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INTEGRATION OF MFA AND LCA METHODOLOGIES: THE ANTHROPOGENIC ALUMINIUM CYCLE IN ITALY

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Abbreviations

AIV	Aluminium Intensive Vehicle
AR	Alumina Refining
ASR	Automotive Shredder Residue
B&C	Building and Construction
BA	Bottom Ash
BAT	Best Available Technology
BM	Bauxite Mining
C&P	Containers and Packaging
CED	Cumulative Energy Demand
CES	Collection of Scrap and Obsolete Products
CF	Carbon Footprint
ConDur	Consumer Durables
DfD	Design for Disassembly
DfE	Design for Environment
DfR	Design for Recycling
ECs	Eddy currents
EE	Electrical Engineering
ELV	End-of-Life Vehicle
LCA	Life Cycle Assessment
LHV	Low Heating Value
M&E	Machinery and Equipment
MAU	Manufacturing
MBT	Mechanical Biological Treatment
MCM	Markov Chain Model
MFA	Material Flow Analysis
MS	Melting of Scrap for Secondary Production
MSW	Municipal Solid Waste
NF	Non-Ferrous
OT	Other process
P	Production
PAS	Primary Aluminium Smelting
PFCs	Perfluorocarbons
PST	Post-Shredder Technology
PVC	Polyvinylchloride
RO	Rolling process
SITC	Standard International Trade Classification

EOL	End-of-Life	**STAF**	Stocks and Flows Project
EX	Extrusion process	**T&D**	Transmission and Distribution
F&M	Fabrication and Manufacturing	**TC**	Transfer Coefficient
FC	Foundry Casting	**Trans**	Transportation
GDP	Gross Domestic Production	**TS**	Treatment of Scrap
GHG	Greenhouse Gas	**U**	Use
GWP	Global Warming Potential	**VCA**	Value Chain Analysis
HS	Harmonized Commodity Description and Coding System	**WM&R**	Waste Management and Recycling
IC	Ingot Casting	**WtE**	Waste-to-Energy
LCA	Life Cycle Assessment		

Abstract

MFA and LCA methodologies were applied to analyse the anthropogenic aluminium cycle in Italy with focus on historical evolution of stocks and flows of the metal, embodied GHG emissions, and potentials from recycling to provide key features to Italy for prioritizing industrial policy toward low-carbon technologies and materials.

Historical trend series were collected from 1947 to 2009 and balanced with data from production, manufacturing and waste management of aluminium-containing products, using a 'top-down' approach to quantify the contemporary in-use stock of the metal, and helping to identify 'applications where aluminium is not yet being recycled to its full potential and to identify present and future recycling flows'. The MFA results were used as a basis for the LCA aimed at evaluating the carbon footprint evolution, from primary and electrical energy, the smelting process and the transportation, embodied in the Italian aluminium. A discussion about how the main factors, according to the *Kaya Identity* equation, they did influence the Italian GHG emissions pattern over time, and which are the levers to mitigate it, it has been also reported.

The contemporary anthropogenic reservoirs of aluminium was estimated at about 320 kg per capita, mainly embedded within the transportation and building and construction sectors. Cumulative in-use stock represents approximately 11 years of supply at current usage rates (about 20 Mt versus 1.7 Mt/year), and it would imply a potential of about 160 Mt of CO_2eq emissions savings. A discussion of criticality related to aluminium waste recovery from the transportation and the containers and packaging sectors was also included in the study, providing an example for how MFA and LCA may support decision-making at sectorial or regional level. The research constitutes the first attempt of an integrated approach between MFA and LCA applied to the aluminium cycle in Italy.

"… The French once displayed Fort Knox-like aluminium bars next to their crown jewels, and the minor emperor Napoleon III reserved a prized set of aluminium cutlery for special guests at banquets. (Less favoured guests used gold knives and forks)."

"… The spelling disagreement [between the international "aluminium" and the American "aluminum"] traces its roots back to the rapid rise of this metal. […] When Charles Hall applied for patents on his electric current process, he used the extra *i*, too. However, when advertising his shiny metal, Hall was looser with his language. […] He dropped the vowel permanently, and with it a syllable, which aligned his product with classy platinum. His new metal caught on so quickly and grew so economically important that "aluminum" became indelibly stamped on the American psyche. As always in the United States, money talks."

– Sam Kean, *The Disappearing Spoon*,
Back Bay Books, New York, USA, 2010 –

1.

Introduction

1.1 Background

In the last decades, social interest toward environmental issues increased significantly achieving a global dimension. Today, global warming and climate change are among environmental threats that generate much concern in social perception and many industrial activities, reports or policy initiatives focus on related indicators (Laurent et al. 2010). Carbon footprint and greenhouse gases emissions are terms entered in the present common speech and although they describe only a part of environmental emergencies, it is certain of topical interest.

For the last fifty years, the average surface temperature on the Earth has increased averagely by 0.13 °C per decade, almost doubling the linear warming trend over the whole century, due to the effect of natural events and human activities, with the latter most likely to have caused most of the global climate change since the second half of the twentieth-century. From 1970 to 2004, anthropogenic greenhouse gas (GHG) emissions increased by approximately 70%, largely from the energy supply, manufacturing, and transportation sectors, amounting at about 60% of global CO_2 emissions (IPCC 2007). As consequence, a dramatic global warming is possible by 2050 that would result across Europe in average temperatures of 4-5 °C warmer than current baseline, a scenario whose effects would severely jeopardize living species. However, if a reduction by a factor 2.5 in GHG emissions was achieved, the proportion of ecosystems severely impaired is predicted to be $< 5\%$ of the current total (IPCC 2007).

Environmental regulations, corporate social responsibility and initiatives for environmental resource protection are among the most pressing drivers for a more sustainable system of production and consumption after for decades cultural attitudes, social behaviours and political factors caused the exploitation of natural resources claiming for a growth and development as absolutes (Gordon et al 2006). Innovation and efficiency-oriented new materials production, designs, and industrial processes show large potential for reduction of environmental burdens (Luttropp & Lagerstedt 2006; Bribián et al. 2011; Anastas & Zimmerman 2003). The article 4th in the *Sixth Programme of Environmental Action* by European Community claims for sustainability in the use of natural resources and in the waste management practices such key levers to preserve the resilience of the earth and guarantee the bearing capacity in the consumption of fossil and renewable fuels. Decoupling the economical growth both to the use of natural resources and waste generation should be achieved by increasing efficiencies of processes and systems, dematerialization measures and initiatives for prevention.

The concept of sustainability derives strictly from the term ‘sustainable development’ (WCED Report 1987; Heijungs et al. 2010), which individuates as social, economic and environmental the three pillars to hold up the goal of sustainability (Huang et al. 2012). However, defining sustainability may determine some contradictions since a time factor must be clearly identified as dynamic features characterizing the exploitation as well as the consumption of materials determine a continuous revision of what is today sustainable (Simpson et al. 2005; Gordon et al. 2006). Simplifying, ‘everything can be sustainable in the short term, but on the longest of timescales nothing can’ (Graedel & Allenby 2010).

Industry, as in its widest definition of human activity, should strive for long-term balance in the carrying capacity and resilience of ecosystems (Ayres & Ayres 2002). In the metals industry specifically, analyses oriented to enhance recycling practices play a strategic role to reduce pressure on natural resources and increase the resilience of metal-based manufacturing and technology. Particularly, primary production of aluminium is one of the most energy-intensive bulk material industries and, considering the widespread use of the metal in the global economy, measures such as decreasing GHG footprint through alternate processing routes, improvements in emissions control, and intensification in the use of secondary (recycled) material will have significant benefits globally. Indeed, despite aluminium being the most abundant metallic element of Earth's crust at about 8.8% by weight (Sanders 2002), its industry is relatively young compared to other major non-ferrous metals such as copper, lead, and zinc, whose use was known since ancient times. Aluminium production has grown steadily since the late 1880s, resulting in it being the most exploited non-ferrous metal since 1900 as shown by Sanders (2002), Johnson et al. (2007), and Graedel & Erdmann (2012). Supplies grew by almost four thousand times during the past century, while production of chromium and nickel, the second and the third high-growth non-ferrous metals, increased by 300 and 100 times, respectively. On a global level, aluminium demand is driven by increasing global population and affluence, urbanization and industrialization: due to its features of lightness, strength and ductility, aluminium is used in many end-use sectors and applications mainly comprising transportation, building and construction, packaging, domestic and office appliances, machinery and equipment, electrical engineering, and chemical, food and agricultural uses (Johnson et al. 2007; Reck & Gordon 2008; USGS 2006; The Aluminium Association 2010).

The aluminium industry has started a voluntary program of continuous improvement (IAI 2012) in order to adopt greener objectives along the metal life cycle. Among these are initiatives for GHG emission reduction such as the cut of 50% of perfluorocarbons (PFCs) emissions by 2020 from 2006 levels; decreasing smelting energy use and water consumption; implementation of environmental management systems including bauxite mining rehabilitation, and the development of sophisticated mass-balance models to identify future global recycling flows (IAI 2012; Martchek 2006). Despite such an improvement, Liu et al. (2011) argued how the achieving of further betterments in the next years could be challenging if new technologies were not discovered and the demand for primary metal grows at current dramatic rates.

The development for strategies aiming at decoupling production and consumption growth from environmental impacts and for achieving sustainable performance of resources' uses, requires to be supported from both a theoretical and practical levels. Thus, indicators and analytical tools have been developed for quantifying and evaluating environmental consequences of processes and systems for providing and implementing sustainability guidance (Finnveden et al. 2009; Graedel & Allenby 2010; Van Caneghem et al. 2010; Wursthorn et al. 2011). Among others, *Material Flow Analysis* (MFA) and *Life Cycle Assessment* (LCA) have been applied extensively (Brunner & Rechberger 2004; Heijungs et al. 2010; Udo de Haes & Heijungs 2007; McMillan 2011), to interpret and evaluate large-scale use of materials on environmental, social, moral, economical or political bases.

The integration of such methods is carried out as much to overpass limits and disadvantages that characterize single analytical tools as to extend the assessment, and making the analysis more accurate by adopting a comprehensive approach that avoids problem-shifting that occurs when focusing on partial systems (Huang et al. 2012; Finnveden et al. 2009 McMillan 2011). Considering fundamental features, dimensions, and limits of MFA and LCA, the two methodologies reveal potentials for integrating in order to improve completeness when approaching environmental issues. Particularly, the former can be regarded as an inventory method for establishing an LCA, although a certain discrepancy may result from tendency of LCA to strive for assessing as many as possible substances, and MFA position to reduce the number of substance and goods investigated to guarantee as much as possible control and comprehensibility.

MFA provides a quantitative picture of resource use at various scales since it refers to a systematic analysis of material stocks and flows characterizing a system defined in space and time (Brunner & Rechberger 2004). It goes beyond basic mass balances of a process and thanks to its feature of including temporal variables, it oriented the contemporary literature towards researches and strategies at approaching urban agglomerates as mines of the future (Graedel 2011; Jacobs 1961). End-of-life (EOL) products and waste streams often have higher exploitable metal concentrations than current average ore grades (Johnson et al. 2007) and are more geographically widespread than geological deposits. Bearing in mind that metals, as elemental materials, can be transformed but not destroyed, MFA applies to anthropogenic resource stocks and flows in order to evaluate the current 'metabolism' of human societies, supporting recycling activities and estimating future needs based on socio-economic trends (Graedel & Allenby 2010).

MFA case studies have often focused on metals for several reasons: their critical role in human societies and technological systems, energy consumption and emissions during their production, relative high recyclability, and potential toxicological effects. Nickel, chromium, copper, iron and aluminium are some of the most thoroughly investigated (Reck et al. 2008; Reck & Gordon 2008; Johnson et al. 2006; Rostkowski et al. 2007; Spatari et al. 2005; Wang & Graedel 2010; Chen et al. 2010; Müller et al. 2011).

LCA is codified by the *International Standard Organisation* series 14040-14044 (ISO 2006) and it aims at evaluating the potential burdens on environment throughout the life cycle of a product, process or system, from raw material extraction to EOL treatments (Finnveden et al 2009). LCA has been subjected to several methodological developments and it is nowadays an assessment tool widely applied to a huge variety of products and processes at various scale levels; therefore, it may not be surprising that the European Directive 2008/98 on waste clearly indicated in a life cycle approach the criteria to confirm or not the European hierarchy for waste management practices (Directive 2008/98/EC).

In this doctoral thesis MFA and LCA methodologies were applied to analyse the anthropogenic aluminium cycle in Italy with focus on historical evolution of stocks and flows of the metal, related GHG emissions from aluminium production, and potentials from recycling. The research constitutes the first attempt of an integrated approach of MFA and LCA applied to the aluminium cycle in Italy.

1.2 Motivations and goals of the study

Metals consumption to provide products and services determine a transfer from natural reservoirs in the lithosphere to in-use stocks in the anthroposphere. Once a metal is in-use, it may be subjected to dissipative phenomena such as corrosion or erosion, recycling processes to new products or deposited in sinks (e.g. landfills) at the end of life. Comparing the amount of natural reservoirs to anthropogenic in-use stocks and waste generation gives a measure of human progress to rely on recycling rather than on primary environmental resources for providing materials and services (Gordon et al. 2006). According to Liu et al. (2011), investigating the dynamics of anthropogenic aluminium cycle allows a systemic approach that supports the detection of environmental-oriented strategies as much as potentials of industrial improvements.

Although, traditionally, aluminium has been one of the largest and valuable heavy industries in Italy together with chemicals and iron & steel (USGS 2009), to date a quantitative estimation of Italian aluminium flows and anthropogenic reservoirs has not been carried out yet; the only paper (Amicarelli et al., 2004) that tried to summarize aggregated aluminium flows in Italy does not give information on metal in-use stocks in society and lacking data affect the metal losses from life cycle steps, and hence an overall perspective of the issue is still missing.

With this intent, the research investigated trends and opportunities in the Italian aluminium cycle by means of stocks and flows modelling. Historical trend series were collected from 1947 to 2009 and balanced with data from production, manufacturing and waste management of aluminium-containing products, using a 'top-down' approach (Gerst & Graedel 2008) to quantify the contemporary anthropogenic reservoirs of the metal within the national boundaries.

The primary motivations for studying anthropological stocks and flows of aluminium in Italy are that, at a national level, such analyses allow us to forecast future domestic metal supply and demand and help to identify 'applications where aluminium is not yet being recycled to its full potential and to identify present and future recycling flows' (IAI 2009). More broadly, the metal life cycle is archetypal of modern resource use generally (IAI 2012), and so a deeper knowledge may help to address successfully the challenge towards a more sustainable use of resources and to guide future national industrial policy and planning. Italy is also a significant player in the global economy of this material; the country has negligible natural reservoirs of aluminium-containing bauxite and imports unrefined forms of the metal. On the other hand, Italy has extensive manufacturing and production of high value-added finished products, with secondary aluminium flows as driving force for the national metal production. The Italian aluminium recycling industry consists of more than fifty refining and remelting plants, and the country places with Germany at the top in Europe for secondary production capacity, and in the third place on a worldwide level after the United States and Japan (CiAl 2010; EAA-OEA 2008).

Secondly, the results of the study were used as a basis for a further analysis aimed at considering the environmental dimension related with flows and stocks of aluminium in Italy. I.e., applying integration between MFA and LCA methodologies it was sought to evaluate how the carbon footprint of the Italian aluminium production changed over years and which benefits would result from exploiting the metal in-use stock. Advantages of adopting a long-term dynamic perspective rely in helping to examine how industrial emissions and consumption of building materials has historically evolved compared to Italy's total GHG emissions, and hence to provide key features to Italy for prioritizing industrial policy toward low-carbon technologies and materials. In addition, the inclusion of aluminium industry into the European carbon markets will imply also economical revenues for a firm from credits: from 2013 the *Emission Trading System* (EU-ETS) will be extended to include new sectors and GHGs, including production plants of aluminium and perfluorocarbons emissions from the aluminium sector (EC 2012).

Besides, the study is intended to help analyses aimed at understanding the global implications of moving aluminium production to countries with different environmental and industrial patterns. Lastly, the outcomes constitute a country-specific set of data for evaluating the global warming potentials of the Italian aluminium production, which integrates and updates the average scores reported in European databases for LCA analysis, supporting consistently future policy and industrial plans.

1.3 Key questions investigated

Following are listed some of the main driving questions of the work to which it is sought to answer through the research carried out. They are divided according to the two general topics treated.

Stocks and flows of the anthropogenic aluminium cycle in Italy

How has the aluminium industry changed over years in Italy?

What is the resulting magnitude of aluminium flows and stocks?

How has Italy related to other countries in terms of trade flows of aluminium?

How might Italy reduce its dependence from net-import of unwrought aluminium?

Which are the main end-use sectors for metal scrap generation?

Which are the main end-use sectors that embed the most relevant amount of the metal?

What are framework and efficiencies of domestic recovery and recycling activities for aluminium-containing waste?

Where does the highest losses of aluminium occur along its life cycle?

What is the contribution of secondary aluminium in the metal industry?

What is the potential to enhance recycling activities for the country?

Which are the main key-issues in which Italy should act?

Which are assumptions and uncertainties affecting the model created?

GHG emissions embodied in the Italian aluminium

How did GHG emissions evolve over years from the domestic aluminium production?

What is the carbon footprint embodied in the aluminium produced and consumed in the country?

What are the contributions of each unit process in the primary aluminium production?

How did GHG footprint related to primary and electrical energy, process, and transportation emissions?

What is the potential of reducing carbon dioxide emissions from exploiting in-use stocks of the metal?
What country-specific global warming potentials can be estimated for primary and mixed production of aluminium?
How did the main Kaya Identity factors influence the Italian GHG emissions pattern?
Which are the levers to mitigate it?
Which are assumptions and uncertainties affecting the model created?

1.4 Structure of the work

This doctoral thesis aims at applying an integration of MFA and LCA to the aluminium life cycle in Italy; in doing so, the work has been structured as follows.
In Chapter 1, motivations and goals of the study have been introduced in the light of the main questions to be answered. Chapter 2 presents MFA and LCA methodologies applied with focus to the case study investigated and a description of the equations set used in the modelling. Discussion of the results for the historical stock and flows and climate change models are reported in Chapter 3 and Chapter 4, respectively. Chapter 5 is dedicated to the discussion of criticality related to aluminium recovery from specific waste flows such as the transportation and the containers and packaging sectors. With the aim to enhance the recycling industry in Italy, this section even provides an example for how MFA and LCA may support decision-making at sectorial or regional level.
Lastly, Chapter 6 outlines the main conclusions and personal considerations should be given to the study.

2.

Methodology

In this Chapter, fundamental basis of MFA and LCA and detailed models developed for the national aluminium cycle investigated are presented. Brunner & Rechberger (2004) discuss how LCA inventory data can be integrated with economy-wide MFA results, and that consistency increases when methods are applied to systems rather than products. The authors cautioned about discrepancies that may arise from the tendency of MFA towards reducing the number of variables, and LCA that instead aims for achieving the greatest completeness of the system under study. Bearing in mind such a recommendation, MFA and LCA were applied to understand the national anthropogenic aluminium 'metabolism', and its relation with the environment.

