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Assessment of Secondary Reserves of Nations: Aluminum, Copper and Zinc
(各国におけるアルミ、銅、亜鉛の二次埋蔵量の評価)

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ABSTRACT

As an outcome of global growth of economies and populations during recent decades, consumption of metals such as aluminum, copper and zinc have increased rapidly, creating vast quantities of metallic materials accumulated in human society. Based on the growth rate of primary production, important metals of aluminum, copper and zinc may be arrived in scarcity in the next about 50 years. Achieving sustainable management of metals demands consideration of not only primary materials in the natural environment but also secondary materials in our society as resources for utilization. This thesis applied our proposed classification framework of secondary resources to aluminum, copper and zinc to investigate the applicability of the framework and to assess the secondary reserves and resource of those metals in the selected major countries. For estimating secondary reserves, we introduced the variable "secondary reserve ratio": the fraction of in-use metal stocks that is technically and economically recoverable. In 2010, our estimates showed the United States has a large amount of secondary reserves of each metal (aluminum: 85 Mt; copper: 44 Mt; zinc; 13 Mt) and we found that those amounts are more than its primary reserves (20 Mt, 35 Mt and 12 Mt, respectively). In Japan, secondary reserves in the year 2010 total 32 Mt for aluminum, 14 Mt for copper and 19 Mt for zinc. The results showed that considerable amount of secondary resources of aluminum, copper and zinc are in landfills, which are potential targets of future extraction of secondary metals through landfill mining.

Our classification framework can provide information about potential short-term and long-term availability of secondary resources of three important metals and can be used for developing waste management and urban mining strategy. It also highlights the need

for integrated management of primary and secondary resources toward sustainability use of aluminum, copper and zinc.

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LIST OF ABBREVIATION

CNMIA	China Non-Metallic Minerals Industry Association
GHG	Greenhouse gases
GDP	Gross Domestic Products
IAI	International Aluminum Institute
ICSG	International Copper Study Group
ILZSG	International Lead and Zinc Study Group
IEA	International Energy Association
ICA	International Copper Association
IWCC	International Wrought Copper Council
PRR	Potential Recyclability Rate
SRR	Secondary Reserve Ratio
UN Comtrade	United Nations International Trade Statistics Database
UNEP	United Nations Environment Programme
WBMS	World Bureau of Metal Statistics
WIO-MFA	Waste Input-Output Material Flow Analysis

1. Introduction

1.1. Sustainability of metals

The word ''sustainability'' has come to mean many things from different views, making it nigh impossible to have a universally agreed definition. In this study, based on definition (Brundland Commission 1987; UN Commission on Environment and Development 1992), sustainability is used in the meaning ''a sustainable metal use meets the needs of the present without compromising the ability of future generations to fulfill their own needs.'' Since pieces of native copper were first hammered into simple tools about 6000 BC, metals have been important part of human activity (Verhoef 2004). Nowadays, metals become essential for the production of almost all products such as automobile, aircraft, computer, or home appliance. National Resources Canada (NRC 1997) reported that current technology of electrical power supply is dependent on copper and aluminum.

With large percentage of savings in energy and reduction of waste compared to primary processing (primary metals), metals can be recycled countless times (secondary metals). For that reason, primary and secondary metals should be included in any consideration of sustainability. Graedel and Cao (2010) stated that sustainability of metals is related to both mineral virgin ore deposits and to the eventual recycling of metals. Maung et al. (2017a) also pointed out that the sustainability of metals demands consideration not only of primary metals in the natural environment but also of secondary metals in society as alternative resources. Using secondary metals slows down the depletion of metal resources, decreasing the demand of primary metals. Extending the life of metal resources is a key element to the concept of sustainability of metals.

1.2. Global metals production

Metals are used almost everywhere around us; there are few products where metals are missing or have not played a important role in their production as they have specific properties that become the foundation of our industrial society. Along with significant population and economic growth of developing countries, a significant increase in global production of key metals continues the trend of the past half-century as shown in Figure 1.1. Economic growth increases the amount of metal uses in our societies. In Figure 1.1, the metals covered are common ones of economic importance: steel, aluminum, copper, zinc, arsenic, cadmium, chromium, gold, lead, mercury, and nickel (USGS 2012). If compared to about 700 million tons in 1980, it surged about 2 times in 2010 to a record about 1.48 billion tons. Figure 1.1. shows how was the situation of global metal production in the last five decades. As a consequence of urbanization and new
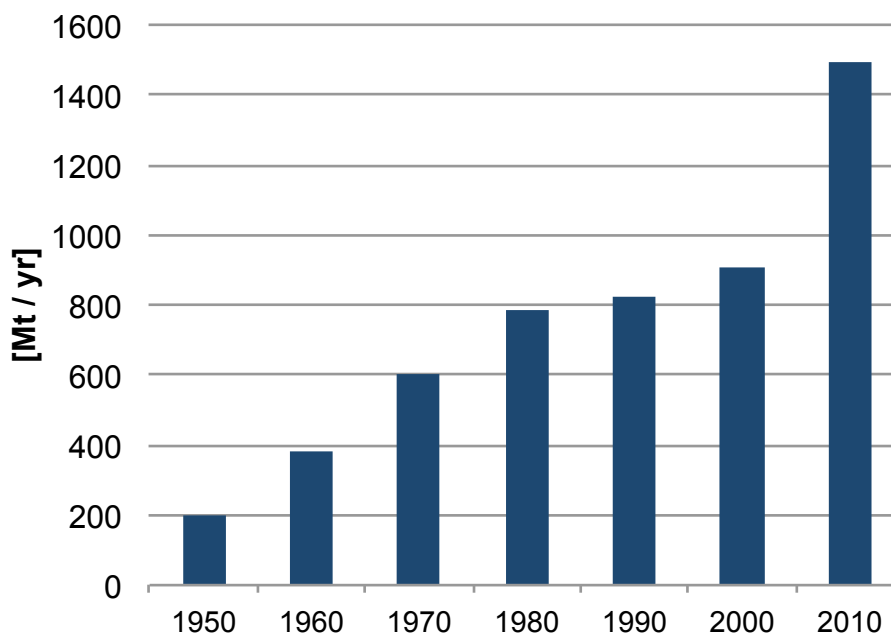
Figure 1.1 World metals production, 1950-2010

infrastructure construction in developing countries, transition in energy technologies and potential rate of electronic product usage, global demand of metals is likely to continue rising further in the future (Van der Voet et al. 2013).

1.3. Role of Aluminum, Copper and Zinc in the industrial society

For manufacturing industries, sustainability and economic growth of nations, non-ferrous metals such as aluminum, copper or zinc are very important. These metals are essential for many products in the construction, mechanical engineering, transport, aerospace, packaging, electricity and energy, electronics and medical devices sectors because of unique characteristics such as low weight (aluminum), high conductivity (copper) or resistance to corrosion and non-magnetic property (zinc). Availability of aluminum, copper and zinc has ever been the basis for economic growth and well-being in society. Traditionally, aluminum, copper and zinc were mostly extracted from natural mineral deposits. Over the past decades, primary production of aluminum, copper and zinc
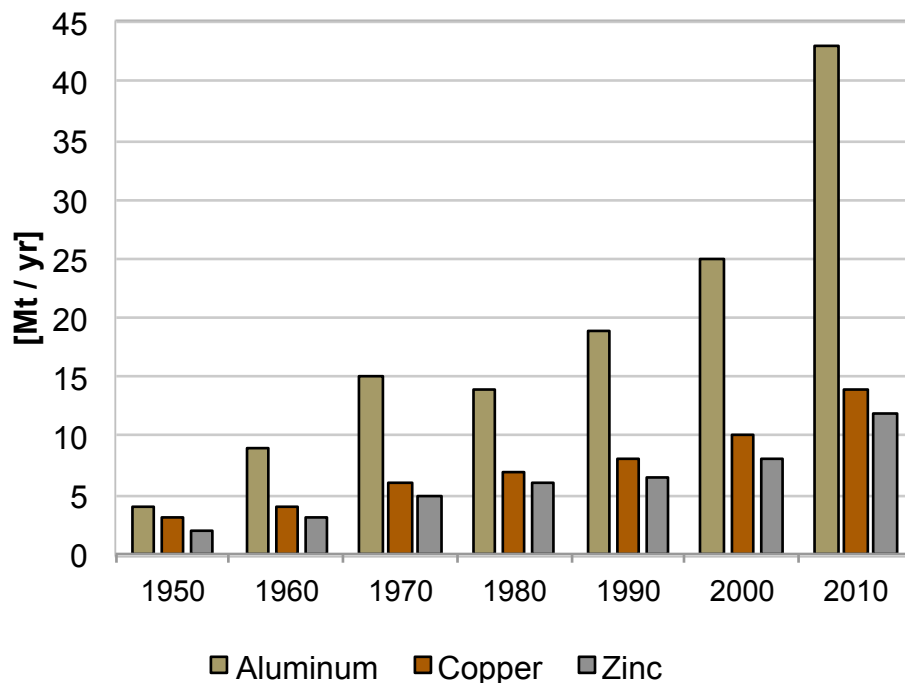
**Figure 1.2 Global production of aluminum, copper and zinc
(IAI, 2015a; ICSG, 2015; WBMS, 2016)**

increased and transferred a considerable amount of aluminum, copper and zinc from the lithosphere to the anthroposphere including their in-use stocks. Historical global primary production for top three most used non-ferrous metals (aluminum, copper and zinc) are shown in Figure 1.2. Global primary production of copper and zinc has more than doubled during the past 30 years, aluminum production more than tripled in the same period. Over the past few decades, global extraction growth rate for aluminum, copper and zinc averaged about 5, 3.4 and 3.4 percent annually. Gordon et al. (2006) found that providing today's developed world technological services for the world's population would appear to require essentially complete extraction of copper ores and essentially complete recycling of copper exiting use. After their lifetimes, anthropogenic stocks of aluminum, copper and zinc go along with generation of EoL scrap from these stocks and
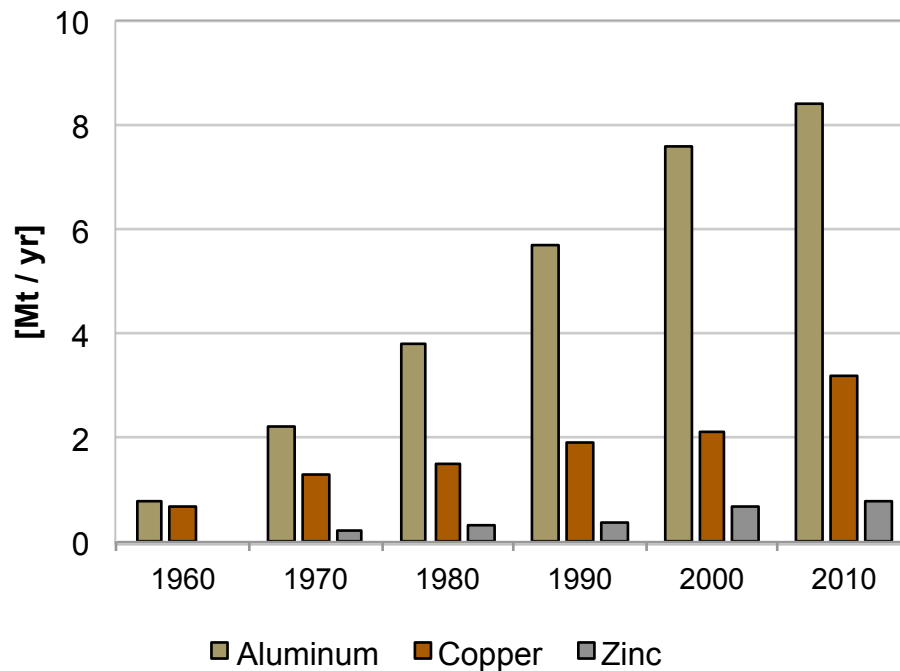
Figure 1.3 Global secondary production of aluminum, copper and zinc (WBMS, 2014; ICSG, 2015; ILZSG, 2016)

availability of secondary metal resources increased considerably. Primary production of aluminum, copper and zinc increased by a factor 3, 2 and 2, respectively while their secondary production increased by a factor of 2 during the past 30 years as shown in Figure 1.3. Secondary production of metals is a very important fraction of global metal production with huge amount produced annually from recycled metals.

With consumption of primary resources, the amount of secondary resources in the technosphere rises continuously. Through secondary production from the technosphere, that provides the possibility for potential replacement of primary resources from the natural environment. Using secondary production of aluminum and copper not only saves finite resources from the natural environment but also offers the possibility of energy saving as show in Table 1.1 (Wright et al. 2002). However, production of secondary zinc is estimated to be about 10% more energy intensive than its primary zinc (Wright et al. 2002).

Table 1.1. Comparison of the energy required (GJ) for 1 ton from primary and secondary sources.

Metal	Aluminum	Copper	Zinc
Primary	250^{*}	68.0^{**}	16.4^{ϕ}
Secondary	15	18.0^{***}	$18.0^{\phi\phi}$

* Based on production from Bauxite

** Based on cathode quality copper. Concentrate derived from open pit mining, shipped to a smelter using a flash smelting technology.

*** Based on cathode quality copper

$^{\phi}$ Based on a average production of 85% from electrolytic zinc, 15% from the Imperial smelting furnace

$^{\phi\phi}$ Based on treating a 35% Zn electric arc furnace dust in a Waelz process

For secondary production, the energy requirement of collecting and preparing the EoL scrap is significantly less than that of primary production (Kellogg 1977; Reuter 1998; Szekely 1996). Thus, production of secondary metal has very favorable energy consumption per ton of metal produced and lower greenhouse gas production than primary metal production. In recent years, serious issues such as social and commercial exploitation of miners, and ecological collapse in some regions have dogged the mining industry. In addition, mining primary metals is water-and energy-intensive and requires large amount of land area, contributing to high environment impact.

1.4. Primary sources and availability of aluminum, copper and zinc (Life expectancies of world reserves)

By definition, primary metals resources are non-renewable and finite. At the beginning of the 1970s, The Club of Rome first drew attentions to the depletion of resources. It was assumed various metal ores would be exhausted within a few decades. In fact, this turned out not to be turn because of some reasons such as discoveries of new deposits, technological advances and etc. However, sooner or later, at the current rate of metal consumption, primary reserves of aluminum, copper and zinc will be exhausted. From a long-term perspective, the continuation of the growing consumption trend is problematic. Resource scarcity and the environmental impacts of metals use might eventually limit growth (Dawkins et al., 2012). Even if primary metals' actual availability is not an issue, environmental and social costs associated with production and uses will restrict the future use (Tilton 2002).

Using data from USGS (2012), Stuermer and Schwerhoff (2013) estimated the availability of aluminum (138 years), copper (43 years) and zinc (21 years), based on reserves divided by annual production.

Alonso and his colleagues (Alonso et al. 2007) examined that for current copper and zinc reserves, the time of depletion could arrive in as few as 50 years. Based on the growth rate of global bauxite production and per-capita consumption, Meyer F. M. (2004) concluded that currently known reserves and the reserve base of high quality bauxite ores will be exhausted within the next 50 years. Kesler (1994) pointed out that zinc's depletion time (reserves/annual use) is no more than 20-30 years. Due to the closing of large mines and the difficulty to find new ones, potentially zinc might be supply-limited (Deaux and Matthew, 2015).

1.5. Urban mining for secondary metals

As the consequence of human activities, we are building up metal stocks that are urban mines in the techno-sphere. Metal scarcity signifies a need for transition on the sustainable use of metals which demands consideration not only primary metals in the natural environment but also of secondary metals in society as alternative resources. This is where urban mining concept comes into play. Around 1980s, Nanjyo (1987) reported that rare earth metals' contents of industrial products often exceeded the grades of raw ores, and they were in the form of refined metals in many cases, so that their reuse demanded less than the massive amount of energy needed to smelt and refine crude ores. Nishiyama (1993) pointed out that the amount of many metal resources that had already been mined exceeded the known primary reserves. Halada et al. (2008) also noted that the consumption of metals will be several times higher than the present metal reserves until the year 2050. Metals are intensively recycled materials. Yoshida & Yoshida (2011) found that urban mines were part of the Japanese move to a circular economy, reducing the amount of virgin material use and promoting more sustainable environment.

Promoting and planning future urban mining requires elucidation of where metals are used, stocked, and lost through their life cycle in society and requires estimation of how much of the in-use stocks might be recovered as secondary resources through recycling. What is the amount of resources that can be recycled by urban mining? In fact, this question has not really been addressed. From the perspective of the suppliers of metal resources, in particular, the metal stocks in society (i.e., artificial or urban mines) will become more important than those of conventional mines (Gordon et al., 2006; Kapur and Graedel, 2006). Previous estimates of in-use stocks of metals have revealed the potential availability of secondary metals (UNEP, 2010). However, these estimates merely indicate potential because the metals are still in service and not all stocks are recoverable.

From this perspective, Hashimoto et al. (2017) proposed a classification framework of secondary resources accumulated in society as well as in the environment, based on a classification of natural resources, that is, a McKelvey diagram. This framework provides useful visualization of available secondary resources classified by different degrees of knowledge related to stocks and economic recoverability. Secondary reserves are the portion of secondary resources that are technologically and economically available for reutilization. Currently, knowledge related to secondary reserves is very limited, but extensive assessments of primary reserves are available.

1.6. Literature review on previous studies of aluminum, copper and zinc

From the perspective of the suppliers of metal resources, in particular, the metal stocks in society (i.e., artificial or urban mines) will become more important than those of conventional mines (Gordon et al., 2006; Kapur and Graedel, 2006). The current

material stocks in society are likely to become either waste or secondary resources in the future. Brunner (1999, 2004) pointed out that most materials that have been exploited in the past are still in use or are "hibernating" in the anthroposphere. Information about the stocks is vital to accurately estimate the future outflow from current stock and to create strategies for waste management and resource reuse. In perspective of a more sustainable use of aluminum, copper and zinc, a better management of anthropogenic stocks becomes compulsory nowadays.

Over the past decades, various researchers have examined aluminum flows and stocks on global, regional and national scale about 40 anthropogenic cycles (Chen and Graedel 2012).

Table 1.2 Previous research on aluminum flows and stocks

Authors	Region & Time Frame	Distinctness
Melo, 1999	Germany 1986-2012	Using fixed lifetime for aluminum finished products
Dahlstrom et al., 2004	UK 1958-2001	Value chain analysis
Bruggink and Martchek, 2004	Global 1960-2020	Recycling and its impacts
USGS, 2005	USA 2002	
Murakami, 2006	Japan 2000	Aggregated metal cycle
Hatayama et al., 2009	Japan, USA, Europe, China 2000-2050	Recycling potential
Jang et al., 2009	Korea 1980-2007	7 cities and 9 provinces

Rauch 2009	World 2000	High correlation of metal stocks with GDP by area
Wang and Graedel, 2010	China 2000-2005	Separation of urban and rural residential
McMillan et al., 2010	USA 1900-2007	Relationship between model results and GDP
GARC, 2011	Global 2007	
Liu and Müller, 2012	Global	Review on LCA
Chen and Graedel, 2012	USA 1900-2009	Comparison of top-down and bottom-up stocks
Chen and Shi, 2012	China 1950-2009	Both stock and flow analysis
Yue et al., 2012	China 1975-2010	Content of social stocks
Billy, 2012	France 1950-2100	Comprehensive data set
Cullen and Allwood, 2013	Global 1950-2050	By end- use goods
Liu and Müller, 2013	Global 1900-2010	Divided by world regions. Includes a comparison to GDP growth
Ciacci et al., 2013	Italy 1947-2009	market share
Buchner et al., 2014	Austria 2010	Breakdown of stocks and scraps

In the earlier studies, copper has been one of the most widely analyzed, with more than 90 anthropogenic cycles available (Chen and Graedel 2012).

Table 1.3 Previous research on copper flows and stocks

Authors	Region & Time Frame	Distinctness
Zeltner et al. 1999	United States 1900-2100	Sustainable copper management
Graedel et al. 2002	Europe 1994	Technological cycle
Spatari et al. 2003	Europe 1994	Regional material flow model
Bertram et al. 2002	Europe 1994	Waste management subsystem
Ayres et al. 2003	United States	Life cycle perspective
Kapur et al. 2003	Asia 1994-1998	Per capita generation of waste
van Beers et al. 2003	Africa 1994	One year stocks and flows
Vexler et al. 2004	Latin American and Caribbean region 1994	Tracing the flow
Graedel et al. 2004	56 countries 1994	Anthropogenic resource cycle
Spatari et al. 2005	North America 1900-1999	Historical inventories of copper stocks and flows
Kapur 2006	India 2000-2100	Scenario of future copper use
Gordon et al. 2006	USA 1900-2000	
Daigo et al. 2007	Japan	Copper and its alloy

	1970-2005	
van Beers and Graedel 2007	Australia	Using GIS inventory
Guo and Song 2008	China 2004	1-year life cycle
Wang et al. 2008	China 1994-2004	One-year snap shot cycles
Daigo et al. 2009	Japan 1950-2005	Dynamic material stock and flow analysis
Gerst 2009	Global 1990-2100	Divided by developing and industrialized regions, 4 scenario analysis
Takahashi et al. 2009	Japan, India, South Korea, Malaysia, Singapore, Taiwan, Thailand, Vietnam, Sri Lanka 1996/1997	Calibrated with previous statistical studies
Rauch 2009	World 2000	High correlation of metal stocks with GDP by area
Yue et al. 2009	China 1995-2005	Copper product lifecycle and its losses
Tanimoto et al. 2010	Brazil 2005	Brazil copper life cycle
Glöser et al. 2013	Global 1910-2010	Dynamic model
Bonnin et al. 2013	France 2000-2009	Special focus on waste stream
Zhang et al. 2014	China 1975-2010	By end-use

Until recently, more than 70 studies on anthropogenic zinc cycle have been conducted (Chen and Graedel 2012).

Table 1.4 Previous research on zinc flows and stocks

Authors	Region & Time Frame	Distinctness
Jolly, 1992	USA 1850-1990	State by state
Spatari et al., 2003	Europe 1994	1 year stock and flows
Gordon et al., 2004	Global 1994	EoL zinc disposed in landfill
Van Beers and Graedel 2007	Cape town, South Africa 2000	Using GIS data
Graedel et al., 2005	54 countries 1994	
Harper et al., 2006	Latin America and Caribbean region 1994	Discard stream losses for future use
Tabayashi et al. 2009	Japan 1970-2005	Detailed end-uses statistics
Rauch 2009	World 2000	High correlation of metal stocks with GDP by area
Graedel and Cao, 2010	49 countries 2000	Trade index
Yan et al., 2013	China 2004-2020	Extrapolating method
Daigo et al., 2014	23 countries 2010-2050	Future demand by scenario
Meylan and Reck, 2016	49 countries 2010	Global values

However, none of those earlier studies focused and discussed secondary reserves of aluminum, copper and zinc that are technologically and economically recyclable stocks. For addressing these issues, we applied the classification framework of secondary resources proposed by Hashimoto et al. (2017) to aluminum, copper and zinc for estimating secondary reserves in major countries.

1.7. Purposes of the study

1.7.1 Aim

The main goal of this thesis is to apply the classification framework of secondary resources to aluminum, copper and zinc for quantifying the secondary reserves and resources in major selected countries and the globe.

1.7.2 Specific objectives

The following objectives tackled in this thesis:

1. To investigate the applicability of the framework for aluminum, copper and zinc

2. To assess the secondary reserves and resources for those metals

1.8. Organization

Following the introduction presented in the current chapter 1 including research background, research objectives and structure of thesis, chapter 2 presents methodology with existing classification framework of secondary resources and improvement in the classification framework of secondary resources, which is the foundation of this work. Chapter 3 investigates the assessment of secondary aluminum reserves and resources of nations, applying the classification framework of secondary resources. This manuscript has been accepted in *Resources, Conservation and Recycling* (Maung et al. 2017b). Chapter 4 examines the assessment of secondary copper reserves and resources of selected major countries. Chapter 4 was published in

Environmental Science & Technology (Maung et al. 2017a). Chapter 5 also investigates the assessment of secondary zinc reserves and resources of selected countries using the classification framework of secondary resources. This manuscript has been under review of *Journal of Industrial Ecology.* Chapter 6 offers the discussion, the overall insights and conclusions reached in combining the findings and closes with overall conclusions including a look into the aims and achievements of this study presented.

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Chapter 2

Methodology and Data

2.1 Existing classification framework of secondary resources

Fig. 2.1 shows the mineral resource classification adopted by the United States Geological Survey (USGS). The horizontal axis represents geological knowledge and the vertical axis show profitability on costs of extracting and material in a given economy at a given time. In this classification system, resources with high profitability are called "reserves." Other occurrences in the classification framework represent "materials that are too low grade or for other reasons are not considered potentially economic" (U.S. Bureau of Mines and U.S. Geological Survey, 1980).

Geological Knowledge

Cumulative Production	IDENTIFIED RESOURCES			UNDISCOVERED RESOURCES	
	Demonstrated		Inferred	Probability Range	
	Measured	Indicated		Hypothetical (or)	Speculative
ECONOMIC	Reserves		Inferred reserves		
MARGINALLY ECONOMIC	Marginal reserves		Inferred marginal reserves		
SUB-ECONOMIC	Demonstrated sub-economic resources		Inferred sub-economic resources		
OTHER OCCURRENCES	Includes nonconventional and low-grade materials				

Profitability

Table 2.1 Classification of mineral resources by the USGS (McKelvey, 1972; U.S. Bureau of Mines and U.S. Geological Survey, 1980)

Based on the normal classification of mineral resources (the so-called McKelvey diagram) (McKelvey, 1972; U.S. Bureau of Mines and U.S. Geological Survey, 1980) as a reference, Hashimoto et al. (2017) proposed the classification framework of secondary resources as shown in Fig. 2.2, assuming that the material stocks in society are artificial or urban mines and when considering their reuse. With similar concepts for the horizontal and vertical axes, the horizontal arrow indicates the level of knowledge: we have more knowledge of the amount of "final products in/after use" stocked in society than the amounts of "wastes in managed landfill sites" or "dissipated material," more knowledge of the amount of wastes or secondary resources that are likely to emerge in a year than the total amount of "final products in/after use" stocked in society. The vertical arrow indicates the possibility of reuse of secondary resources based on different degrees of profitability—economic, marginally economic, sub-economic, and other conditions at the time of the estimation. The secondary resources under economic and marginally economic conditions are classifiable as secondary reserves, which are considered technologically and economically recoverable at the time of estimation. Conversely, the secondary resources in the sub-economic and other categories are regarded as unrecoverable at the time of the estimation for technological, economic, and other relevant reasons. One advantages of their approach is to use types of secondary material stocks for horizontal axis, which enable us to clearly understand existence forms of secondary resources. Another advantage is the time of emerging as wastes or secondary resources, which informs actual availability of secondary resources on a yearly basis.

Knowledge

Time of emerging as waste or secondary resources

	Storage	Inventories and final products in/after use		Wastes in managed landfill sites	Dissipated materials
		Emerging in a year	Not emerging in a year		
Economic	Secondary reserves in a year		Secondary reserves in future		
Marginally economic		Marginal secondary reserves in a year	Marginal secondary reserves in future		
Sub-economic		Subeconomic secondary resources in a year	Subeconomic secondary resources in future	Subeconomic secondary resources	Subeconomic secondary resources
Other		Unrecoverable materials	Unrecoverable materials	Unrecoverable materials	Unrecoverable materials

(Left-side axes: Profitability; Probability of reuse)

Table 2.2 Classification of secondary resources with a particular emphasis on the stages of reuse

2.2 Improvements in classification framework of secondary resources

Daigo et al. (2009) explored the possible final destinations of uncollected materials: landfill, mixing into the steel cycle, and export in mixed metals and estimated the proportion of uncollected materials going to each final destination in 2000. Based on global aluminum cycle, Liu and his colleagues (2012) analyzed the amount of aluminum unrecyclable EoL products sent to landfills and the amount of processing loss and other repository. Gordon et al. (2004) estimated the amount of zinc losses in EoL zinc disposed in landfill in individual countries and world total. Graedel et al. (2011a) found that not all the EoL products collected for recycling returns to the original metal cycle and some products are recycled or lost into the cycle of another metal (as with copper

wire mixed into steel scrap) as a result of inefficient separation. Reck (2009) illustrated the relationship of stainless steel and its alloying elements nickel, chromium, and iron (showing also the connection of nickel and copper). In order to reflect actual conditions of final destinations of unrecoverable materials, the category of "unrecoverable materials (mixed metal loss)" was added to the original classification framework of secondary resources shown in Fig. 2.2 because it is difficult to identify categories for lost metals (i.e., "final products in/after use," "wastes in managed landfill sites," or "dissipated materials"). Furthermore, storage and inventories were excluded from the original classification table because it is difficult to get data to quantify. Although landfill mining concepts are emerging and demonstrated recently, wastes in managed landfill sites were classified as subeconomic secondary resources or unrecoverable materials (other) and were excluded from our reserve estimates. In fact, a certain fraction of landfilled secondary resources might be economic, but it remains a small fraction.

