumberger (2016)	Zn	6.5	%		Swiss fly ahes	Switzerland
Funari et al. 2014	Zn	36000	kg/a	Thermo recycling	Two incinerator plants. Flow FA. ± 200	Italy
Average		**36504**	mg/kg			
Boesch et al. 2014, supp. Material.	Fly Ash	20	kg fly ash/ton waste			Switzerland

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