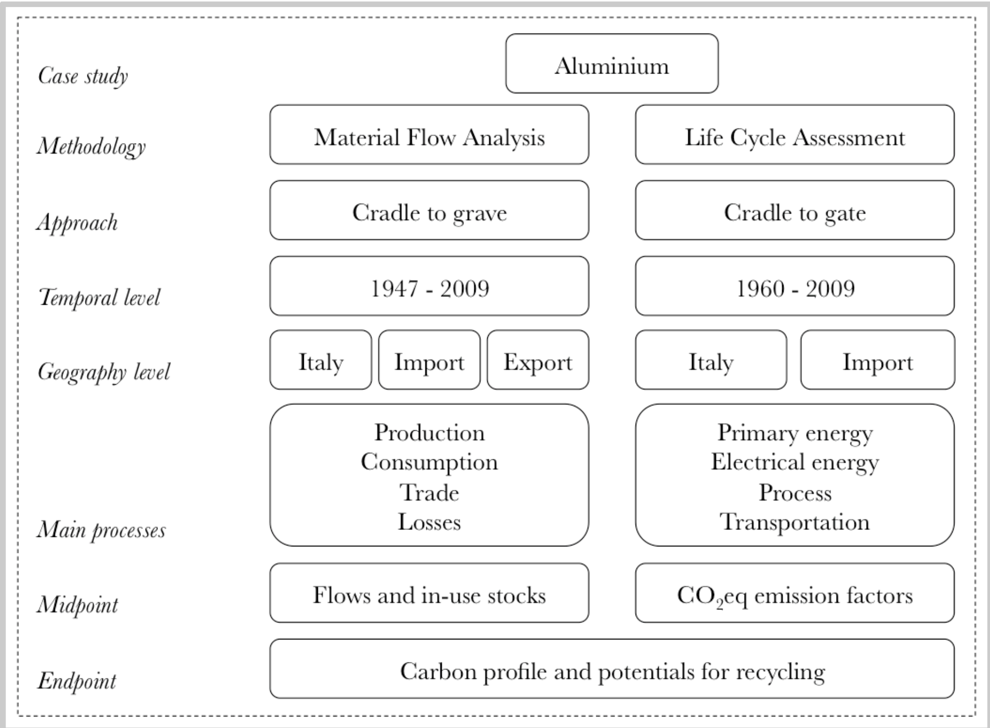
2.1 The dynamic MFA model

Determining in-use stocks in human society by MFA approaches has distinct advantages when analysing resource flows trends. Indeed, in-use stocks allow describing the relationship between final products entering the usage stage and old scrap generation, and they are more suitable for long-term analyses because of the trend to remain stored in anthropogenic reservoirs (Müller et al. 2011).

MFA techniques have been applied previously to aluminium at various scales to track the metal along its whole life cycle, however, only a minority of studies utilizing a dynamic approach aimed at quantifying stocks and flows over different time spans. Martchek (2006) proposed a global quantitative model for aluminium, and the year-2009 updated the *International Aluminium Institute* mass flow model estimated the global in-use stock accumulated since 1880s at 660 Mt. Boin & Bertram (2005) created a detailed aluminium mass balance for European smelting and refining, with particular attention to metal losses during each step; Chen et al. (2010) applied *Substance Flow Analysis* to analyse aluminium stocks and flows changes in China in three representative years, while Liu et al. (2011) performed a dynamic aluminium anthropogenic cycle in the United States, integrated with an environmental analysis to estimate the potential of de-carbonizing the metal cycle. A critical review of anthropogenic cycles of the elements with typical features of aluminium analysis is provided in (Chen & Graedel 2012b), while a comparison among in-use stocks for Japan, U.S., Europe and China by dynamic MFA was carried on by Hatayama et al. (2009): extrapolation to the future was estimated by using population and *Gross Demand Production* (GDP) changes. The economic dimension was also included in Dahlstrom & Ekins (2007), which combined MFA and *Value Chain Analysis* (VCA) to quantify aluminium flows through the United Kingdom. To analyse the potential recycling of aluminium, several MFA studies focused specifically on the presence of alloying elements as these can inhibit or alter the recycling of aluminium as contaminants in re-melting operations (Hatayama et al. 2007), and the metal scrap generation (Melo 1999; Nakajima et al. 2007). To decrease the uncertainty that affects assumptions on average lifetimes for scrap generation can be obtained through complementary methods as the *Markov chain modelling* (MCM): Eckelman & Daigo (2008) applied MCM to analyse the global technological lifetime of copper. Although that methodology provides a suitable tool to investigate complex material flows systems, it results particularly helpful when there is geographic differentiation (Eckelman & Daigo 2008). Since this study focuses on a single nation, it is assumed that spatial distribution within the country be uniform.

The *Stock and Flows Project* (STAF) framework divides the life cycle of a substance into four main stages (**Figure 3**): each stage embeds the main sub-processes and balances for material inputs and outputs. Chen et al. (2010) reported a highly detailed description of the STAF methodology, which is briefly reviewed here.

Figure 1 – Scheme for MFA and LCA integration followed in the study: brief comparison of similarities and differences between the two methodologies.

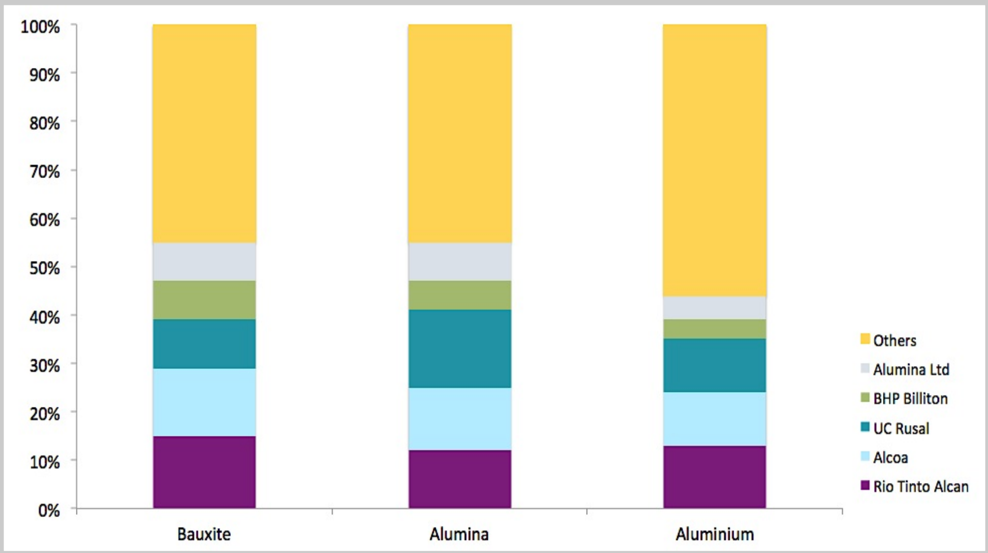


2.1.1 Description of the anthropogenic aluminium cycle

Aluminium is the third element in the earth's crust, amounting at about 8-9% by weight; despite its abundance, anyway, the metal is rarely found free in nature, but often combined in mineral forms with oxygen and silicon. Among aluminium-containing rocks, bauxite ore presents highest concentrations of aluminium hydroxide, and it has constituted the starting material for the industrial production of aluminium since the first commercial process developed. Indeed, the process route remained almost the same until today, and it begins with the extraction of metallurgical grade alumina from the bauxite ore through the *Bayer Process*, and it ends with the production of primary aluminium by means of the *Hall-Héroult* electrolysis step. The Bayer Process, specifically, converts bauxite minerals into alumina through a series of four main steps, namely: digestion of ore with caustic soda, liquor stream clarification, precipitation and calcination of alumina hydrate. During the refining process, about two tons of red mud as by-product are generated per ton of alumina produced, which still constitute a relevant load for the environment (Tan & Khoo 2005). In the Hall-Héroult process alumina is then dissolved in a molten cryolite bath and aluminium is obtained by electrolysis and deposited on a carbon cathode, while oxygen consumes the carbon anode. Operational melting point is between 920-980°C, since the system cryolite plus alumina and impurities it determines a eutectic that decreases single melting temperatures of pure compounds (1012°C for cryolite and about 2000°C for alumina). In spite of decreasing the operational temperature, the electrolysis step is the most energy intensive stage in the aluminium cycle, requiring about 20 kWh per kg of metal produced. The largest producers of virgin aluminium are China (30%), Russia (10%), Canada (8%), the United States (7%) and Australia (5%). The aluminium industry is consolidated across the world, and in the 2006 less than ten companies were controlling around 55% of the global market. Globally, about forty bauxite mines supply less than 300 aluminium production sites, and about 35% of total production is controlled by companies Rio Tinto Alcan, Alcoa and UC Rusal (Carbon Trust 2011).

Production (P) is the first stage of STAF Project framework and it describes operations from mining to the smelting process, including bauxite mining, alumina refining, primary aluminium smelting by electrolysis of alumina, and ingot casting. Generally, primary aluminium alloys smelted are cast into ingot for energy savings and easiness of shipping to fabrication and manufacturing plants.

Figure 2 – Market share of the top five aluminium producers in the year 2006. Source: Carbon Trust (2011) – modified.



Fabrication and Manufacturing (F&M) comprises the conversion of a smelter output into intermediate products, alloys and semi-finished products, which are then used to obtain final products for each end-use market. The resulting products may be divided into wrought aluminium alloys (metal sheets, plates, foils as much as wires, tubes and bars, obtained by rolling and extruding processes) and sand and die casting techniques. Because high purity aluminium is too soft and weak for most of the common mechanical application, but its resistance to acidic attack is useful for some particular purpose such as the storage of inorganic acid (e.g. nitric acid), aluminium is usually added with impurities to improve strength to weight ratio and makes metal alloys suitable for a wide variety of end uses markets. Typical alloying elements are Si, Fe, Cu, Mn, Mg, Cr, Zn, V and Zr (Sanders 2002). Generally, refiners are defined industrial plants that produce standard cast alloys and deoxidation aluminium for steel production from cast and wrought scrap, while remelters manufacture wrought alloys from wrought scrap only (UNEP-IRP 2011; Boin & Bertram 2005). A further distinction regards cast and wrought alloys: the formers have a maximum content of 20% alloying elements (e.g. Si, Mg, Cu) and the silicon is more than 5%. The latters have a maximum content of 10% alloying elements, with the silicon < 1% (UNEP-IRP 2011). Finishing processes for wrought products and casts may include esthetical or functional treatments; some examples are coating, thin film deposition, painting and anodic oxidation (Sanders 2002).

Aluminium final products enter the Use (U) stage depending on the main end use market of pertinence: namely, building and construction, transportation, mechanical and equipment, electrical engineering, containers and packaging, and other miscellaneous appliance types. The Use stage does not include sub-processes, but instead contains an in-use stock where aluminium-containing products are stored during their lifetimes in the dynamic model. In-use stock can be quantified through a top-down or a bottom-up method. The former estimates the mass balance between material or substance flows entering the use phase and waste generated leaving out. The cumulative entity of such stock is calculated by integrating year by year the mass balance determined. The bottom-up method, instead, relies on inventories of all products and services containing the material or substance of interest and combining them with census information for a given area. Albeit the bottom-up method gives information on spatial distribution of stocks at local level, it may be affected by a lack and uncertainty about metal content in waste and extension of landfills (Gordon et al 2006). A top-down approach was hence preferred even due to the availability of data regarding census information about metal production, manufacturing, trades, etc., that is generally high at national or global level (Rostkowski et al. 2007).

Waste Management and Recycling (WM&R) includes the sub-processes of scrap collection, treatment and melting for secondary aluminium production, and disposal of unrecovered material. Several factors may affect the efficiency of aluminium-containing products collection: separate waste grouping by consumers, co-operations between aluminium associations, legislators and local communities play an active role (EAA-OEA 2008). In this study, the collection of new and old scrap has been considered into WM&R since both are valuable inputs for the aluminium recycling industry: generally, new scrap indicates that material has not yet reached the use stage (it may come from P or F&M processes), while old scrap is the common term to name EOL products.

Two other types of flows are included within the model. First, import and export flows of aluminium-containing products exchanged between Italy and the rest of the world as in a world more and more globalized, trade flows of materials and products may count significantly for material accumulation or depletion in a country's economy. Traditionally, metals trade between countries has been dealt with the evaluation of ores and pure refined forms only, thus limiting the analysis since a metal can be exchanged as constituent of intermediate or finished products as well. The inclusion of such 'hidden' trade metals (Johnson et al. 2006) embedded in products is a crucial aspect of the present analysis. The second type of flows represents the depletion of minerals and ores as much as aluminium losses to air, water and soils due to corrosion, dissipation, deposition or similar transformations.

Figure 3 – STAF model adapted to the aluminium life cycle MFA used for this study. Source: Ciacci et al. (2013).

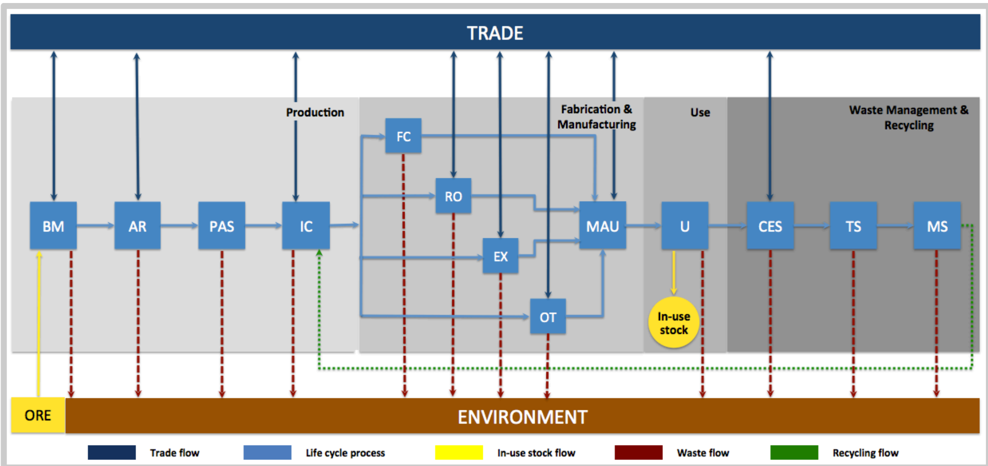


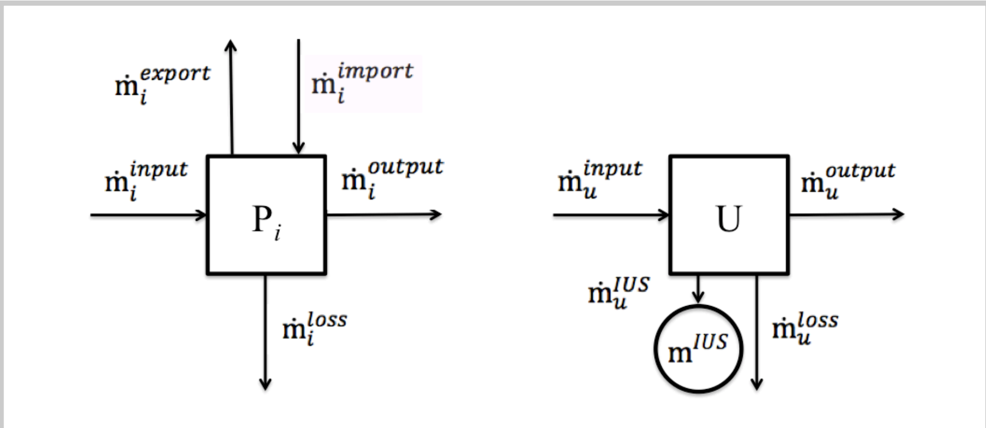
Table 1 – Main sub-processes included in each stage.

Production	**Fabrication & Manufacturing**	**Use**	**Waste Management & Recycling**
BM – Bauxite mining	***FC*** – Foundry castings	***U*** – Use	***CES*** – Collection of scrap and obsolete products
AR – Alumina refining	***RO*** – Rolling process		***TS*** – Treatment of scrap
PAS – Primary aluminium smelting	***EX*** – Extrusion process		***MS*** – Melting of scrap for secondary production
IC – Ingot casting	***OT*** – Other processes		
	MAU – Manufacturing		

2.1.2 Accounting MFA equations

As shown in **Figure 3**, life cycle processes of a substance are linked by materials flows or fluxes: a rigorous definition distinguishes between imports and exports for flows of fluxes crossing system boundaries, and inputs and outputs for those entering or leaving a process. Quantifying flows, fluxes and stocks is a fundamental task in MFA study, which can be satisfied, in accordance with the law of the conservation of matter, by a material balance of flows for every single process (Brunner & Rechberger 2004). In **Figure 4** the main material flows accounted for each process in the model, and the Use stage, are depicted.

Figure 4 – Mass balance for a generic process of the model created (on the left), and the Use stage (on the right).



Remark:

ṁ – generic mass flow of a substance in t/y; IUS – in use stock

For a general process P_i, hence, the resulting mass balance equation can be described as:

$$\dot{m}_{i,j}^{input} + \dot{m}_{i,j}^{import} = \dot{m}_{i,j}^{output} + \dot{m}_{i,j}^{export} + \dot{m}_{i,j}^{loss} \qquad \textbf{(A1)}$$

Where *i* is the index for a generic life cycle process, and *j* is the index for the year of reference. Specifically, in the case of aluminium, *i* may refer to BM, AR, PAS, IC, FC, RO, EX, OT, MAU, U, CES, TS, MS, while *j* covered years from 1947 to

2009. Mass flow parameters ($\dot{m}$) indicate the quantity of substance entering and leaving (or imported and exported by) a process. The mass balance assumes in first approximation that temporary stocks of a substance are negligible in a long term; i.e. mass flows leaving a process do equal the substance consumption. Whether that assumption is not valid, an accounting term for accumulation or depletion of temporary stocks should be added to equation **A1**. For more detail, see (Chen et al 2010).