Knowledge

	Final products in/after use		Wastes in managed landfill sites	Dissipated materials
	Emerging in a year	**Not emerging in a year**		
Economic	Secondary reserves in a year	Secondary reserves in future		
Marginally economic	Marginal secondary reserves in a year	Marginal secondary reserves in future		
Sub-economic	Sub-economic secondary resources in a year	Sub-economic secondary resources in future	Sub-economic secondary resources	Sub-economic secondary resources
Other	Unrecoverable materials (other)	Unrecoverable materials (other)	Unrecoverable materials (other)	Unrecoverable materials (other)
	Unrecoverable materials (mixed metal losses and other repositories)			

Profitability

Table 2.3 Framework for classification of secondary resources

2.3 Characterizing metal cycle

In a simplified metal life cycle, there are four main processes: production, fabrication and manufacturing, use, and waste management as shown in Fig. 2.3. Sub-processes of metal production include mining and milling, smelting and refining. In fabrication, the refined metal is used for plating, casting, and other uses such as catalysts or as an alloying element. The types of first uses differ from metal to metal. The output of fabrication process are semi or intermediate products that are used in manufacturing to make parts and finished products. Finished products can be categorized into major end-use sectors such as construction, transportation, infrastructure and machinery. And in detail, they will again differ metal by metal. Finished products enter the use phase where they remain in service until they reach their end-of-life. The lifetime of metal products
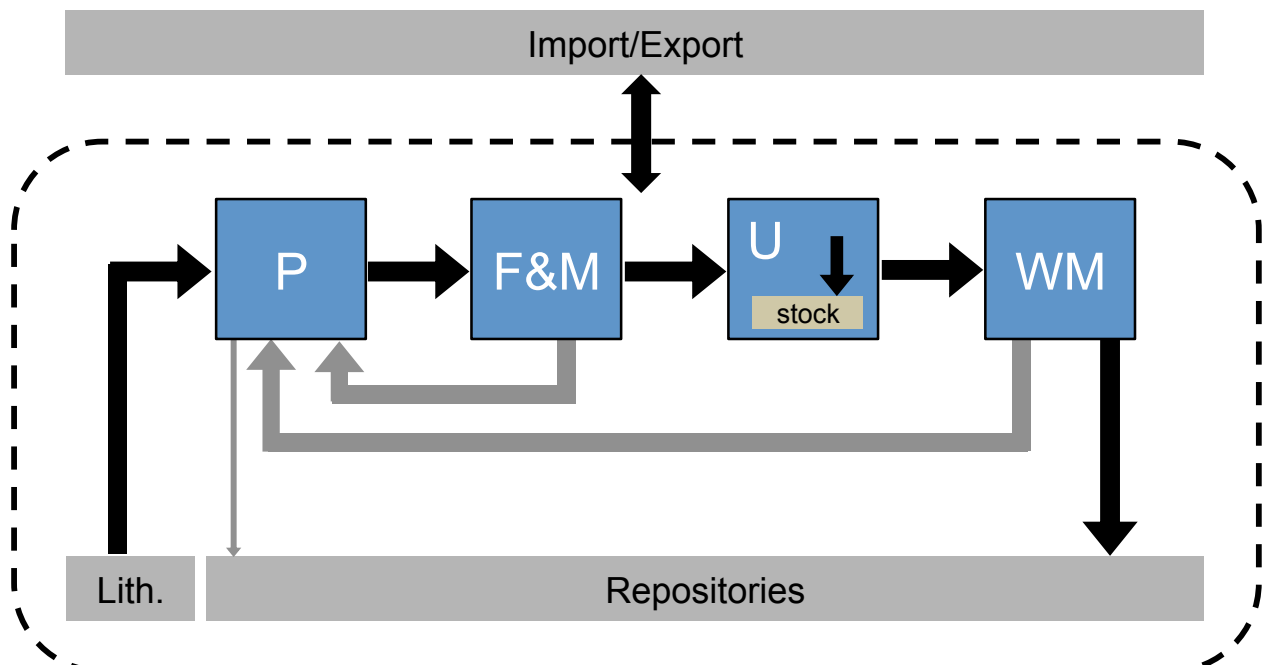
Figure 2.1. A generic metal life cycle adapted from Graedel et al. (2004). P: Production, F&M: fabrication and manufacturing, U: use, WM: waste management, Lith: Lithosphere

can vary from year to decades and more. They are part of the in-use stocks as long as metal are in use. When products in use reach their end-of-life, they are either discarded to landfills or collected for recycling. Not all the material collected for recycling will return to the original metal cycle. Some metals will be recycled with other metal recycling streams as a result of inefficient separation.

2.4. Defining the Life Cycle of Aluminum, Copper and Zinc

2.4.1 Aluminum
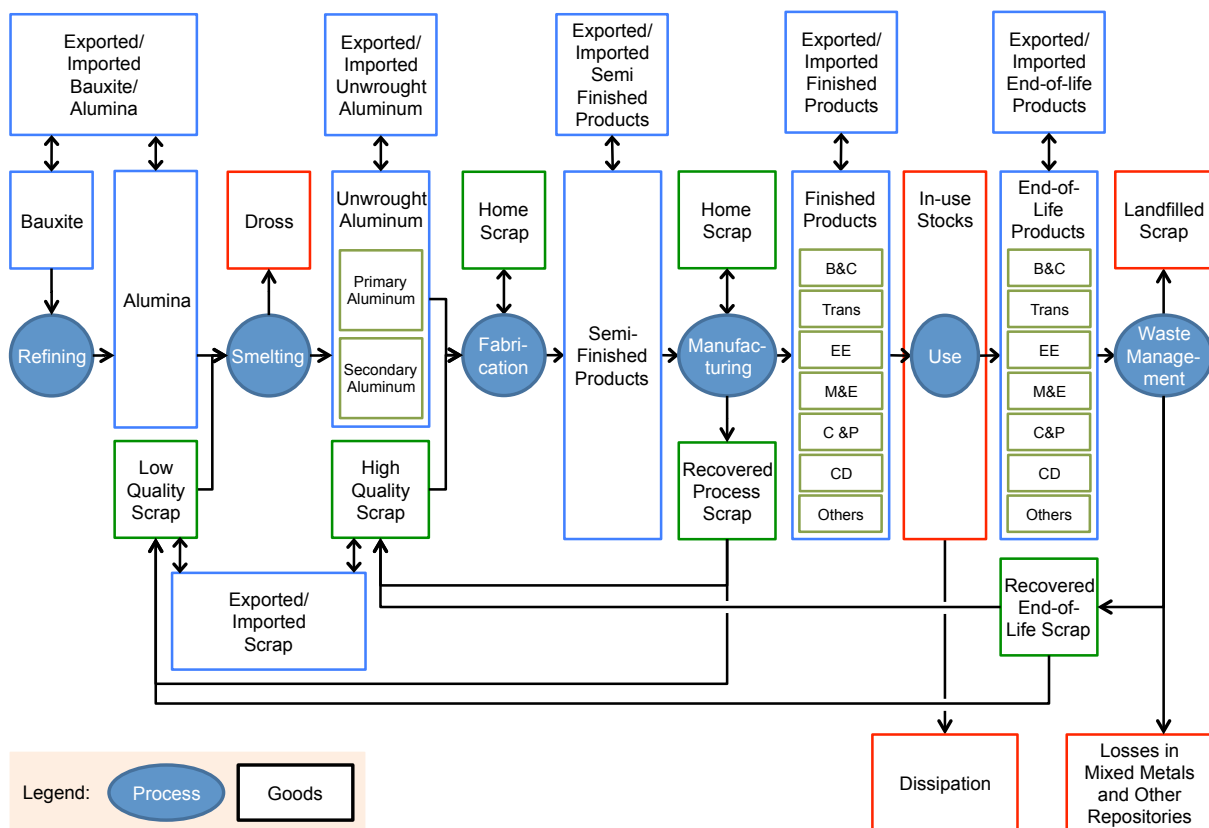
Figure 2.2. Structure of aluminum flow model used for this study. Seven categories of end-use products are: Building and Construction (B&C), Transportation (Trans), Electrical Engineering (EE), Machinery and Equipment (M&E), Containers and Packaging (C&P), Consumer Durables (CD) and Other Uses (Others). Processes and Goods are portrayed respectively as ovals and boxes.

Our aluminum flowchart was developed based on the anthropogenic aluminum cycle developed in Liu and Müller (2013), considering primary production, fabrication of semi-finished goods, manufacturing of finished products, use, and waste management, as shown in Figure 2.2. The mass balance principle was applied in all processes in our model.

2.4.2 Copper.

Glöser et al. (2013) provided a detailed description of the copper life cycle through primary production, fabrication of semi-finished goods, manufacturing of finished products, stock in use, and waste management and recycling. Our copper flows and stocks model consists of five main processes, as shown in Figure 2.3: refinery, fabrication, manufacturing, use, and waste management.

These processes produce goods and wastes of different kinds: refined copper, semi-finished products, and finished products, as well as home scrap, process scrap, and EoL scrap. The amounts of imports and exports of these goods and wastes were also considered in our analysis. The finished products were classified into five end-use sectors based on international copper statistics: building and construction, infrastructure, industrial equipment, transport, and consumer products (Figure 2.3; see Table S2 in SI for details of classification in Appendix B). These finished products exist in society for varying periods of time as in-use stock. During the use stage, a small portion of in-use copper stocks is dissipated in the environment. After their lifetime, a part of EoL scraps are collected and recycled: high-quality and low-quality scraps are used, respectively, for production of semi-finished products and refined copper. Other EoL scraps are disposed of in landfill sites or lost in other recycling loops (i.e., losses in mixed metals).
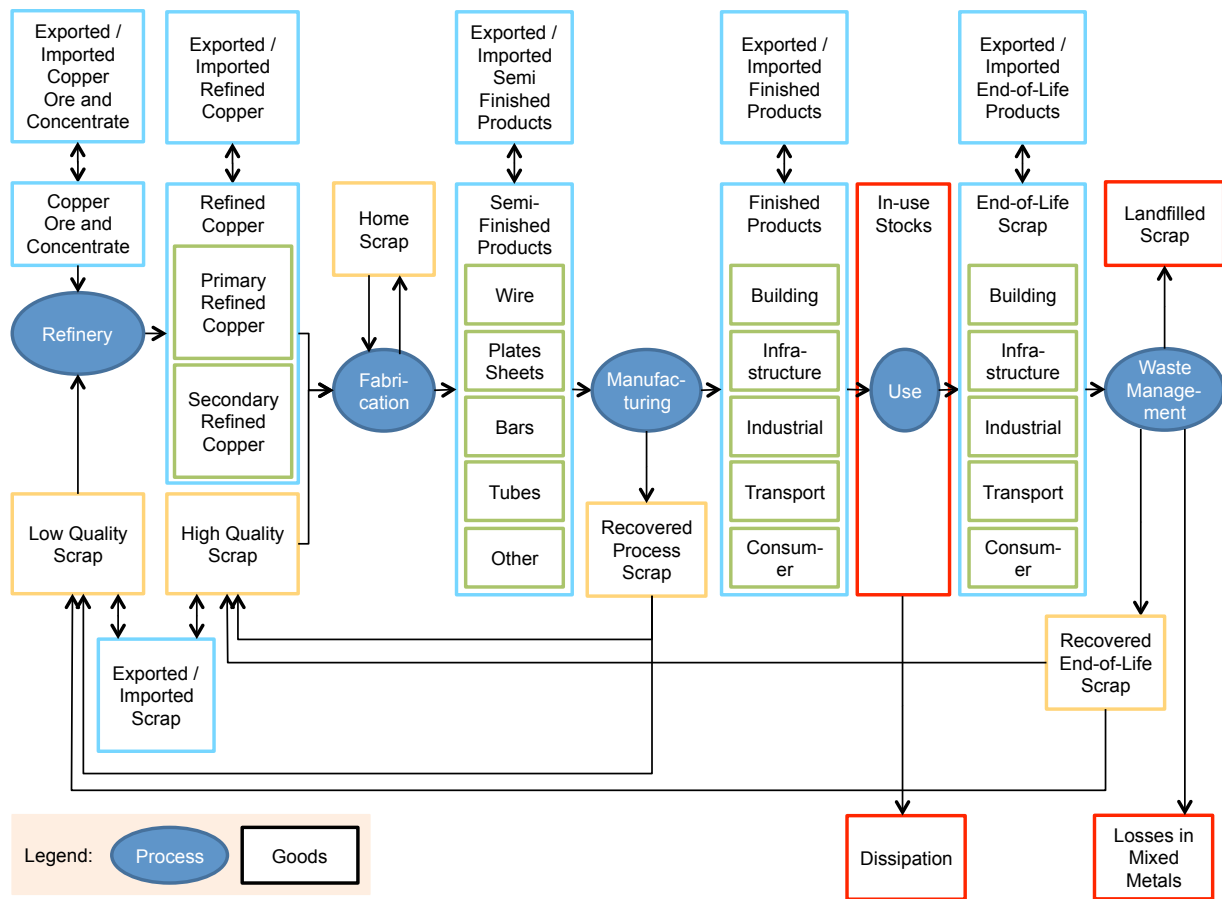
Figure 2.3. Structure of copper flows and stocks model.

2.4.3 Zinc

Based on generic zinc cycle in the Gordon et al. (2004), our zinc flowchart was simplified and developed, considering primary refined zinc production, fabrication of semi-finished goods, manufacturing of finished products, as well as stock in use, and waste management and recycling as shown in Figure 2.4. Mass balance principle was complied in all processes and in our model. Processes and Goods are portrayed respectively as ovals and boxes .
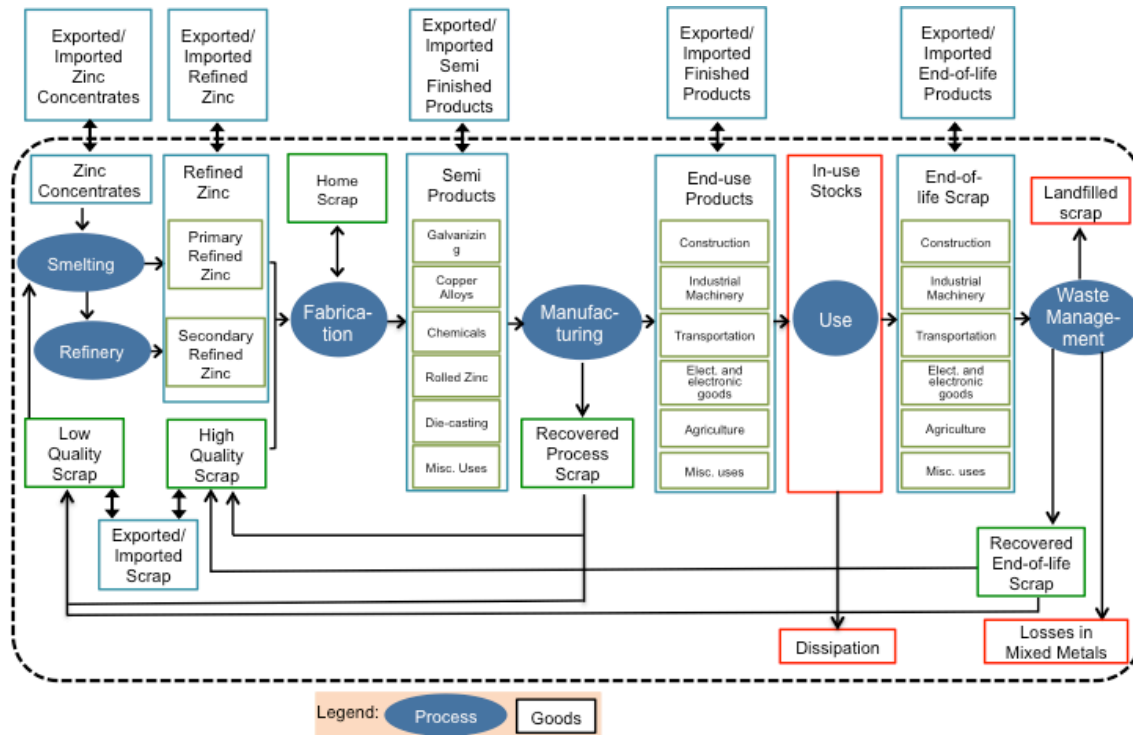
.**Figure 2.4.** Structure of zinc flow model. Six categories of end-use products are: Construction, Industrial Machinery, Transportation, Electrical and Electronic goods, Agriculture and Miscellaneous Uses. The dashed line represents the system boundary

2.5 Estimation of Secondary and Marginal Secondary Reserves for Aluminum, Copper, and Zine

The amount of secondary reserves in a year and in the future in Table 2.3 were estimated using the following equations.

$$SR(t,c)=\Sigma_i S(t,c,i) \times SRR(t,c) \qquad (1a)$$

$$SRe(t,c)=\Sigma_i GEoLS(t,c,i) \times SRR(t,c) \qquad (1b)$$

$$SRn(t,c)=SR(t,c)-SRe(t,c) \qquad (1c)$$

Therein, $SR(t,c)$ represents the secondary reserves in country c in year t. Also, e and n denote stocks emerging in a year and not emerging in a year, respectively. $S(t,c,i)$

denotes the stocks of finished product i in country c in year t. $SRR(t,c)$ is the secondary reserve ratio in a country c in year t. Finally, $GEoLS(t,c,i)$ denotes the generated EoL scrap of finished product i in/after use in country c in year t.

The amounts of marginal secondary reserves in a year and in the future in Table 2.3 were estimated similarly using the following equations.

$$MSR(t,c)=\Sigma_i S(t,c,i) \times (SRR(t_{high},c)-SRR(t,c)) \quad (2a)$$

$$MSRe(t,c)=\Sigma_i GEoLS(t,c,i)\times(SRR(t_{high},c)-SRR(t,c)) \quad (2b)$$

$$MSRn(t,c)=MSR(t,c)-MSRe(t,c) \quad (2c)$$

In those equations, $MSR(t,c)$ denotes marginal secondary reserves of country c in year t; t_{high} represents the year in which SRR was highest. The SRR is the fraction of stocks in/after use that is technologically and economically recoverable. The difference between $SRR(t_{high},c)$ and $SRR(t,c)$ is the fraction that was technologically and economically recoverable in year t_{high} but not economically recoverable in year t. Therefore, it is marginal.

2.6. Estimation of sub-economic secondary resources and unrecoverable materials for Aluminum, Copper and Zine

The amounts of sub-economic secondary resources and unrecoverable materials (others) in the column of final products in/after use were estimated as

$$SSRUMe(t,c)=\Sigma_i GEoLS(t,c,i)-SRe(t,c)-MSRe(t,c) \quad (3a)$$

$$SSRUMn(t,c)=\Sigma_i S(t,c,i) - SSRUMe(t,c) - SR(t,c) - MSR(t,c), \quad (3b)$$

where $SSRUM(t,c)$ represents sub-economic secondary resources and unrecoverable materials (others) in products in/after use in country c in year t.

For dissipated materials of aluminum and copper, equation (5) is used and equation (6) is used for those of zinc. The amounts of sub-economic secondary

resources and unrecoverable materials (others) in the column of wastes in managed landfill sites and dissipated materials in Table 2.3 were estimated respectively as

$$SSRUMw(t,c)=\Sigma_{t'}SSRUMe(t',c) \times LFR(t',c) \qquad (4)$$

$$SSRUMd(t,c)= \Sigma_i GEoLS(t,c,i) \times DpR(t',c,i) \qquad (5)$$

$$SSRUMd(t,c)= \Sigma_i S(t,c,i) \times DpR(t',c,i) \qquad (6)$$

where w and d denote wastes in managed landfill sites and dissipated materials, respectively. Also, $LFR(t',c)$ is the landfill ratio in country c in year t'; $DpR(t',c,i)$ is the dissipation ratio of finished products i in country c in year t'. In the row of others in Table 2.3, unrecoverable materials (mixed metal losses and other repositories) were estimated as

$$SSRUMm(t,c) = \Sigma_{t'}SSRUMe(t',c) \times RMMLOR (t',c) \qquad (7)$$

where m represents the mixed metal loss, and where $RMMLOR (t',c)$ denotes the ratio of mixed metal losses and other repositories in country c in year t'.

2.7 Estimation of the Secondary Reserve Ratio

2.7.1 Aluminum

The secondary reserve ratio SRR is the fraction of stocks in/after use that are economically and technologically recoverable. For this study, the ratio was estimated by dividing the amount of recovered EoL scrap by the total amount of generated EoL scrap (i.e., the EoL recycling rate reported by Graedel et al. (2011)).

$$SRR(t,c) = REoLS(t,c)/\Sigma_i GEoLS(t,c) \qquad (8)$$

In that equation, $REoLS(t,c)$ signifies the amount of recovered EoL scrap in country c in year t. $REoLS(t,c)$ was estimated using the following equations.

$$REoLS(t,c) = \Sigma_i [GEoLS(t,c,i) \times EoLRR(t,c,i)] \qquad (9)$$

In those equations, *EoLRR(t,c,i)* is the EoL recycling rate of finished product *i* in country

c in year *t*.

2.7.2 Copper and Zinc

For copper and zinc, the following calculation equations are used to estimate *SRR*. The

SRR was estimated by dividing the amount of recovered EoL scrap by the total amount

of generated EoL scrap (i.e., the EoL recycling rate in Graedel et al. (2011)):

$$SRR(t,c) = REoLS(t,c)/GEoLS(t,c) \tag{10}$$

Therein, *REoLS(t,c)* is the amount of recovered EoL scrap in country *c* in year *t*.

REoLS(t,c) was estimated using the following equations.

$$REoLS(t,c) = US(t,c) - RPS(t,c) + ES(t,c) - IS(t,c) \tag{11}$$

$$US(t,c) = HQS(t,c) + LQS(t,c) = [SP(t,c) - RC(t,c)] + [RP(t,c) - OC(t,c)] \tag{12}$$

$$RPS(t,c) = \Sigma_i \, SC(t,c,i) \times (1 - FE(t,c,i)/100) \tag{13}$$

Therein, *US(t,c)* is the amount of utilized scrap in country *c* in year *t*; *RPS(t,c)* is the

amount of recovered process scrap in country *c* in year *t*; *ES(t,c)* and *IS(t,c)* respectively

represent the amounts of exported and imported scrap in country *c* in year *t*; and

HQS(t,c) and *LQS(t,c)* respectively denote the amounts of utilized high and low quality

scraps in country *c* in year *t*. In addition, *SP(t,c)* is the amount of semi-finished product

production in country *c* in year *t*; *RC(t,c)* is the amount of refined copper consumed in

country *c* in year *t*; *RP(t,c)* is the amount of refined one produced in country *c* in year *t*;

OC(t,c) is the amount of metal ore consumed in country *c* in year *t*; *SC(t,c,i)* represents

the amount of semi-finished product *i* consumed in country *c* in year *t*; and *FE(t,c,i)*

denotes the fabrication efficiency of semi-finished product *i* for the production of finished

products in country *c* in year *t* (%).

2.8. Data Sources

2.8.1 Aluminum

Statistical data from World Bureau of Metals Statistics (1960–2010) and U.S. Geological Survey (1960–2010) were used for production of aluminum ore (bauxite), alumina, primary aluminum, and secondary aluminum during 1960–2010. The United Nations Comtrade database (2015) was used for data of exports and imports of bauxite, alumina, unwrought aluminum, semi-finished products and finished products and scrap. Aluminum content data were used to ascertain the aluminum contents of traded commodities (See S3 in the Supplementary Materials in Appendix A). For comparison with our estimated secondary reserves, data related to primary reserves from U.S. Geological Survey (2012) were used.

Fabrication efficiency by end-use sector and average regional lifetime were adapted respectively from GARC (2011) and Liu and Müller (2013) (See S4 in the Supplementary Materials in Appendix A). Landfill ratio $LFR(t',c)$ used in equation (4), dissipation ratio $DpR(t',c,i)$ used in equation (5) and the ratio of mixed metal losses and other repositories $RMMLOR (t',c)$ used in equation (6) were used from Liu et al. (2012), Ciacci et al. (2015) and Liu et al. (2012), respectively (See S4 in the Supplementary Materials in Appendix A). EoL recycling rate $EoLRR(t,c,i)$ used in equation (8) were from GARC model (collection + recovery)(IAI, 2015).

With the availability of reported domestic shipment data of aluminum semi-products (see S5 in Supplementary Materials in Appendix), which plays an important role in this study, we target 19 countries (Argentina, Australia, Austria, Belgium, Brazil, China, France, Germany, India, Italy, Japan, Netherlands, Norway, Russia, South Africa,

Spain, Switzerland, the United Kingdom, and the United States). For the global analysis, global product in use data from GARC model (IAI, 2015) was used.

2.8.2 Copper

International Copper Study Group (ICSG, 2010) data were used for production of copper ore and concentrates, refined copper, and semi-finished products from 1960 to 2010. UN Comtrade data (UN Comtrade 2014) were used for exports and imports of copper ore, refined copper, semi-finished products, finished products, and scraps. Copper content data were used to ascertain the copper content of the traded commodities (see Table S4 in the SI in Appendix B). All of these data were used to estimate the consumption of copper ore, refined copper, semi-finished products, finished products, and scraps. For countries with high levels of consumption of finished products in 1960 (Germany, Italy, Japan, Spain, and the United States), consumption of finished products before 1960 was estimated by extrapolating their consumption trends during the 1960s.

Statistics provided by the International Copper Association (ICA) and the International Wrought Copper Council (IWCC)(ICA&IWCC, 2014) were used to determine the market share of finished products (the five end-use sectors in Figure 4.1). Because of limited data availability, data for 2013 were used for all the years analyzed (see Table S5 in the SI in Appendix B).

Data on country-specific and time-series fabrication efficiencies and country-specific mean lifetimes of copper products are very limited. Therefore, global average fabrication efficiencies and mean lifetimes (Gloser et al. 2013) were used for all countries and years analyzed with some exceptions (see Table S5 in the SI in Appendix B).

2.8.3 Zinc

International Lead and Zinc Study Group (ILZSG, 2016) Data were used for production of zinc concentrates, primary refined zinc, and secondary refined zinc from 1960 to 2010 and also for exports and imports of zinc concentrates and refined zinc. UN Comtrade (2016) data were also used for exports and imports of semi-finished products and finished products and scraps. Zinc content data were used to determine the zinc content of the traded commodities (see Table S2 in the Supplementary Material in Appendix C). To compare with primary zinc reserves, U.S. Geological Survey data (2012) was used for all countries in the study.

World Bureau of Metals Statistics (WBMS, 2013) data were used for principal end-uses of zinc in Germany, France, Italy, Japan and United States from 1982 to 2010 (see Figure S1 in the Supplementary Material) and the data before 1982 was estimated by extrapolating their usage trends during the 1980s. China Non-ferrous Metals Industry Association (CNMIA, 2005) data was used for principal end-uses from 1990 to 2004 (see Figure S1 in the Supplementary Material in Appendix C). In China, principal end-uses in 2004 were assumed to be the same until 2010. The regional models of allocation of semi-fabricated to end-use products (Meylan and Reck, 2016) was used in this study.

Country-specific data on zinc product lifetime are very limited. Therefore, in this study, average global lifetime and average fabrication efficiency by end-use sector has been adapted from Meylan and Reck (2016) and Van Genderen (2014), respectively (Table S3 in the Supplementary Material in Appendix C). Landfill ratio $LFR(t',c)$ used in equation (9), dissipation ratio $DpR(t',c,i)$ used in equation (10) and mixed metal loss ratio $MMLR(t',c)$ used in equation (11) were used from Gordon et al. (2004), Meylan and

Reck (2016) and our assumption, respectively (Table S4 in the Supplementary Material in Appendix C).

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Chapter 3

Results and Discussions

**3.1**. **Assessment of Secondary Aluminum Reserves of Nations**

3.1.1. Aluminum Stocks

Figure 3.1 presents trends of aluminum stocks in 19 countries during 1980–2010. The total aluminum stocks in the United States in 1990 were estimated as 83 million tonnes and 152 million tonnes in 2010: about 80% increase in 20 years. In China, the total aluminum stocks were approximately 10 million tonnes in 1990. The stocks for 2010 were 97 million tonnes: an approximately ten-fold increase in 20 years. China's aluminum stocks skyrocketed particularly after the Open and Reform Policy was adopted in 1978. Furthermore, the aluminum stocks of Japan were increased from 24 million tonnes in 1990 to 41 million tonnes in 2010, with respective increases of 70%. Emerging economies such as those of Brazil, India, and South Africa have shown rapidly increasing trends. In the case of Russia, the total aluminum stocks in 1980 were 29 million tonnes. Stocks in 2010 were 14 million tonnes, about 50% decrease over 30 years because of the post-1990s transition period. It is noteworthy that economically developed nations must have had a certain amount of aluminum that had been accumulating in 1962. Therefore, our estimates are somehow underestimations.

On a per-capita basis in 2010, Norway had the highest per-capita aluminum stocks with 566 kg, followed by 500 kg in Switzerland and 485 kg in the United States, as depicted in Fig. 3.2. Among Brazil, Russia, India, and China, the so-called BRICs countries, Russia had a higher amount of per-capita aluminum stocks with 98 kg/capita than Brazil with 48 kg/capita, India with 9 kg/capita, and China with 72 kg/capita.
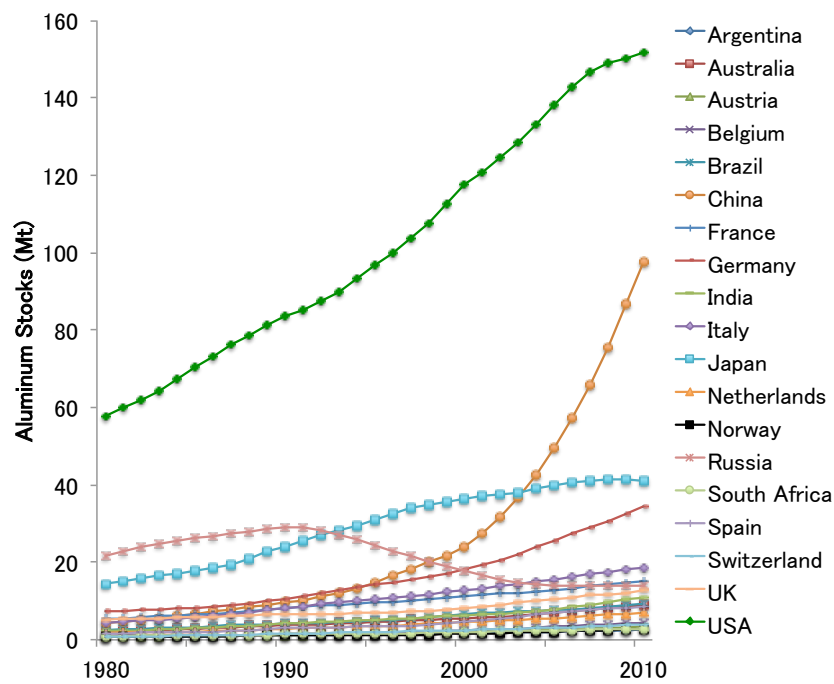
Figure 3.1. Historical patterns of aluminum stocks of finished product in/after use in 19 countries.
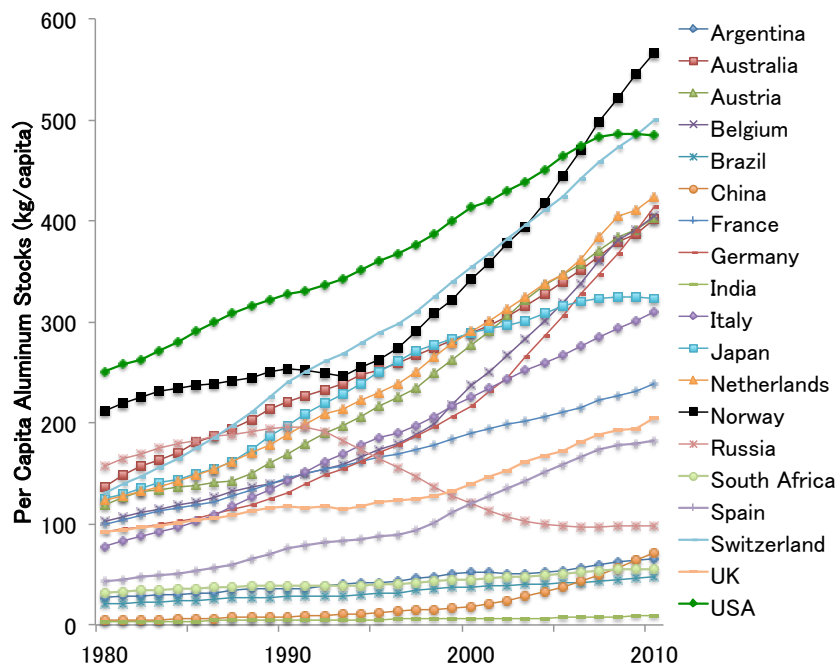
Figure 3.2. Historical patterns of per-capita aluminum stocks of finished product in/after use in 19 countries.

As shown in Table 3.1, our values of aluminum stocks in the study generally agree with estimated results of previous studies. Our values are higher than those estimated by McMillan et al. (2010) with 97.6 million tones of stocks in the United States for 2007 and Hatayama et al. (2009) with 32 million tonnes of stocks in Japan for 2003. Although the two studies rely on different data sources, the difference is most likely attributable to the lifetime of finished products. For China, Italy, and Japan in 2010, our estimates of aluminum stocks and its per capita are higher than that of Liu and Müller (2013).

Table 3.1. Aluminum stocks estimated in earlier studies and this study

Country	Year	Stocks (Mt)	Per-capita stocks (kg/capita)	Method	Reference
China	2010	97.4	72	Top-down	This study
	2010		64 (-11%)	Top-down	Liu and Müller, 2013
	2009	88.9 (3%)		Top-down	Yue et al. 2011
	2005	48.8 (-2%)	37.3 (-1%)	Bottom-up	Wang and Graedel, 2009
Italy	2010	18.8	311	Top-down	This study
	2010		264 (-15%)	Top-down	Liu and Müller, 2013
	2009	19.1 (5%)	320 (6%)	Top-down	Ciacci et al. 2012
Japan	2010	41.2	324	Top-down	This study
	2010		318 (-2%)	Top-down	Liu and Müller, 2013
	2003	32 (-16%)	250 (-17%)	Top-down	Hatayama et al. 2009
	2000	34.8 (-4%)		Top-down	Murakami, 2006
U.S.	2010	151.5	485	Top-down	This study
	2010		530 (9%)	Top-down	Liu and Müller, 2013
	2009	151 (1%)	491.2 (1%)	Top-down	Chen and Graedel, 2012
	2007	97.6 (-33%)		Top-down	McMillan et al. 2010

Note: Percentage in brackets are differences between previous estimates and this study for the same year.

Although a similar lifetime and share of end-use sectors were used in both studies, the differences most probably derive from explicit consideration of the aluminum contents of traded commodities. This fact might be explained to a large degree by the different data sources of aluminum contents of traded commodities and data handling methods from monetary values to physical values (i.e., their aluminum content data are from industry surveys and various literature; ours are from the use of waste input–output material flow analysis (WIO-MFA)). See also S6 in Supplementary Materials in Appendix A for the historical evolution of aluminum stocks by finished products in these 19 countries.

3.1.2. Secondary Reserve Ratio

In this study, one fundamentally important parameter for determining the amount of secondary aluminum reserves is the secondary reserve ratio (*SRR*). As shown in Figure 3.3, almost all countries' *SRRs* are gradually increasing, except some countries. Japan and France have the highest reserve ratios (about 70%) and Belgium has the lowest reserve ratio (about 40%) in 2010. China's *SRR* increased from 2007 to 2008 because of the big change in EoL recycling rates of machinery & equipment, consumer durables, and other uses in GARC model (IAI, 2015). Graedel et al. (2011) estimated that the global average EoL recycling rate of aluminum was higher than 50% during 2000–2005, although the rate varies among countries and aluminum-containing products, which is consistent with Figure 3. Norgate and Rankin (2002) estimated the 32% in the United States, which is lower rate than our SRR value 49% in 2000. Chen (2013) reported that in the United States, the estimates of EoL recycling rate during 1980–2009 exhibited a similar historical trend with aluminum prices. Its EoL
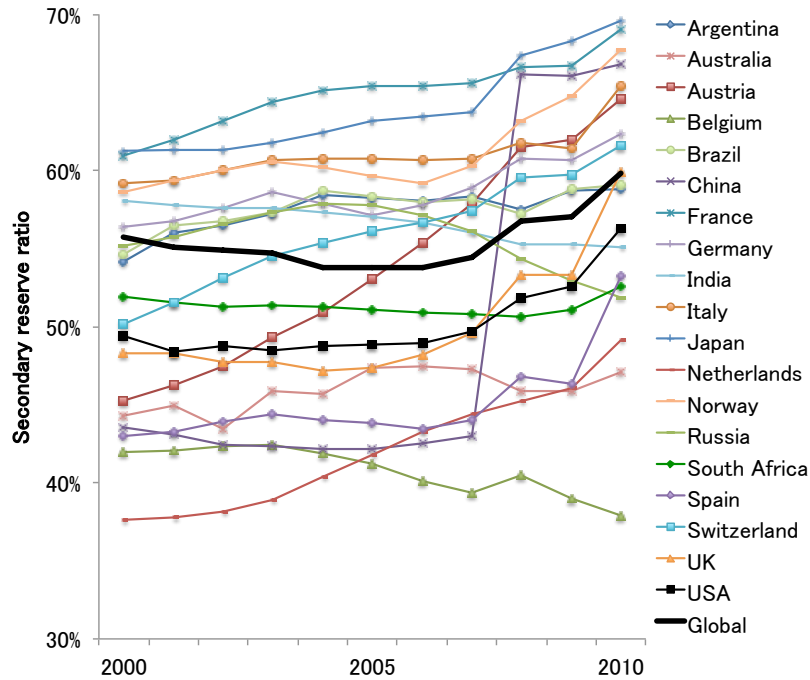
Figure 3.3. Estimated secondary reserve ratios

recycling in previous study was slightly higher than our value. Henstock (1996)

estimated that the EoL recycling rate in the UK was 31% in 1988, which is quite lower

than our value 45% in the same year. Unfortunately, it is not possible to compare our

estimates with those of other target countries in this study because very few earlier

studies of EoL recycling rates have been reported in the literature. According to

sensitivity analysis results (see S7 in Supplementary Materials), *SRR* was affected by

the change in market share and mean lifetime of finished products because it changes

the amount of EoL scrap generation, which has an direct effect on the quantity of

recovered EoL scrap by end use (see equation (8)). However, the changes in *SRRs*

were small (mostly less than ±2%).

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3.1.3. Secondary Aluminum Reserves and Their Comparison with Primary Aluminum Reserves (Bauxite reserves)

As portrayed in Fig. 3.4, Australia has the largest primary reserves of aluminum ore (bauxite reserves): 5,400 million tonnes, accounting for about 20% of world bauxite reserves, followed by Brazil with 3,400 million tonnes and China with 750 million tonnes among the target countries in 2010 (USGS, 2015). Australia was the leading producer of bauxite in the world with 30% of global bauxite production. In terms of secondary reserves (secondary reserve + marginal secondary reserve in Table 2.3), it has been estimated that the United States has the largest secondary aluminum reserves of 85 million tonnes, followed by 65 million tonnes in China and 29 million tonnes in Japan. Countries such as Japan, Germany, and France have large secondary aluminum reserves but no primary reserves, although countries such as Australia, Brazil, and India have large primary reserves but only small secondary reserves. According to the distribution of primary and secondary aluminum reserves as portrayed in Fig. 3.4, the quantity of secondary aluminum reserves in Japan is roughly equivalent to the primary aluminum reserves in the United States. It is noteworthy that, for the United States, its secondary reserves are larger than its primary reserves. Therefore, domestic reserves of bauxite only are inadequate to meet long-term demand in the United States and we need to consider appropriate management of secondary reserves of aluminum accumulated in products and infrastructure in society. The global secondary reserves were estimated 413 million tonnes, which is only about 1.5% of global primary reserves of 28,000 million tonnes (USGS, 2012).
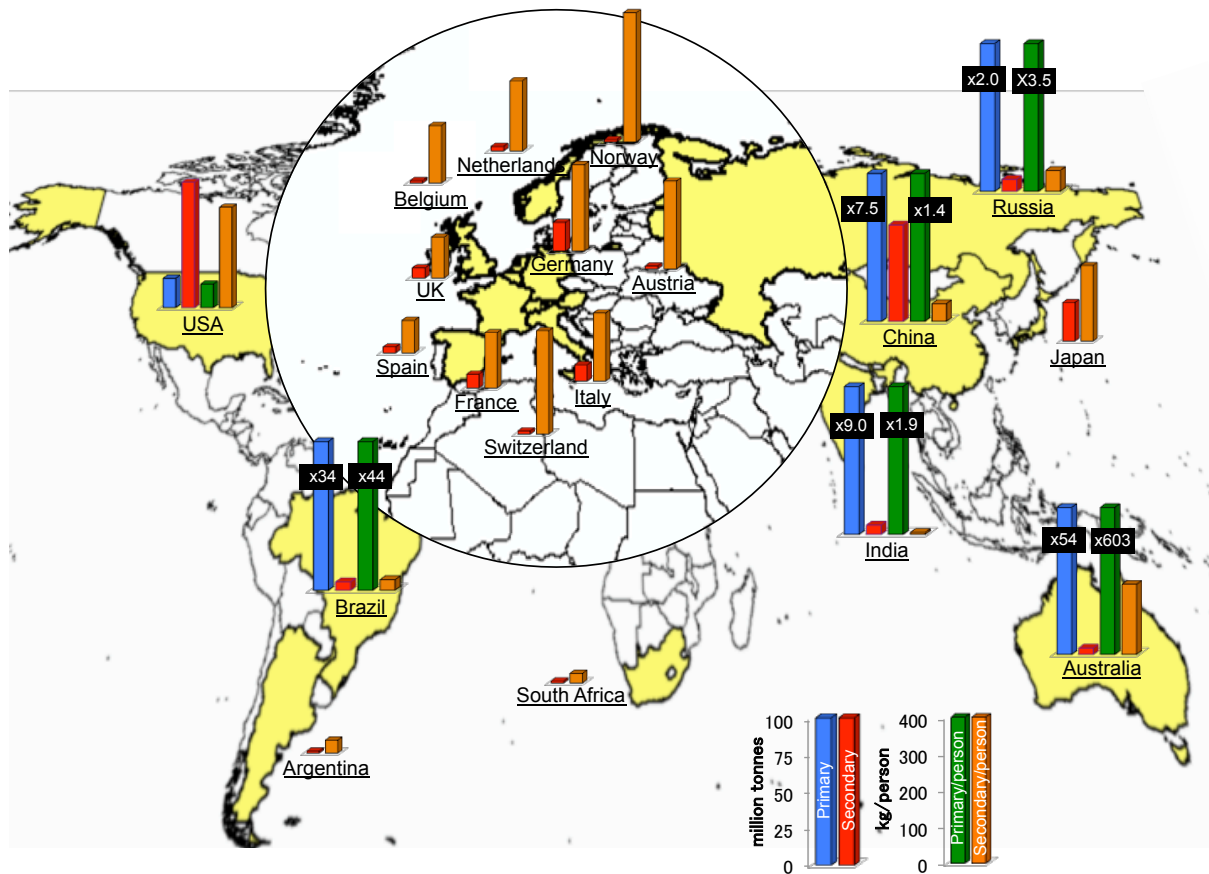
Figure 3.4. Distribution of primary and secondary aluminum reserves in 19 countries.

On a per-capita basis, Australia has the highest primary reserves, with about 241,000 kg/person followed by about 17,000 kg/person in Brazil and about 700 kg/person in India. These numbers are substantially higher than the per-capitasecondary reserves, of which the highest was about 383 kg in Norway, followed by 308 kg in Switzerland and about 273 kg in the United States. These values are more than 10 times higher than those in economically developing countries such as Brazil and India.

Secondary aluminum reserves are expected to become important in countries with no primary aluminum reserves, although the total quantity of primary aluminum reserves in the world is much larger than that of secondary reserves. However, having

knowledge related to the sizes of both primary and secondary aluminum reserves is useful for planning the stable supply of aluminum on a national scale and beyond. In assessing the amounts of secondary reserves, the value of *SRR* is a crucial factor. Actually, SRR can be raised through enhanced EoL recycling. To achieve this, all key stakeholders such as scrap collectors, aluminum producers, and decision makers should be involved, creating positive effects on the domestic economy.

The availability of primary reserves of bauxite is limited not only by the quantity that is economically and technologically extractable but also by geopolitical settings in the region. Moreover, supply chains of primary resources are most likely affected by domestic political changes in resource rich countries. For secondary reserves of aluminum, the availability of aluminum stocks in use for reutilization is affected by the quantity of EoL products entering the waste management system and the capacity of recycling. It is also important to note that finished products in some end-use sectors remain in use for decades (Liu and Müller, 2013).

3.1.4. Application of Classification Framework for Secondary Aluminum Resources

The classification framework for secondary resources was applied to aluminum for France, Japan, and the United States in 2010 as representatives of countries in Europe, Asia, and North America.

As described in Table 3.2, France has 10 million tonnes (44%) of economically recoverable secondary aluminum, whereas Japan and the United States have 29 million tonnes (48%) and 85 million tonnes (37%). Higher share of economically recoverable secondary aluminum in France and Japan was observed throughout the study years (see Fig. 3.3). No marginally economic secondary reserves was estimated for these

countries because their secondary reserve ratios in 2010 were the highest during past 10 years (see Fig. 3.3). The amount of the yearly available secondary aluminum (economic and marginally economic secondary aluminum reserves emerging in a year) represents about 40% of its recent annual aluminum consumption in France, 75% in Japan, and 45% in the United States, which is crucially important to note for urban mining strategies.

The United States has large amounts (44 million tonnes) of secondary aluminum resources as wastes in managed landfill sites. They are about four times bigger than France's amount of secondary aluminum reserves (10 million tonnes). In Japan, considerable amounts of secondary aluminum resources are also accumulated in landfill sites (10 million tonnes). These deposits in the landfill sites will be used potentially for the future extraction of secondary aluminum through landfill mining. Wagner and Raymond (2015) demonstrated that landfill mining for metals (i.e., steel, aluminum, copper, brass, silver and etc.) can be very profitable without financial support from government. Therefore, we may be able to put the numbers of secondary reserves in the column of wastes in managed landfill sites in the future. It is also interesting to note that 12-14% of their secondary aluminum resources have been accumulated in France, Japan, and the United States as mixed metal losses and other repositories. As shown in Table 3, the total amounts of dissipated materials were estimated about 2% of secondary resources for three countries.

As shown in Table 3.2(d), 950 million tonnes of aluminum has been consumed at the global level, 413 million tonnes (43%) of which are economically extractable. About 121 million tonnes (13%) are in landfill sites; 54 million tonnes (6%) are dissipated and

85 million tonnes (9%) are lost in mixed metals and other repositories. To increase

resource use efficiency, these kinds of flows should be decreased.

The classification framework provides more detailed insights of size and location

of secondary aluminum resources, which is very useful for policy setting of future

aluminum waste management and resource reutilization. In addition, to meet the

expected long-term aluminum demand in the major countries, the quantity of secondary

reserves as in the classification framework can be regarded as the major domestic

potential exploitable aluminum resources towards the security of aluminum supply and a

circular economy in the future. Moreover, to smooth out the short-term fluctuating

imbalances between the supply and demand of recovered secondary aluminum, there

should be stockpiling of aluminum EoL products to provide a stable supply of secondary

aluminum. In this study, subeconomic and other resources were not distinguished.

Table 3.2. Classification of secondary aluminum resources in (a) France, (b) Japan, (c) the United States, and (d) Globally in 2010

(a) France	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	495 kt 2%	9,767 kt 42%			
Marginally economic	0 kt 0%	0 kt 0%			
Subeconomic & others	235 kt 1%	4,627 kt 20%	4,384 kt 19%	455 kt 2%	Total
Mixed metal losses	3,174 kt 14%				23,136 kt 100%

(b) Japan

	Products in/after use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	1,450 kt 2%	27,230 kt 45%			
Marginally economic	0 kt 0%	0 kt 0%			
Subeconomic & others	634 kt 1%	11,900 kt 20%	10,194 kt 17%	1,461 kt 2%	Total
Mixed metal losses	7,382 kt 12%				60,250 kt 100%

(c) United States

	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	2,899 kt 1%	82,347 kt 35%			
Marginally economic	0 kt 0%	0 kt 0%			
Subeconomic & others	2,254 kt 1%	64,036 kt 28%	44,302 kt 19%	4,813 kt 2%	Total
Mixed metal losses	32,081 kt 14%				232,732 kt 100%

(d) Globally

	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	12,245 kt 1%	400,849 kt 42%			
Marginally economic	0 kt 0%	0 kt 0%			
Subeconomic & others	8,215 kt 1%	268,905 kt 28%	120,559 kt 13%	54,436 kt 6%	Total
Mixed metal losses	84,665 kt 9%				949,873 kt 100%

However, as Ciacci et al (2015) pointed out, "a distinction between temporary and permanent stocks of elements might reveal some opportunities for future recovery." It will be further useful if potentially recyclable (subeconmic or temporary) and potentially not recyclable (others or permanent) resources can be classified.

3.2. Assessment of Secondary Copper Reserves of Nations

3.2.1 Trends in Copper Stocks of Finished Products in Use. Trends of copper stocks of finished products in use are depicted in Figure 3.5. The United States had the largest copper stocks throughout the entire period, but China's stocks increased dramatically after 1990 in response to economic growth driven by its "open-door policy" in the 1990s. Both countries reached 60–70 Mt of copper stocks in 2010. Japan ranked third, with a stock of about 25 Mt, showing a constant or slightly declining trend in recent years because of a declining population and a long recession. Italy and Germany had the fourth largest copper stocks, with about 20 Mt each in 2010.

On a per-capita basis, Italy had the highest copper stock, with more than 300 kg/person, followed by several other countries at about 200 kg/person, which are in the range of 140–300 kg/person reported in the literature (Gerst and Graedel 2008) (see Figure 3.6). Italy's per-capita stock increased after 1990 because of high per-capita consumption of finished products during 1990–2010, in contrast with the declining consumption trends observed in other economically developed countries after 2000 (see Figure S1 in the SI in Appendix B). The declining consumption trends led to stable per-capita copper stocks for Germany, Japan, and the United States during the past 20 years. Furthermore, China's per-capita copper stocks are still considerably less than those in economically developed countries, even though its total stocks are increasing rapidly.

Zeltner et al. (1999) reported that the copper stock in use in the United States was about 70 Mt in 1990, which is greater than our estimate of about 50 Mt in 1990 and close to the estimated value in 2010 (see Figure 3.5). The discrepancy might be a result of the use of a longer lifetime for long-term products and longer time-series data to estimate stocks in 1990 in Zelter et al. (1999) Gordon et al.(2006) showed, based on an estimate by Spatari et al. (2005) that per-capita copper stock in use in the United States was about 240 kg/person in 1999, which is almost equivalent to our estimate. Daigo et al. (2009) estimated that Japan's copper stock in use was about 20 Mt in 2005, which is slightly less than our estimate.
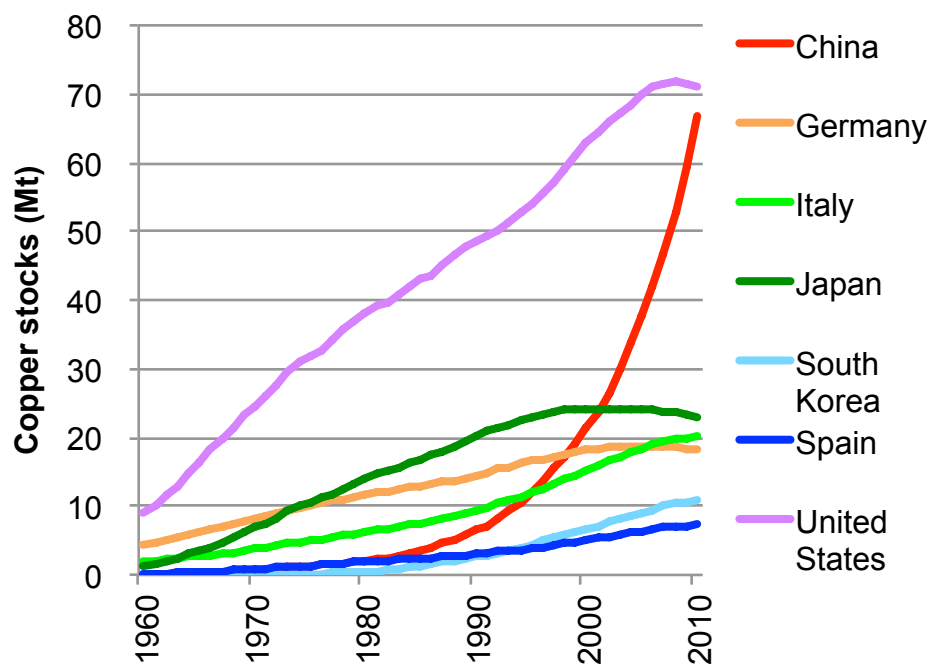
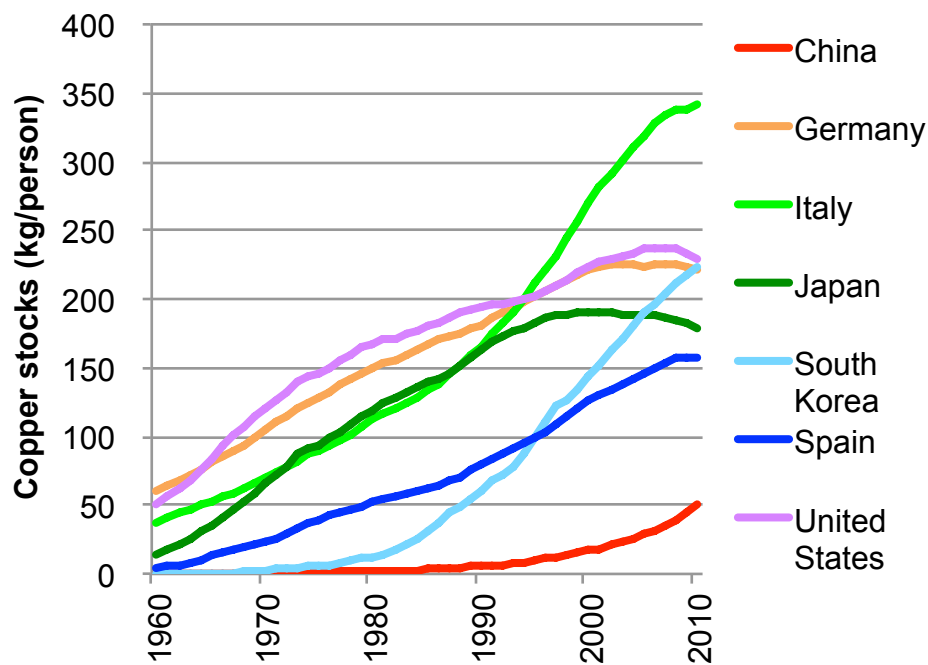Figure 3.5 Trends of copper stocks of finished products in use

Figure 3.6 Trends in per-capita copper stocks of finished products in use.

Our sensitivity analyses of market share, fabrication efficiency, lifetime, and copper content of traded finished products showed that copper stocks were most affected by the change in fabrication efficiency (see Figure S2 in SI in Appendix B). A 10% increase in fabrication efficiency increased copper stocks by about 10% in all countries examined in this study. A 10% increase in the copper content of traded finished products had negative impacts on copper stocks for net exporters of finished products (China, Germany, Italy, Japan, and South Korea).

In this study, constant copper contents of traded finished products were assumed. However, for example, there must be differences in copper content of machinery produced in different countries in different years. Therefore, this assumption might affect the stock results for some countries – Italy's high per-capita stock might be caused by this.

3.2.2 Secondary Reserve Ratio in the 2000s. Secondary Reserve Ratio (SRR) is the most important variable in this model. Figure 3.7 shows the five-year moving average of estimated $SRR(t,c)$ in equation (10). Estimated annual $SRR(t,c)$ fluctuated because the estimated recovered EoL scraps, $REoLS(t,c)$, fluctuated while the estimated generated EoL scraps, $GEoLS(t,c)$, changed gradually (mostly increased). As shown in equation (11), $REoLS(t,c)$ was estimated based on utilized scrap, $US(t,c)$, and recovered process scrap, $RPS(t,c)$. The fluctuations in $REoLS(t,c)$ are derived mainly from fluctuations in $US(t,c)$, which are strongly affected by the precision or consistency in statistical data. Even using a five-year moving average, China's $SRR(t,c)$ values were out of range (negative). Germany's and South Korea's were more than 100% in some years.