Since material goods and related flows are generally easier to identify and quantify than single substances embodied, the amount of a substance contained in a good was calculated according to equation (**A2**). It must be pointed out that good is here intended as 'economic entity of matter with a positive or negative economic value' (Brunner & Rechberger 2004).

$$\dot{m}_{i,j} = \dot{g}_{i,j} \cdot C_{i,j} \qquad (\mathbf{A2})$$

Where $\dot{g}_{i,j}$ is a generic good containing the substance of interest, and $C_{i,j}$ is the concentration of such substance in the good itself, at process and time defined. Equation **A2** was therefore applied even to imports and exports of aluminium-containing products. The difference between import and export of the substance in the *i*-th year and by the *j*-th process it results in the net-import quantification (**A3**).

$$\dot{m}_{i,j}^{net-import} = \dot{m}_{i,j}^{import} - \dot{m}_{i,j}^{export} \qquad (\mathbf{A3})$$

Losses of aluminium were calculated according to the next two equations (**A4** and **A5**). Respectively, the former estimates the amount of the metal theoretically lost from every process, with the exception of the Use stage. The latter, instead, was applied to the quantification of aluminium consumed in the steel industry for deoxidation purposes, which constituted the metal loss characterizing the Use stage.

$$\dot{m}_{i,j}^{loss} = \dot{m}_{i,j}^{input} \cdot r_{i,j}^{loss} \qquad (\mathbf{A4})$$

$$\dot{m}_u^{loss} = \dot{s}_j \cdot D_j^{steel} \quad \textbf{(A5)}$$

Where $r_{i,j}^{loss}$ is the loss rate of aluminium for the *i*-th process in the *j*-th year; $\dot{s}_j$ is the amount of steel produced in Italy in the *j*-th year, and D_j^{steel} is the share of aluminium consumed for deoxidation purposes per ton of steel produced in Italy in the *j*-th year. A further distinction was also considered in terms of deposited and dissipated losses. The formers include in the model created the aluminium losses from the phase bauxite mining, alumina refining, collection and treatment of scrap, while the remaining processes losses were counted as dissipated.

Lastly, the in-use stock constitutes an anthropogenic reservoir of a substance embedded within the Use stage to which is associated a potential for supporting secondary material production. Through a top-down approach the annual additions to the in-use stock of aluminium were estimated: different statistical distribution can be applied to simulate the yearly waste and obsolete products generation ($\dot{m}_{u,j}^{output}$) depending on the lifetime of a good: indeed, it is unlikely that a product discarding does remain constant during the lifetime, but rather it is reasonable that the trend may increase until an average value and then gradually declines (Melo 1999). In the model, the normal distribution was chosen to represent the probability density function of the lifetime distribution for every end use markets. Equations **A6** and **A7** summary the main mass balance for the Use stage.

$$\dot{m}_{u,j}^{IUS} = \dot{m}_{u,j}^{input} - \dot{m}_{u,j}^{output} - \dot{m}_{u,j}^{loss} \quad \textbf{(A6)}$$

$$m^{IUS} = \sum_{j=1}^{k} \dot{m}_{u,j}^{IUS} \quad \textbf{(A7)}$$

Where $\dot{m}_{u,j}^{IUS}$ is the annual mass flow accumulated within the cumulative in-use stock m^{IUS}, over the *k*-th years considered.

The accounting MFA equations were applied to estimates flows, fluxes, and stocks of aluminium along the whole anthropogenic life cycle. In the next Chapter a detailed description of sources investigated for the data collection is reported.

The resulting information was used to fill a Data Spreadsheet model in Microsoft Excel®, overall structured as shown in **Table 2** and **Table 3**. Detailed results are reported in Appendix A.

Table 2 – Simplified Data Spreadsheet structure used for the MFA model. Detail for any i-th process, with the exception of the Use stage.

Year	**Process *i*-th**				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947	$\dot{m}_{i,1947}$	$\dot{m}_{i,1947}^{loss}$	$\dot{m}_{i,1947}^{input}$	$\dot{m}_{i,1947}^{net-import}$	$\dot{m}_{i,1947}^{output}$
1948	$\dot{m}_{i,1948}$	$\dot{m}_{i,1948}^{loss}$	$\dot{m}_{i,1948}^{input}$	$\dot{m}_{i,1948}^{net-import}$	$\dot{m}_{i,1948}^{output}$
1949	$\dot{m}_{i,1949}$	$\dot{m}_{i,1949}^{loss}$	$\dot{m}_{i,1949}^{input}$	$\dot{m}_{i,1949}^{net-import}$	$\dot{m}_{i,1949}^{output}$
…	…	…	…	…	…
…	…	…	…	…	…
2009	$\dot{m}_{i,2009}$	$\dot{m}_{i,2009}^{loss}$	$\dot{m}_{i,2009}^{input}$	$\dot{m}_{i,2009}^{net-import}$	$\dot{m}_{i,2009}^{output}$

Table 3 – Simplified Data Spreadsheet structure used for the MFA model. Detail for the Use stage.

Year	**Use stage**			
	Scrap	***Loss***	***Input***	***Stock change***
1947	$\dot{m}_{u,1947}^{output}$	$\dot{m}_{u,1947}^{loss}$	$\dot{m}_{u,1947}^{input}$	$\dot{m}_{u,1947}^{IUS}$
1948	$\dot{m}_{u,1948}^{output}$	$\dot{m}_{u,1948}^{loss}$	$\dot{m}_{u,1948}^{input}$	$\dot{m}_{u,1948}^{IUS}$
1949	$\dot{m}_{u,1949}^{output}$	$\dot{m}_{u,1949}^{loss}$	$\dot{m}_{u,1949}^{input}$	$\dot{m}_{u,1949}^{IUS}$
…	…	…	…	…
…	…	…	…	…
2009	$\dot{m}_{u,2009}^{output}$	$\dot{m}_{u,2009}^{loss}$	$\dot{m}_{u,2009}^{input}$	$\dot{m}_{u,2009}^{IUS}$

2.2 The LCA model

2.2.1 Carbon footprint

LCA is an evaluation method of environmental burdens associated to a product, process or service, and internationally standardized by ISO series 14040 and 14044. Its holist perspective covering all components of a product life cycle, from raw material acquisition to final disposal, even known as 'from cradle to grave', together with the possibility to evaluate a product or service as absolute or by comparing alternative options, made LCA one of the most used analytical tool for environmental assessing. As of today, many impact assessment methods used to calculate the environmental performance results of a system do exist in literature. Impact categories that may be selected are vary and extensive, and they often differ for characterisation factors and accounting method employed, making the cross comparison of results from different studies vain and conceptually incorrect. However, every impact assessment method seeks to convert mass, or economical, information into an environmental impact in term of contribution to one or more categories finally, and possibly, associated to three main damage spheres: human health, ecosystem quality and resource depletion.

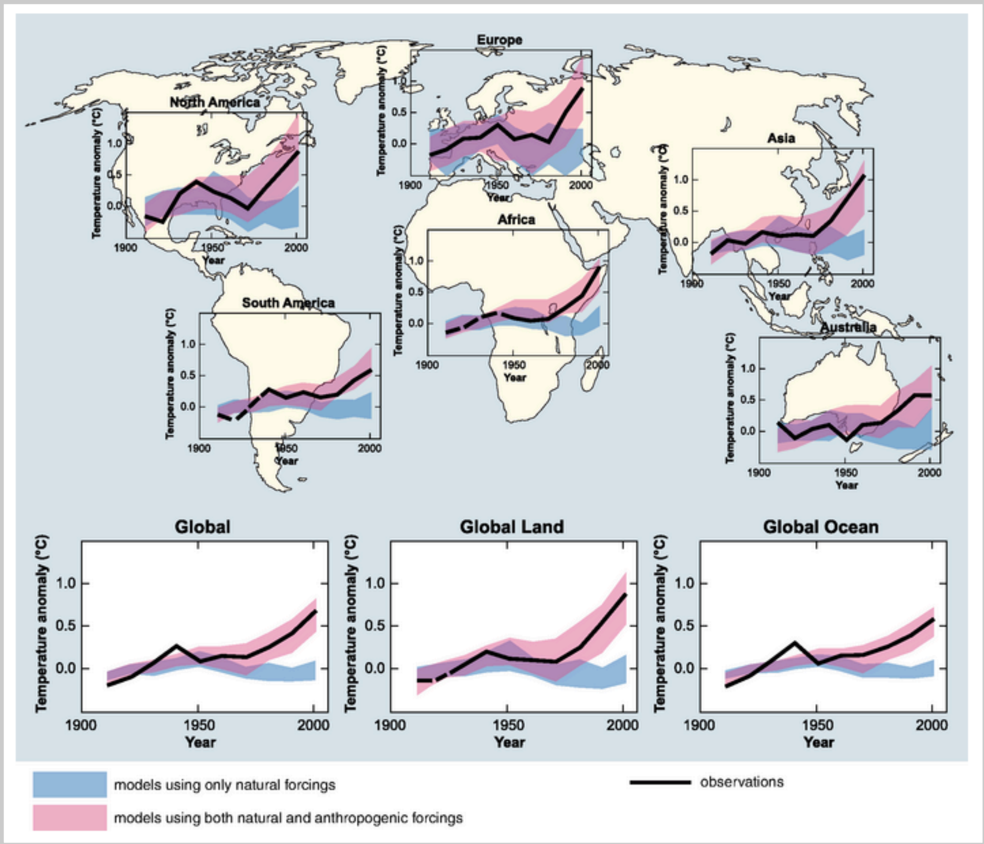
Among others, and being of high concern today, climate change has been orienting part of LCA studies toward an increasing demand for carbon footprint (CF) analysis. CF, or carbon profile, is the total amount of carbon dioxide (CO_2) and similar GHG emissions related to a product life cycle. In other words, a CF is an LCA 'with the analysis limited to emissions that have an effect on climate change' (EC 2007). The GHG species internationally indicated to be responsible for global warming effects and regulated by the *Kyoto Protocol* are, beyond carbon dioxide, methane (CH_4), nitrous oxide (N_2O), sulphur hexafluoride (SF_6), hydrofluorocarbons (HFCs), and perfluorocarbons (PFCs). In literature, several studies dealt with a LCA application to aluminium and its uses for assessing the contribution to climate change category (Mayyas et al. 2012; McMillan 2011; Liu et al. 2011; Du et al. 2010; Laurent et al. 2010; Norgate & Haque 2010; Gunasegaram & Tharumarajah 2009; Bertram et al. 2009; Tan & Khoo 2005;

Norgate & Rankin 2001). As aforementioned, aluminium industry has adopted strategies and initiatives for direct and indirect GHG emissions reduction, and from 2013 the metal production will be under the EU Emission Trading Scheme.

According to the main framework defined by ISO series and consisting in four phases, i.e. goal and scope definition, life cycle inventory, life cycle impact assessment and interpretation (ISO 2006), LCA was applied to assess the CF of aluminium production in Italy using a cradle to gate perspective. The quantification of CF is done using the *Global Warming Potential* (GWP) indicator, as defined by the *Intergovernmental Panel on Climate Change* (IPCC). When assessing environmental burdens for GHG emissions over decadal time periods, it must be borne in mind that the evaluating process is dynamic: values for GWP strictly depend on the time horizon considered, as radiative efficiency depends on the lifetimes and concentrations of GHGs in the atmosphere. GWP at 100 years (GWP_{100}) were assumed consistently with reporting under the *United Nations Framework Convention on Climate Change* (UNFCCC), (IPCC 1995). As detailed below, GWP_{100} values were linked with national-level emissions associated with MFA results for aluminium flows and in-use stocks from bauxite mining (BM), alumina refining (AR), primary aluminium smelting (PAS) and ingot casting (IC).

The model was created calculating carbon dioxide emission factors for the main aluminium production processes causing global warming effects: primary and electrical energy consumption, process-related CO_2eq leakage, and the transportation. A distinction between domestic and foreign production was also considered, and the system boundaries include GHG emissions from aluminium-containing products imported 100% allocated to Italy, while exports from the country were assumed counted during the production stage. Similarly, transportation-related CO_2 emissions were considered only for imports. This allows for consideration of both consumption and production perspectives associated with the life cycle of aluminium in Italy: trade flows play a role in explaining trend in emissions profile of a country since they imply a shift in emissions transfers between the location of production and that of consumption, with rebounds in climate policies' structure (Peters et al. 2011).

Figure 5 – Comparison between natural and anthropogenic forcings to global warming. Source: IPCC (2007).



The accounting method used for the carbon profile recalls the concept of Scope introduced by the *GHG Protocol*. System boundaries investigated cover direct emissions from fuel combustion for the production of electricity, physical or chemical processing, and transportation of materials, which draw a parallel with Scope 1; indirect emissions from the consumption of imported and internally produced electricity and indirect emissions from production of imported aluminium-containing products are analogous to Scopes 2 and Scope 3.

In the next section is a detailed description of GHG emissions accounting method for primary and electrical energy, process and transportation adopted in the study. The equations set was applied to each stage of the primary aluminium production; the results were calculated per ton of aluminium-containing product in output (i.e. bauxite, alumina, or aluminium).

2.2.2 Accounting LCA equations

2.2.2.1 Primary energy

Primary energy estimates the energy content in fossil and renewable fuels input a system and GHG emissions occur when those fuels are directly combusted for heat generation. Equation **A8** was applied to each process of the aluminium production stage

$$\mathrm{CF}_j^P = \sum \mathrm{LHV}_k \cdot \mathrm{FC}_{j,k} \cdot \mathrm{EF}_{j,k,l}^P \cdot \mathrm{GWP}_{j,l}^{100} \qquad (\mathbf{A8})$$

Where CF_j^P is the cumulative carbon footprint from primary energy in the *j*-th year, and LHV_k is the Low Heating Value for the *k*-th fuel source, which does include coal, oil, diesel, natural gas and propane gas. $\mathrm{FC}_{j,k}$ is the index for the consumption of every *k*-th fuel per ton of aluminium-containing product in the *j*-th year; $\mathrm{EF}_{j,k,l}^P$ is the emission factor expressed in mass unit of *l*-th GHG species per energy intensity of fuel consumed in primary energy production, and $\mathrm{GWP}_{j,l}^{100}$ is the term for the global warming potential at 100 years for the *l*-th GHG species in the *j*-th year. As reminded in the paragraph 2.2.1, GHG species considered are CO_2, CH_4, N_2O, SF_6, HFCs, and PFCs.

2.2.2.2 Electrical energy

The intensive feature for power consumption characterizing the primary aluminium production required counting properly the GHG emissions from electricity generation. In the model, both on-site power generation and grid electricity are considered and the carbon footprint was estimated using equation **A9**. Anyway, a distinction is done when considering percentages by sources of fuel type consumed during the conversion to electrical energy.

$$\mathrm{CF}_j^E = \frac{\mathrm{EC}_j}{(1-\lambda_j)} \cdot \sum \mathrm{Q}_{j,m} \cdot \mathrm{EF}_{j,l,m}^E \cdot \mathrm{GWP}_{j,l}^{100} \qquad (\mathbf{A9})$$

Where CF_j^E is the cumulative carbon footprint from electrical energy in the *j*-th year; EC_j is the electricity consumption; λ_j is the term to model country losses from power transmission and distribution; $Q_{j,l}$ is the percentage of electricity generated by the *m*-th fuel source in the year *j*. Fuel sources for the generation of electrical energy considered in the case study were hydro, coal, oil, natural gas, nuclear, and renewable. $EF_{j,l,m}^E$ indicates the emission factor expressed in mass unit of *l*-th GHG species emitted per unit of electrical energy produced, and $GWP_{j,l}^{100}$ is the term for the global warming potential at 100 years for the *l*-th GHG species in the *j*-th year. Similarly to primary energy accounting method, GHG species considered are CO_2, CH_4, N_2O, SF_6, HFCs, and PFCs.

2.2.2.3 Process

Accounting equation for Process was modelled to consider the contribution to climate change from PFCs emissions since carbon dioxide release from fuel consumptions were already include in Primary energy. In the case study, the following equation (**A10**) was applied to tetrafluoromethane (CF_4) and hexafluoroethane (C_2F_6) leakages from the primary aluminium smelting phase (PAS) only.

$$CF_j^{PAS} = \sum EF_{j,l}^{PAS} \cdot GWP_{j,l}^{100} \qquad (\mathbf{A10})$$

Where CF_j^{PAS} is the cumulative carbon footprint from PAS emissions in the *j*-th year; $EF_{j,l}^{PAS}$ is the term that expresses emission factors in mass unit of *l*-th GHG species emitted per unit of product (i.e. primary aluminium in the study), and $GWP_{j,l}^{100}$ is the term for the global warming potential at 100 years for the *l*-th GHG species in the *j*-th year. As aforementioned, in the anthropogenic aluminium model, GHG species for Process were CF_4 and C_2F_6.

2.2.2.4 Transportation

GHG emissions from transportation were modelled according to the type of freight operational process (ship, rail and lorry) and average distance covered at every aluminium life cycle stage. Equation **A11** was used for estimating the carbon intensity for domestic transportation of aluminium-containing products as well as trade flows. Parameter S_j was included only for freight imports from world regions and weighted under UNCTSD records.

$$CF_j^T = \sum S_{j,n} \cdot D_{j,n} \cdot EF_{j,l}^T \cdot GWP_{j,l}^{100} \qquad (\mathbf{A11})$$

Where CF_j^T is the cumulative carbon footprint from the transportation in the *j*-th year; $S_{j,n}$ is the percentage of freight transportation from the *n*-th country to Italy in the year *j*, and $D_{j,n}$ is the distance covered during the transportation from the *n*-th country to Italy. Lastly, $EF_{j,l}^T$ is the emission factor expressed in mass unit of *l*-th GHG species emitted per t*km, and $GWP_{j,l}^{100}$ is the term for the global warming potential at 100 years for the *l*-th GHG species in the *j*-th year. Again, GHG species counted were CO_2, CH_4, N_2O, SF_6, HFCs, and PFCs.

Even in this case, the resulting information was used to fill a Data Spreadsheet model in Microsoft Excel®; **Table 4** shows the overall structure.

Table 4 – Simplified Data Spreadsheet structure used for the LCA model.

Year	Domestic/Foreign primary aluminium production stage			
1960	$CF_{1960}^{X,BM}$	$CF_{1960}^{X,AR}$	$CF_{1960}^{X,PAS}$	$CF_{1960}^{X,IC}$
1961	$CF_{1961}^{X,BM}$	$CF_{1961}^{X,AR}$	$CF_{1961}^{X,PAS}$	$CF_{1961}^{X,IC}$
1962	$CF_{1962}^{X,BM}$	$CF_{1962}^{X,AR}$	$CF_{1962}^{X,PAS}$	$CF_{1962}^{X,IC}$
…	…	…	…	…
…	…	…	…	…
2009	$CF_{2009}^{X,BM}$	$CF_{2009}^{X,AR}$	$CF_{2009}^{X,PAS}$	$CF_{2009}^{X,IC}$

Remark:

X = Primary energy (P), Electricity (E), Process (PAS), Transportation (T).

Source of data, inventories, assumptions and uncertainties, and the outcomes resulting from the accounting methods for MFA and LCA models here described are reported in detail in Chapter 3 and Chapter 4, respectively.

3.

Stocks and flows of the anthropogenic aluminium cycle in Italy

The stocks and flows model described in Chapter 2 was applied to analyse the historical evolution of the anthropogenic aluminium cycle in Italy over the years 1947-2009. Since MFA is based on models that consider all stocks and flows entering and leaving a process over a given space and time, the data collection covered historical information for production, use, trade and losses for each sub-process of the model, including both aluminium products (such as sheets) and aluminium contained in multi-material products. These were then combined with information on the diffusion and consumption of items themselves, including raw materials, unwrought metal, semi-finished and finished products or commodities in order to express the data in mass of aluminium embedded. Data inventories and discussion of the results are followed reported. Supporting materials is in Appendix A. Full paper has been presented in (Ciacci et al. 2013) and the final publication is available at www.elsevier.com.