Graedel et al. (2011) concluded that the global average EoL recycling ratio of copper was greater than 50% during 2000–2005. Gloser et al.)2013) also presented an EoL recycling rate in the range of 40–50% for 2000–2010, which is in line with our estimates. At the regional level, Spatari et al. (2002) and Goonan (2009) reported that EoL recycling ratios were approximately 40% in North America during 1900–1999 and 43% in the United States in 2004, respectively. Our estimate is somewhat larger in the mid-2000s, but it is close to these values. Daigo et al. (2009) created a diagram of copper flows in Japan in 2005 and reported an EoL recycling ratio of about 60%, which closely approximates our estimate. Ruhrberg (2006) assessed the EoL recycling ratio in Western Europe and concluded that it was 60–70% in 1999, depending on the estimation approach used. Our estimates for Germany, Italy, and Spain are comparable with this range and all show declining trends during the time period. In Italy, the production of refined copper and semi-finished products and the associated use of copper scraps decreased in 2000s, whereas the amount of EoL scrap increased, which

explains the declining trend in its EoL recycling ratio. In Germany, the production of secondary copper decreased during 2000–2010 according to the World Bureau of Metal Statistics (WBMS, 2010) and its reported values are similar to our estimated values of scrap use in refinery. Our estimates also showed that scrap use in the fabrication of semi-finished products decreased during this period. These trends can explain the declining SRR for Germany.

The SRR in this study is the fraction of in-use copper stocks that are technologically and economically recoverable: it does not indicate the potentially recoverable fraction. Ciacci et al. (2015) investigated that potential recyclability rate (PRR) for copper was about 95% on a global market share basis. It is larger than our estimates of SRR and the SRR can be increased to this number.

As noted previously, Germany's SRRs in the early 2000s were greater than 100%, as
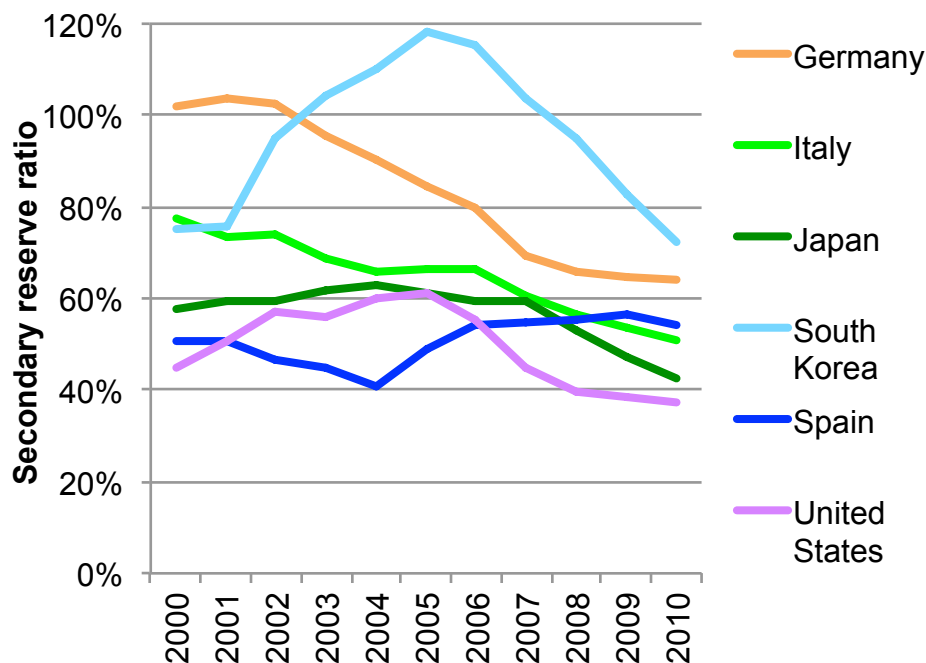
Figure 3.7 Five-year moving average of estimated secondary reserve ratios

were South Korea's in the mid-2000s. In addition, China SRRs were negative (as low as –280%) during 2000–2010, which means that the provision of process and EoL scraps was greater than the use of these scraps. Graedel et al. (2004) and Wang et al. (2008) added "phantom" flows when input flows did not equal output flows for each process and showed large "phantom" flows for waste management processes in China's copper cycles. This phenomenon can result from lack of transparency in traded second-hand products or illegal (not reported) scrap trade. Their actual amounts were estimated as probably higher than the reported (Janz and Bilitewski 2009). That strongly affects the SRRs. For our estimates of secondary copper reserves in the following section, we used the global average for China (a maximum of 50% during 2000–2010 and 43% in 2010) (Gloser et al. 2013). For Germany and South Korea, we used 80% as the maximum SRR.

Our sensitivity analyses showed that SRR was affected most by the change in fabrication efficiency (see Figure S2 in the SI in the Appendix B). Fabrication efficiency directly changes the amount of process scrap generation, which in turn changes the .amount of EoL scrap use (see equations (15) and (13)). A 10% increase in fabrication efficiencies increased SRR by more than 10% for South Korea, by 7–8% for Italy and Spain, and by 4% for Germany, Japan, and the United States.**3.2.3 Estimated Secondary Copper Reserves and Their Comparison with Primary Copper Reserves.** Figure 3.8 presents the estimated secondary copper reserves for our study countries along with the countries with the five highest primary reserves in 2010 (China and the United States are in both groups). According to the U.S. Geological Survey (USGS 2011), Chile has the largest primary copper reserves with 150 Mt in 2010, followed by Peru with 90 Mt and Australia with 80 Mt. Our estimates show that the

United States (44 Mt) and China (33 Mt) have the largest secondary copper reserves. Germany, Italy, and Japan have similar secondary reserves (about 15 Mt). Integrated management of both primary and secondary copper resources is important in China and the United States because they have similar amounts of primary and secondary reserves. We estimated global secondary copper reserves of about 175 Mt by multiplying global copper stocks in use in 2010 (about 350 Mt) and the highest EoL recycling ratio during 2000–2010 (50%) (Gloser et al. 2013). This amount represents about 30% of global primary reserves of 630 Mt in 2010 (USGS, 2011). The current secondary reserves at the global level are therefore still much smaller than primary reserves.

On a per-capita basis, Chile has the highest primary reserves, with about 8,700 kg/person followed by about 3,600 kg/person in Australia, and about 3,000 kg/person in Peru. These amounts are considerably larger than the per-capita secondary reserves in the study countries, where the greatest average was 250 kg/person in Italy, followed by about 180 kg/person in Germany and South Korea. Although the total amount of primary copper reserves in the world is much greater than that of secondary copper reserves, secondary reserves are expected to serve an important role in countries with no primary reserves. Knowing the amounts of both primary and secondary reserves is expected to be important for planning the national copper supply. SRR is an important factor in increasing the amount of secondary reserves. The amount of secondary reserves can be increased if SRR can be increased through enhanced EoL recycling. Key stakeholders such as scrap collectors, copper producers, and policymakers are expected to serve important roles in improving SRR. Increasing secondary reserves also has positive effects on the domestic economy.
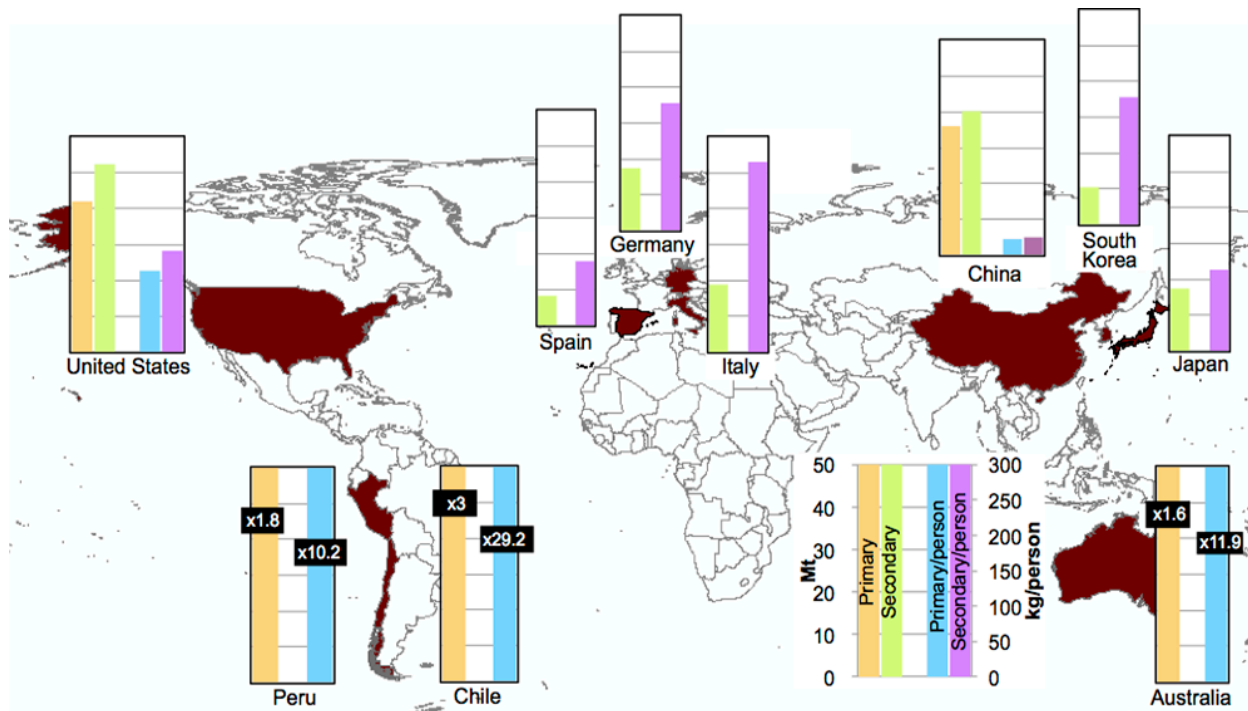
Figure 3.8. Distribution of primary and estimated secondary copper reserves in 2010. Primary reserves are shown for the countries with the five largest primary reserves; secondary reserves are shown for the study countries. China and the United States are in both groups.

In terms of the timing of extraction and production, notable differences exist between primary and secondary reserves. For primary reserves, it is possible to increase extraction and production through the expansion of mining and refining capacities. However, increasing extraction and production from secondary reserves is not easy because the annual maximum amount of available secondary reserves is restricted by the amount of EoL scrap generated. In other words, the availability of secondary copper reserves for recycling is limited. It is also noteworthy that the finished products in some end-use sectors remain in use for decades (Gloser et al. 2013).

3.2.4 Application of Classification Framework for Secondary Copper Resources.

We applied the classification framework for secondary copper resources to Italy, Japan, and the United States in 2010 as representatives of major countries in Europe, Asia, and North America and as countries for which our calculations were completed in a consistent manner. We also applied the framework globally using values estimated by Glöser et al. (2013).

Italy, Japan, and the United States respectively have secondary copper resources of about 25.9 Mt, 37.5 Mt, and 101.5 Mt (see Table 3.3). Italy has 10.3 Mt (40%) of economically recoverable secondary copper, whereas Japan and the United States have 9.8 Mt (26%) and 26.4 Mt (26%), respectively. The reason for Italy's higher share of economically recoverable secondary copper is that its estimated SRR was higher throughout the analyzed years. The secondary reserves including marginally economic secondary copper for those countries were estimated, respectively, as 15.7 Mt (61%), 14.5 Mt (39%), and 43.7 Mt (43%). The yearly available secondary copper (economic and marginally economic resources emerging in a year) was estimated as about 1–2% for each country: a small amount of secondary copper is useful on a yearly basis, which is an important consideration for urban mining. However, the amount of yearly available secondary copper for Japan, for example, represents more than 70% of its recent annual copper consumption, presenting many implications for domestic industry and resource supply security.

The United States has a large amount (23.0 Mt) of secondary copper deposited as waste in landfill sites: almost equal to Italy's total amount of secondary copper (25.9 Mt) and the United States' economic secondary resources (26.4 Mt). Japan has similarly large amounts of such resources in landfill sites (10.9 Mt). These deposits represent

potential targets for the future extraction of secondary copper through landfill mining. As shown in Table 3.3(d), about 550 Mt of copper has been extracted at the global level, 150.5 Mt (27%) of which are economically extractable and 24.5 Mt (4%) are marginally economic secondary resources. About 130.0 Mt (24%) are in landfill sites and 175.0 Mt (32%) will be disposed of, dissipated, or lost in mixed metals. These flows must be decreased to use the resource more efficiently.

For a long-term perspective, primary and secondary copper resources are more important than reserves. According to Mudd et al. (2012), Chile has the largest primary copper resources with 658 Mt in 2010, followed by the United States with 170 Mt and Peru with 168 Mt. As presented in Table 4.1, the United States has about 90 Mt of secondary copper resources in all (dissipated materials and mixed metal loss were excluded), which is about half of its primary copper resources. Globally, about 480 Mt of secondary copper resources have been identified, which is comparable to Chile's primary copper resource.

Table 3.3 Classification of secondary copper resources for (a) Italy, (b) Japan, (c) the United States, and (d) globally in 2010

a) Italy	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	300 kt 1%	10,000 kt 38%			
Marginally economic	200 kt 1%	5,200 kt 20%			
Sub-economic & other	200 kt 1%	4,400 kt 17%	4,200 kt 16%	500 kt 2%	Total
Mixed metal loss	900 kt 4%				25,900 kt 100%

b) Japan

b) Japan	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	500 kt 1%	9,300 kt 25%			
Marginally economic	200 kt 1%	4,500 kt 12%			
Sub-economic & other	400 kt 1%	8,100 kt 22%	10,900 kt 29%	1,200 kt 3%	Total
Mixed metal loss	2,400 kt 6%				37,500 kt 100%

c) United States

c) United States	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	900 kt 1%	25,500 kt 25%			
Marginally economic	600 kt 1%	16,700 kt 16%			
Sub-economic & other	900 kt 1%	26,500 kt 26%	23,000 kt 23%	2,400 kt 2%	Total
Mixed metal loss	5,049 kt 5%				101,500 kt 100%

d) Globally

| d) Globally	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	5,200 kt 1%	145,300 kt 26%			
Marginally economic	800 kt 0%	23,700 kt 4%			
Sub-economic & other	6,000 kt 1%	169,000 kt 31%	130,000 kt 24%	35,000 kt 6%	Total
Mixed metal loss	35,000 kt 6%				550,000 kt 100%

Overall, our proposed classification framework provides a better understanding of the current size of available secondary resources and waste deposits. The framework highlights the need for integrated management of primary and secondary resources. Moreover, it is applicable to other important metals as well as non-metallic resources to move toward the sustainable use of resources. Projection of future secondary reserves remains an interesting topic for future research, especially for metals which currently have low SRR (EoL recycling rate (Graedel et al. 2011)) but have high recycling potential. Moreover, using the potential recyclability rate (PRR) (Ciacci et al. 2015), future assessments can differentiate sub-economic secondary resources from unrecoverable materials (other).

3.3. Assessment of secondary zinc reserves of nations

3.3.1. Analysis of Zinc Stocks in/after use

Figure 3.9 shows the trends of zinc stocks in six major countries with top zinc consumption. The United States' zinc stocks were the largest throughout the study period, follwed by Japan and China. The zinc stocks of the United States in 1990 were estimated as 16 Mt and 27 Mt in 2010, a 68% increase in 20 years. The zinc stocks in Japan were increased from 10 Mt in 1990 to 21 Mt in 2010, with a two-fold increase. Until 1990s, the zinc stocks of China were the lowest among the six major countries and were approximately estimated at 4 Mt in 1990 and 26 Mt in 2010, about more than eight-fold increase that is characterized by a moderate growth until the beginning of the 1990s and a rapid growth after that.
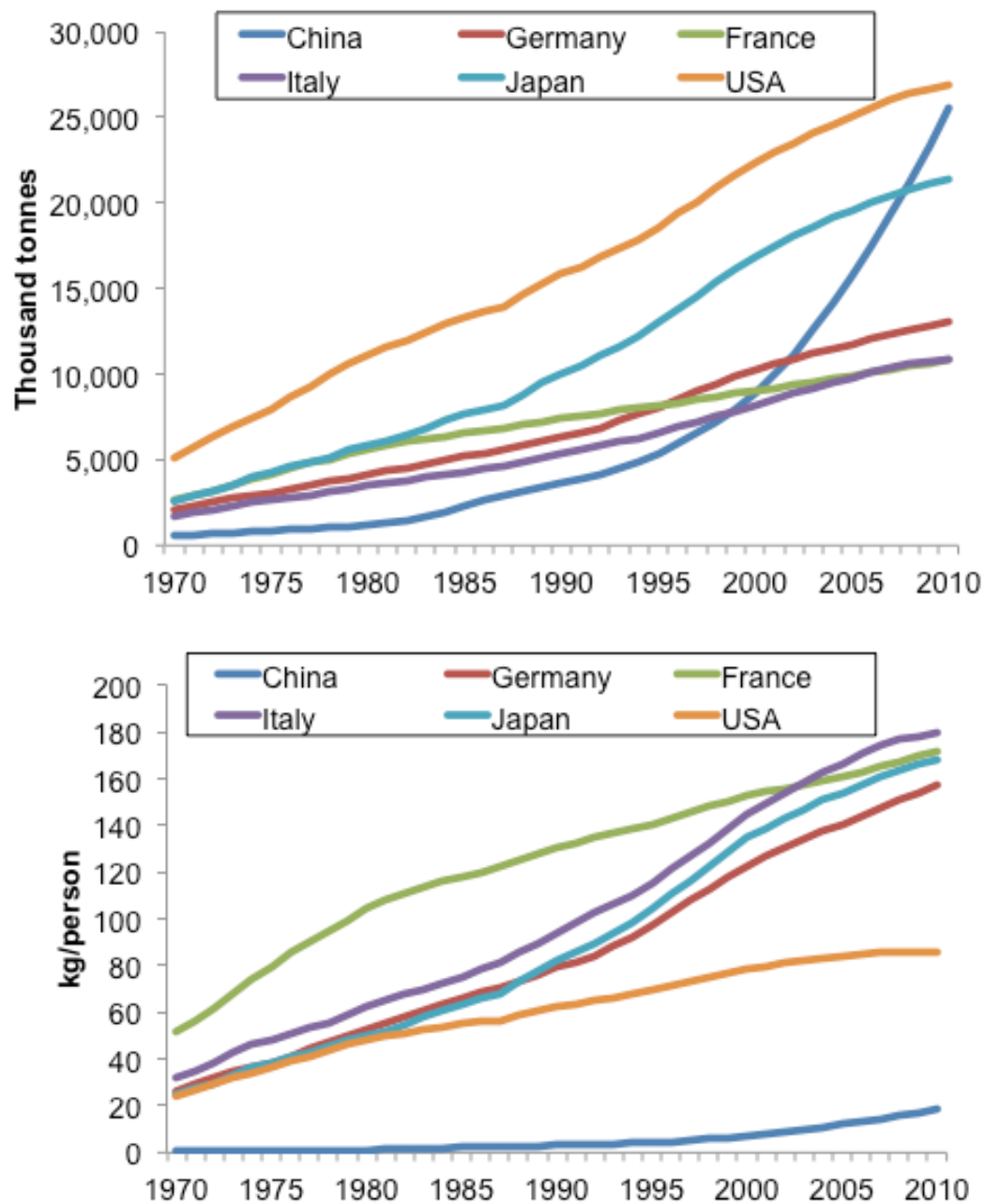
Figure 3.9 Historical patterns of zinc stocks and its per capita

Furthermore, the zinc stocks of France, Germany and Italy were increased respectively

from 7, 6 and 5 Mt in 1990 to 11, 13 and 11 Mt in 2010, with an increase of 57%, 100%

and 100%. Aside from China, in all of developed countries, the trends of growth in zinc

stocks have show a sign of slowdown in recent years. It is noteworthy that the developed nations must have had a certain amount of zinc which has already been accumulating in 1962: therefore, our estimates are somehow underestimated.

On a per-capita basis in 2010, Italy had the highest per capita zinc stocks with 179 kg with subsequent 171 kg in France and 168 kg in Japan as depicted in Fig. 3.9. Among the developed countries, the United States' per capita zinc stocks was estimated as the lowest despite holding their largest zinc stocks. Even though its total stocks are increasing rapidly, per capita zinc stocks in China was also the lowest throughout the study period as shown in Figure 3.9.

Jolly (1992) estimated that the zinc stocks in-use in the United States was about 23 Mt in 1990, which is greater than our value of about 17 Mt and the same to our estimate in 1999 as shown in Figure 5.1. One possible reason is the use of the longer time-series statistical data to estimate in-use zinc stocks in Jolly (1992). Considering only die-casting, galvanized sheets and other galvanized products in Japan, Tabayashi et al. (2009) estimated that the zinc stocks that is recyclable were about 3.3 Mt in the 2005 which are smaller than our results. In the present study, zinc alloy, rolled zinc, zinc oxides and other uses were included in the zinc stocks but they were ignored completely in the estimate of Tabayashi et al. (2009). This discrepancy is attributable to the use of the different definitions of zinc stocks and time-series data. In the present study, we defined zinc stock as all the zinc in use stage and did not distinguished if it would be recycled or not.

After reviewing 54 studies of metal in-use stocks, Gerst and Graedel summarized that per capita zinc in-use stocks in the more developed countries range from 80–200 kg which are very close to our estimates. In 1990, Jolly (1992) also

estimated U.S. per-capita zinc stocks of 92 kg which is larger than our per-capita results of 65 kg. Using concentrations of zinc in the main in-use reservoirs and geographic information system (GIS) data sets, van Beers and Graedel (2006) estimated per capita in-use zinc stocks of 205 kg in Australia which is also bigger than our values of Italy, France, Japan and Germany. Figure 3.10 shows the sensitivity analysis result of the zinc stock estimates for the China, Germany, France, Italy, Japan and the United States. The main contribution to zinc stocks came from the data on fabrication efficiency by end-use sectors.
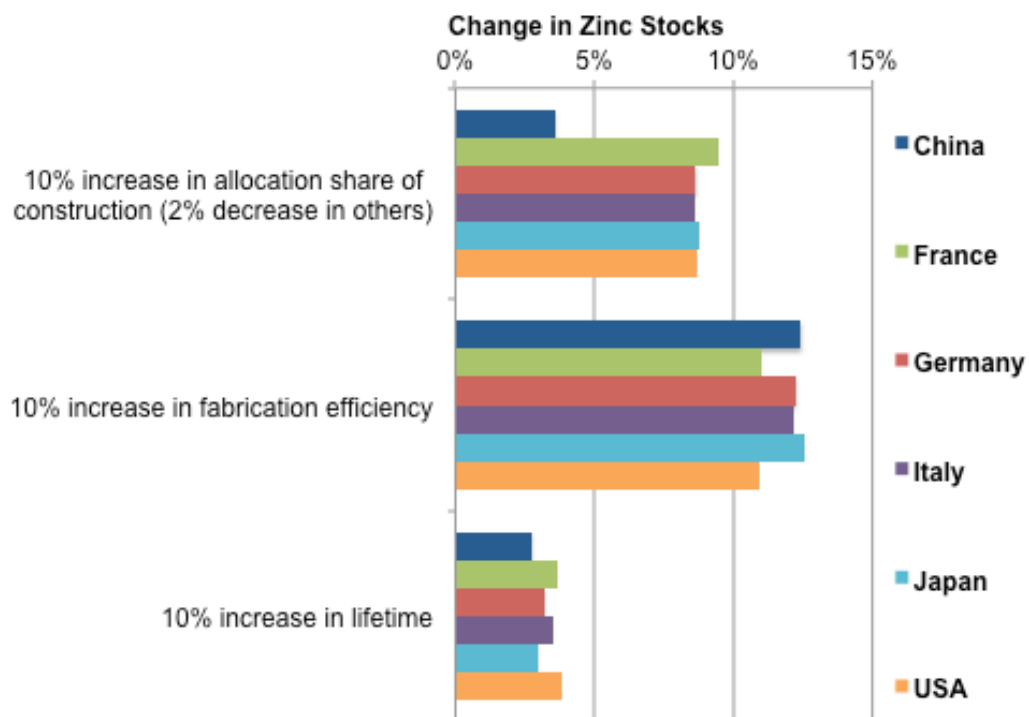
Figure 3.10 Sensitivity analysis of the zinc stock estimates

3.3.2. Trends in Secondary Reserve Ratio
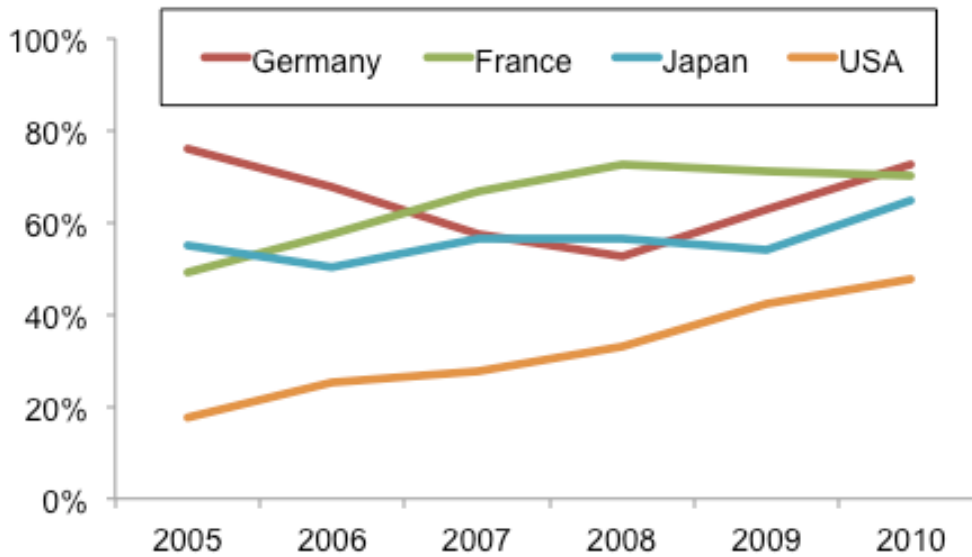
Figure 3.11. Three-year moving average of estimated secondary reserve

In this model, Secondary Reserve Ratio (SRR) is the most important variable for determining the amount of secondary zinc reserves. Figure 3.11 shows the three-year moving average of estimated *SRR(t,c)* in equation (10) in Germany, France, Japan and United States.

Estimated secondary reserve ratios, *SRR(t,c)* in equation (10), were fluctuated because the estimated recovered EoL scraps, *REoLS(t,c)*, were fluctuated. As shown in equation (11) and (12), *REoLS(t,c)* was estimated based on utilized scrap, *US(t,c),* and recovered process scrap, *RPS(t,c)*. The fluctuations in *REoLS(t,c)* are mainly driven from the fluctuations in *US(t,c)*, which are largely affected by the accuracy of statistical dataset and other factors. During 2005–2010, secondary reserve ratios for almost all major countries are in the range of about 25 and 80 percent. Despite using a three-year moving average, *SRR(t,c)* of Italy and China were out of range (negative).

Norgate and Rankin (2002) reported that recycling rate were 36% at the global scale and 40% in the United States that closely approximates our estimated value around 2008. At the country level, Meylan and Reck (2016) revealed that the estimates of EoL recycling rate in 2010 are 33% at global scale, 44% in Germany, 48% in France, 43% in Japan and 37% in the United States, respectively that are smaller than our estimates of Germany, France, Japan and the United States in 2010. Graedel et al. (2011) estimated that the global average EoL recycling rate of zinc was greater than 50% during the 2000–2005 periods In most cases, our estimates are comparable to these values during the 2005–2010 period. Alonso et al. (2007) explained that the resource prices influence demand and recycling rate. In this study, the estimated SRR is the fraction of in/after-use zinc stocks that are technologically and economically recoverable, not meaning the potentially recoverable fraction. Ciacci et al. (2015) provided that using global market share basis, potential recyclability rate (PRR) of zinc was about 77% that are almost identical ratios in our estimate of 2010. As described previously, China and Italy's SRRs were negative (as low as -200%) during 2005–2010, indicating that the amount of process and EoL scraps was less than the use of these scraps as in equation (13). Graedel et al. (2005) added additional flows to achieve mass balance of the contemporary zinc cycle and showed a dotted-line box of additional flows for zinc production process. Furthermore, this situation can be attributable to lack of transparency in traded second-hand products or illegal (not reported) EoL scrap trade. The actual amounts were estimated as probably higher than the reported amounts (Janz and Bilitewski, 2009). It strongly affects the SRRs.

For estimated secondary zinc reserves in the following section, the global average EoL recycling rate (maximum of 41% during 2000-2009 and 33% in 2010) was used for China and Italy (Meylan and Reck 2016).

Figure 3.12 shows the sensitivity analysis result of the SRR estimates for Germany, France, Japan and the United States. It indicated that SRR results are mainly sensitive to the change in end-use allocation share.
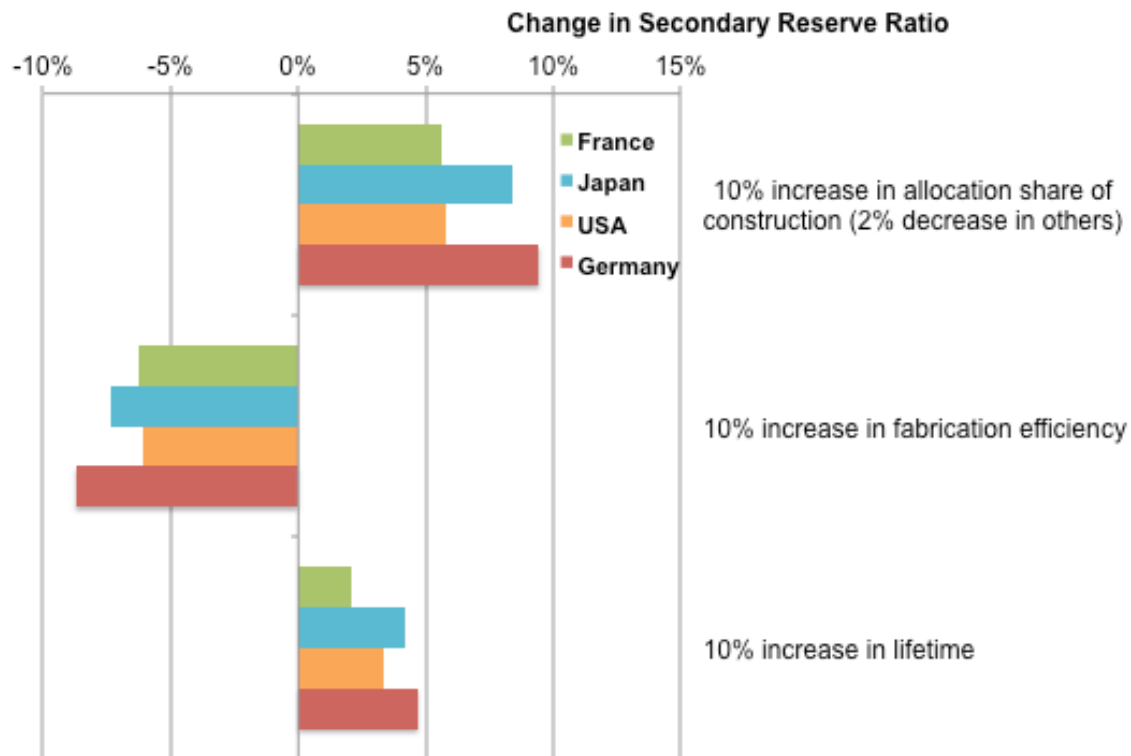
Figure 3.12. Sensitivity analysis of the SRR estimates ratios

3.3.3. Estimated Secondary Zinc Reserves and Their Comparison with Primary Zinc Reserves

Figure 3.13 presents the estimated secondary zinc reserves in our study countries along with the countries with the top four primary reserves in 2010 (the United States are in both groups). USGS (2011) reported that Australia has the largest primary zinc reserves of 53 Mt which is approximately 21% of global zinc reserves, followed by China with 42 Mt (17%) and Peru with 23 Mt (9%) in 2010. Our results show that the United States has the amount of the estimated secondary zinc reserves of 12.8 Mt, 13.9 in Japan and 9.5 Mt in Germany. China, Italy, and France have in the range of 5 and 10 Mt of secondary zinc reserves in 2010. Integrated management of both zinc resources is important for China and the United States because they have both amounts of primary and secondary zinc reserves whereas the countries such as Japan, Germany, France and Italy have only secondary zinc reserves. Countries with only secondary zinc reserves need to install inclusive policy setting for improving domestic recycling infrastructure to handle the generated amount of EoL zinc scraps, reducing not only risks of supply chain disruption of raw material (geopolitical, economic or social limits) but also dependencies on depleting zinc resources. In 2010, our total estimated amount of secondary zinc reserves for the study countries is 60 Mt, which represents about 24% of global primary zinc reserves of 250 Mt in 2010 (USGS, 2011). The quantity of secondary zinc reserves of Japan and Germany is similar to that of the primary zinc reserves of Peru and the United States.

On a per-capita basis, Australia has the highest primary reserves of about 2,300 kg/person with subsequent about 780 kg/person in Peru, and about 38 kg/person in the United States. It is interesting to note that these values are significantly greater than the

per-capita secondary zinc reserves in the study countries, where the largest number is about 128 kg/person in France with subsequent about 126 kg/person in Japan and about 125 kg/person in Germany. The amounts of per-capita primary zinc reserves are 20-fold greater than per-capita secondary zinc reserves of China.

Necessarily, secondary reserves of zinc are expected to play an important role in countries with no primary zinc reserves. Understanding the sizes of both primary and secondary zinc reserves becomes essential for informing and establishing comprehensive mechanisms for sustainable zinc metal governance and use within the boundary and beyond. In determining the ups and downs in the secondary zinc reserves, the role of SRR variable is important. The amount of secondary reserves can be increased through making SRR elevated that is promotion of recycling. It requires that technological competency and infrastructure be in place.

The difference between primary reserves and secondary reserves of zinc: For primary reserves, the availability of primary zinc reserves is limited not only by the quantity that is economically and technologically extractable but also geopolitical settings in the area. Moreover, supply chain of primary resources are most likely affected by domestic political changes in the resource-rich countries. For secondary zinc reserves, the availability of zinc stocks in use for reutilization is regulated by the quantity of EoL products entering waste management system. In other words, it explains that the potential recovery of secondary zinc reserves is determined by product lifetime. The zinc products remain in use until the end of their lifetime, varying from a few years to some decades (see Table S3 in Appendix C).
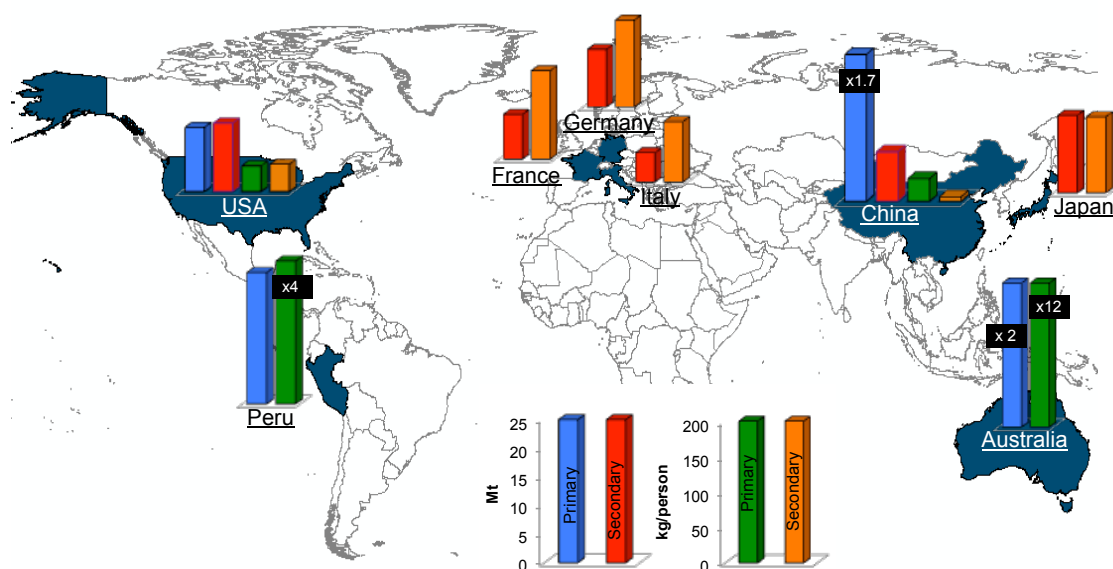
Figure 3.13 Distribution of primary and estimated secondary zinc reserves in 2010. Primary reserves are shown in the countries with the top four primary reserves; secondary reserves are shown in the study countries. United States and China are in both groups.

3.3.4. Application of Classification Framework for Secondary Zinc Resources

The classification framework for secondary resources was applied to zinc for the United States, Japan and Germany in 2010 as representatives of major countries in North America, Europe and Asia and as countries for which our results were done in a consistent manner.

As presented in Table 3.4, the United States, Japan, and Germany have respective total amount of secondary zinc resources of about 73 Mt, 44 Mt and 21 Mt. Germany has 10 Mt (46%) of economically recoverable secondary zinc whereas Japan and United States have 14 Mt (32%) and 13 Mt (17%). Germany's higher share of economically recoverable secondary zinc was observed throughout the study years. The

secondary zinc reserves including marginally economic secondary reserves were estimated at 10 Mt (49%) in Germany. The percentage of the yearly available secondary zinc (economic zinc resources emerging in a year) was between 0.5% and 1% for each country: such an amount of secondary zinc is available on a yearly basis, that is an important information for recovery of these resources. However, the amount of yearly available secondary zinc represents more than 38% of its recent annual zinc consumption in Japan, 33% in the United States and 48% in Germany, respectively, motivating those countries to tap actively their urban mines. The United States has a large amount (15 Mt) of secondary zinc resources as wastes in managed landfill sites, that is comparable to the Japan's secondary zinc reserves (14 Mt). Similar amount of secondary zinc resources is also in landfill sites (7 Mt) in Japan. In the future, these deposits present potential sources for extraction of secondary zinc through landfill mining activities. Therefore, the future extractability of secondary zinc mainly depends on the current management of these deposits

The application of the presented classification framework provides sizes and locations of secondary zinc resources in detail that is useful for industry and policy actors to maximize access to valuable secondary zinc sources for zinc waste management and resource reutilization. To promote recycling, quality, grade and design of EoL zinc scrap and such are key elements. Beyond these requirements, quantitative assessment of secondary zinc reserves presented by the classification framework play fundamental information for system-wide management strategies of secondary zinc resources toward circular economy. To meet their zinc demand, the estimates of secondary zinc reserves and resources as in the classification framework can be

regarded as the major domestic potential exploitable zinc resources towards the security

of zinc supply and circular economy in the future.

Table 3.4. Classification of secondary zinc resources in (a) the United States, (b) Japan, (c) Germany in 2010

a) United States

	Products in/after use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	325 kt 0%	12,461 kt 17%			
Marginally economic	0 kt 0%	0 kt 0%			
Subeconomic & others	357 kt 0%	13,711 kt 19%	15,459 kt 21%	10,224 kt 14%	Total
Mixed metal losses	20,001 kt 28%				72,539 kt 100%

b) Japan

	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	305 kt 1%	13,572 kt 31%			
Marginally economic	0 kt 0%	0 kt 0%			
Subeconomic & others	166 kt 0%	7,392 kt 17%	7,504 kt 17%	5,655 kt 13%	Total
Mixed metal losses	9,908 kt 22%				44,265 kt 100%

c) Germany

	Products in use		Waste in managed landfill sites	Dissipated materials	
	Emerging in a year	Not emerging in a year			
Economic	243 kt 1%	9,182 kt 43%			
Marginally economic	12 kt 0%	476 kt 2%			
Subeconomic & others	54 kt 0%	3,054 kt 14%	2,166 kt 10%	3,835 kt 18%	Total
Mixed metal losses	2,254 kt 11%				21,279 kt 100%

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Chapter 4

Conclusions and future work

4.1 Summary and discussion of the findings

The proposed framework identifies not only the amount of secondary reserves but also the quality of secondary resources. The framework presented and applied by this study provides a standardized method for assessing stocks of secondary reserves and resources, in a way that is somewhat comparable with how geologic mineral resources/reserves are defined. Various research groups are actively researching how copper and other metals flow through the economy and build up in secondary stocks. However, this is the first study to date that has attempted to systematically apply the principles for geologic resource estimation to quantify economic categories to secondary reserves/resource.

Based on historical metal flow studies, a novel method was used for a top-down account of stocks of aluminum, copper and zinc. In 2010, total stocks of aluminum, copper and zinc was estimated at 152 Mt, 70 Mt and 27 Mt in the United States; 97 Mt, 67 Mt and 26 Mt in China; and 41 Mt, 25 Mt and 21 Mt in Japan. It was proven that our approach delivers reasonable results and has merits in comparison to other top-down and bottom up approaches for these metal stocks. In three metals, growing trends of China' stocks skyrocketed particularly after the Open and Reform Policy adopted in 1970s. During the past 20 years, Germany, France, Japan and the United States faced the stable per-capita stocks due to their declining consumption trends.

The secondary reserve ratio SRR, the fraction of stocks in/after use that are economically and technologically recoverable was estimated for determining the amount of secondary reserves in this works. During 2005-2010, trends of SRRs of three metals

were above 50% in almost all countries except the United States, meaning large portion of EoL products are entering other final destinations such as landfills and mixed metal losses. In both Japan and the United States, SRRs of aluminum were larger than that of copper and zinc. In Germany, copper's SRRs were bigger than those of aluminum and zinc.

Among major countries, the result shows that the estimated amount of secondary aluminum reserves was 85 Mt in the United States, 65 Mt in China and 29 Mt in Japan in 2010, respectively. On the per-capita secondary reserves, the highest was 383 kg in Norway followed by about 308 kg in Switzerland and about 273 kg in the United States.

To estimate secondary copper reserves, five year moving average of SRRs was estimated in the 2000s. Our estimates show that the United States (44 Mt) and China (33 Mt) have the largest secondary copper reserves. Germany, Italy, and Japan have similar secondary reserves ($\sim$15 Mt). We also estimated global secondary copper reserves of ~175 Mt which represents ~30% of global primary reserves of 630 Mt in 2010. These amounts are considerably larger than the per-capita secondary reserves in the study countries.

Our results show that the United States has the amount of the estimated secondary zinc reserves of 12.8 Mt, 13.9 in Japan and 9.5 Mt in Germany. On a per-capita basis, Australia has the highest primary reserves of about 2,300 kg/person with subsequent about 780 kg/person in Peru, and about 38 kg/person in the United States. It is interesting to note that these values are significantly greater than the per-capita secondary zinc reserves in the study countries, where the largest number is about 128 kg/person in France with subsequent about 126 kg/person in Japan and about 125

kg/person in Germany. The amounts of per-capita primary zinc reserves are 20-fold greater than per-capita secondary zinc reserves of China.

Overall, our classification framework provides better understanding of the size and locations of available secondary resources and wasted deposit so far and in the future for integrated management of primary and secondary resources. Secondary reserves of aluminum, copper and zinc in the United States were the largest, followed by China and Japan, meaning that these countries need to focus on effective management of wastes and secondary resources (EoL products) in the near future and far future. For a long-term perspective, it highlights the need for integrated management of primary and secondary copper reserves.

The classification framework of the secondary resources employed in this study prove to be practical tool in the assessment of secondary reserves and would be very useful on urban mining and landfill mining. The numbers in the frameworks presented in this study may become useful to policy setting that enhance the efficiency and effectiveness of EoL products management in the study countries. The types, forms, and the amount of metals stocked in society are required for the appropriate management of waste and effective reuse of secondary resources. However, urban mining is technologically far more challenging than mining in the geosphere.

4.2. Overall achievement of the research objectives

Based on previous work derived from the conceptual framework utilized to classify primary resource and reserve and exemplified by the McKelvey diagram, these findings present metal stocks over time in different major countries, application of the modified classification framework for estimating secondary reserves and resources and

compared with their primary reserves. The framework presented and applied in this study provides a standardized method for assessing stocks of secondary reserves and resources, in a way that is somewhat comparable with how geologic mineral resources/reserves are defined. These findings help to fill the gap in the field of industrial ecology, meaning potentially closing the loop in the circular economy from theory to practice. The findings can be summarized by revisiting the research objectives.

1. To investigate the applicability of the framework for aluminum, copper and zinc

It was found that this classification framework of secondary resources could successfully be applied to aluminum, copper and zinc in not only individual country but also global scale. The framework was applied in representatives of major countries in Europe, Asia, and North America and countries with calculation in a consistent manner.

2. To assess the secondary reserves and resources for those metals

Secondary Reserve Ratio (SRR), which is used to estimate the secondary copper reserves in specific country and year, is the most important variable in the study. Using SRRs, secondary aluminum reserves and resources were estimated in France, Japan, the United States and globe. In Italy, Japan, the United States and globe, secondary copper reserves and resources were assessed. Secondary zinc reserves and resources were estimated in Germany, Japan and the United States.

4.3. Implications of the findings

In order to do the validity of results, research has to include clear knowledge on the amount of secondary reserves that are likely to emerge in a year and in the future. Research on estimating secondary reserves is still in its starting point and hence little is known on it. In this study, like the existing primary resource classification, new modified classification framework for secondary metals resources was presented with very useful information, namely the estimated amount of secondary reserves and resource (including time dimension) that will cause tremendous impacts on the secondary production of metals and will reduce reliance on primary metal sources.

The findings of this study can contribute to the significant topic currently debated around resource scarcity, depletion and the availability of secondary resources in the future. This new information on secondary reserves (in a year and the future) and secondary resources in final destinations may become very useful in urban mining that focuses on the availability of secondary resources by economic feasibility. With the growing interest of decision makers and international organizations in sustainable waste and resource management, the understanding of secondary reserves, defined as the fraction of copper stocks in use that are technically and economically recoverable and the amount of waste in managed landfill sites, dissipated materials and mixed metal losses are becoming crucial. This research offers a sound, robust classification framework of secondary resources and allocation methods for use in this discourse. The significance of the results for long-term metal supply could be explored further. Hence, its contribution to existing literature is both methodically and with regard to content referring to the spatial distribution of stocks in use, secondary reserves and resources.

4.4. Limitations of the study

First, it was assumed that the market share of finished products for 2013 (the five end-use sectors in Chapter 4) was the same for all the years analyzed, due to limited data availability. The regional models of allocation of semi-fabricated to end-use products was used for all the years analyzed In the Chapter 5. However, market share of finished products may be different year by year depending on many factors.

A second limitation, data on country-specific and time-series fabrication efficiencies and country-specific mean lifetimes of copper products are very limited. Therefore, global average fabrication efficiencies and mean lifetimes were used.

Third, there should be severe difference in the composition of machinery products in different countries and metal content in export flows compared to import flows. In our works, the same content in both imports and exports was used.

Fourth, to allocate the amount of waste in landfills, landfill ratio in a year was used for all the study years in Chapter 3, 4 and 5. Practically, it seems different in every year.

4.5. Future works

The classification framework, allocation methods on it and SRR presented in this study can be used for further case studies of other sub-national territories, countries, global regions and the entire globe. It is also applicable to other important metals as well as non-metallic resources to track on moving toward the sustainable use of resources.

Moreover, projection of future secondary reserves remains an interesting topic for future research. In addition, future assessment can differentiate sub-economic secondary resources from unrecoverable materials (other), using the potential recyclability rate (PRR) (Ciacci et al. 2015).

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Ciacci, L.; Reck, B.K.; Nassar, N.T.; Graedel, T.E. Lost by design. 2015**.** *Environ. Sci. Technol.*, 49, 9443-9451.

APPENDICE A: SUPPLEMENTARY MATERIAL: SECTION 3.1

S1. List of variables used in the paper

$AC(t',c,i)$	aluminum consumption for finished product i in country c in year t'
$AS(t,c,i)$	aluminum stocks of finished products i used in country c in year t
$DcR(y,c,i)$	discard ratio of finished product i in country c after y years
$DpR(t',c,i)$	dissipation ratio of finished product i in country c in year t'
$DSSP(t',c)$	domestic shipment of semi products in country c in year t'
$EXFP(t,c,i)$	exported finished products i in country c in year t
$EoLRR(t,c,i)$	End-of-life recycling rate of finished product i in country c in year t.
$FE(t,c,i)$	fabrication efficiency of semi-finished product i for the production of finished products in country c in year t
$FP(t',c,i)$	finished product i production in country c in year t'
$GEoLS(t,c,i)$	generated EoL scrap of finished product i in/after use in country c in t
$IMFP(t,c,i)$	imported finished products i in country c in year t
$IMSP(t',c)$	imported semi-finished product in country c in year t'
$LFR(t',c)$	landfill ratio in country c in year t'
$MS(t,c,i)$	market share of semi-finished products for finished product i in country c in year t
$MSAR(t,c)$	marginal secondary aluminum reserves in country c in year t
$MSARe(t,c)$	marginal secondary aluminum reserves emerging in a year in country c in year t
$MSARn(t,c)$	marginal secondary aluminum reserves not emerging in a year in country c in year t
$REoLS(t,c)$	recovered EoL scrap in country c in year t
$RMMLOR(t',c)$	ratio of mixed metal losses and other repositories in country c in year t'
$SAR(t,c)$	secondary aluminum reserves in country c in year t
$SARe(t,c)$	secondary aluminum reserves emerging in a year in country c in year t
$SARn(t,c)$	secondary aluminum reserves not emerging in a year in country c in year t
$SC(t,c,i)$	semi-finished product i consumption in country c in year t
$SR(y,c,i)$	survival ratio of finished product i in country c after y years
$SRR(t,c)$	secondary reserve ratio in country c in year t
$SSARUMe(t,c)$	sub-economic secondary aluminum resources and unrecoverable materials (others) emerging in a year in the column of products in/after use in country c in year t
$SSARUMn(t,c)$	sub-economic secondary aluminum resources and unrecoverable materials (others) not emerging in a year in the column of products in/after use in country c in year t
$SSARUMw(t,c)$	sub-economic secondary aluminum resources and unrecoverable materials (others) in managed landfill sites
$SSARUMd(t,c)$	sub-economic secondary aluminum resources and unrecoverable materials (others) in the dissipated materials
$SSARUMm(t,c)$	amount of unrecoverable materials (mixed metal losses)
$TSC(t',c)$	total semi-product consumption in country c in year t'

S2. Estimation of aluminum stocks of products in use and generated end-of-life scrap

The amount of aluminum stocks of finished products in use was estimated using the following equation.

$$AS(t,c,i) = \Sigma_{t'} AC(t',c,i) \times SR(t-t',c,i) \qquad (S1)$$

In that equation, $AS(t,c,i)S_{(t,c,i)}$ stands for the aluminum stocks of finished product i in use in country c in year t, $F^{Con}_{(t',c,i)}$ $AC(t',c,i)$ signifies the aluminum consumption for finished product i in country c in year t', and $SR(y,c,i)RR_{(t-t',c,i)}$ denotes the survival ratio of finished product i in country c after y years.

The aluminum consumption $AC(t',c,i)$ was estimated using the following equation.

$$AC(t',c,i) = \Sigma_t FP(t',c,i) + IMFP(t',c,i) - EXFP(t',c,i) \qquad (S2)$$

Therein, $FP(t',c,i)$ denotes the amounts of finished product i production in country c in year t', $IMFP(t',c,i)$ is the amounts of aluminum contained in imported finished product i in country c in year t'. $EXFP(t',c,i)$ is the amounts of aluminum contained in exported finished product i in country c in year t'

The production of finished products $FP(t',c,i,)$ was estimated using the following equation.

$$FP(t',c,i) = SC(t,c,i) \times FE(t',c,i)/100$$
$$= (TSC(t',c) \times MS(t',c,i)/100) \times FE(t',c,i)/100 \qquad (S3)$$

In that equation, $SC(t,c,i)$ is the amount of semi-finished product i consumption in country c in year t. $TSC(t',c)$ represents the amount of total semi-finished product consumption in country c in year t'. $MS(t',c,i)$ is the market share of semi-finished product for finished product i in country c in year t' (%). $FE(t',c,i)$ signifies the fabrication efficiency of semi-finished product i for the production of finished products in country c in year t' (%).

The total semi-finished product consumption $TSC(t',c)$ was estimated using the following equation.

$$TSC(t',c) = DSSP(t',c) + IMSP(t',c) \qquad (S4)$$

Therein, $DSSP(t',c)$ denotes the amount of domestic shipment of semi-finished product in country c in year t', $IMSP(t',c)$ is the amount of imported semi-finished product in country c in year t'.

The survival ratio $SR(y,c,i)$ is calculable using the following equation.

$$SR(y,c,i) = EXP[-\{y/Yci\}^{dci} \cdot \{\Gamma(1+1/dci)^{dci}\}] \qquad (S4)$$

where y denotes the age of finished products, Yci signifies the average lifetime of finished product i in country c, Γ is the gamma function, dci is the parameter of distribution range of finished product i in country c.

In addition, generated end-of-life scrap was calculated as

$$GEoLS(t,c,i) = \Sigma_{t'} AC(t',c,i) \times DcR(t-t',c,i) \qquad (S5)$$
$$DcR(y,c,i) = SR(y,c,i) - SR(y+1,c,i) \qquad (S6)$$

where *GEoLS*(*t*,*c*,*i*) stands for generated end-of-life scrap of finished product *i* in/after use in country *c* in year *t* and *DcR*(*y*,*c*,*i*) is the discard ratio of finished product *i* in country *c* after *y* years.

S3. Aluminum contents of traded commodities

SITC Rev. 1	Items	Unit	Aluminum content	Source
2833	Bauxite and concentrates of aluminum	t/t	0.455	Roskill, 2005
51365	Aluminum oxide and hydroxide	t/t	0.444	Assumption
28404	Aluminum waste and scrap	t/t	0.750	METI, 2015
6841	Aluminum and aluminum alloys, unwrought	t/t	0.950	JAA, 2000
68421	Bars, rods, angles, shapes and wire of aluminum	t/t	0.990	SLMI, undated
68422	Plates, sheets and strip of aluminum	t/t	0.990	SLMI, undated
68423	Aluminum foil	t/t	0.990	SLMI, undated
68424	Aluminum powder and flakes	t/t	0.990	SLMI, undated
68425	Tubes, pipes & banks, hollow bars of aluminum	t/t	0.950	JAA, 2000
68426	Tube and pipe fittings of aluminum	t/t	0.990	SLMI, undated
69213	Tanks, etc. for storage or manuf. use of aluminum	t/t	0.990	SLMI, undated
69222	Casks, drums, etc. used for transport of aluminum	t/t	0.990	SLMI, undated
69232	Compressed gas cylinders of aluminum	t/t	0.990	SLMI, undated
69313	Wire, cables, ropes etc. not insulated, aluminum	t/t	0.190	Assumption
69333	Gauze, netting, grill, fencing wire of aluminum	t/t	0.990	SLMI, undated
69343	Expanded metal of aluminum	t/t	0.990	SLMI, undated
695	Tools for use in the hand or in machines	t/MY	0.013	NIES, 2000
696	Cutlery	t/MY	0.074	NIES, 2000
69723	Domestic utensils of aluminum	t/t	0.990	SLMI, undated
6979	Other household equipment of base metals	t/MY	0.192	NIES, 2000
6981	Locksmith wares	t/MY	0.074	NIES, 2000

6988	Miscell. articles of base metal	t/MY	0.051	NIES, 2000
69894	Articles of aluminum, n.e.s.	t/t	0.990	SLMI, undated
71	Machinery, other than electric	t/MY	0.012	NIES, 2000
72	Electrical machinery, apparatus and appliances	t/MY	0.010	NIES, 2000
73	Transport equipment	t/MY	0.050	NIES, 2000
81	Sanitary, plumbing, heating and light fixtures	t/MY	0.012	NIES, 2000
82	Furniture	t/MY	0.009	NIES, 2000
86	Scientific & control instr., photogr. gds., clocks	t/MY	0.012	NIES, 2000
89	Miscellaneous manufactured articles, n.e.s.	t/MY	0.010	NIES, 2000
95	Military firearms and ammunition therefor	t/MY	0.055	NIES, 2000

S4. Parameters used

		B&C	Trans	EE	M&E	C&P	CD	Others	Reference
Fabrication efficiency (%)		90	80	75	75	85	80	80	GARC, 2011
Mean lifetime (year)*	Europe	50	13	20	15	1	8	10	Liu and Muller, 2013
	North America	75	20	20	30	1	12	10	
	Developed Asia & Oceania	40	10	20	20	1	10	10	
	China	40	15	20	20	1	12	10	
	Rest of the world	50	15	20	20	1	12	10	
Dissipation ratio (%)			0.01						Ciacci et al. (2015)
Landfill ratio (%)**		58							Liu et al. (2012)
Ratio of mixed metal loss and other repositories (%)		42							Liu et al. (2012)

* Weibull distribution function was used with the distribution width of 6, based on Tasaki et al. (2001).