3.1 Data collection

3.1.1 Sources of data

Several sources of data were utilized. ASSOMET (2012), the *Italian Association of Non-Ferrous Metal Industry*, provided information for domestic primary production, semi-finished and castings products. Bauxite and alumina production volumes were obtained from the *US-Geological Survey* database (USGS various years), and from metals statistical reports (Metallstatistik various years). As reported by these sources, Italy did not mine bauxite ore in significant amounts from 1990s. The *United Nations Commodity Trade Statistics Database* (UNCTSD 2012) was used to determine all the registered trade flows of aluminium-containing products and commodities exchanged by Italy with the world countries from 1962 to 2009. No data were available for prior years. The inventory of aluminium-containing products was assembled at primary forms, semi-finished and final products levels, according to the *Standard International Trade Classification* (SITC), and the *Harmonized Commodity Description and Coding System* (HS) for commodities classes (Chen & Graedel 2012a).

Waste flows from the WM&R stage were modelled using a top-down approach by applying a statistical distribution (normal distribution) to the lifetime of products in each end-use sector. Lifetimes for every sector were estimated from European averages (EAA-OEA 2008) and values used in the study are reported in **Table 5**. C&P are usually consumed within one year (Hatayama et al. 2007); consequently, no statistical distribution was applied to this end-use sector. MATLAB R2011a software was used to perform the simulation.

Losses of aluminium to the environment generally occur along the entire aluminium life cycle, and some of these losses are irreversible. Tailing and slags, oxidation processes, dissipative uses of aluminium, and corrosion phenomena (Reck and Gordon 2008; Chen et al. 2010) are examples that were included in the study. Also, the use of aluminium for deoxidation purpose in the steel industry (the only kind of loss associated with the use stage) and the deposition of waste in landfills comprise other losses of the metal considered for the analysis. The main data source for losses was the *European Aluminium Association* (EAA 2008) that reported averages for the European community. Complementary information was obtained from the *Kirk-Othmer Encyclopedia* (Sanders 2002), and Kippenberger (2001); Dr. Philip Hunt from the *International Steel Statistics Bureau* (ISSB) also provided the amounts of steel production data for Italy with estimates of aluminium dissipative consumption for deoxidation. The amounts of aluminium applied to every end-use sector, which are required to simulate the waste generation rates, were mainly made available for the study by ASSOMET. Reports by Metallstatistik (various years), EAA (2008) and CiAl (2005) were also consulted to complete the inventory.

Table 5 – Lifetimes and standard deviations assumed for this study and the U.S. data reported in Liu et al. (2011). Source: Ciacci et al. (2013).

	Lifetime (Years)						**In-use stock**	
	Italy				**USA**		**Italy**	**USA**
Market	***Low***	***High***	***Mean***	***Dev***	***Mean***	***Dev***	***%***	***%***
Trans	7	20	14	5	20	5	36.1	39.2
B&C	30	50	40	14	50	15	37.8	32.9
M&E	20	30	25	7	30	5	15.3	7.6
ConDur	9	13	11	3	12	5	5.1	7.5
EE	20	30	25	7	25	5	4.6	10.9
C&P	1	1	1	0	1	0	0.8	1.2
Other	9	14	12	4	15	3	0.3	0.6

Remarks:

Trans: Transportation; B&C: Building and construction; M&E: Machinery and equipment; ConDur: Consumer durables; EE: Electrical engineering; C&P: Containers and packaging.

U.S. data are reported in Liu et al. (2011). Estimations for Italy were based on average European data (EAA-OEA 2008). Low and high values were used for the sensitivity analysis. Normal distribution was used for simulating the old scrap generation. The last columns summarize percentage of in-use stock by end-use sector for the two countries.

Based on these data, the dynamic model was built to provide results for stocks and flows of aluminium in Italy over the years 1947-2009; the results were calculated in terms of absolute quantities and per capita amounts. Italian population data were gathered from the *National Institute for Statistics* (ISTAT 2012). Cumulative results were aggregated for different decades and, eventually, the model was designed to integrate with new and more accurate data, even for future updating, in order to carry on a continuous improvement of the analysis of aluminium life cycle in Italy.

3.1.2 Inventory and assumptions

Bauxite ore was assumed the only natural source of aluminium. The metal content in bauxite was estimated to be 29.0%, based on the compositions of ores grade from Australia (90%) and Guinea (10%), which are the main providers for EU countries, including Italy (EAA-OEA 2008; UNCTSD 2012). Percentages have been estimated according to the average magnitude of mass trade flows. Recent trends show a shift towards China as primary bauxite provider for Italy; future analysis should consider relative variations in the ore grade composition. Since the UNCTS database provides registered trade flows in terms of commodities, in order to calculate the total amount of aluminium crossing the national borders some estimates of the metal content, embedded within each semi-finished or final product, were necessary to collect: in some cases proxy data from the U.S. were assumed consistent for our goal (Chen & Graedel 2012a). Values considered for the aluminium contents depict a metal purity at 99.5% for primary aluminium, 95.0% for metal plates, sheets, and strip, and 99.0% for aluminium foil as rolled products. Extrusion items. Castings resulted in 90.0% of aluminium content.

Loss rates for the Production and F&M sub-processes have been reported for various years by EEA (2008). BM and AR counted for 13.0% and 17.0%, 1.2% resulted for PAS and 0.6% for IC; ranges of values were instead used for RO, EX, OT, and FW at 3.1-4.6%, 2.2-2.7%, 2.6-3.7% and 2.6-3.7% respectively. Losses data for the foundry casting were assumed equal to U.S. proxies 6.5% (Chen & Graedel 2012a) since no more consistent information was found.

In terms of aluminium-containing raw material input a sub-process, a range of 1.923-1.925 tons was assumed to produce 1 ton of liquid aluminium from alumina. Intervals 1.006-1.032 and 1.008-1.013 tons were used for aluminium consumption to obtain 1 t of rolled and extruded product respectively. Eventually, 1.8 kg of aluminium per ton of steel was used as metal loss rate during the usage stage for deoxidation purpose (EAA 2008). Missing data in historical series have been dealt with interpolation procedures: a linear regression was chosen for range of values comprised between certain data, while in some cases they were approximated equal to the last datum available.

The annual amount of aluminium utilized in each end-use sector was obtained from *Metallstatistik* reports for decades 1954-1994 (Metallstatistik various years), while for the most recent years data were provided by ASSOMET. Linear interpolation was applied to missing data from 1995 to 1999. The lifetime values chosen for the estimation of old scrap generation are highly uncertain. Considering the wide range of aluminium-containing products and consistency with the main goal, we decided to focus at a more general level by assuming time spans for end-use sectors reasonably realistic for the country and near to similar case studies in literature (Blomberg 2007; Chen et al. 2010; Wang & Graedel 2010). Rates for collection of old scrap and obsolete products by end-use sector, leaving the U stage and entering the WM&R, were assumed accordingly to professional expertise at 80%, or quantifying at 20% the total output from CES not recovered. Of course, over years an increase in collection of scrap efficiency should be expected: anyway, since historical trends were missing, we assumed constant rates as first approximation. However, some recovery rates by sector were available for year 2006. Particularly, CiAl (2006) reported percentages for B&C, T, and C&P resulting in 96%, 95% and 55% respectively. These values were used to check robustness of the model created, as described later.

Recovery rate from treatment (TS) of old scrap was assumed at average 95%, while melting (MS) was differentiated by end-use sector according to EAA (2008) data. As aforementioned, new scrap is quantitatively recovered and recycled during F&M, hence the recovery rate from collection was considered equal to 100%, while TS was assumed similar to old scrap. Melting rate of new scrap is 99,1%, resulting from metal losses due to re-melting process adding 0.15% due to oxidation of aluminium. Average European recovery rates for MS of old scrap were estimated at 97.5% for the B&C sector, 94% for Transportation, 96.3% for C&P, 95% for Machinery & Equipment, Electrical Engineering, and Consumer Durables (EAA 2008), all high rates of recovery compared to other basic materials such as plastic.

3.1.3 Uncertainty and sensitivity analyses

Determining stocks and flows of aluminium in an entire national economy is an ambitious goal that requires several approximations, some with a high degree of uncertainty, which may influence interpretation of the final results. The main sources of uncertainty are connected with the interpolation of data to fill gaps over the study period and changes in of the aluminium content in various product types. Despite the large number of aluminium-containing products considered, aggregate inflows of aluminium into the Italian economy were consistently less than outflows in the model. This is likely due to the hidden net-import of obsolete products (such as second-hand vehicles); such products are not covered by the UNCTSD and no other reliable sources of information exist. **Table 6** summaries aluminium contents, recovery and loss rates for every life cycle phase used in the study.
A sensitivity analysis was performed aiming to bound uncertainties over the simulation results. Previous research has discussed implications of using different statistical distributions to estimate the scrap generation (Chen & Shi, 2012; Müller et al. 2011; Spatari et al. 2005; Melo 1999): overall, the choice of the distribution does not seem to affect estimates in-use stocks as much as the partitioning of products to end use sectors with relatively long lifetimes. The present model uses only normal lifetime distributions but varies the mean lifetimes for each end-use sector, as well as net-imports of aluminium, which are recognized as major sources of uncertainty (Müller et al. 2011; Chen & Shi 2012). The sensitivity analysis was conducted by running the scrap generation model using alternatively lower and upper bound data for market lifetimes (as reported in **Table 5**) and the aluminium content embedded in each commodity traded. In the latter case, the 1947-2009-years standard deviations for aluminium content vary on average 33% for bauxite and alumina, 2% for unwrought aluminium, 1% and 6% for semi-finished and final products respectively, and 10% for EOL products and scrap traded. The sensitivity analysis results are reported in **Table 7** and **Figure 13**.

Table 6 – Aluminium content, recovery and loss rates for each life cycle process assumed in the study. Values are in percentage except were specified otherwise. Ranges of values mean linear interpolation was applied between extremes listed. Source: Ciacci et al. (2013).

Life cycle process	Content	Recovery	Loss
BM	29.0	87.0	13.0
AR	52.1	83.0	17.0
PAS	99.5	1.923-1.925	1.15
		(t alumina/t liquid Al)	
IC	100.0	99.4	0.6
FC	90.0	93.5	6.5
RO	95.0-99.0	1.006-1.032	3.05-4.62
		(t Al/t product)	
EX	97.0-98.5	1.008-1.013	2.19-2.67
		(t Al/t product)	
OT	99.0	n.a.	2.62-3.65
FW	n.a.	n.a.	2.62-3.65
U	n.a.	n.a.	0.0018
			(t Al/t steel produced)
CES (new)	n.a.	100.0	0.0
CES (old)	n.a.	80.0	20.0
TS	n.a.	95.5	0.5
MS (new)	n.a.	99.1	0.9
MS (old)	n.a.	95.0	5.0

Remarks:

RO includes different types of products: plates and sheets, strips and foils.

Extrusion covers rods, profiles, tubes and pipes, bars and wires. Ranges reported reflect minimum and maximum aluminium content.

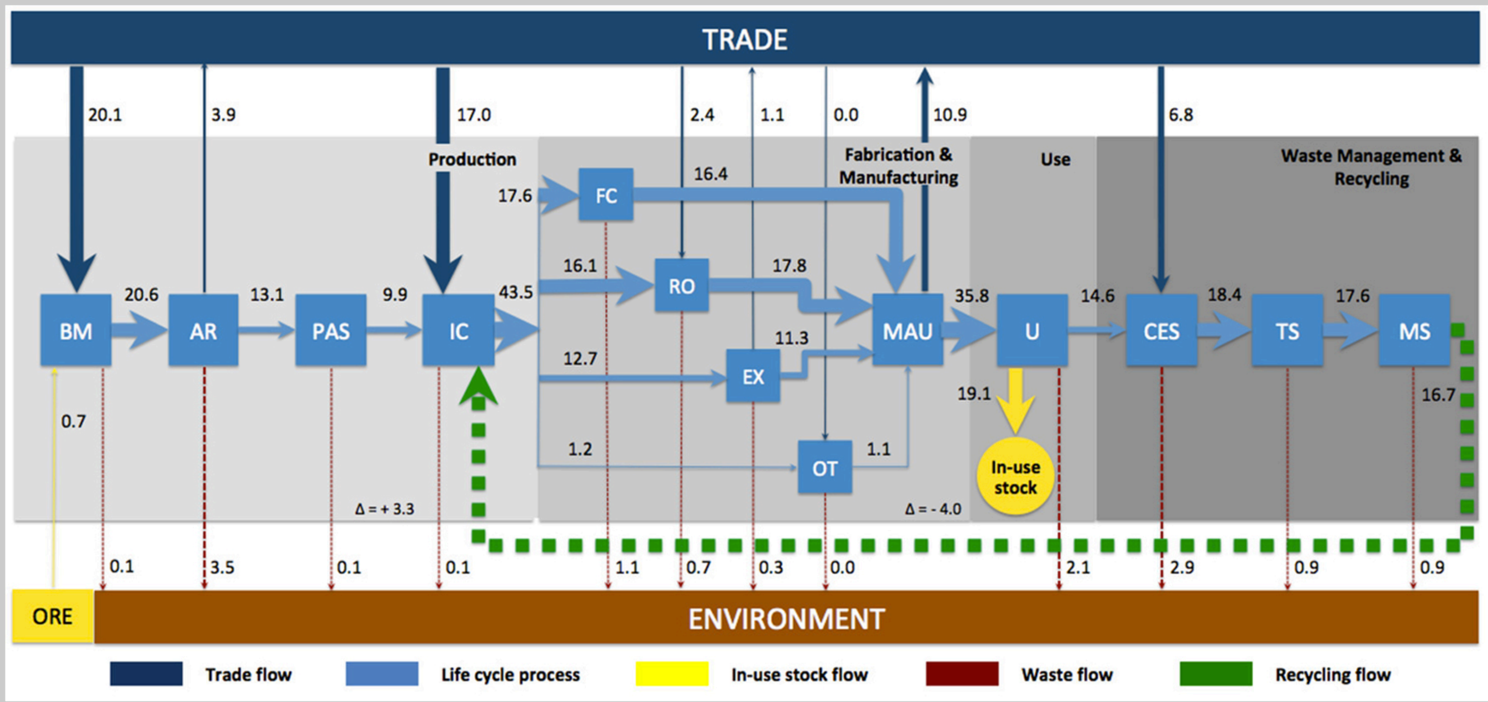
U stage includes only dissipative use of aluminium for deoxidation purpose in the steel industry.

N.a. – not applicable.

3.2 Results and discussion

3.2.1 Flows analysis

Figure 6 – Aluminium life cycle in Italy: cumulative flows and stocks, years 1947-2009. Values are in Mt. Source: Ciacci et al. (2013)



Remarks:
BM: Bauxite mining; AR: Alumina refining; PAS: Primary aluminium smelting; IC: Ingot casting; FC: Foundry casting; RO: Rolling mill; EX: Extrusion; OT: Other; MAU: Manufacturing; U: Usage; CES: Collection of scrap; TS: Treatment of scrap; MS: Melting of scrap.

Figure 6 shows the cumulative aluminium life cycle in Italy over years 1947-2009. Italy has negligible national production of bauxite ores and it is compelled to rely on imports of primary material (20.1 Mt). The ingot casting process is the core of the Italian aluminium cycle as it encompasses both primary and secondary metal production: flow analysis reveals Italy is a net importer of unwrought aluminium for a cumulative total of 17.0 Mt, which amounts at 40% of national aluminium production. The lack of primary aluminium supplies has enhanced the secondary production from metal scrap and waste management (16.7 Mt). In 2010, the total amount of processed aluminium scrap within the country exceeded 800,000 tons, whereof only a half sourced domestically (CiAl 2010). The import/export ratio has increased and net-imports make up the third largest trade category per volume traded, after primary resources and unwrought metal (**Figure 7**).

Figure 7 – Import and export flows of aluminium over years 1962 – 2010 by life cycle stages series. Values are in metric tons. Source: Ciacci et al. (2013).

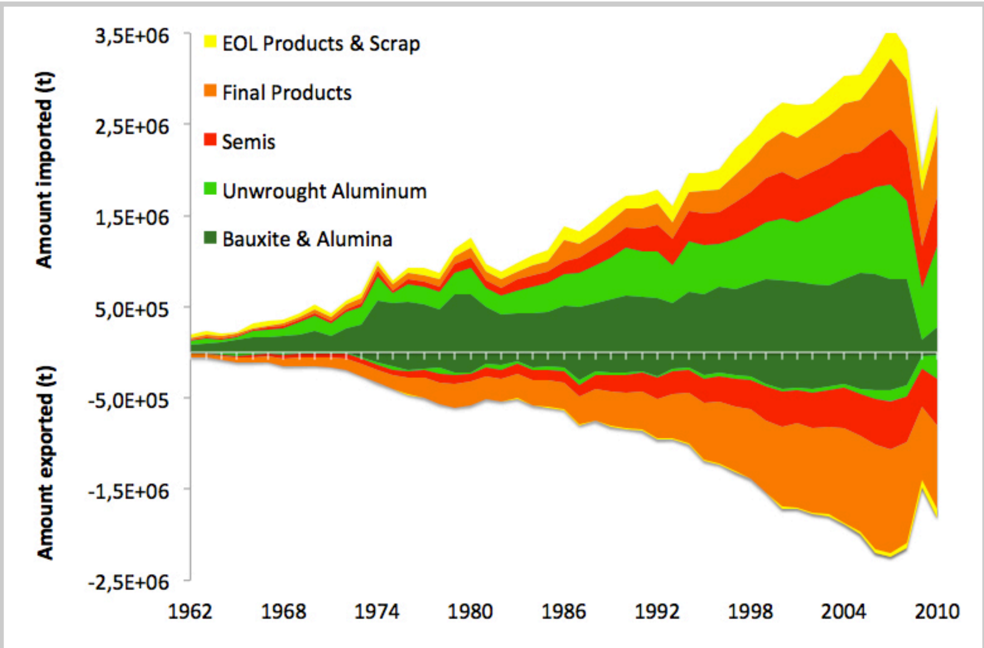
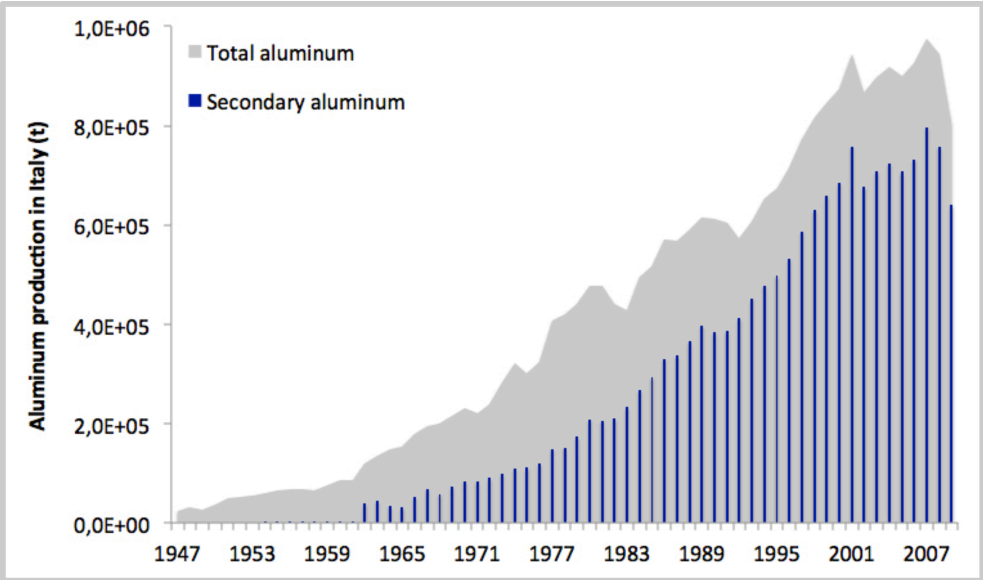


Figure 8 depicts the historical shares of secondary aluminium in total metal production. Metal recycling trend increased until last decade when a plateau at about 80% was achieved; the results are in accordance with value of 77% reported

by CiAl (2005). Considering the cumulative amounts of internal production for primary and secondary aluminium, which is a measure of import dependence, about 63% of the total metal has been recycled in Italy from 1947 to 2009.