** This was originally landfill ratio in 2009 and was used for 1962-2010.

Note: Building and Construction (B&C), Transportation (Trans.), Electrical Engineering (EE), Machinery and Equipment (M&E), Containers and Packaging (C&P), Consumer Durables (CD), and Other Uses (Others).

S5. Data sources of aluminum domestic end-use shipment and total consumption for 19 countries

No.	Country	Domestic end-use shipment	Total consumption
1.	Argentina	1996–2010 (6)	1960–1995 (1,3)
2.	Australia	1981–2009 (2), 1960–1980 (7,8)	
3.	Austria	1962–1997 (1)	1960–1961 (1), 1998–2009 (1,3)
4.	Belgium	1962–1997 (1)	1960–1961 (1), 1998–2009 (1,3)
5.	Brazil	1960–2009 (2)	
6.	China	1960–2009 (2)	
7.	France	1962–1997 (1)	1960–1961 (1), 1998–2009 (1,3)
8.	Germany	1960–2006 (1)	2007–2009 (3)
9.	India	1960–2009 (2)	
10.	Italy	1962–1994 (1)	1960–1961 (1) 1995–2009 (4)
11.	Japan	1960–2009 (2)	
12.	Netherlands	1962–1970 & 1982–1997 (1)	1960–1961 (1), 1971–1981 & 1998–2009 (1,3)
13.	Norway	1978–1998 (1)	1960–1977 (1), 1999–2009 (1,3)
14.	Russia	1960–2009 (2)	
15.	South Africa	1960–2009 (2)	
16.	Spain	1969–1997 (1)	1960–1968 (1), 1998–2009 (1,3)
17.	Switzerland	1962–1997 (1)	1960–1961 (1), 1998–2009 (1,3)
18.	U.K.	1962–1997 (1)	1960–1961 (1), 1998–2009 (5)
19.	U.S.	1960–2009 (2,9)	

Notes and sources: (1): (Metallgesellschaft, 1889–2007) (Data for 2008–2010 were assumed the same as 2007 if not available); (2) (GARC, 2011); (3) (WBMS various years); (4) (ASSOMET, 2003–2010); (5) (Alfred, 2011); (6) (CAIAMA, 2002–2011); (7) (Commonwealth of Australia, 1960); (8) (Govett and Larsen, 1981); (9) The reported shipment in GARC after 2001 was deducted by 15% as the share of Canadian shipment (GARC, 2011).

S6. Historical trend of aluminum stocks by finished products in 19 countries

(a) Argentina
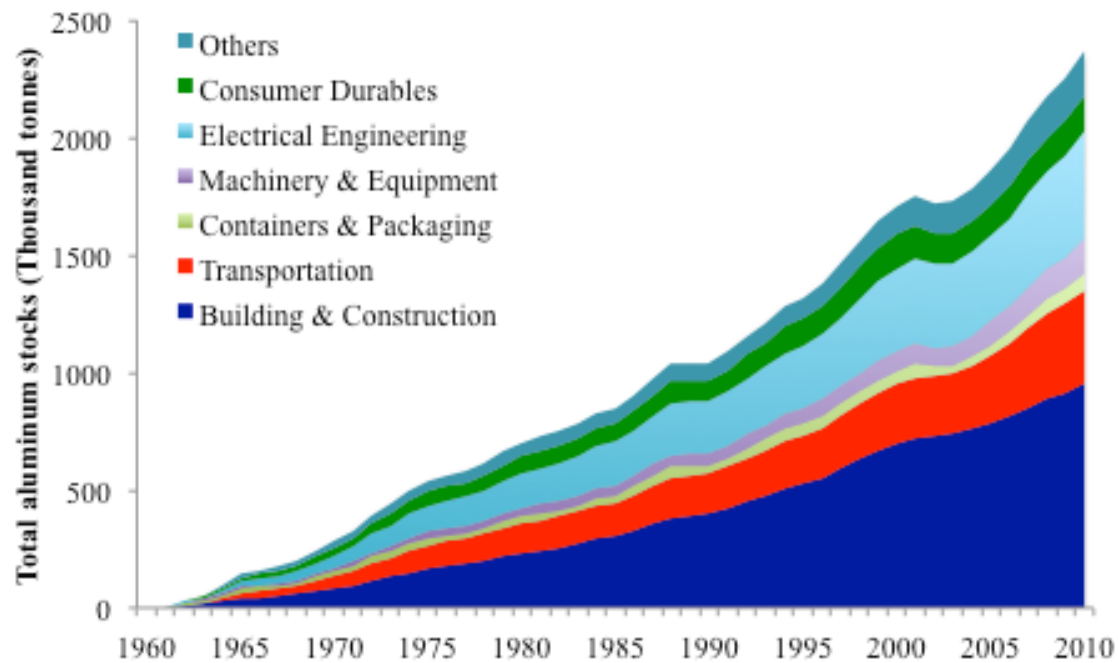
(b) Australia
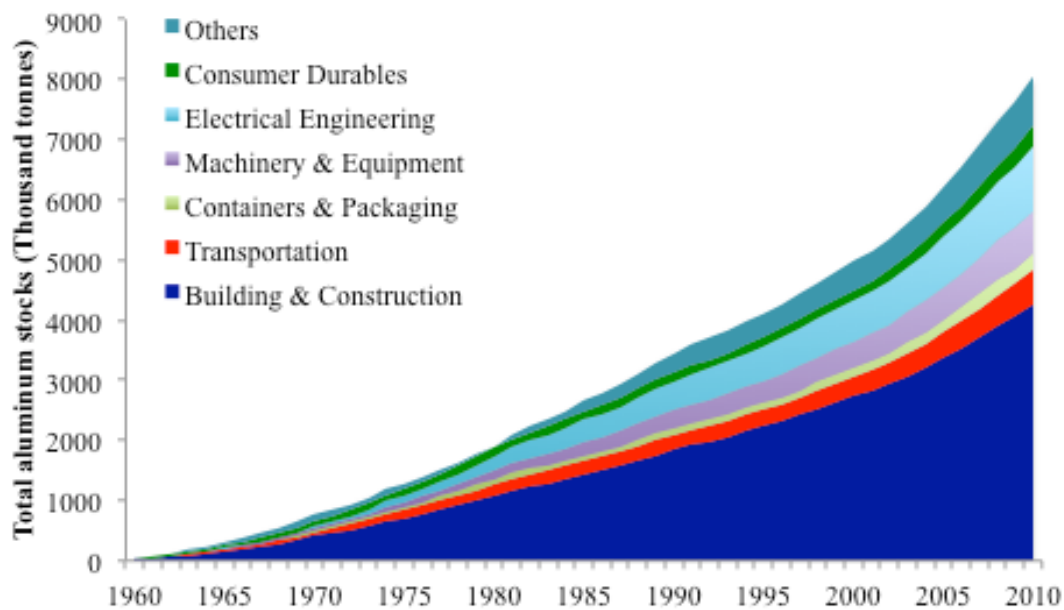
(c) Austria
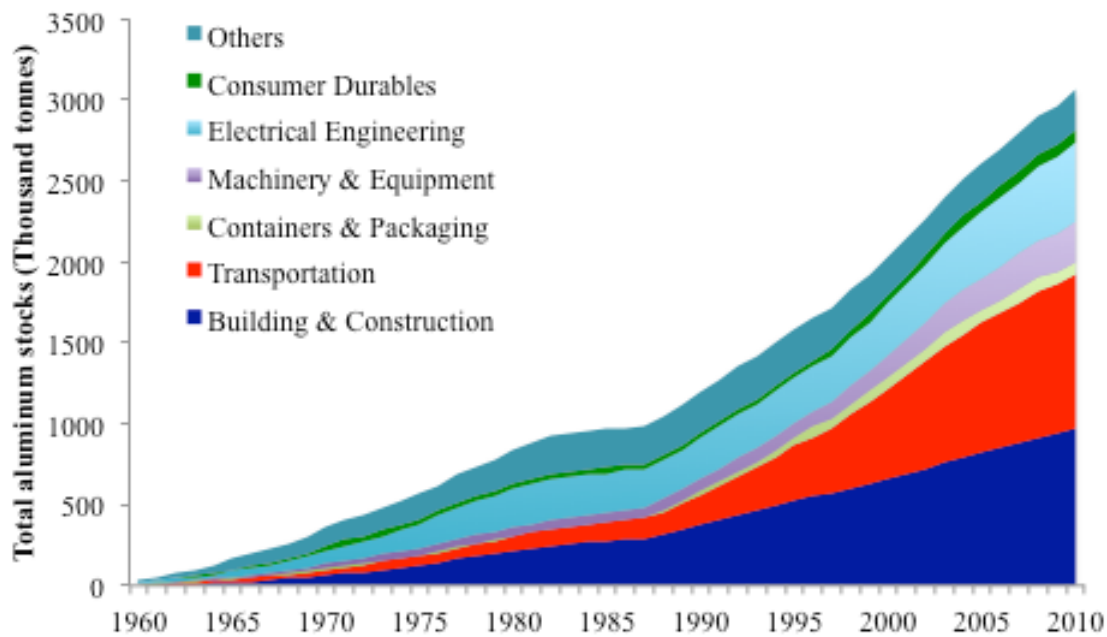
(d) Belgium
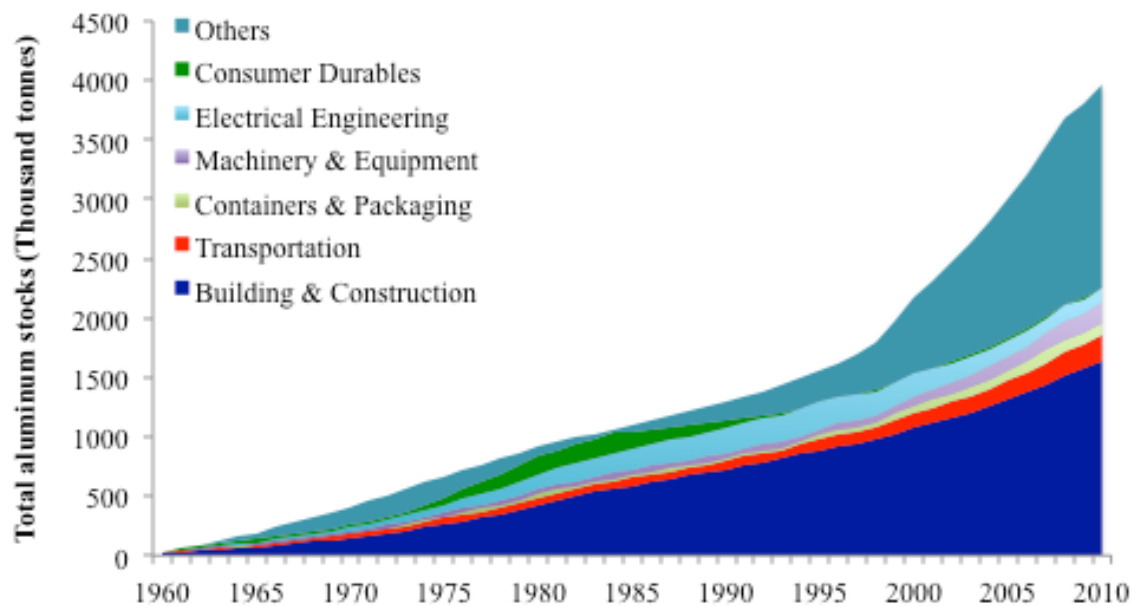
(e) Brazil
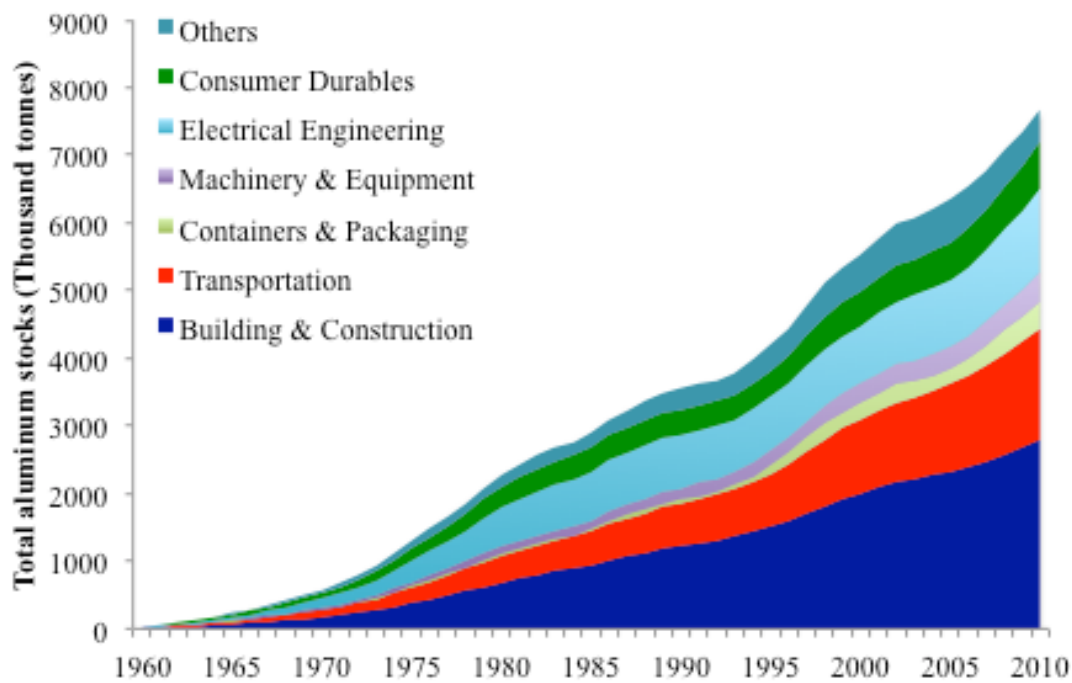
(f) China
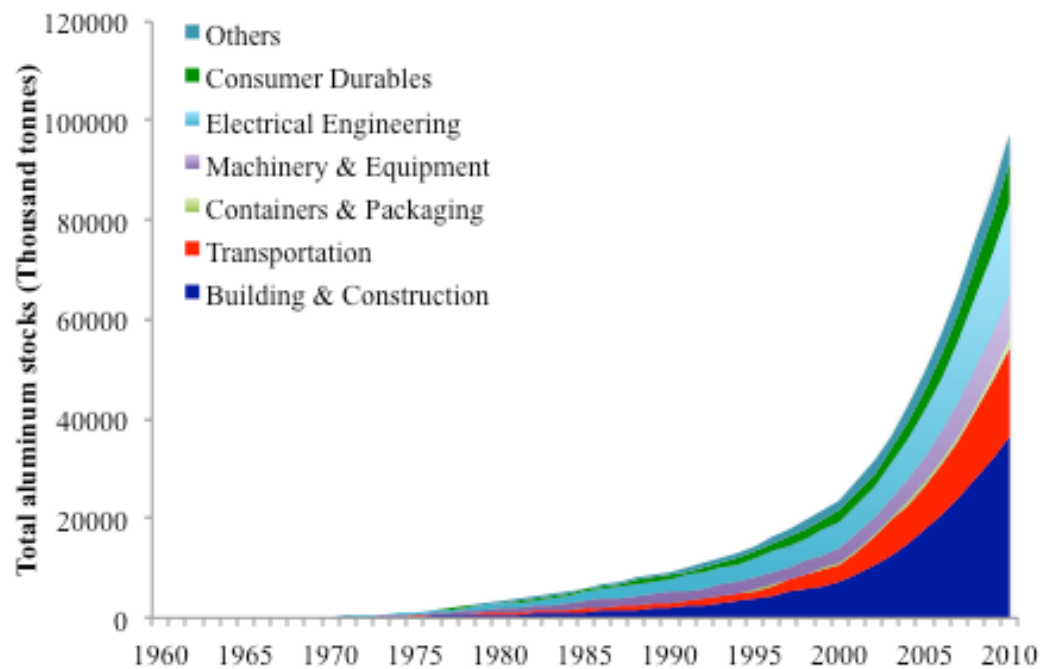
(g) France
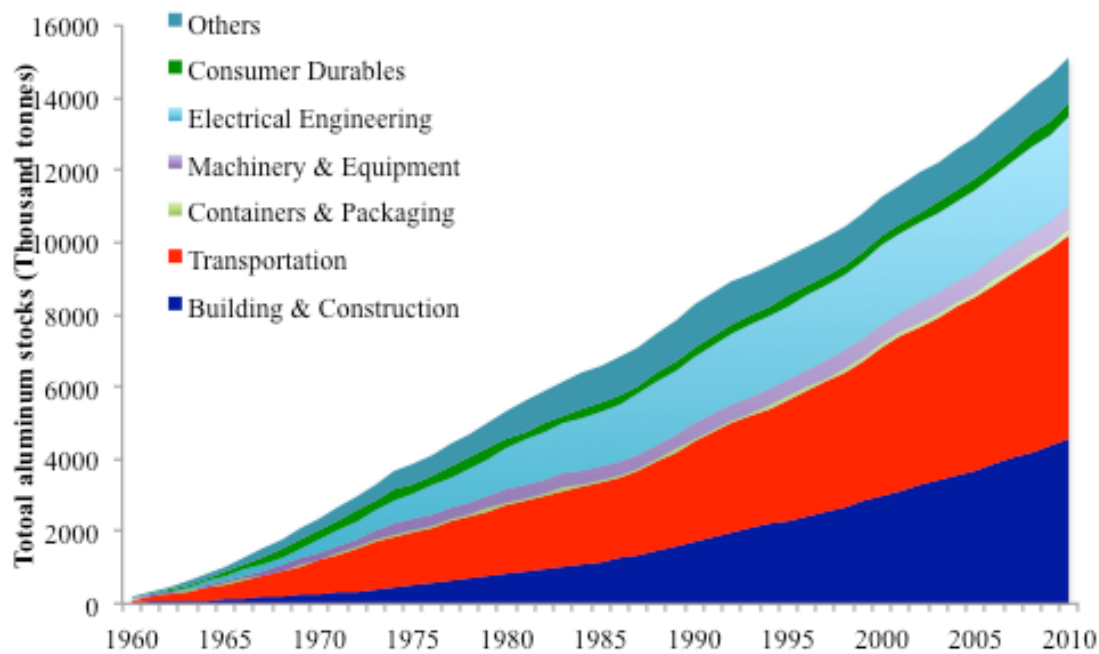
(h) Germany
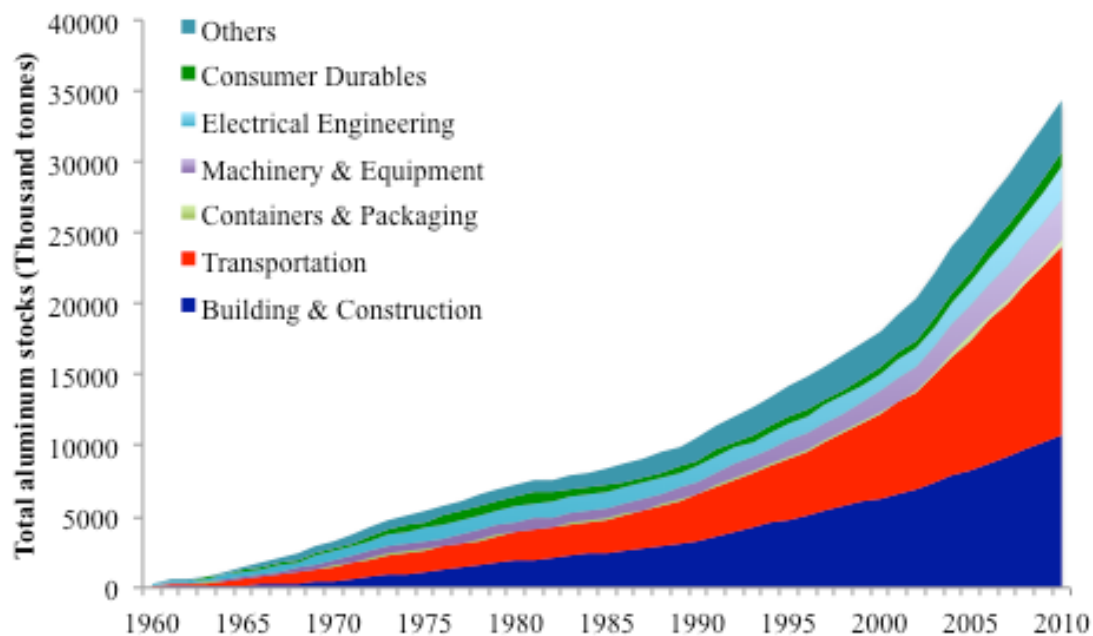
(i) India
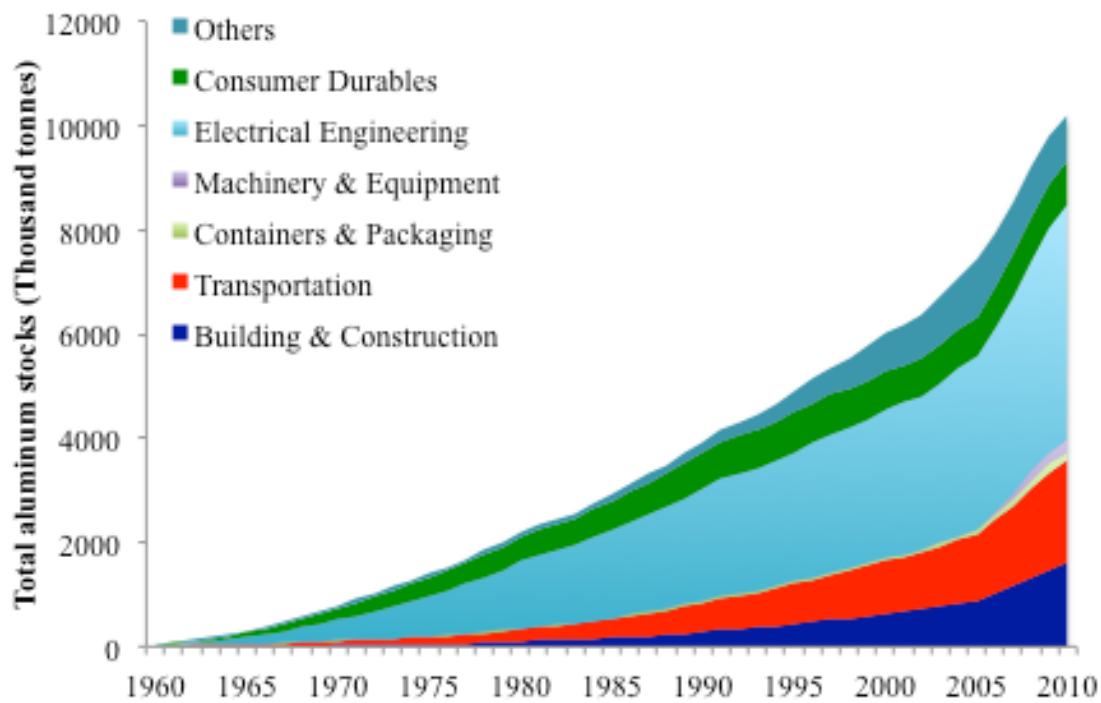
(j) Italy
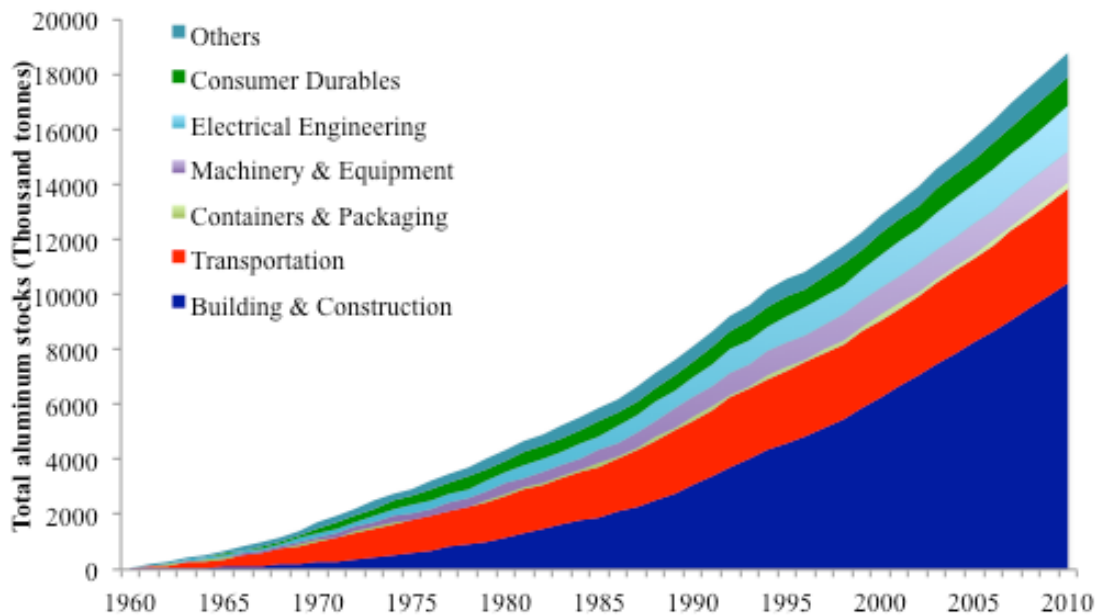
(k) Japan
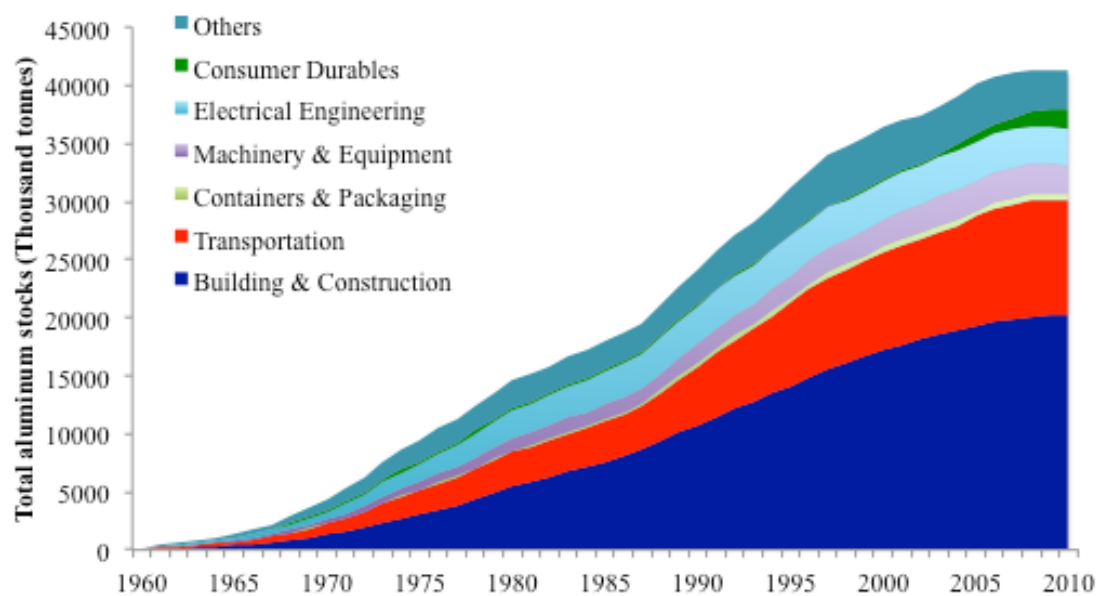
(l) Netherlands
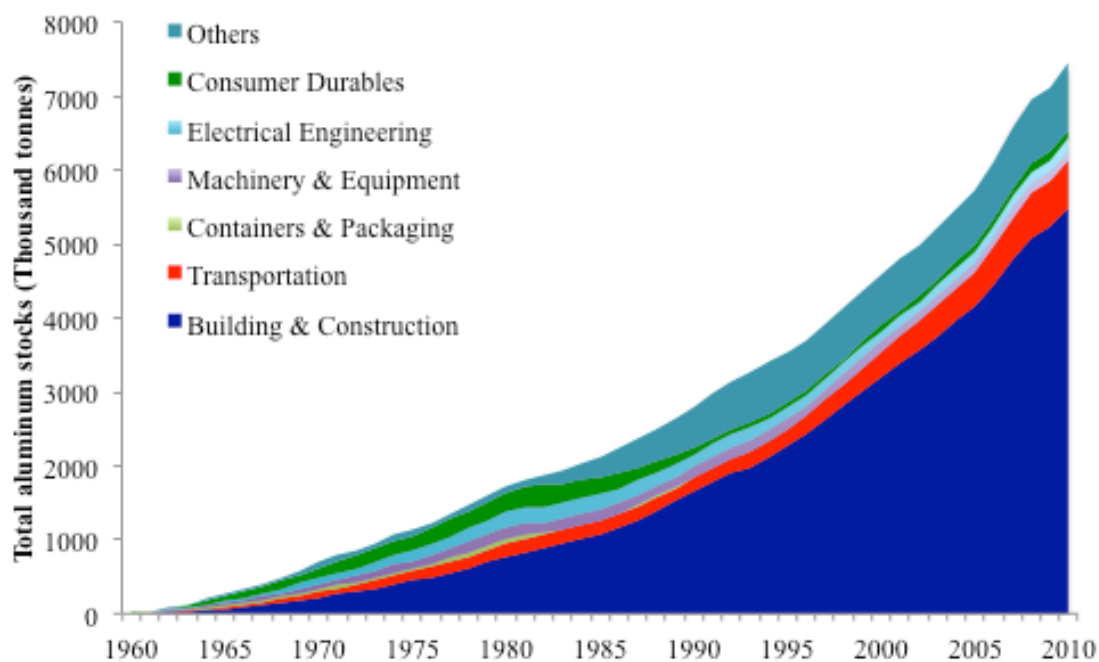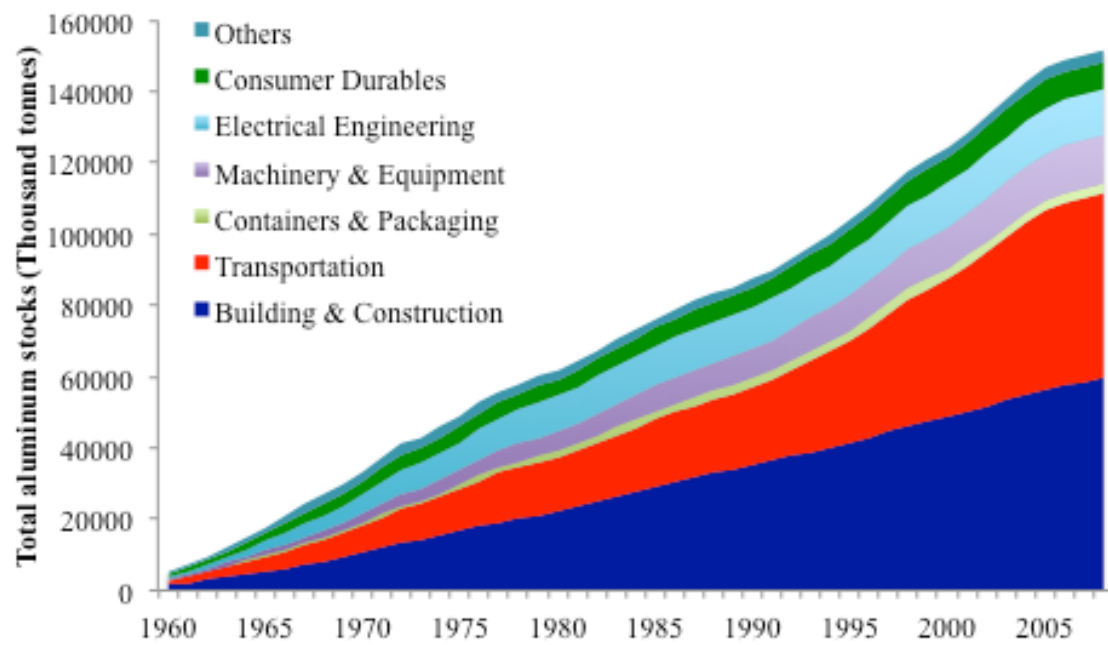
(m) Norway
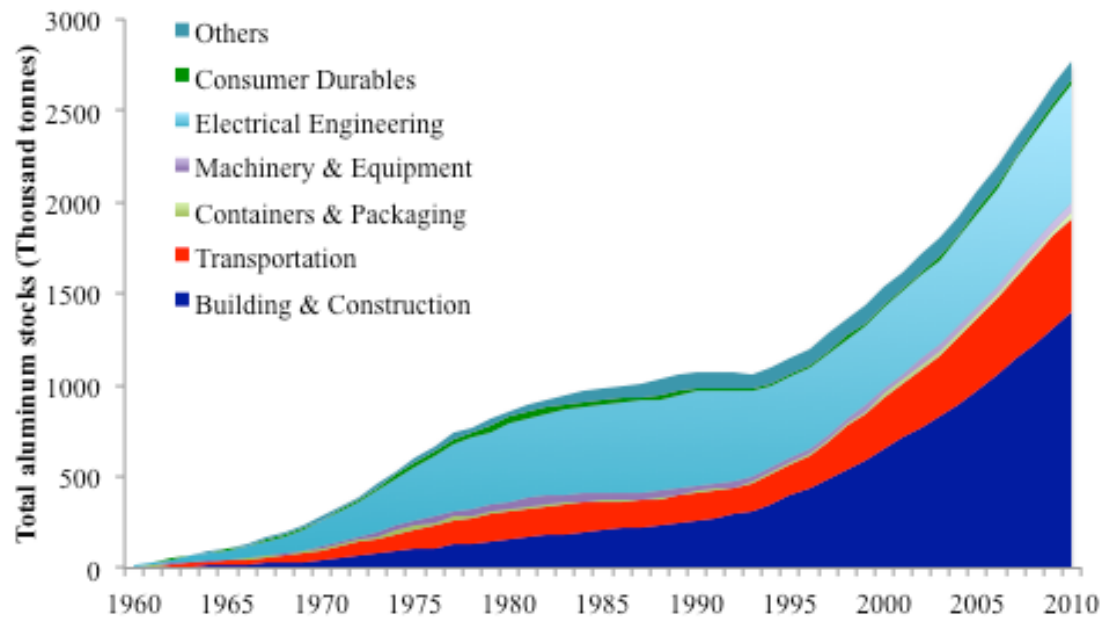
(n) Russia
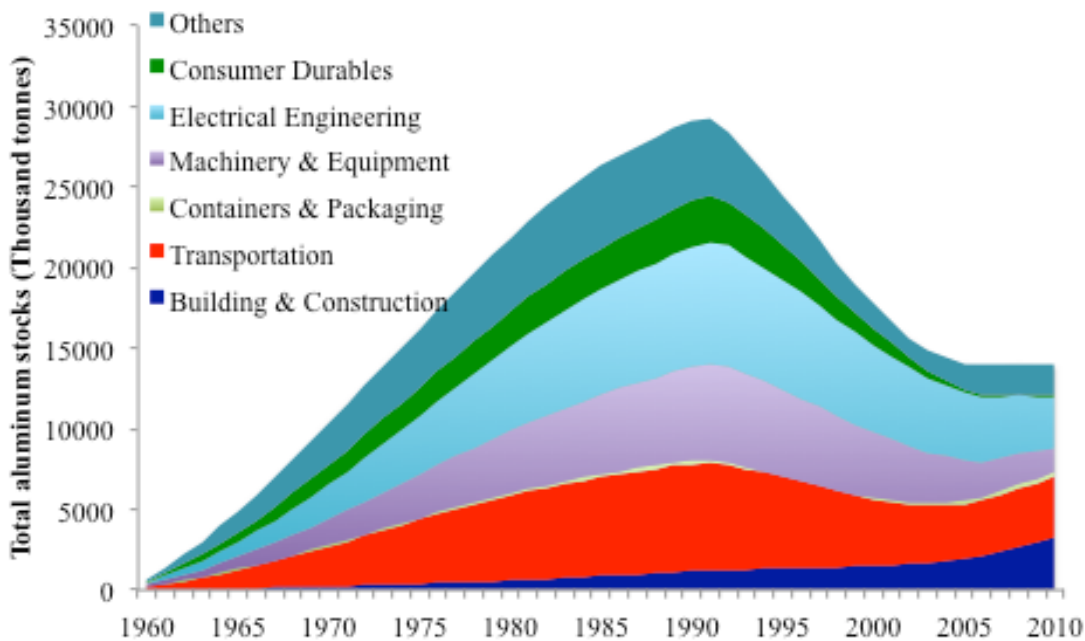
(o) South Africa
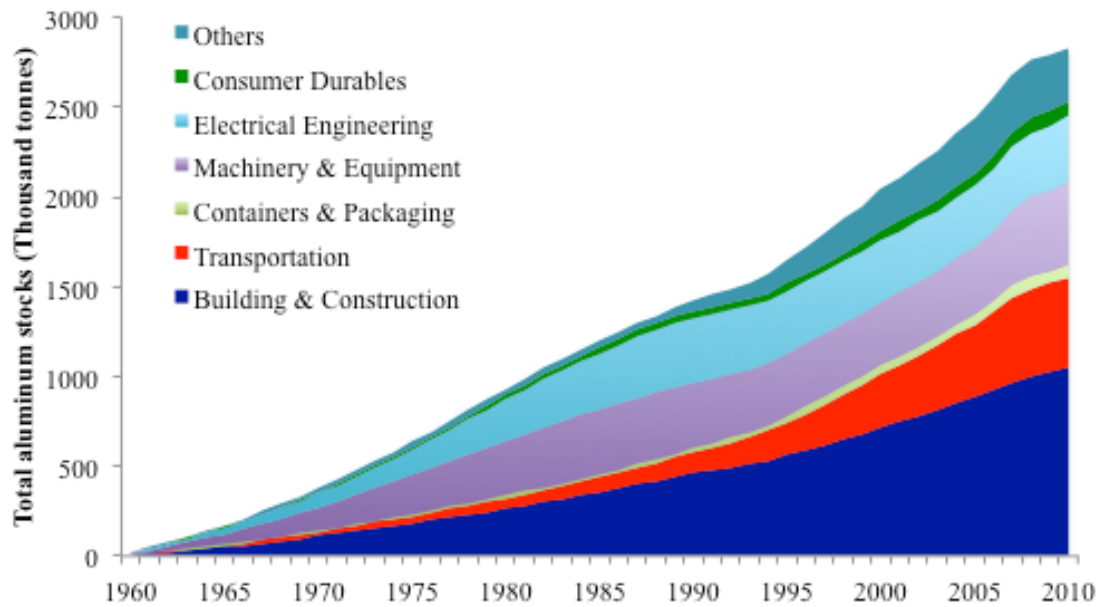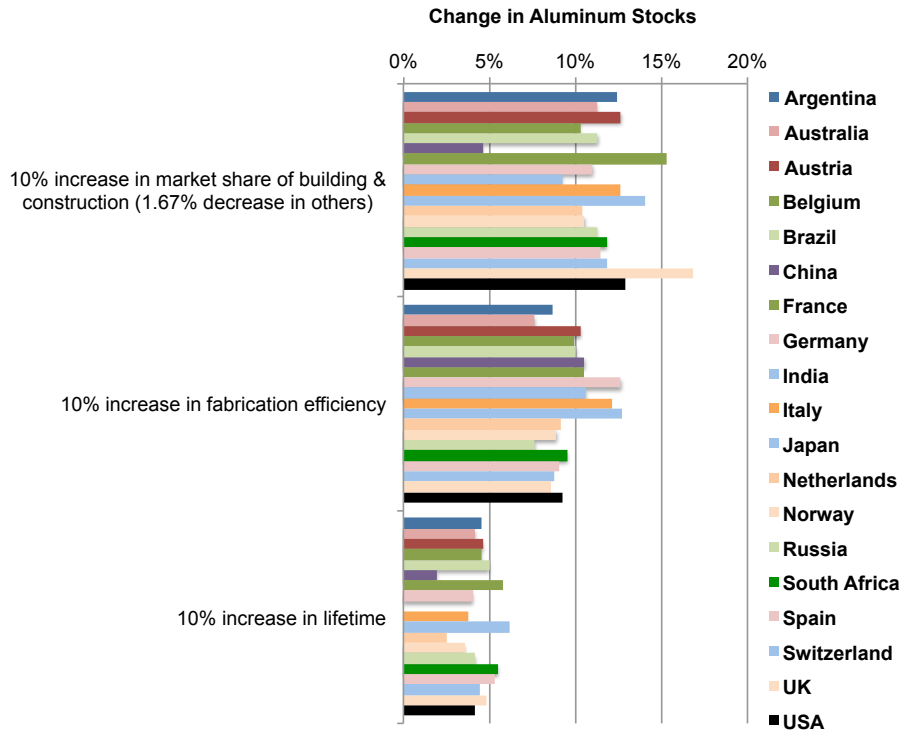