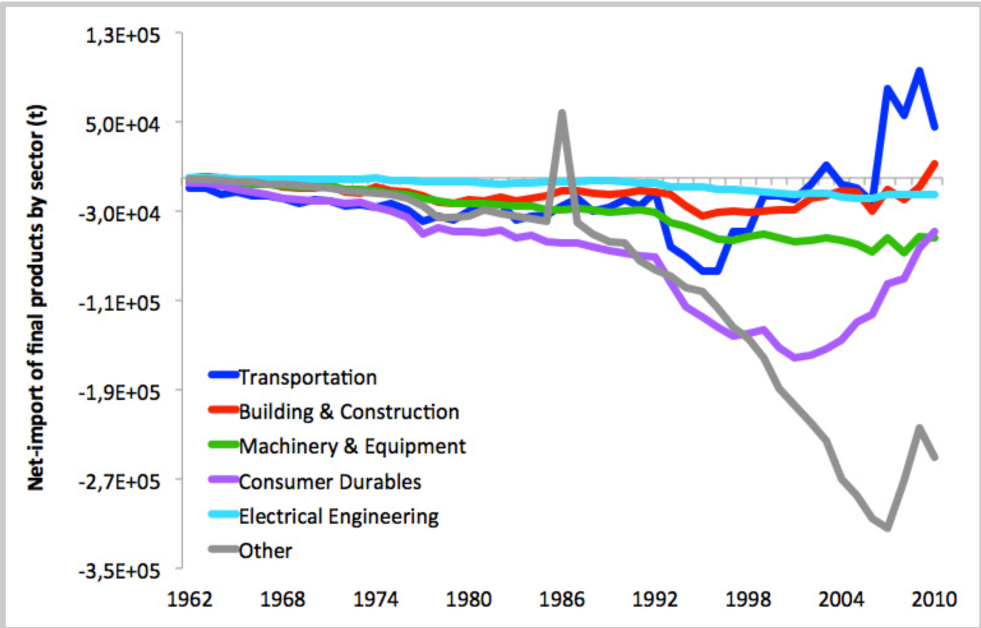
Figure 8 – Amounts of secondary (new and old scrap input) and total aluminium production in Italy. Values are in metric tons. Source: Ciacci et al. (2013).



About 44 Mt of aluminium ingots feed the fabrication stage and were disaggregated among foundry castings, mill and extruded products. The former two wrought products classes count for more than 78% of input due to the large use of recycled aluminium into the B&C and Trans sectors (CiAl 2005). 80% of final products from manufacturing enter the national market, while the remaining share is exported. This value-added feature of Italian manufacturing of aluminium-containing products has been being a typical feature of national industry since its birth during the years of economic boom (known also as 'years of economic miracle') during the 1960s (**Figure 7**).

Long-term analysis of trade in final products revealed interesting trends for the national aluminium flows (**Figure 9**). Although Italy has a long tradition as net exporter of road vehicles, the trend reversed in 2007, and at the same time, drastically increased the export of 'articles of aluminium', resulting in the most important for absolute amount traded: since this class includes trades of nails, screws, bolts, rivets, cotters, grill, netting and fencing of aluminium wire, it seems that Italy be shifting from a production of high added value final products towards a less added one. Despite the total amount net-exported being increased recently, the shift might lead the country to compete with developing countries that typically produce similar final products but face lower costs. Whether this trend will be confirmed, a decreasing in net-export flows of aluminium for Italy in the next years might be expected.

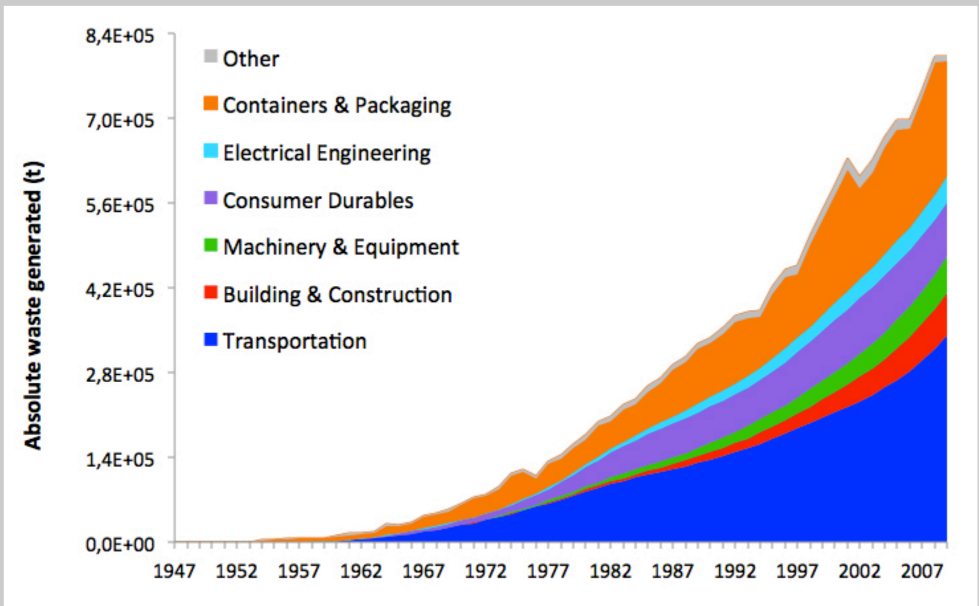
Figure 9 – Total net-import of final aluminium-containing products by end-use sector, years 1962 – 2010. Positive values express net imports, while negative ones net exports. Values are in metric tons. Source: Ciacci et al. (2013).



Aluminium scrap and obsolete products leaving the use phase amount to 14.6 Mt (or 37% of input) and the fraction is supplemented by 6.8 Mt from net-import flows. **Figure 10** shows the waste generation rate by end use sector in Italy: Trans and C&P sectors provide more than two thirds of obsolete aluminium scrap and discards, and for both about 90% of scrap generation is concentrated between 1980-2009. These results are in accordance with the situation described by McMillan et al. (2010). The ConDur, M&E, and EE sectors have similar trends that show a progressive increase in old scrap generation from about 15% to 45-

50% in last thirty years. B&C scrap has been almost negligible historically since aluminium from construction and demolition activities only started to be significant in the 1990s. In this sense, B&C scrap generation can be expected to increase in coming years due to renovation and rebuilding practices. While the recovery rate from the Transport sector is near to one hundred per cent, for C&P the fraction collected is around 50% (CiAl 2005).

Figure 10 – Generation rate by end-use sector for aluminium-containing old scrap. Values are in metric tons. Source: Ciacci et al. (2013).



Aluminium waste from building and construction, engineering sectors, and commercial activities is usually recovered from big shredding plants, such as for automotive waste (Staudinger & Koleian 2001; Kumar & Sutherland 2008; Gesing 2004; Ciacci et al. 2010), or, depending on size and quality of the fractions, they may be also collected and sent directly to foundries. Instead, recovery of aluminium waste from packaging includes processing of separate collection fractions and mixed municipal solid waste. Metal scrap is usually mixed with other materials suitable for recycling after concentration steps: depending on local waste management, aluminium can be discarded with other metals only, or metals, glass and plastics ('multi-materials heavy collection'), or metals and plastics ('multi-materials light collection'). Aluminium is sorted and separated in pre-treatment plants before being sent to foundries: about 60% commonly use eddy currents (ECs) technology (CiAl 2010), while remaining separation of metal scrap is done manually or by alternative sorting systems depending on physical properties and contamination of impurities (e.g., air separation, float/sink separation, spectrographic technologies), (Gaustad et al. 2012). Aluminium scrap that is not separately collected ends up in general MSW, disposed of through landfilling or incineration, in some cases following mechanical-biological treatments (ISPRA 2011). From ISPRA statistics (2011), over the last decade about 45% of aluminium containers and packaging have been collected for recycling plus 6.5% treated for energy recovery.

About 90% of aluminium containers and packaging are used in the food industry to produce cans for drinks (60%) and boxes, trays, and tubes for carrying solid food (40%), (CiAl 2010): this fraction has the shortest lifetime and a full recovery should be achieved to help satisfy domestic secondary material. Prevention is one of the key lever to act: one a side, aiming to improve and enhance the social awareness towards correct separate collection practices, and, on another, to claim and support integration of ecodesign and design for environment principles within the production stage. Indeed, although lightweighting of certain products (e.g. cars), for less material usage and lower energy inputs, seems already among the main goals of producers, simplifying the joining connections and limiting the number of materials used for composites are not yet, but they may help to fulfil a close-loop recycling for aluminium. At the same time, national and industrial measures should be taken to drastically reduce the amount of untreated waste to landfills, and to strengthen separating efficiencies in pre-treatment plants.

The importance to close the loop for the aluminium cycle is underlined by the current interest to treat Waste-to-Energy (WtE) bottom ash for aluminium removal, in order to obtain an aluminium-free inert fraction suitable for concrete production. Some European countries have already adopted technologies oriented to aluminium separation from ash, while Italy treats only 20% of total ash (Crillesen & Skaarup 2006). Aluminium recovery from conventional ash is commonly limited to 50% due to significant losses by oxidation; however,

advanced and innovative technologies may lead to higher efficiencies (70%-80%), (CiAl-DIIAR 2010).
Finally, in terms of material losses, the model indicates that alumina refining is the process responsible for the largest amount, followed by collection of scrap and the use of aluminium in the steel industry mainly for deoxidation purpose (**Figure 6**). However, the latter is probably destined to slow in Italy as the national steel industry has already achieved a production plateau of around 8-9 t per capita (Müller et al. 2011). Alumina refining losses are due to the leaching residue, generally called 'red mud'. EU countries, thanks to higher ore grades, generally process lower amounts of bauxite per ton of alumina produced than the other world countries: European red mud production is about 700 kg/t alumina compared to 1,140 kg/t of worldwide average (EAA 2008).

3.2.2 Stocks analysis

Natural stocks of bauxite ores in Italy were found in Sardinia and Puglia regions, although USGS statistics reported more than 85% of bauxite extraction occurred until years 1970s. Currently no significant amounts are extracted in the country, and last 16-years of statistics on bauxite mine reserves by USGS do not mention Italy in the top ten countries that together contain more than four fifth of world reserves (USGS various years). Recent explorations estimated about 7 Mt of bauxite resources at the Olmedo mine (Sardinia), with an average 65% of aluminium content (Mameli et al. 2007). However, the underground mining operations required make this deposit economically infeasible to exploit currently, depending on several variables as cost of drilling, global ore prices, and potential for flooding. This leaves the in-use stock as the major domestic resource that can help Italy to meet its long-term metal demand.

Figure 11 – Absolute by end-use sector (left hand axis) and per capita (right hand axis) in-use stocks evolution. Source: Ciacci et al. (2013).

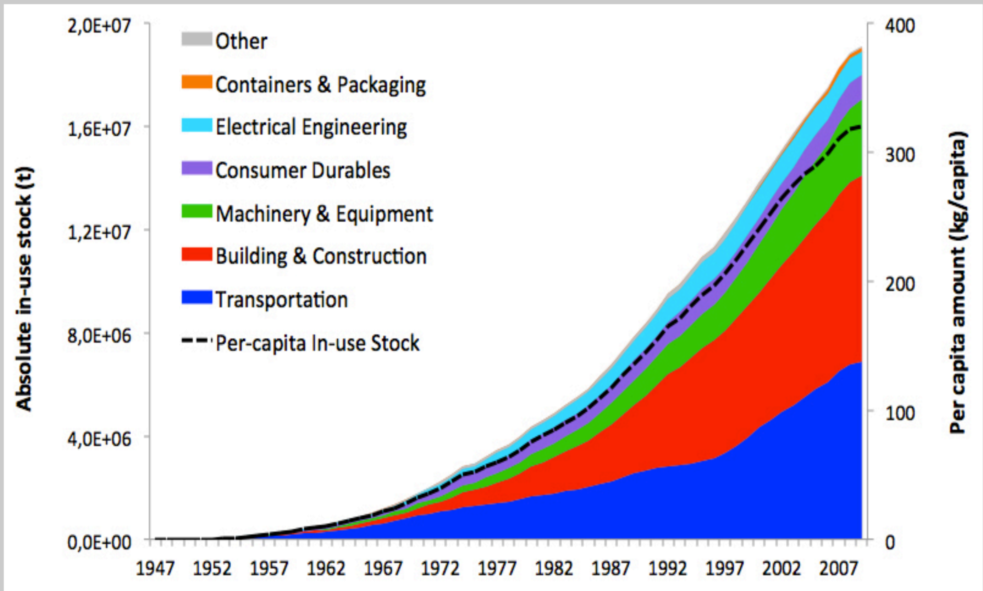


Figure 11 shows the results of absolute in-use stock, with EOL scrap generation as the other face of the same coin. A comparison may help to illuminate the importance of recycling activities. Absolute aluminium in-use stock amounts to about 20 Mt, or 320 kg per capita in 2009. This cumulative in-use stock represents approximately 11 years of supply at current rates: comparing the cumulative in-use stock with last ten-years average flow inputs the use phase (19.1 Mt versus 1.7 Mt/year), and considering the metal in-use stock is bigger than the cumulative aluminium recycled over the past 62 years, it can be inferred how this anthropogenic reservoir be an exploitable source for potential recycling in the future. The main pool of aluminium embedded in the B&C sector (7.2 Mt), followed by Trans (6.9 Mt). Among the remaining end-use sectors, engineering applications play the major role: M&E, EE and ConDur contribute with 4.8 Mt of aluminium. C&P and the Other markets are negligible as in-use stock amount is about 1 Mt.

The results are in line with the per capita stocks over similar time span: Chen & Shi (2012) estimated 60 kg per-capita for China (60 kg), while Liu et al. (2011) and Chen & Graedel (2012a) 490 kg for the United States. The in-use stock results for

Italy have a reasonable proximity to reported values for the U.S.; however, it is important to consider possible reasons for lower level of per capita aluminium in-use stock (**Table 5**). One obvious area to explore is the structure of end-use demand. Sectors with longer lifetimes contribute over a longer period to per capita stocks than shorter-lived sectors. Comparing Italian rates with European and World averages, proportionally the country uses higher amounts of rolled and extruded aluminium-containing products in the B&C sector (about 48% versus 43%), but significantly lower ones in EE (2% versus of 4%), Trans (on average, 12% against 26%), and M&E (10% versus 14%). In particular, the EE sector involves aluminium as material for cables in the electricity distribution network, and the decreasing amount of aluminium applied to the sector over the last decade (ASSOMET several years) can be interpreted as a decrease in the replacement of obsolete power lines.

Considering the Trans sector, aluminium content in Italian vehicles has been estimated to range between 3% and 9% (Amicarelli et al. 2004; Santini et al. 2011), with an average of 5.7%. This rate is slightly lower than those reported for the U.S. and Japanese vehicles, which generally amount at about 6.3% (Fuse et al. 2007; Staudinger & Koleian 2001), or even more (Kumar & Sutherland 2008). These data are in accordance with international reports (IAI 2009; ATG 2008) that report aluminium content per vehicle traditionally higher in North American vehicles than European ones. Moreover, transportation is the only sector with significant levels of in-use stock and old scrap generation (**Figure 10** and **Figure 11**). There are several possible reasons for this: (i) vehicles, especially automobiles, are widely recognized as a symbol of economic wealth and social status and they are widely spread in most developed countries; (ii) relatively shorter lifetimes relative to B&C and engineering applications, which guarantees an early return of the metal into flows to recycling; and (iii) increases in the aluminium content of new vehicle production as consequence of policies that aim at vehicle lightweighting for emissions reduction and energy savings (Gesing 2004; GHK/BioIS 2006; Passarini et al. 2012). Whether in past years mainly castings aluminium-containing products were used for transportation vehicles, today aluminium covers a wide range of applications that move from sheet components to car body structure itself (Liu et al.

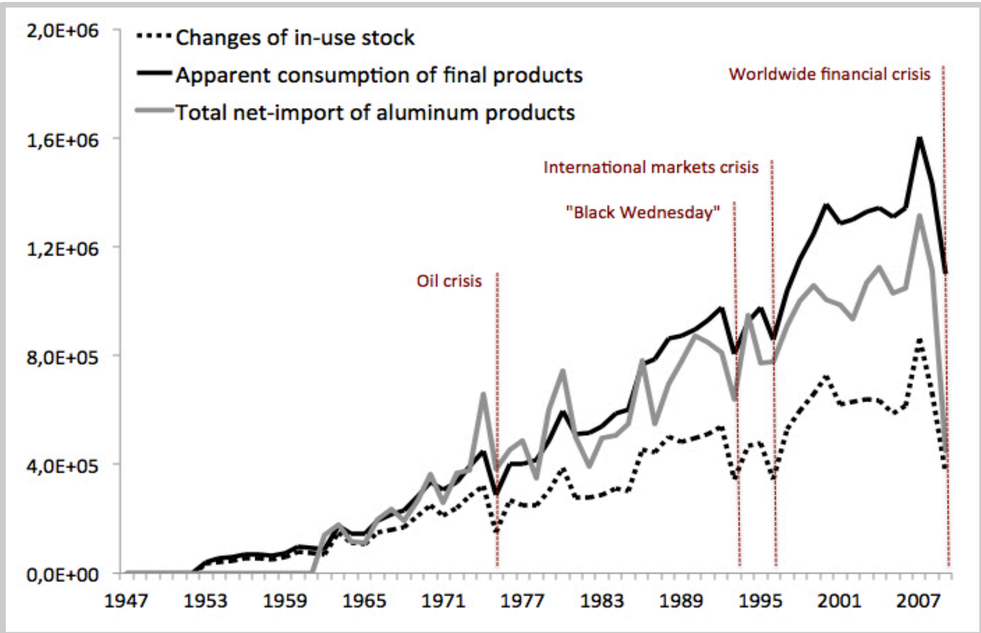
2011; Bertram et al. 2009; Kim et al 2011). Finally, (iv) car recycling is a profitable industry and EOL vehicles are subjected to defined legislations that intensify metal recovery practices.