(p) Spain
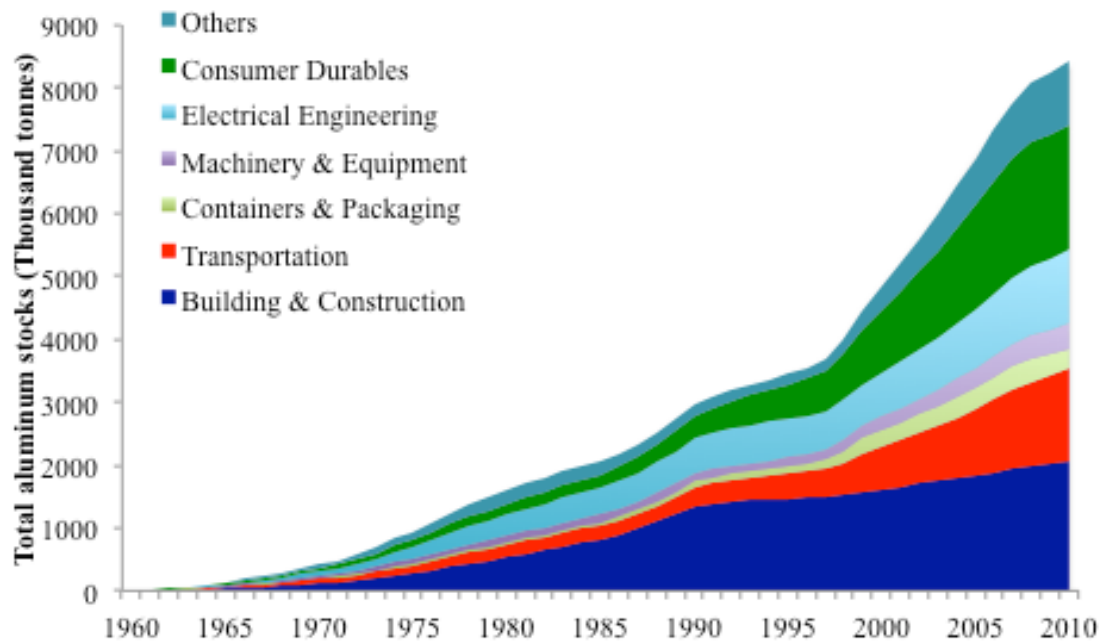
(q) Switzerland
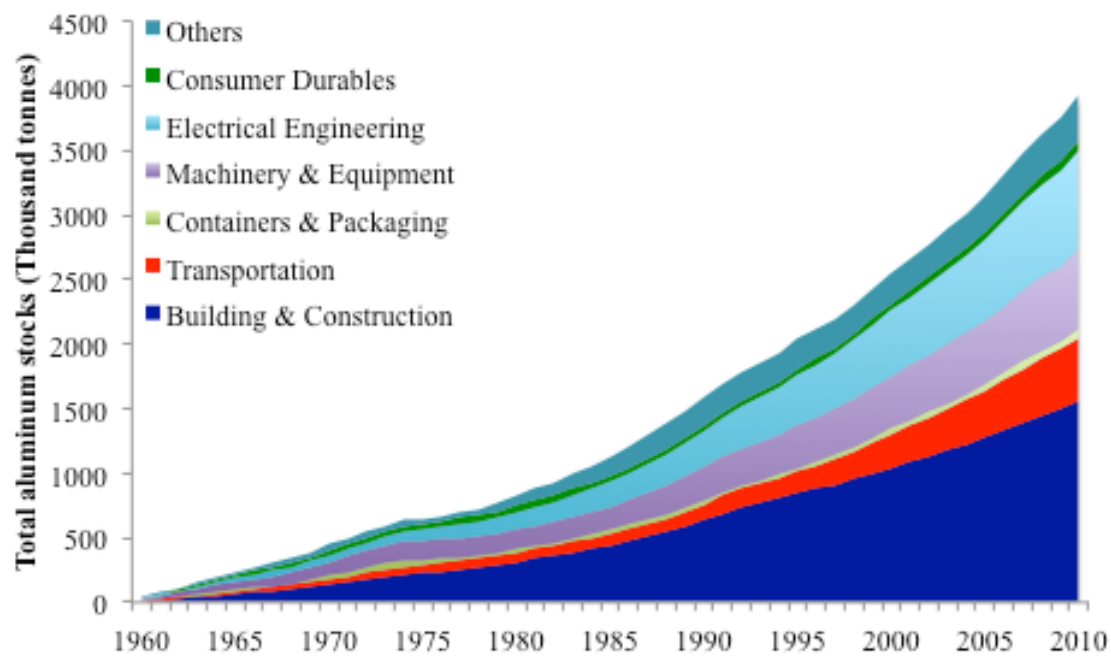
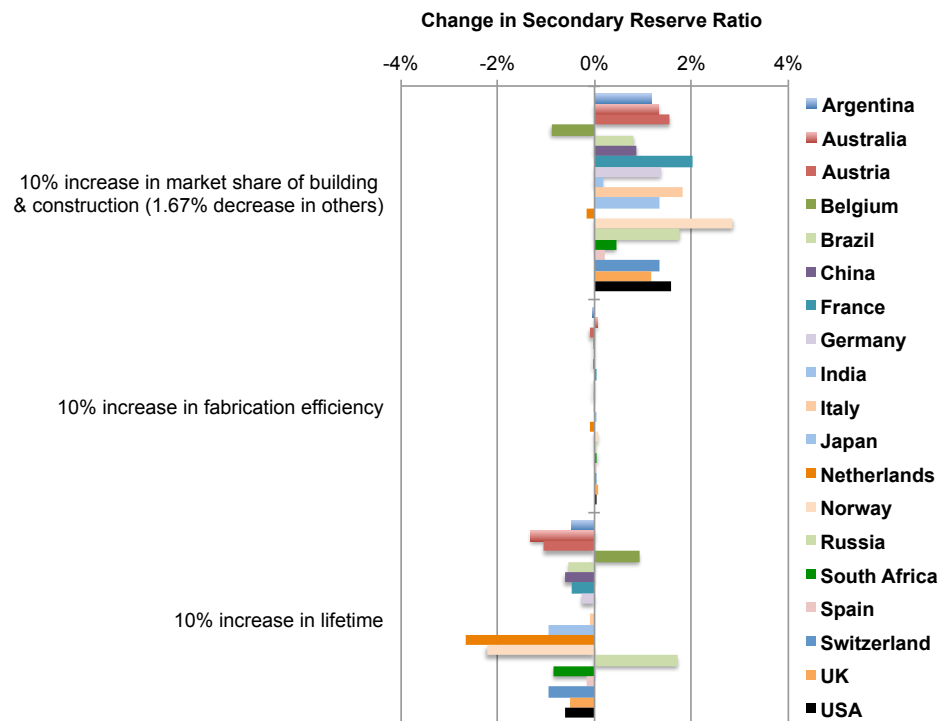## (r) The United Kingdom
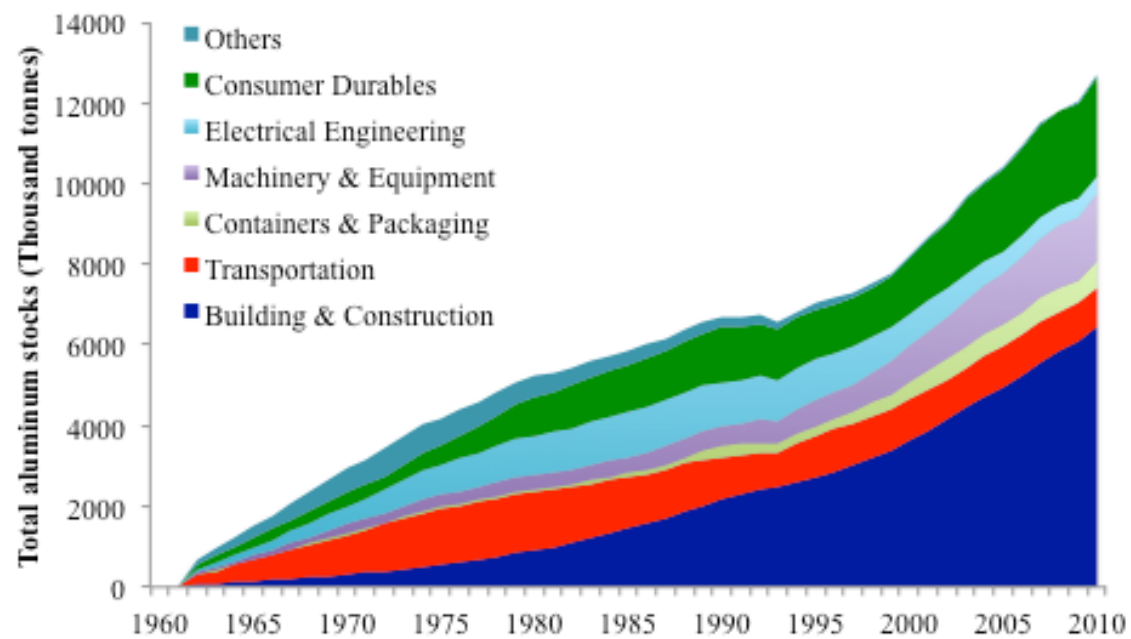
(s) The United States

S7. Sensitivity analysis results

(a) Aluminum stocks (AS)

Change in Aluminum Stocks

(b) Secondary reserve ratio (SRR)

Change in Secondary Reserve Ratio

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APPENDICE B: SUPPLEMENTARY MATERIAL: SECTION 3.2

Table S1. Top five copper using countries (population at least 40 million)

	1	2	3	4	5
Refined copper usage in 2010 [1]	China 7,393 kt	United States 1,770 kt	Germany 1,312 kt	Japan 1,060 kt	South Korea 828 kt
Refined copper usage per capita in 2010 [1]	South Korea 19 kg/capita	Germany 16 kg/capita	Italy 10 kg/capita	Japan 8.3 kg/capita	Spain 7.6 kg/capita
Cumulative refined copper usage during 1962–2010 [1,2]	United States 108,883 kt	China 91,321 kt	Japan 57,431 kt	Germany 49,584 kt	Italy 22,712 kt
Semis production in 2010 [1]	China 10,093 kt	United States 2,211 kt	Germany 1,656 kt	Japan 1,458 kt	South Korea 1,327 kt
Semis production per capita in 2010 [1,2]	South Korea 31 kg/capita	Germany 20 kg/capita	Italy 19 kg/capita	Japan 11 kg/capita	China 8 kg/capita

Table S2. Classification of end-use sectors of finished products [3]

End-use sector	Example	
Building and Construction	Plumbing	Water distribution, heating, gas and sprinkler
	Building plant	Aircon tube
	Architecture	Roofs, gutters, flashing, decor., builders h/w
	Communications	Comms wiring in buildings
	Electrical power	Power distrib., earth, grounds, light, wire device
Infrastructure	Power utility	Power transmission and distribution network
	Telecommunications	Telecom network
Industrial Equipment	Electrical equipment	Industrial transformers and motors
	Non-electrical equipment	Valves, fittings, instruments, and in plant equipment
Transport	Automotive electrical equipment	Harnesses, motors
	Automotive non-electrical equipment	Radiators and tubing
	Other transport equipment	Railroad, shipping, and marine
Other products	Consumer and general products	Appliances, instruments, tools, and other products
	Cooling equipment	Aircon and refrigeration
	Electronic equipment	Industrial/commercial electronics and PCs
	Diverse	Ammunition, clothing, coins, and other

Table S3. List of variables used in the paper

$CC(t,c,i)$	copper consumption for finished product i in country c in year t
$CS(t,c,i)$	copper stocks of finished products i in use in country c in year t
$DcR(y,c,i)$	discard ratio of finished product i in country c after y years
$DpR(t,c,i)$	dissipation ratio of finished product i in country c in year t
$EFP(t,c,i)$	exported finished products i in country c in year t
$ES(t,c)$	exported scrap
$FE(t,c,i)$	fabrication efficiency of semi-finished product i for the production of finished products in country c in year t
$GEoLS(t,c,i)$	generated EoL scrap of finished product i in use in country c in year t
$HQS(t,c)$	utilized high-quality scraps in country c in year t
$IFP(t,c,i)$	imported finished products i in country c in year t
$IS(t,c)$	imported scrap
$LFR(t,c)$	landfill ratio in country c in year t
$LQS(t,c)$	utilized low-quality scraps in country c in year t
$MMLR(t,c)$	mixed metal loss ratio in country c in year t
$MS(t,c,i)$	market share of semi-finished products for finished product i in country c in year t
$MSCR(t,c)$	marginal secondary copper reserves emerging in a year in country c in year t
$MSCRe(t,c)$	marginal secondary copper reserves not emerging in a year in country c in year t
$MSCRn(t,c)$	marginal secondary copper reserves in country c in year t
$OC(t,c)$	copper ore and concentrates consumption in country c in year t
$RC(t,c)$	refined copper consumption in country c in year t
$REoLS(t,c)$	recovered EoL scrap in country c in year t
$RP(t,c)$	refined copper production in country c in year t
$RPS(t,c)$	recovered process scrap in country c in year t
$SC(t,c,i)$	semi-finished product i consumption in country c in year t
$SCR(t,c)$	secondary copper reserves in country c in year t
$SCRe(t,c)$	secondary copper reserves emerging in a year in country c in year t
$SCRn(t,c)$	secondary copper reserves not emerging in a year in country c in year t
$SP(t,c)$	semi-finished product production in country c in year t
$SR(y,c,i)$	survival ratio of finished product i in country c after y years
$SRR(t,c)$	secondary reserve ratio in country c in year t
$SSRUMe(t,c)$	sub-economic secondary resources and unrecoverable materials (others) emerging in a year in the column of products in use in country c in year t
$SSRUMn(t,c)$	sub-economic secondary resources and unrecoverable materials (others) not emerging in a year in the column of products in use in country c in year t
$SSRUMw(t,c)$	sub-economic secondary resources and unrecoverable materials (others) in managed landfill sites
$SSRUMd(t,c)$	sub-economic secondary resources and unrecoverable materials (others) in the dissipated materials

SSRUMm(*t*,*c*)	amount of unrecoverable materials (mixed metal losses)
TSC(*t*,*c*)	total semi-products consumption in country *c* in year *t*
US(*t*,*c*)	utilized scrap in country *c* in year *t*

Note S1. Estimation method of copper stocks of finished products in use and generated end-of-life scrap

The amount of copper stocks of finished products in use was estimated using the following equation.

$$CS(t,c,i) = \Sigma_{t'} CC(t',c,i) \times SR(t-t',c,i) \tag{S1}$$

Therein, $CS(t,c,i)S_{(t,c,i)}$ signifies the copper stock of finished product i in use in country c in year t; $CC(t',c,i)$ denotes the copper consumption for finished product i in country c in year t'; and $SR(y,c,i)$ represents the survival ratio of finished product i in country c after y years.

$CC(t',c,i)$ was estimated using the following equation.

$$CC(t',c,i) = \Sigma_{t'} SC(t',c,i) \times FE(t',c,i)/100 - EFP(t',c,i) + IFP(t',c,i) \tag{S2}$$

Therein, $SC(t',c,i)$ stands for the amount of semi-finished product i consumed in country c in year t'; $FE(t',c,i)$ signifies the fabrication efficiency of semi-finished product i for the production of finished products in country c in year t' (%); and $EFP(t',c,i)$ and $IFP(t',c,i)$ respectively denote the amounts of copper contained in exported and imported finished product i in country c in year t'.

$SC(t',c,i)$ was estimated using the following equation.

$$SC(t',c,i) = TSC(t',c) \times MS(t',c,i)/100 \ . \tag{S3}$$

Therein, $TSC(t',c)$ is the total amount of semi-product consumed in country c in year t' and $MS(t',c,i)$ denotes the market share of semi-finished products for finished product i in country c in year t' (%).

$SR(y,c,i)$ is calculable using the following equation.

$$SR(y,c,i) = EXP[-\{y/Yci\}^{dci} . \{ \Gamma(1+1/dci)^{dci}\}] \tag{S4}$$

Therein, y is the age of the finished products; Yci is the average lifetime of finished product i in country c; Γ is a gamma function; and dci is a parameter of the distribution width of finished product i in country c.

The generated end-of-life scrap was calculated using the following equation.

$$GEoLS(t,c,i) = \Sigma_{t'} CC(t',c,i) \times DcR(t-t',c,i) \tag{S5}$$

$$DcR(y,c,i) = SR(y,c,i) - SR(y+1,c,i) \tag{S6}$$

Therein, $GEoLS(t,c,i)$ stands for generated end-of-life scrap of finished product i in use in country c in year t and $DcR(y,c,i)$ is the discard ratio of finished product i in country c after y years.

**Table S4. Copper contents of traded commodities**

SITC Rev.1	Items	Unit	Copper content
28311	Ores and concentrates of copper	t/t	0.272[4]
28312	Copper matte	t/t	0.625[5]
28402	Copper waste and scrap	t/t	0.750[6]
68211	Blister copper and other unrefined copper	t/t	0.9850[5]
68212	Refined copper including remelted	t/t	0.9999[5]
68213	Master alloys of copper	t/t	0.9999[5]
68221	Bars, rods, angles, shapes, wire of copper	t/t	0.9999[5]
68222	Plates, sheets, and strips of copper	t/t	0.9999[5]
68223	Copper foil	t/t	0.9999[5]
68224	Copper powders and flakes	t/t	0.9999[5]
68225	Tubes, pipes and blanks, hollow bars of copper	t/t	0.9999[5]
68226	Tube and pipe fittings of copper	t/t	0.650[7]
69212	Refined copper including remelted	t/t	0.9999[5]
69312	Wire, cables, ropes, etc. not insulated of copper	t/t	0.9999[5]
69332	Gauze, netting, grill, fencing wire of copper	t/t	0.9999[5]
69342	Expanded metal of copper	t/t	0.9999[5]
69412	Nails, tacks, staples, spikes, etc. of copper	t/t	0.65[7]
69422	Nuts, bolts, screws, rivets, washers of copper	t/t	0.65[7]
695	Tools of use in the hand or in machines	t/MY	0.006[8]
696	Cutlery	t/MY	0.003[8]
69712	Domestic stoves, etc. of copper	t/t	0[9]
69722	Domestic utensils of copper	t/t	0.9999[5]
6979	Other household equipment of base metals	t/MY	0.002[8]
6981	Locksmith wares	t/MY	0.003[8]
69862	Springs and leaves for springs of copper	t/t	0.9999[5]
6988	Miscell. articles of base metal	t/MY	0.004[8]
69892	Articles of copper, n.e.s.	t/t	0.9999[5]
71	Machinery, other than electric	t/MY	0.006[8]
72	Electrical machinery, apparatus, and appliances	t/MY	0.008[8]
73	Transport equipment	t/MY	0.009[8]
81	Sanitary, plumbing, heating, and light fixtures	t/MY	0.012[8]
82	Furniture	t/MY	0.001[8]
86	Scientif & control instrum, photogr gds, clocks	t/MY	0.002[8]
89	Miscellaneous manufactured articles, n.e.s.	t/MY	0.004[8]
95	Firearms of war and ammunition therefor	t/MY	0.020[8]

Table S5. Market Share (%) and fabrication efficiency (%) of semi-finished products and mean lifetime (years) of finished products

|		Building & Construction	Infrastructure	Industrial equipment	Transport	Other products
Market share (%)[3]	China	24	17	11	12	36
	Germany	38	10	14	15	23
	Italy	38	10	14	15	23
	Japan	30	12	12	14	33
	South Korea	25	15	15	14	31
	Spain	38	10	14	15	23
	United States	39	12	9	14	26
Fabrication efficiency (%)[10]		90	88	85	82	76
Mean lifetime (years)*	Japan[11]	38.7	17.5	12.0	11.0	11.4
	United States[12]	32.5	50	20	20	13.5
	Others[10]	40	30	17.5	17.3	8.25

* In the Weibull distribution function, the distribution width was set as 2 based on Tasaki et al.[13].

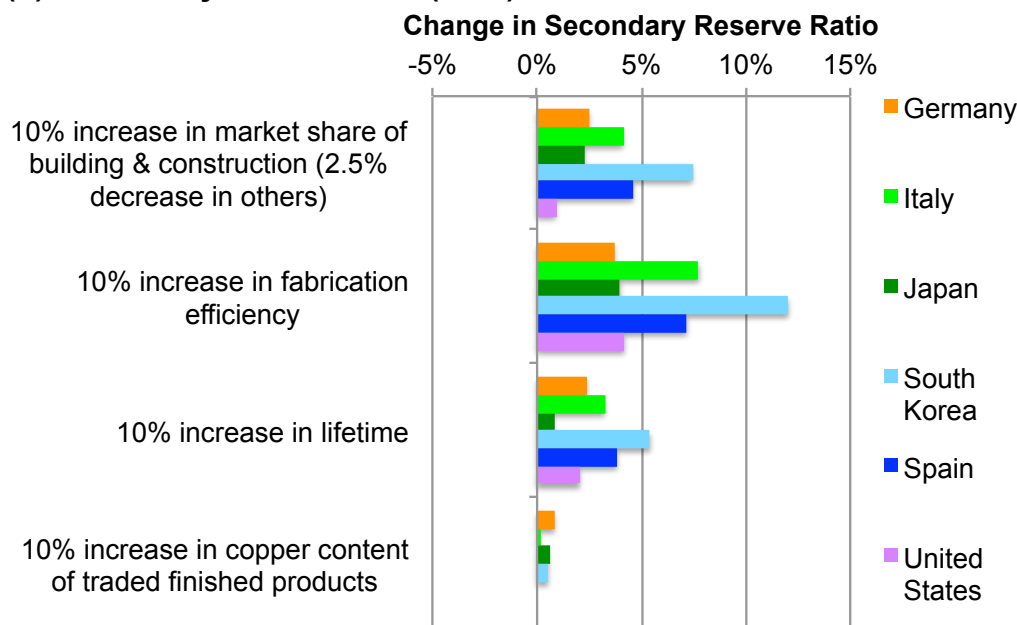**Figure S1. Trends in per-capita consumption of finished products**
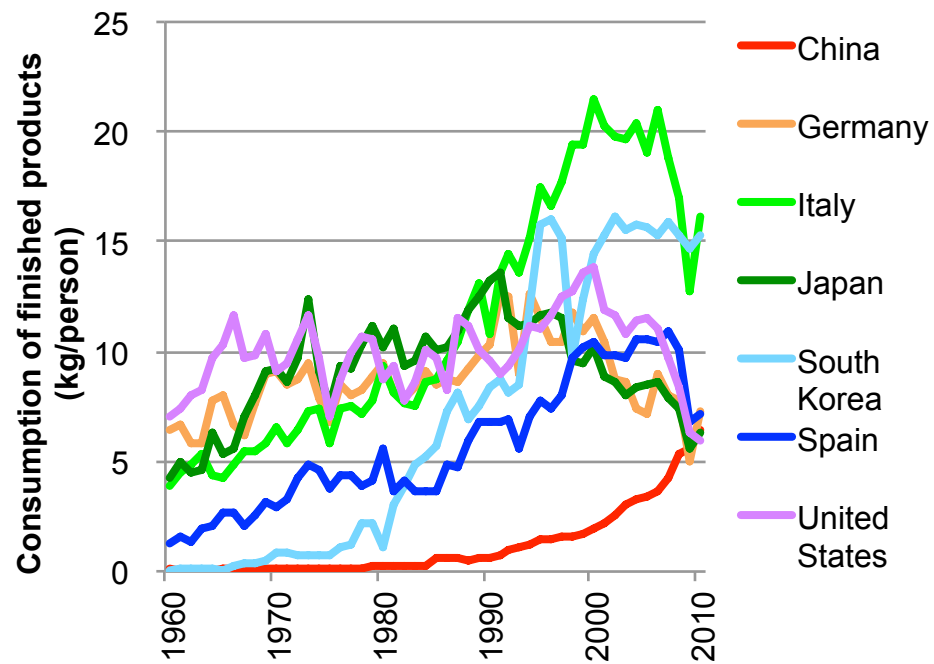
Figure S2. Sensitivity analysis results
(a) Copper stocks (CS)
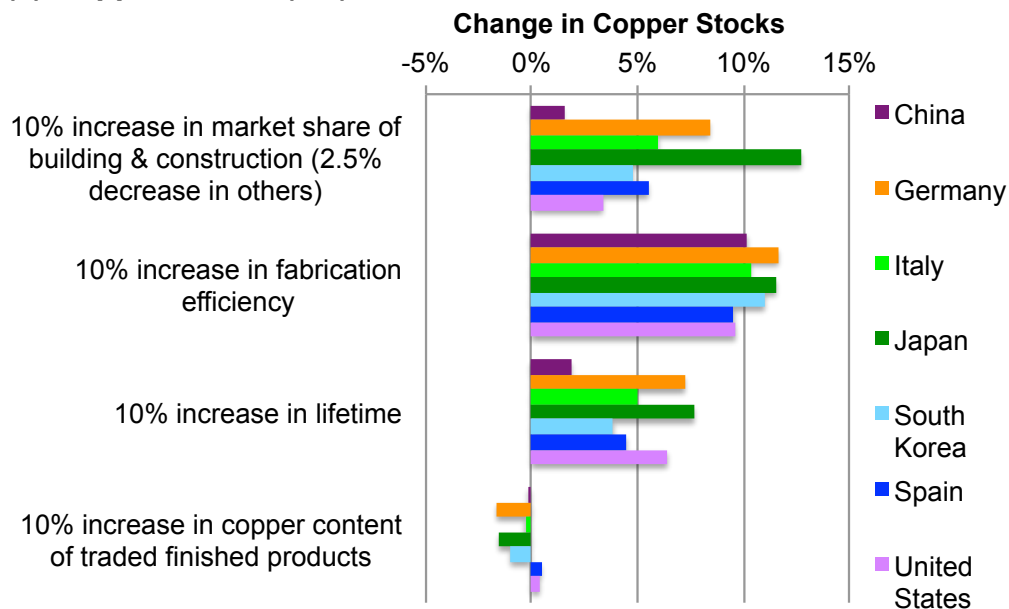
(b) Secondary reserve ratio (SRR)

Note: A 10% increase in fabrication efficiency would bring more then 95% fabrication efficiency for some finished product categories. Ruhrberg[14] approved that 95%+ of fabrication efficiency can be achieved in some finished products categories based on expertise gathered from the ICSG industry advisors and copper associations.

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Figure S1. Principal end-use of zinc for the six countries
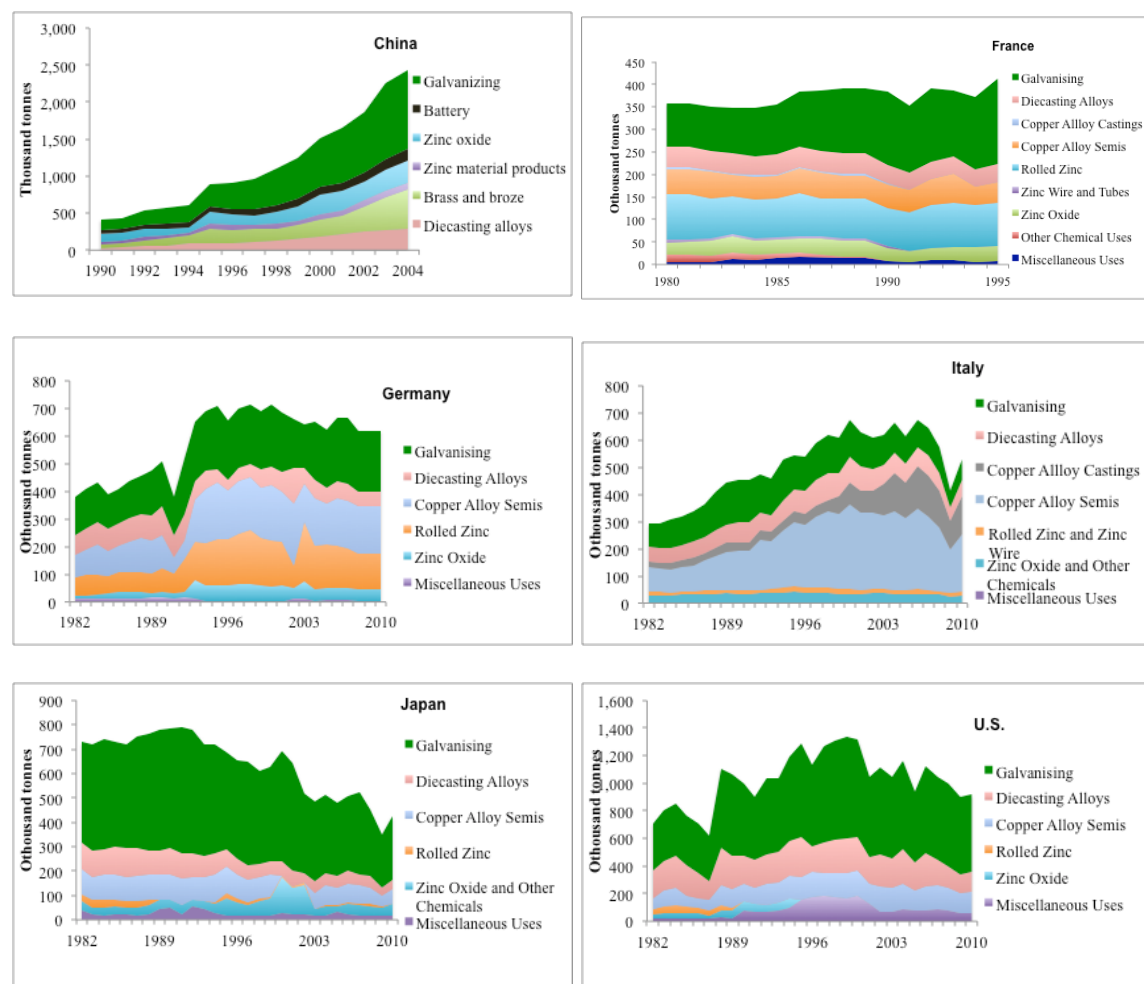
Note: Based on data availability of each country, scale of x-axis is different. Data for 1998–2010 were assumed the same as 1997 if not available.

Source: China (CNMIA 1990-2004); France (WBMS 1982-1997); Germany (WBMS 1982-2008); Italy (WBMS 1982-2010); Japan (WBMS 1982-2010) and U.S (WBMS 1975-2010).

Figure S2. Top zinc consumption countries in 2010 (Meylan and Reck 2016).
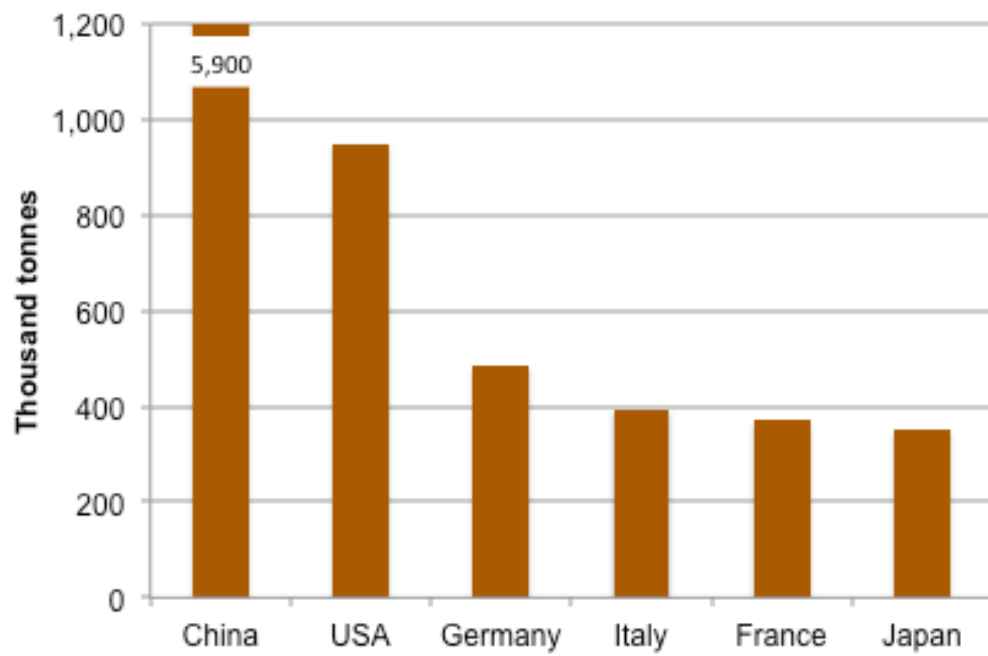
Table S1. List of variables used in the paper

$ZC(t',c,i)$	zinc consumption for finished product i in country c in year t'
$ZS(t,c,i)$	zinc stocks of finished products i used in country c in year t
$DR(y,c,i)$	discard ratio of finished product i in country c after y years
$DpR(t',c,i)$	dissipation ratio of finished product i in country c in year t'
$IFP(t,c,i)$	imported finished products i in country c in year t
$PEU(t',c,i)$	principal end uses of semi products i in country c in year t'
$FE(t,c,i)$	fabrication efficiency of semi-finished product i for the production of finished products in country c in year t
$GEoLS(t,c,i)$	generated EoL scrap of finished product i in use in country c in year t
$HQS(t,c)$	high-quality scrap used in country c in year t
$IMS(t,c)$	imported scrap amount
$EXS(t,c)$	exported scrap amount
$LFR(t',c)$	landfill ratio in country c in year t'
$LQS(t,c)$	low-quality scrap used in country c in year t
$MMLR(t',c)$	mixed metal loss ratio in country c in year t'
$AR(t,c,i)$	the allocation ratio from semi products to finished product i in country c in year t'
$MSZR(t,c)$	marginal secondary zinc reserves emerging in a year in country c in year t
$MSZRe(t,c)$	marginal secondary zinc reserves not emerging in a year in country c in year t
$MSZRn(t,c)$	marginal secondary zinc reserves in country c in year t
$ZCC(t,c)$	zinc concentrates consumption in country c in year t
$RZC(t,c)$	refined zinc consumption in country c in year t
$REoLS(t,c)$	recovered EoL scrap in country c in year t
$ZCP(t,c)$	zinc concentrates production in country c in year t
$RPS(t,c)$	recovered process scrap in country c in year t
$SC(t,c,i)$	semi-finished product i consumption in country c in year t
$SZR(t,c)$	secondary zinc reserves in country c in year t
$SZRe(t,c)$	secondary zinc reserves emerging in a year in country c in year t
$SZRn(t,c)$	secondary zinc reserves not emerging in a year in country c in year t
$SP(t,c)$	semi-finished product production in country c in year t
$SR(y,c,i)$	survival ratio of finished product i in country c after y years
$SRR(t,c)$	secondary reserve ratio in country c in year t
$SSZRUMe(t,c)$	sub-economic secondary zinc resources and unrecoverable materials (others) emerging in a year in the column of products in use in country c in year t
$SSZRUMn(t,c)$	sub-economic secondary zinc resources and unrecoverable materials (others) not emerging in a year in the column of products in use in

	country *c* in year *t*
SSZRUMw(*t,c*)	sub-economic secondary zinc resources and unrecoverable materials (others) in managed landfill sites
SSZRUMd(*t,c*)	sub-economic secondary zinc resources and unrecoverable materials (others) in the dissipated materials
SSZRUMm(*t,c*)	amount of unrecoverable materials (mixed metal losses)
US(*t,c*)	scrap used in country *c* in year *t*

Note S1. Estimation of zinc stocks of products in use and generated end-of-life scrap

Zinc stocks of final products in use were estimated using the following equation (S1).

$$ZS(t,c,i) = \Sigma_{t'} ZC(t',c,i) \times SR(t-t',c,i) \tag{S1}$$

In that equation, $ZS(t,c,i)S_{(t,c,i)}$ stands for the zinc stocks of finished product i in use in country c in year t, $F^{Con}_{(t',c,i)}$ $ZC(t',c,i)$ signifies the zinc consumption for finished product i in country c in year t', and $SR(y,c,i)RR_{(t-t',c,i)}$ denotes the survival ratio of finished product i in country c after y years.

Zinc consumption $ZC(t',c,i)$ in equation (S1) was estimated in the following equation (S2):

$$ZC(t',c,i) = \Sigma_{t'}PEU(t',c,i) \times FE(t',c, i)/100 - EFP(t',c,i) + IFP(t',c,i) \tag{S2}$$

Therein, $PEU(t',c,i)$ denotes the principal end-uses i in country c in year t'; $FE(t',c,i)$ signifies the fabrication efficiency of semi-finished product i for the production of finished products in country c in year t' (%); $EFP(t',c,i)$ and $IFP(t',c,i)$ are the amounts of zinc in exported and imported finished products i in country c in year t'.

$$PEU(t',c,i) = PEU(t',c) \times AR(t',c,i)/100 \tag{S3}$$

In that equation, $PEU(t',c)$ represents principal end-uses of semi-product in country c in year t'. $AR(t',c,i)$ is the allocation ratio from semi products to finished product i in country c in year t' (%).

The survival ratio is calculable using the following equation.

$$SR(y,c,i) = EXP[-\{y/Yci\}^{dci}. \{\Gamma(1+1/dci)^{dci}\}] \tag{S4}$$

where y denotes the age of finished products, Yci signifies the average lifetime of finished product i in country c, Γ is the gamma function, dci is the parameter of distribution range of finished product i in country c.

In addition, generated end-of-life scrap was calculated as

$$GEolS(t,c,i) = \Sigma_{t'} ZC(t',c,i) \times DR(t-t',c,i) \qquad \text{(S5)}$$

$$DR(y,c,i) = SR(y,c,i) - SR(y+1,c,i), \qquad \text{(S6)}$$

where $DR(y,c,i)$ is the discard ratio of finished product i in country c after y years.

Table S2. Zinc contents of traded commodities

Life process	SITC Rev.1	Items	Zinc content	Source
Galvanizing	67481	Plates under 3mm coated ex tin not h.c. or all.	4 %	Spatari, S., et al. (2003), Naito, W., et al. (2007)
	67701	Iron/steel wire not high carbon or alloy steel	4 %	Spatari, S., et al. (2003), Naito, W., et al. (2007)
	67702	Iron/steel wire of high carbon steel	4 %	Spatari, S., et al. (2003), Naito, W., et al. (2007)
	674	Universals, plates and sheets of iron or steel	4 %	Spatari, S., et al. (2003), Naito, W., et al. (2007)
	67483	Plates/sheets <3mm coated of alloy steel	4 %	Spatari, S., et al. (2003), Naito, W., et al. (2007)
Die casting	6861	Zinc and zinc alloys, Unwrought	95 %	IZA, 2002
Brass	68212	Refined copper including remelted	35 %	CDA (2012) and Twarog, D.L. (2000)
	68221	Bars, rods, angles, shapes, wire of copper	35 %	CDA (2012) and Twarog, D.L. (2000)
	68222	Plates, sheets and strip of copper	35 %	CDA (2012) and Twarog, D.L. (2000)
	68225	Tubes, pipes and blanks, hollow bars of copper	35 %	CDA (2012) and Twarog, D.L. (2000)
Zinc semis	68621	Bars, rods, angles, shapes, sections/wire of zinc	100 %	Yang. Y.M., et al. (2014)
	68622	Plates, sheets, strip, foil, powders, flakes of zinc	100 %	Yang. Y.M., et al. (2014)
	68623	Tubes, pipes, blanks/fittings, hollow bars of zinc	100 %	Yang. Y.M., et al. (2014)
Chemicals	51351	Zinc oxide and peroxide	80 %	Edwards and Baker (1999)
	51412	Chlorides and oxychlorides	36 %	Edwards and Baker (1999)
	51421	Sulphides (incl.polysulphides)	67 %	Goodwin, F.E. (2006)
	51424	Sulphates (incl.alums) and persulphates	23 %	Goodwin, F.E and Updated by Staff (2012)
	51435	Salts of metallic acids	18 %	Meylan and Reck, (2016)
Construction	6913	Fin. structural parts of zinc	100%	Assumption
	69331	Gauze, netting, grill, fencing wire of iron steel	4 %	Spatari, S., et al. (2003), Naito, W., et al. (2007)
	69897	Articles of zinc, n.e.s.	100 %	Meylan and Reck, (2016)
Transportat	7125	Tractors, other than road tractors	0.6 %	Roskill (1997)

ion	7321	Passenger motor cars, other than buses	0.9 %	Spatari, S., et al. (2003), AGA (2013), USEPA (2013)
	7322	Buses, including trolleybuses	0.6 %	JOGMEC (2007)
	7323	Lorries and trucks, including ambulances, etc.	0.6 %	Campestrini and Mock (2011)
	7324	Special purpose lorries, trucks and vans	0.6 %	USEPA (2013)
	7325	Road tractors for tractor trailer combinations	0.6 %	Roskill (1997)
	73291	Motorcycles, auto cycles, etc.& side cars	0.9 %	Cherry, C.R., et al. (2009)
Electrical and electronic products	71912	Air conditioning machines	2 %	Environment Canada (2000)
	71915	Refrigerators not domestic & oth refrig equip.	2 %	Environment Canada (2000)
	72501	Domestic refrigerators, electrical	2 %	Environment Canada (2000)
	72502	Domestic washing machines whether or not elec.	2 %	Environment Canada (2000)
	71962	Mach. for cleaning or filling containers	2 %	Environment Canada (2000)
	71715	Textile bleaching, washing, dressing, etc. Mach.	2 %	Environment Canada (2000)
	7143	Statistical machines cards or tapes	2 %	Environment Canada (2000)
	7296	Electro mechanical hand tools	2 %	Environment Canada (2000)
	72503	Electro mechanical domestic appliances nes	2 %	Environment Canada (2000)
	72504	Electric shavers & hair clippers	2 %	Environment Canada (2000)
	72505	Electric space heating equipment etc.	2 %	Environment Canada (2000)
	72491	Electrical line telephone & telegraph equipment	2 %	Environment Canada (2000)
	72492	Microphones, loudspeakers & amplifiers	2 %	Environment Canada (2000)
	72499	Other telecommunications equipment	2 %	Environment Canada (2000)
	7241	Television broadcast receivers	2 %	Environment Canada (2000)
	89112	Acc. of gramophones, tape recorders & sound rec.	2 %	Environment Canada (2000)

Industrial and metal working machinery	71521	Converters, ladles, ingot moulds & castings	2 %	Spatari, S., et al. (2003)
	71522	Rolling mills & rolls, for metalworking	2 %	Spatari, S., et al. (2003)
	71523	Gas operated welding, cutting etc. Appliances, Machinery and mechanical appliances, nes	2 %	Spatari, S., et al. (2003)
	7198	Machinery and mechanical appliances, nes	2 %	Spatari, S., et al. (2003)
	72992	Electr. Furnaces, welding & cutting apparatus	2 %	Spatari, S., et al. (2003)
Scrap	2820	Iron and steel scrap	4 %	Spatari, S., et al. (2003); Naito, W., et al. (2007)
	28401	Ash and residues bearing non ferrous metals	51 %	BGRIMM (2010)
	28402	Copper waste and scrap	10 %	Meylan and Reck, (2016)
	28407	Zinc waste and scrap	95 %	Meylan and Reck, (2016)

Table S3. Fabrication efficiency (%) of semi-finished products and mean lifetime (year)

of finished products

		Cons	Trans	IE	E&EG	Agri	Misc	Reference
Fabrication efficiency (%)		85	85	85	85	85	85	Van Genderen (2014)
Mean lifetime (year)*	China	20	12	10	15	14	14	Meylan and Reck, (2016)
	France	60	12	10	15	14	14	
	Germany	60	12	10	15	14	14	
	Italy	60	12	10	15	14	14	
	Japan	60	12	10	15	14	14	
	United States	60	12	10	15	14	14	

* Weibull distribution function was used with the distribution width of 2, based on Tasaki et al. (2001).
Note: Construction (Cons), Transportation (Trans.), Industrial Machinery (IE), Electrical and Electronic Goods (E&EG), Agriculture (Agri), and Miscellaneous Uses (Misc).

Table S4. Landfill ratio (%), dissipation ratio (%) and mixed metal loss ratio

	Landfill ratio (%)*	Mixed metal loss ratio (%)	Dissipation ratio (%)
China	47%	53%	
France	49%	51%	
Germany	49%	51%	0.4% for galvanizing uses; 20% for zinc in tire
Italy	21%	79%	
Japan	43%	57%	
United States	44%	56%	
Sources	Gordon et al. (2004)	Own assumption	Meylan and Reck (2016)

*This was originally landfill ratio in 1994 and was used for 1962-2010.

Note: All zinc chemicals going to agriculture (end use) are dissipated in the same year they enter use.

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