Beside natural and in-use stocks, metal losses to the environment have implications for recycling; aluminium losses to the environment include both dissipated and deposited stocks (Chen et al. 2010). The former includes flows to the environment generally due to irreversible changes in metal properties that happens consequently to transformation along the metal life cycle as oxidative and corrosive phenomena. Deposited stocks instead consist of material accumulation that embeds metal form that may be exploited in the future, such as residues and tailings during mining operations, deposits or landfilled EOL products. These deposited stocks may constitute an alternate secondary resource reservoir. For the scope of the study, we considered deposited losses only those occurring from bauxite mining, alumina refining, collection and treatment of scrap: cumulative amount was estimated at about 7.5 Mt versus 6 Mt of dissipated losses.

Figure 12 includes the yearly net-addition to stock, which is calculated as the difference between domestic production plus net-imports and scrap generated. A constant accumulation of aluminium-containing products in the country is displayed. According to Brunner & Rechberger (2004) this accumulation is a typical metabolism of developed countries, which tend to increase materials inflows and in-use reservoirs in contrast with non-developed ones that largely export raw materials.

The resulting trend is a progressive accumulation of aluminium with variations primarily due to economic conditions. In accordance with Tilton (1990) metal demand is highly correlated with economic cyclical trends, and contractions in the growth rate appear periodically. The cumulative trends depict declines whose cause can be identified with historical financial crises: in the year 1973 was the oil crisis, with a subsequent stock market crash over a two-year period; in years 1992-1993 speculative attacks in the *Exchange Rate Mechanism* occurred in Europe (also known as 'Black Wednesday'); in 1993-1996 some international markets crisis were registered; and finally, a deep decrease in 2009 as consequence of the latest worldwide financial crisis. In particular, year 1973 was the turning point for some metals consumption in several OECD countries, and since then aluminium experienced the most rapid growth during the previous decade (10% annually), the decline in growth was more severe after 1973 (less than 1%), even though its consumption continued to expand (Tilton 1990).

Figure 12 – Net addition to stock (change in in-use stock), apparent consumption of final products, and total net-import of aluminium products. Values are in metric tons. Source: Ciacci et al. (2013).



3.2.3 Sensitivity analysis results

The sensitivity analysis results do not reveal any significant model sensitivity and therefore suggest robust results (**Table 7**). The strongest variability results are in the Transportation sector (**Figure 13**), and they are mainly due to the large share of aluminium-containing products partitioned to this sector. A detailed disaggregation among Transportation sub-categories (e.g., cars, light trucks, heavy trucks, train, air), each with associated lifetime distributions and aluminium concentrations, may help to decrease model sensitivity.

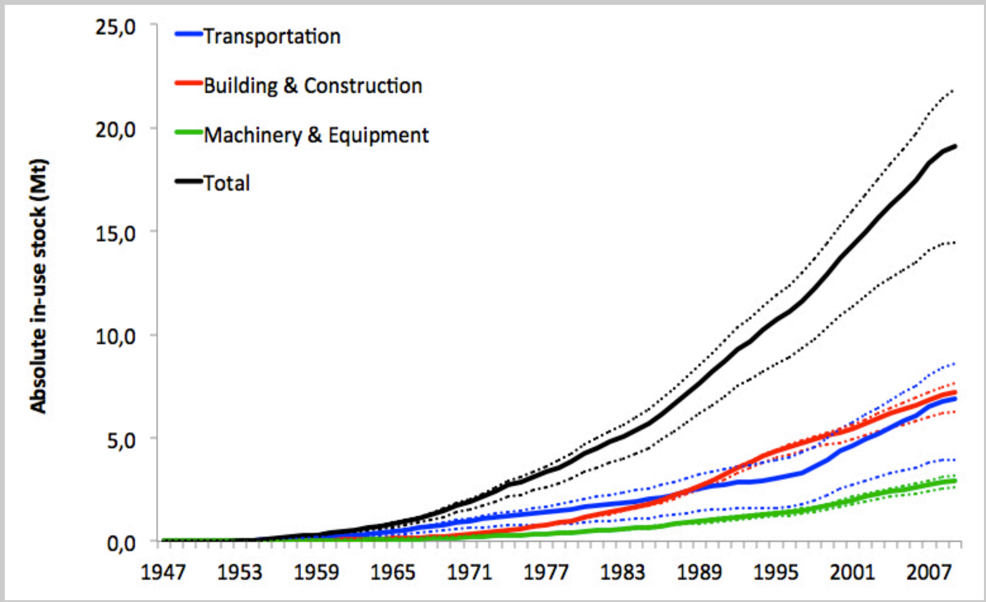
Table 7 – Cumulative results of the sensitivity analysis. Discussion was performed using the mean values only. Values are in Mt. Source: Ciacci et al. (2013).

Life cycle	Sensitivity	Production	Loss	Input	Net-import	Consumption
BM	***mean***	***0.7***	***0.1***	***0.8***	***20.1***	***20.6***
	high	0.7	0.1	0.8	23.0	20.6
	low	0.7	0.1	0.8	17.2	20.6
AR	***mean***	***17.1***	***3.5***	***20.6***	***-3.9***	***13.1***
	high	17.1	3.5	20.6	-4.0	13.1
	low	17.1	3.5	20.6	-3.9	13.2
PAS	***mean***	***9.9***	***0.1***	***10.0***	***0.0***	***9.9***
	high	9.9	0.1	10.0	0.0	9.9
	low	9.9	0.1	10.0	0.0	9.9
IC*	***mean***	***26.5***	***0.1***	***27.5***	***17.0***	***43.5***
	high	24.6	0.1	25.5	17.1	41.8
	low	29.8	0.1	30.9	16.8	46.6
FC	***mean***	***16.4***	***1.1***	***17.6***	***0.0***	***16.4***
	high	16.4	1.1	17.6	0.0	16.4
	low	16.4	1.1	17.6	0.0	16.4
RO	***mean***	***15.4***	***0.7***	***16.1***	***2.4***	***17.8***
	high	15.4	0.7	16.1	2.4	17.8
	low	15.4	0.7	16.1	2.4	17.8
EX	***mean***	***12.4***	***0.3***	***12.7***	***-1.1***	***11.3***
	high	12.4	0.3	12.7	-1.1	11.3
	low	12.4	0.3	12.7	-1.1	11.3
OT	***mean***	***1.1***	***0.0***	***1.2***	***0.0***	***1.1***
	high	1.1	0.0	1.2	0.0	1.1
	low	1.1	0.0	1.2	0.0	1.1
MAU	***mean***	***46.7***	***0.0***	***46.7***	***-10.9***	***35.8***
	high	46.7	0.0	46.7	-11.2	35.5
	low	46.7	0.0	46.7	-10.6	36.1
CES	***mean***	***11.7***	***2.9***	***14.6***	***6.8***	***18.5***
	high	9.2	2.3	11.5	7.2	16.4
	low	15.6	3.9	19.5	6.5	22.1
TS	***mean***	***17.6***	***0.9***	***18.5***	***0.0***	***17.6***
	high	15.6	0.8	16.4	0.0	15.6
	low	21.0	1.1	22.1	0.0	21.0
MS	***mean***	***16.7***	***0.9***	***17.6***	***0.0***	***16.7***
	high	14.8	0.8	15.6	0.0	14.8
	low	20.0	1.1	21.0	0.0	20.0

		Scrap	Loss	Input	Stock
U	***mean***	***14.6***	***2.1***	***35.8***	***19.1***
	high	11.5	2.1	35.5	21.8
	low	19.5	2.1	36.1	14.5

*IC includes input of secondary aluminium from remelting of scrap.

Figure 13 – Absolute in-use stocks: cumulative and main end use sectors detail. Dashed lines delimit variability ranges resulting from the sensitivity analysis performed. Source: Ciacci et al. (2013).



3.3 Final considerations

The dynamic MFA model applied here is the first attempt to describe the anthropogenic aluminium cycle in Italy over time. The analysis revealed high potential to enhance recycling activities for the country, based on dynamic quantification in-use stocks that will supply secondary resources. About 90% of the anthropogenic aluminium reservoir is embedded in the transportation sector, building and construction, and machinery and equipment; recovery and recycling initiatives should hence focus on these markets. Moreover, collection of scrap is a key-issue in which Italy could act to decrease metal losses and consequently to increase the old scrap pool suitable for secondary production. Increasing support to separate collection and initiatives oriented to aluminium recovery specifically would allow Italy to increase its reliance on domestic material, and may also allow a decline of net-import of unwrought aluminium. This will be important to Italian industry and consumers, as on a worldwide scale, aluminium demand is expected to triple at least by 2050 (IAI 2009; Liu et al. 2011).

Assumptions and uncertainties are inherent in the results, and many variables play a role in developing scenarios for future trends for metal demand, such as variations in the aluminium content, demographic growth, and the trade in aluminium-containing products. Therefore, findings should not be extrapolated to other materials due to structural differences that usually characterized distinct materials life cycles in human society.

Finally, the dynamic MFA results here presented constituted a basis for integrating LCA methodology: in the next Chapter an environmental analysis related with aluminium flows and stocks is performed to consider energy consumption and carbon emissions across the metal cycle.

4.

GHG emissions embodied in the Italian aluminium

In this Chapter an evaluation of the carbon footprint evolution embodied in the Italian aluminium has been carried out. As described in the Chapter 2, GWP_{100} indicators were linked with national-level emissions associated with MFA results for aluminium flows and in-use stocks from bauxite mining, alumina refining, primary aluminium smelting and ingot casting. Cumulative carbon dioxide emission values from primary and electrical energy, process and the transportation were estimated per ton of aluminium produced over years 1960 to 2009; consolidation of the model was carried out at domestic (national) and foreign (import from world partners) levels for aluminium production. A discussion about how the main factors, according to the *Kaya Identity* equation, they did influence the Italian GHG emissions pattern over time, and which are the levers to mitigate it, was also included. Data inventories and discussion of the results are followed reported. Supporting materials is presented in Appendix B.

4.1 Data collection

4.1.1 Sources of data

Sources investigated at national level were the *Istituto Superiore per la Protezione e la Ricerca Ambientale* (ISPRA 2012) for records of GHGs emission from primary and electrical energy, and process inputs; TERNA (2010) for statistics of electricity generation and distribution. Expert elicitation allowed filling the gap regarding the historical evolution of power sources for electricity usage by the Italian smelters. At worldwide level, sources of data included statistics from the IAI (2012), EAA (2008), IEA (2010) over years 1980 to 2009. Complementary information was also assumed from McMillan (2009), McMillan & Koleian (2009), Leroy (2009), and Liu et al. (2011).

Environmental loads associated to bauxite were assessed assuming aluminium production as unique use of the mineral. Despite bauxite is also used as refractory material in the construction sector, its primary appliance is as source of aluminium and such as carbon emissions were 100% allocated to the metal, i.e. the accounting method was performed considering the whole mass of bauxite and alumina and not just in terms of aluminium content. Similar assumptions were followed for alumina refining step. Metal content into alumina was estimated at 52% from theoretical chemical composition and 1.5% of other metal oxides, but even in this case GHG emissions were fully allocated to aluminium production.

According to the scope of the study, we scanned the exporting partners of Italy at a world region level. The UNCTS Database (UNCTSD 2012) was consulted in order to identify, over years 1960 to 2009, from which world regions Italy imported primary forms of aluminium as bauxite ore and alumina, and unwrought aluminium from primary aluminium smelting. The investigation revealed that Italy imported the major fraction of bauxite ore from Australian mines, and the remaining portion from Guinea. For imports of aluminium hydroxide, European countries (mainly France and Germany) were the main providers for Italy in recent decades; prior to this, the percentage from European sources decreases by about 50% as almost half of alumina imports were from Africa, North America and

Oceania. Unwrought aluminium flows into Italy were also primarily from European countries (besides the already cited France and Germany, also the Netherlands), although complementary world regions included North America before 1980s and Africa (primarily Mozambique) in the last decade.

The distribution of import sources for aluminium-containing commodities was used to model the carbon profile from metal production outside Italian boundaries. Briefly, the parameters sensitive to geographical macro-area differences were consumption of primary energy by fuel type, electrical power mix generated by source and relative consumptions, power transmission and distribution losses, and distances covered during transportation from exporting countries to Italy. Finally, a linear interpolation was applied to estimate intermediate values between certain years.

4.1.2 Inventory and assumptions

4.1.2.1 Primary energy

Several sources in literature report fuel types and consumptions at life cycle stages (Leroy 2009; IAI 2012; EAA 2008). Inventory of fuel types is restricted to fossil fuels only, including oil, coal, diesel, natural gas and propane gas. Quantification of carbon footprint per ton of bauxite, alumina and aluminium produced were calculated accordingly to the equation **A8**. Quantities for $\mathrm{FC}_{i,j}$ in BM, AR, PAS and IC were modelled according to IAI (2012) and EAA (2008) and Leroy (2009). Fuel consumption was assumed to be independent from domestic or foreign production, while different values for GHGs emissions were used: ISPRA (2012) reported yearly GHGs emissions from fuel combustion by non-ferrous metals sector. Instead, $\mathrm{EF}_{P,j}$ for foreign production were calculated assuming values reported by Liu et al. (2011) for coal, oil, diesel, natural and propane gases.

4.1.2.2 Electrical energy

Domestic production

Historical percentage distributions by source of the Italian electricity grid mix was estimated according to TERNA data. Such values cover only electrical power produced within the country, leaving out import by foreign countries. Anyway, over years Italy has become a great importer of electricity from foreign countries (mainly France), requiring in recent years about 14% of gross electrical energy consumed. Overall, the emission factors from electricity generation including imports from France are on average 7% lower than the Italian average, due to France's energy policy reliance on nuclear plants and on the limiting exploitation of thermal fuels at less than 10%; thus, France has a 2007-2009 average at GHG emissions factor of 89 g CO_2/kWh compared with 375 g CO_2/kWh for Italy. Aiming to properly quantify the GHG emissions from electricity, we included the import from France net of transmission and distribution losses.

The grid electricity model detailed above was applied to bauxite mining, alumina refining and ingots castings stages, while for primary aluminium smelting a specific electric power scenario was developed. Currently, in Italy the main country's producer of primary aluminium is Alcoa Italia S.p.A. (USGS 2009), and from a direct expert elicitation it resulted the electricity mix inputs the only one smelting plant for primary aluminium, located in Sardinia, nowadays does not differ significantly from the national grid. However, considering the scope of the study, a glance on historical trend-series requires to consider changes in the distribution for electrical power used by smelters.

Briefly, the history of primary aluminium industry within the country reflects the smelters' skill for the electricity self-production: although nowadays it is not carried out anymore, in the past decades the electricity self-generation covered part of the power consumed by the smelters, and before 1960s about the whole electricity input in primary aluminium production was self-generated by means integrated hydro-sourced plants. Over years, the production capacity of aluminium plants increased and complementary power sources were required to integrate beside the hydro one: whether thermal sources, mainly fuel oil, were chosen at first, after the oil crisis a progressive shift towards the electricity national grid mix was then preferred. As of today, this solution is the most adopted in the country.

Foreign production

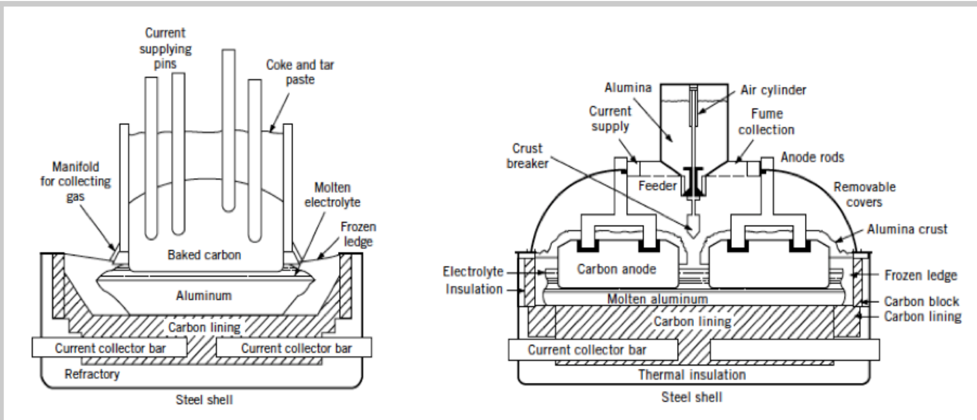
Electrical energy used in bauxite mining, alumina refining and ingot casting was modelled according to general world region production mixes: Africa, North and South America, Asia, Europe and Oceania. EIA (2010) and World Databank (2012) provided historical information of electrical power distribution sources and country transmission and distribution (T&D) losses; IAI statistics published historical data for last 30-years distribution by source of electricity power used, and electrical energy consumptions by smelters (PAS) in each macro-area (IAI 2012). Values before the year 1980 were assumed equal to the last percentage distribution. Electrical energy carbon emissions from foreign aluminium production were calculated summing each singular contribution from geographical areas determined according to equation **A9**. Consolidation was then performed for every aluminium life cycle stage by the yearly percentage distribution of exporting partners. In fact, the UNCTSD records were used to disaggregate the Italian import of aluminium-containing products for every macro-area and annually. Such allocation was followed also for the CF from the transportation, as described in the respective section below.

4.1.2.3 Process

Around 90% of total process CO_2eq emissions occur during the aluminium smelting due to fossil fuel combustion and PFCs emissions. The Bayer process for alumina refining does not produce carbon dioxide directly, but some emissions may result from calcium carbonate calcination. Most of CO_2 emissions are primary energy-related (IAI 2006). Sources of carbon dioxide emissions in PAS include electrolysis reaction of the carbon anode with alumina, and the Boudouard reaction between CO_2 and the carbon anode with following carbon monoxide

oxidation; beside direct electrolytic processes, other sources of CO_2 emissions are auxiliary processes such as the prebake anode consumption, the baking of green anodes, and the use of sodium carbonate for scrubbing process (IAI 2006).
When anode effect events do occur, the use of cryolite (Na_3AlF_6) in primary aluminium production causes emission of PFCs gases tetrafluoromethane and hexafluoroethane, which are characterized by GWP_{100} factors of 6,500 and 9,200 kg CO_2eq per kg of gas respectively. Anode effect is a failure process condition generated by a low level of alumina content within the electrolytic pots. That insufficient amount of Al_2O_3 determines a consumption of the electrolytic bath, resulting in the consequently emissions of PFCs species. The anode effect causes a rapid increase in voltage in the cell, which overcomes the normal operating values. A direct relationship does exist between time duration of the anode effect and PFCs release from the smelter (IAI 2010). From the IAI 2010 survey, the Prebake technology may currently range between less than 1 to about 4 tCO_2eq/t Al depending on the alumina feed configuration: manual side feed is the worst one, while the point centre alumina feed seems to guarantee the best performance; the bar broken centre feed configuration shows PFCs emission rates similar to Söderberg technologies, at about 1 tCO_2eq/t Al (IAI 2010). About 90% of primary aluminium produced in Europe follows the Prebake technology, with the remaining 10% utilizing Söderberg anodes (Leroy 2009). In **Figure 14** is shown a general electrolyzing cell with Prebaked and Söderberg anodes, respectively.

Figure 14 – General electrolyzing cells with Prebaked (left) and Söderberg (right) anodes. Source: Sanders (2002).



Due to their relatively high GWP factors and IPCC recommendations for cutting global GHG emissions, initiatives for PFCs emissions reduction were adopted over last two decades, and leading to about 75% decrease of total emissions from primary aluminium production, with a PFCs emissions reduction to about 0.6 kg CO_2eq/kg Al in 2009 (IAI 2011). Despite such an improvement and considering the IAI voluntary 50% reduction of GHG emissions by 2020 on 2006-year basis, Liu et al. (2011) described how further reductions in the coming years could be challenging if new technologies are not discovered and the demand for primary metal grows at current dramatic rates (Liu et al. 2011).

4.1.2.4 Transportation

Assumed values for transportation are reported in **Table 8**. Emission factors in tCO_2eq per t*km were obtained from the Ecoinvent database 2.2. Average fuel efficiency of different transport modes from 1985 to 2009 was collected from Eurostat (2012), whilst a linear extrapolation estimated older years. The equation **A11** was applied for the estimation of the results.

Table 8 – Average distances assumed for trade flows from world region to Italy.

World macro-region	Distance (km)
Europe	1000
North America	7000
South America	8500
Africa	6000
Oceania	8500
Asia	8000

4.1.3 Limits and sensitivity of the study

The study considers primary aluminium production using a 'cradle-to-gate' perspective, which is by definition a partial view of the whole metal life cycle (ISO 14040). LCA studies with a cradle-to-gate approach are common in the metal industry (Norgate et al. 2007; Ramakrishnan & Koltun 2004), mainly due to the focus on metal production and the energy and emissions intensity required to transform mineral concentrates into metals (Norgate et al. 2007; Liu & Müller 2012).

Extending the system boundaries to cover all life cycle stages is not a straightforward task; for example the use phase is commonly omitted because of allocating procedures to properly assess the environmental costs and benefits from multi-material appliances that contain aluminium. In their review, Liu & Müller (2012) pointed out how such a concern is a current question, and efforts of the LCA community should be addressed towards standardized and harmonized methods.

The choice to focus on GHG emissions only may obscure important trends for other categories of environmental impact; however, such a decision is justified by recognizing the midpoint indicator for climate change is among the most common metrics used to assess environmental performance by the aluminium industry itself (Liu & Müller 2012), and by indicating them as a current link for connecting sustainability concerns from environmental, economical and social perspectives. On the other hand, historical data series for detailed input and output inventories of the national aluminium industry are often missing, and thus consequently limited the study to GHG emissions on a more practical level.

The robustness of the model used for the investigation was tested by a sensitivity analysis. Emission factors for the CF were considered the main source for uncertainties as data for the Italian aluminium flows and stocks have been checked in Chapter 3 and in (Ciacci et al. 2013). Here, an uncertainty range of 15% was applied to each GHG-related process, and the model was run using alternatively upper and lower values. The sensitivity results, attesting a good reliability of the model, are displayed in the **Figure 18**.

4.2 Results and discussion

Tables 9-14 list the average carbon profile values for the domestic and foreign aluminium production at each cycle stages. Due to the importance of PAS in the context of life cycle carbon profile, this stage is where most efforts to decrease the climate change effects of primary aluminium production were concentrated: CO_2eq emissions reduction led to a maximum cut up to 44% for the Italian metal production, and 48% for the international one respect to the most impacting decades, i.e. 1980-1989 and 1960-1969, respectively.
The national trend for PAS carbon dioxide emissions shows an increase until the end of 1980s, as a consequence of the shift towards thermal sources for electrical energy fed to the Italian smelters, as discussed previously. From the beginning of 1990s, instead, carbon emissions from power usage decreased due to (i) phasing out fuel oil consumption for electricity generation, which started to include renewable sources too, and (ii) initiatives oriented to a strong PFCs emission reduction (IAI 2012). Outside of Italy, trends for imported production in the PAS stage depended on shifts in the world partners providing aluminium-containing products to the country: the preference for European states to the detriment of extra-European ones, with the former characterized by a less dependence from thermal fuels and a shorter distance for transportation, seems to have resulted in a better environmental performance for primary aluminium production than in other regions (Liu & Müller 2012).
The ecoinvent database 2.1 process *'Aluminium, primary, at plant/RER U'* determined a CF result of 12.4 t CO_2eq/t aluminium over the years 1995-2002, which is comparable but higher than the 10.2 t CO_2eq/t result found in this study for the domestic production of the same years. The average percentage contribution at each stage resulting from the main carbon-related processes is displayed in **Tables 9-12** and **Figure 15**; decadal average contributions are displayed over the years 1960-2009. Overall, the results reflect the trend of the primary aluminium industry in upgrading to standardized production procedures worldwide, and they confirm how the sustainability challenge are faced by all the nations and industrial actors involved (IAI 2012). On a percentage basis, the primary energy consumption

constitutes the largest source for CO_2eq emissions, particularly during ore mining operations, alumina refining process and casting of metal ingots. Electrical energy use contributes more than half of the global warming potential during the PAS, which is the most carbon-intensive stage (**Table 13**). The remaining portion is covered by direct process emissions in the forms of PFCs, a result that justifies initiatives aimed at a reduction in emissions. **Table 14** reports the cumulative results for domestic and foreign CO_2eq emission factors by decades along the aluminium cradle-to-gate.

Figure 15 – Percentage distribution among the four CO_2-related processes at every life cycle stage. The detail level includes a comparison in terms of national primary aluminium production (Domestic production), and primary aluminium produced from world partners and imported by Italy (Foreign production).

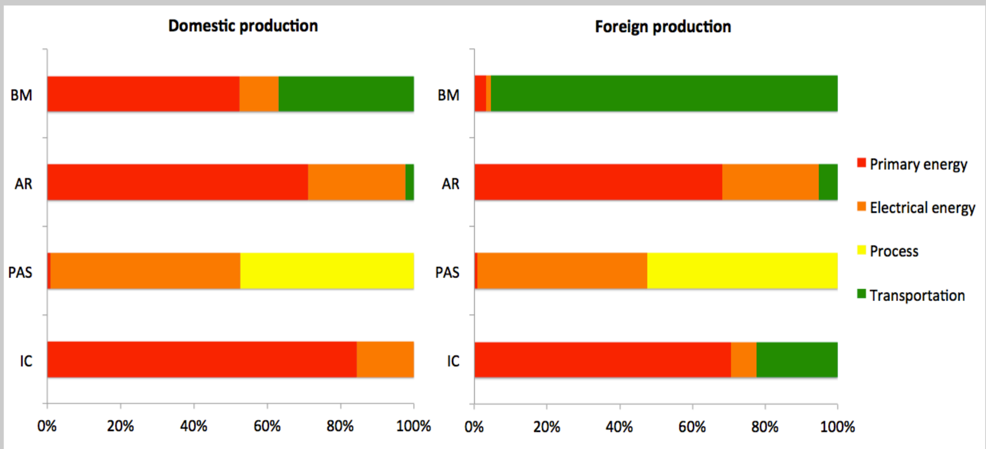


Table 9 – Average CO_2eq emission factors from primary energy at domestic and foreign level of detail.

	Primary energy kgCO_2eq/t of bauxite, alumina, aluminium							
	Domestic				Foreign			
Years	**BM**	**AR**	**PAS**	**IC**	**BM**	**AR**	**PAS**	**IC**
1960 – 1969	3.88	681	95.0	76.2	3.86	650	90.7	182
1970 – 1979	3.88	681	95.0	76.2	3.86	650	90.7	182
1980 – 1989	3.88	682	95.0	76.2	3.86	674	90.7	182
1990 – 1999	3.87	713	94.6	74.3	3.86	749	89.5	172
2000 – 2009	4.00	769	87.1	75.4	3.86	769	80.0	94.1

Table 10 – Average CO_2eq emission factors from electrical energy at domestic and foreign level of detail.

	Electrical energy kgCO_2eq/t of bauxite, alumina, aluminium							
	Domestic				Foreign			
Years	**BM**	**AR**	**PAS**	**IC**	**BM**	**AR**	**PAS**	**IC**
1960 – 1969	0.54	212	3190	4.58	1.74	324	5740	7.82
1970 – 1979	0.88	306	5360	7.38	1.75	304	5260	8.55
1980 – 1989	0.93	291	6660	7.81	1.77	267	5160	7.81
1990 – 1999	0.86	240	6590	7.91	1.77	225	3930	7.41
2000 – 2009	0.79	271	6110	42.0	1.62	240	5670	50.1

Table 11 – Average CO_2eq emission factors from process PAS at domestic and foreign level of detail.

	Process kgCO_2eq/t of bauxite, alumina, aluminium							
	Domestic				Foreign			
Years	**BM**	**AR**	**PAS**	**IC**	**BM**	**AR**	**PAS**	**IC**
1960 – 1969	-	-	7250	-	-	-	7250	-
1970 – 1979	-	-	7250	-	-	-	7250	-
1980 – 1989	-	-	7250	-	-	-	7250	-
1990 – 1999	-	-	2840	-	-	-	6160	-
2000 – 2009	-	-	983	-	-	-	1060	-

Table 12 – Average CO_2eq emission factors from the transportation at domestic and foreign level of detail.

	Transportation kgCO_2eq/t of bauxite, alumina, aluminium							
	Domestic				Foreign			
Years	**BM**	**AR**	**PAS**	**IC**	**BM**	**AR**	**PAS**	**IC**
1960 – 1969	2.72	25.6	-	-	115	69.5	-	72.9
1970 – 1979	2.72	25.6	-	-	115	69.5	-	61.8
1980 – 1989	2.72	25.0	-	-	116	68.8	-	49.1
1990 – 1999	2.75	20.0	-	-	117	35.5	-	35.5
2000 – 2009	2.78	15.2	-	-	118	23.2	-	38.6

Table 13 – Average CO_2eq emission factors by decades: domestic and foreign primary aluminium production stages.

	kgCO_2eq/t Al							
	Domestic				Foreign			
Years	**BM**	**AR**	**PAS**	**IC**	**BM**	**AR**	**PAS**	**IC**
1960 – 1969	28.3	2120	10700	81.2	480	2410	13400	191
1970 – 1979	29.6	2340	12900	84.0	480	2360	12900	192
1980 – 1989	29.9	2310	14200	84.5	481	2330	12800	191
1990 – 1999	29.6	2250	9680	82.7	485	2330	10400	180
2000 – 2009	30.0	2440	7300	118	490	2380	6970	145

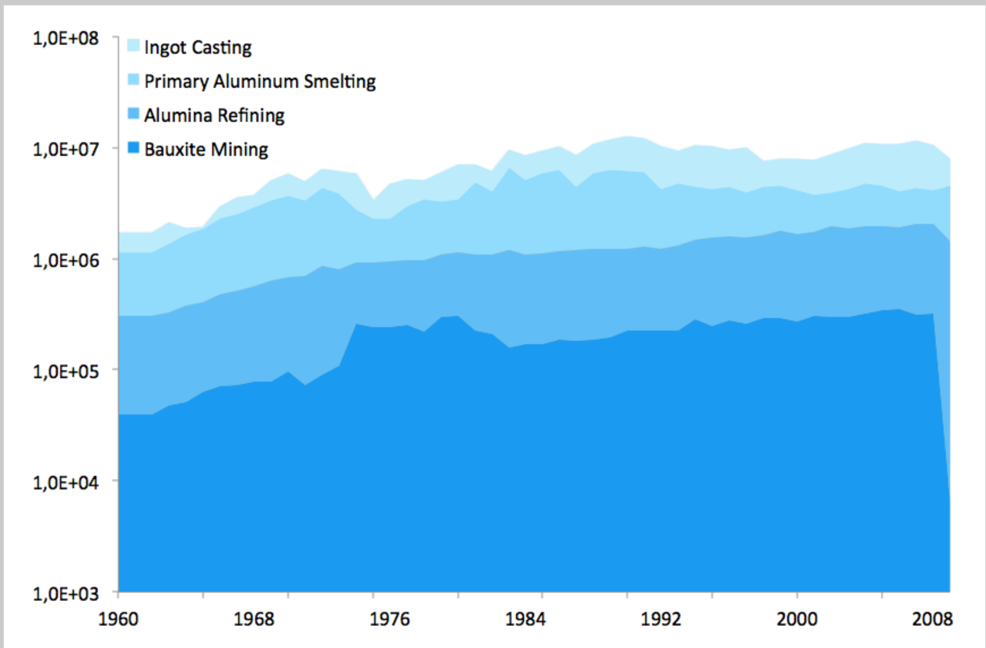
Table 14 – Cumulative results for CO_2eq emission factors by decades: domestic and foreign primary aluminium production (cradle-to gate).

	tCO_2eq/t Al	
Years	**Domestic**	**Foreign**
1960 – 1969	12.9	16.5
1970 – 1979	15.4	15.9
1980 – 1989	16.7	15.8
1990 – 1999	12.0	13.4
2000 – 2009	9.9	10.0

4.2.1 The Italian carbon profile from primary aluminium production's evolution

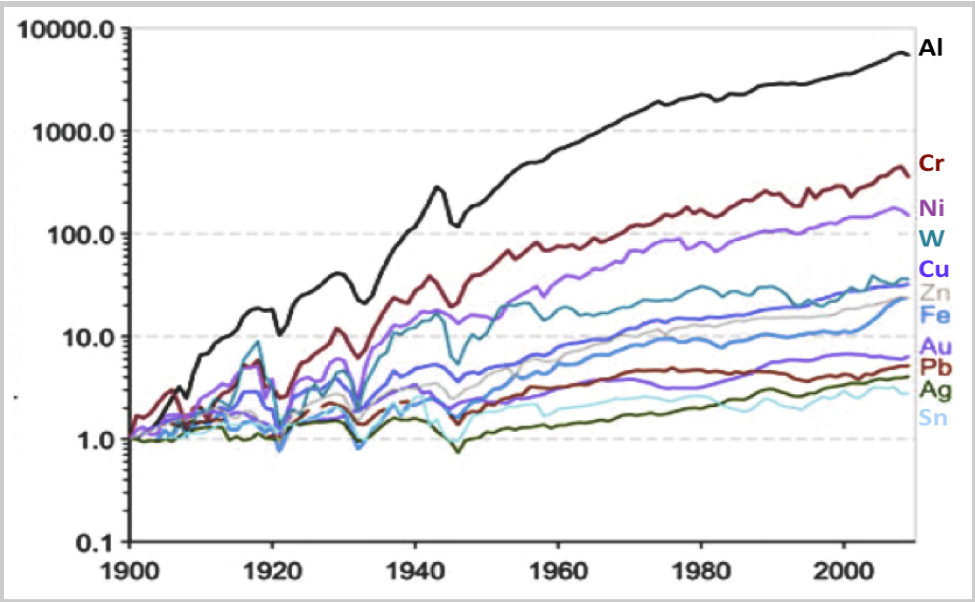
Cumulative amount of CO_2eq emissions from the Italian primary aluminium production was estimated at 129.1 Mt. **Figure 16** shows the historical evolution of GHG emissions resulting from the aluminium cradle-to-gate production in Italy. From 1960-2009, carbon dioxide emissions increased about six hundred percentage as a consequence of the nearly exponential growth in the aluminium metal industry in the last century (**Figure 17**, Johnson et al. 2007; Graedel & Erdmann 2012).

Figure 16 – The historical evolution of carbon intensity from the cradle-to-gate primary aluminium production in Italy on a regular logarithmic scale. Trends were obtained allocating carbon dioxide emissions from the domestic metal production as much as the foreign one. Values are in MtCO_2eq.



All the cradle-to-gate processes displayed have been producing a continuous increase in GHG emissions. Carbon dioxide emissions from AR derive almost completely from domestic production, as traditionally Italy has exported aluminium hydroxide. Contribution analysis revealed that the increase in metal production is the main responsible for increasing total emissions, rather than evolution of CO_2eq emission factors. In fact, AR emission factors were constant until the mid of 1990s when they increased slightly, perhaps as a consequence of natural resources depletion in terms of aluminium grade in ores and minerals forms.

Figure 17 – Mine production since 1900. Source: Graedel & Erdmann (2012) – modified.



Similar trends for PAS and IC stages are displayed until, indicatively, the beginning of 1990s. In those years the social sensitivity towards the environmental concern of anthropogenic activities grew and the first pollution control initiatives took place. During the last decade of 20th century, primary aluminium smelters strongly decreased their carbon intensity, largely through PFCs emissions restriction. Moreover, in contrast to AR, the IC stage traditionally saw Italy as importer of aluminium ingots from world partners, and about two orders of magnitude separate the CF between foreign and domestic production. The former includes the direct contribution from IC stage as much as PAS carbon intensity since import of primary aluminium does occur such as casted ingots rather than in liquid form.

Therefore, net-import of metal ingots increased from the mid of 20th century to today; beside, CO_2eq emission factors for IC shows a shift from primary energy to electrical energy carriers since the beginning of 2000s. Foreign PAS, instead, attested a decreasing in energy consumptions.

The characteristic feature of being a net-importer of aluminium determines for Italy a disproportion between emissions from production and those associated with the consumption. Although this is quite often in many developed countries, and Europe is one of the largest net-importer (Carbon Trust 2011), comparing the cumulative CF from aluminium production and consumption in Italy over the decade 2000-2009, it emerges that GHG emissions from the country production should increase by 140% to balance the impact of final consumption. And, whether the comparison is extended to the whole NF metals sector, the percentage change has been quantified at more than 270% (Carbon Trust 2011).

Figure 18 – Historical trends for the Italian cradle-to-gate apparent consumption, and the annual net-addition to carbon profile. Dashed line depicts the sensitivity analysis ranges.

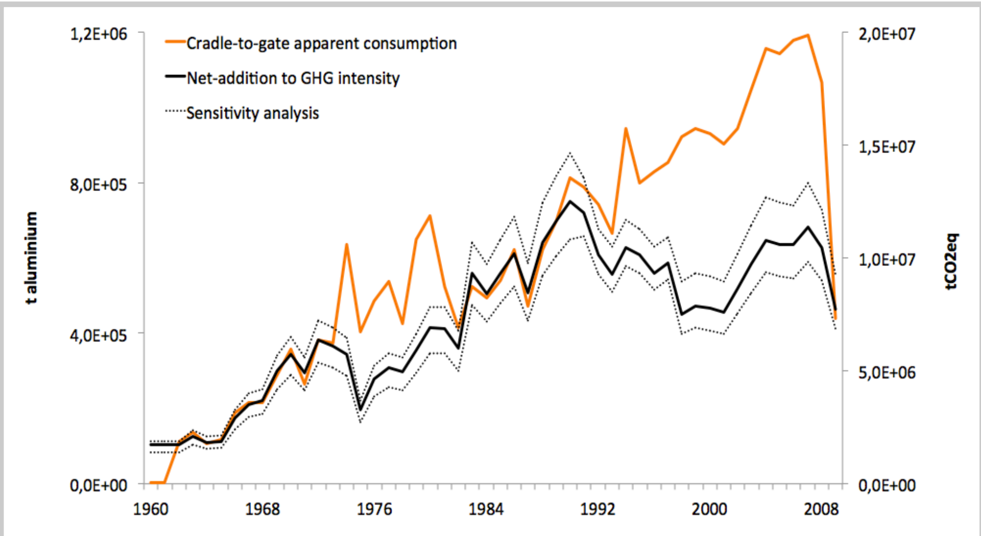


Figure 17 compares historical trends for the cradle-to-gate aluminium apparent consumption, calculated as net-consumption between domestic production, net-import and scrap generated, and the annual net-addition to CF. The two lines

inspire some reflections regarding the sustainable development in industrial production. Till the 1990s, the coupling between economical growth and environmental pollutions appears clearly in the aluminium industry, but such a growth cannot be unlimited in a world with limited resources and bearing capacity. Rather than implying an economic downturn to aim at the environmental sustainability, the awareness of such limits should orient to a decoupling process. Leaving out the effects associated with the 2007-year financial crisis, the historical evolution of carbon intensity decoupled from the cradle-to-gate apparent consumption laying the foundations for a model of sustainable development based on eco-efficient production routes and a 'green' management of natural resources and polluting emissions reduction.

The results achieved by the aluminium industry are just the beginning of the path towards the sustainability, but not the end: the goal of creating environmental-oriented anthropic societies requires aiming at on-going improvements in energy consumptions and renewable sources' enhancing, closing-loop material flows, researching for innovations and technological progresses, and preventing policies. The next section shed a light on potential benefits that would result from exploiting the anthropogenic aluminium in-use stock as supplying material for the secondary metal production.

4.2.2 Potential for recycling and benefits from secondary metal flows

Secondary metal production is the most efficient way to strongly reduce the greenhouse gases release and base national economy on circular material flows as claimed by Industrial Ecology principles. Anthropogenic in-use stock embeds aluminium-containing products suitable to support recycling-oriented initiatives. Debate on strategies for achieving a long-term carbon leakage reduction is of topic interest at present, and among the major ones explored are (i) enhancing efficiencies and non-destructive recycling processes, extending service lifetime, optimising material usage or substitution (Allwood & Cullen 2009; Milford et al.

2011) through, for instance, *ecodesign* and *Design for Environment* (DfE) methods, and consumers' initiatives asking for a 'green demand' of commodity aluminium, and driving a diffusion of carbon profiles for products on market (Carbon Trust 2011).

Overall, respect to primary aluminium, the carbon profile for the secondary metal production strongly decreases consequently to the reduction in energy consumption by avoiding the electrolytic process in PAS. On average, about 95% of energy requirement is cut off when recycling waste and obsolete products: assuming 5% of the whole carbon intensity used in primary aluminium production, we assed (i) the evolution of the Italian aluminium production mix (primary plus secondary), and (ii) the potential for carbon dioxide emissions reduction whether a close-loop recycling consistently based on the in-use stock will be persecuted in the next years. In accordance with at national level survey (CiAl-DIIAR 2010), a 78% recycling rate was assumed in first approximation (i.e. about 1,282 kg of aluminium old scrap are required to obtain 1 ton of secondary metal ingot). About 45% of metal losses is constituted by the fraction of aluminium that is vaporized at high temperature or kept in a vitreous compound during the remelting process (Ferrão et al. 2006; Passarini et al. 2012).

EAA (2008) reported inventory for aluminium scrap mass flows modelling, and records for primary and electrical energy from the remelting process were here used to quantify the carbon profile from secondary aluminium production. Historical values were estimated by linear interpolation between years 1998 and 2005, and assumed constant with time elsewhere.

Although Italian primary aluminium production is currently confined to one single plant, secondary aluminium production is widely operating in the country, and Italy shares with Germany the first place in Europe per number of refiners and remelters and capacity of production (EAA 2008). Secondary metal production is a driving force of the aluminium industry in Italy, and imports of scrap increased constantly over the past several decades. As a consequence, the results show a strong reduction in terms of carbon footprint resulting from the aluminium production mix. The model developed was consolidated by weighting the carbon dioxide emissions with primary and secondary amounts of aluminium produced within Italy.

A strong difference in carbon profile resulted between Italy and the average of EU-countries: while the former achieved in recent years near 80% of secondary aluminium in total metal production, the latters have only half of whole aluminium produced from recycling flows. A direct consequence is the lower carbon intensity score for the country that requires national-specific estimates for quantifying the CO_2eq emissions from secondary production.

The model determined the annual emission factors from aluminium scrap, which were then used to weight the contribution to climate change from the Italian production mix of aluminium. In **Table 15** the average scores for primary, secondary and the mix production are summarized. Weighting the country CF between primary and secondary aluminium domestically produced, 1960-2009 aggregate estimation achieves about 140 Mt CO_2eq, respect to 129.1 Mt CO_2eq from primary aluminium only. Comparing the results for years 1995-2002 with the European averages it emerges the latter would overestimate the carbon leakage from the Italian aluminium production, being they quantified at 8.7 tCO_2eq/t Al and 1.4 tCO_2eq/t Al, respectively from the ecoinvent process '*Aluminium, production mix, at plant/RER*' and '*Aluminium, secondary, from old scrap, at plant/RER*', respect to 3.0 and 0.9 tCO_2eq/t Al estimated in this study. As stated above, reasons must be sought in the share of secondary aluminium in the total metal production: while in 2000 the Italian ratio had achieved about 78%, the European average laid around 30% (Swiss Center for Life Cycle Inventories 2009). At 2009, the values seem to have decreased at 2.0 and 0.3 tCO_2eq/t Al, respectively.

In 2009, the CO_2eq emissions from the national aluminium industry were 1.8 Mt: comparing it with the total anthropogenic aggregate GHGs release in Italy (about 491 MtCO_2eq, UNFCCC 2011), it results the metal production is responsible for 0.37%. Liu et al. (2011) estimated at 0.53% the ratio for the U.S. aluminium industry, while an average 1% is attributed at worldwide level by the IEA (2010). The lower contribution for the Italian aluminium industry to the national CF is very likely determined by the significant amount of secondary metal flows entering the production phase. Indeed, if Italy had all primary aluminium production, the ratio would increase at 1.6% of cumulative anthropogenic CO_2eq emissions.

Table 15 – Average CO_2eq emission factors from the Italian aluminium production: primary, secondary and the resulting mix estimated by weighting the mass contribution from primary and secondary aluminium (see also Table in Appendix B).

	tCO_2eq/t Al		
Years	**Primary**	**Secondary**	**Mix**
1960 – 1969	12.9	0.8	9.4
1970 – 1979	15.4	0.9	9.8
1980 – 1989	16.7	0.9	8.2
1990 – 1999	12.0	0.9	4.2
2000 – 2009	9.9	0.5	2.4

Furthermore, if the anthropogenic aluminium in-use stock had been exploited quantitatively at the end of the year 2009, the potential GHG emissions savings would have resulted in about 160 $MtCO_2eq$. The estimation is calculated as difference between the avoided carbon dioxide emissions from an equivalent amount of primary aluminium and the CO_2eq from secondary aluminium production. Despite being a mere speculation, that simple calculation gives an idea about the magnitude of carbon reduction embodied in the national in-use stock as it represents more than one third of the total GHG emissions occurred in 2009.

Finally, linking such an estimation for national carbon savings with the EU's priority to contrast global warming, by promoting green technologies and developing strategy to reduce GHGs emissions cost-effectively (i.e. EU-ETS), it might derive relevant financial revenues for actors involved in primary and secondary aluminium production (EC 2012). Particularly, whether the primary aluminium industry might be likely affected by smelters' ability to secure electricity contracts with low carbon leakage in order to preserve competitiveness in the EU-ETS (Reinaud 2008), the secondary aluminium production might reflect less dependence from the potential increasing in electricity prices consequently to carbon emissions' cap in power generation.

However, the role of appropriate price on carbon emissions as condition to promote green technological innovations for global warming contrasting must be discussed carefully. Appropriate price implies a balance between environmental burdens from greenhouse release and marginal costs for reducing such emissions.

In addition, due to lacks for standardized procedures when accounting international trade of aluminium, which generate as seen a disproportion between CF of production and consumption of aluminium in a country, a relevant portion of GHG emissions cannot be covered by the EU-ETS scheme; at European level, about two third of carbon dioxide emissions are missed (Carbon Trust 2011). Nordhaus (2011) argued cap-and-trade approaches be a solution to propose an efficient price for limiting carbon emissions, especially for orienting innovations development. Rather, the author indicates how internationally harmonized carbon tax might give more reliable information.
Aiming at analysing which factors influenced the CF of aluminium industry over years, in the next section the Kaya Identity (Kaya 1990) equation is applied, being it adopted by the UNFCCC and the IPCC as supporting tool for the planning of strategies oriented to a reduction of climate changing emissions.

4.2.3 How did factors influence the carbon profile over time and which are the levers to mitigate it?

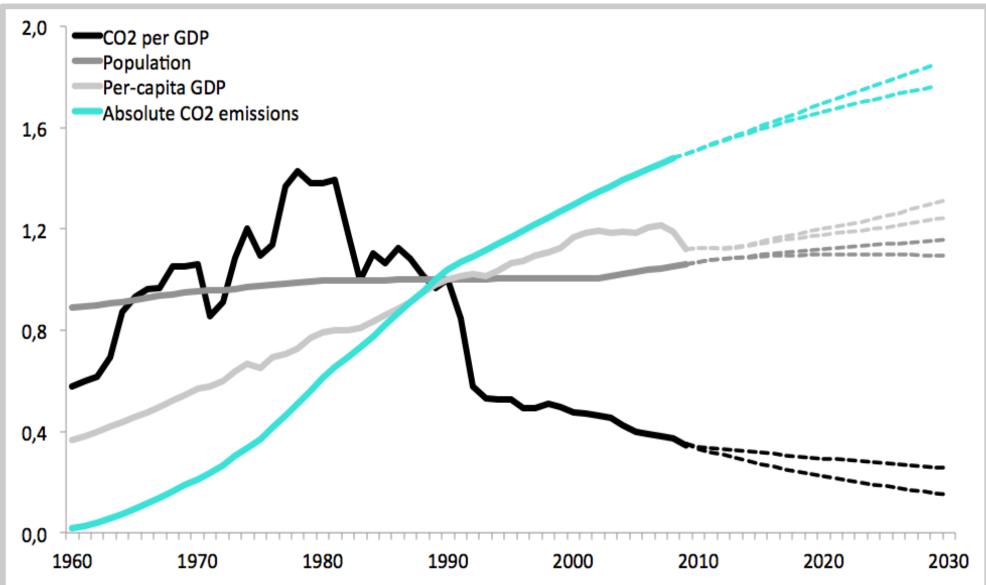
Using the Kaya Identity equation (Kaya 1990) it is investigated how the main drivers addressed the CF of the Italian aluminium production industry over the study period. In fact, the Kaya Identity equation weaves CO_2 emissions with factors that consider the diffusion of technological innovations, energy savings and decarbonizing of fuels, and demographic trends, according to the equation **A12** (Rapauch et al 2007):

$$F_j = P_j \times \frac{G_j}{P_j} \times \frac{F_j}{G_j} \qquad (\mathbf{A12})$$

Where F_j is the total CO_2 emission from the aluminium industry, P_j is the Italian population, and G_j is the Italian GDP in the *j*th-year. The logarithmic transformation of the Kaya Identity expresses the annual rates of factors equation, which allows for comparing a single variable's influence on GHG intensity.

Figure 19 shows historical trend-lines for Kaya Identity factors and absolute CO_2 emissions. Scores are normalized at 1990-year values. Before 1990s, GHG emissions increased due to per-capita GDP, population growth and carbon intensity from primary aluminium industry. After year 1990, absolute CO_2 emissions slowed down consequently to a drastic decreasing in GHG outflows from primary aluminium industry, caused from both an increase in secondary metal flows for recycling and measures to contrast climate change emissions. Dashed lines in **Figure 19** estimate projections for next years. Although long-time forecast be arguable and an unexpected factor might change quickly the current situation, extrapolation to the next two decades may help to support decision-making strategies. Forecasting trend-lines assume upper and lower uncertainty values from literature sources for the Italian demographical development (ISTAT 2011) and the GDP (ERS-USDA 2011); instead, CO_2 emissions range was estimated assuming an upper value equal to the annual average of last decade (– 3%), and a lower one from an arbitrary and precautionary choice at – 0.5%.

Figure 19 – Evolution of the Kaya Identity factors from the Italian aluminium production industry.



On a hand, projections seem to share expectations for a decreasing in CO_2 emissions, although very likely at lower rates than in past years. The results are in line with the warning for claiming innovations in the technosphere such a lever for future improvements presented in Liu & Müller (2011). Indeed, being the per-capita GDP an index of economical wellness for a country, technical and technological progress is the main factor that would enhance mitigation in climate change emissions from the aluminium industry. The *European IPPC Bureau* and the *Technical Working Group* (TWG) for the Non-Ferrous Metals Industries (EC-JRC 2001) indicates technological improvements depend on certain variables and factor, including raw materials composition, the type and amount of contaminants, gas collection and abatement solutions, and energy requirements. Particularly, high-efficiency advanced equipment and scientific knowledge, decarbonized energy-sources for power production and renewable primary fuels adequately supported by taxing measures for carbon emissions should inspire and orient future researches and policies in the aluminium industry. At national level, specifically, Italy applied in 2002 the *Strategia d'azione ambientale per lo sviluppo sostenibile in Italia* (Environmental action strategy for sustainable development in Italy, CIPE 57/2002), in line with the *European VI Environment Action Programme*, and claiming, among others, for a 6.5% carbon emissions reduction, against a year 1990 baseline, over the time lapse 2008-2012, increasing renewable energy sources for power production, implementing waste to energy plants at the expense of landfill disposal practices, and prevention and reduction policies against the global warming. Despite national environmental policies have showed some progress, lacks in its implementation still remain (EC 2011).

4.3 Final considerations

The analysis of GHG emissions embodied in the Italian aluminium revealed relevant potentials for decreasing the contribution to climate change embodied in the anthropogenic aluminium in-use stock. The research constitutes a first study dealing with a historical perspective of the Italian aluminium industry and as such it may provide support to the country for orienting industrial policy toward the goal of a low carbon society.

Data availability and collection determined the main assumptions and uncertainties affecting the results: future works will aim at checking and implementing the sources in number and quality. Moreover, the extension of impact assessment to additional environmental categories, beside the inclusion of further aluminium life cycle stages, would allow to improve the glance on the whole metal performance to the environment.

However, the outcomes pointed out the 'virtual' carbon emissions associated with international trade flows of aluminium are not negligible but they may cover up to two third of embedded leakage from a country's consumption, and the need of standardized procedures to properly account them in carbon markets. Also, the analysis allowed to set historical indicators for the climate change category at national level, which does integrate and updates average scores generally published in international LCA-oriented databases.

Finally, since the Italian aluminium industry is mostly based on secondary metal flows aiming at giving contribution to enhance the aluminium recycling industry, in the next Chapter a discussion about criticalities influencing the metal recovery is carried out through two case studies regarding the transportation and containers and packaging end-use markets.

5.

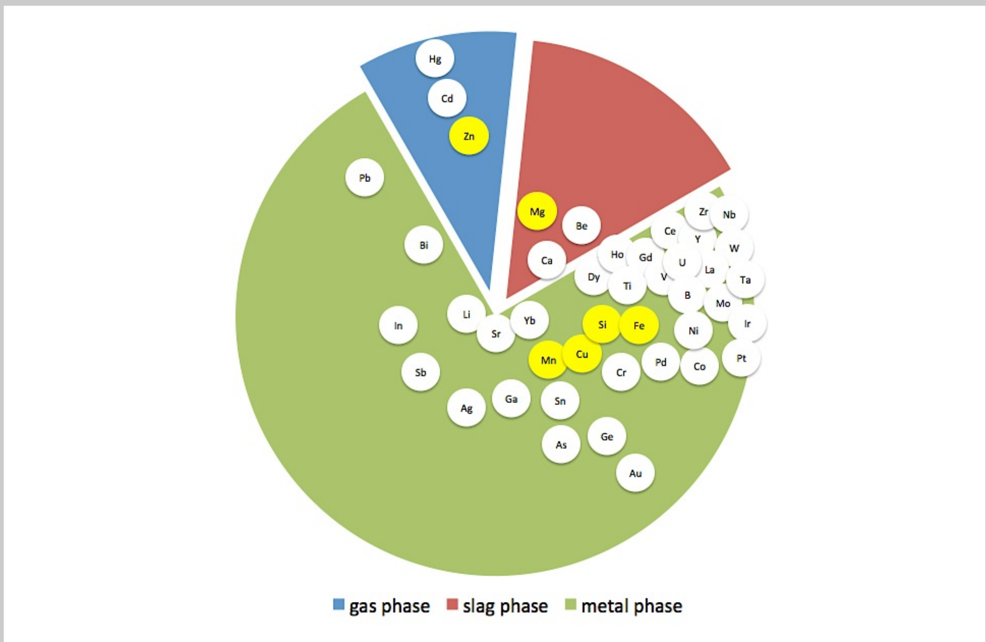
Enhancing the aluminium recycling industry: a contribution from two Italian case studies

The results from MFA and LCA models presented in Chapter 3 and Chapter 4 identified and quantified significant potentials for reducing the contribution to climate change from the exploitation of the anthropogenic aluminium in-use stock in the next years. National aluminium industry is mostly based on secondary metal flows and Italy is at first places in Europe for numbers of refining and remelting plants (EAA 2008): therefore, the achievement of a society based on secondary flows of the metal seems to be likely to happen.

However, recycling is becoming more and more challenging as products are today multifunctional and sophisticated than in the past, and despite aluminium be among the most common used metals today, its recycling rate has been averagely estimated around 50% (UNEP-IRP 2011; McMillan 2011): this means that, before being lost, a common unit of aluminium is reused two or three times only, disproving the credo of infinite recyclability for metals (Reck & Graedel 2012; Eckelman & Daigo 2008).

Reasons can be sought in thermodynamic limitations that may affect the grade of re-processing: removing impurities and elements from aluminium alloys is a great challenge. For instance, the 3000 series of aluminium alloys contain manganese, which is retained during the metal remelting operations but making the melt not fitting any other aluminium-based alloys (Reck & Graedel 2012). Avoiding similar bottlenecks requires a successful system of separation, collection and processing steps as much as to integrate design for recycling principles in the production stage. EOL management chain has smaller issues of scale and economical returns may be not adequate to cover the adoption of further sensing and sorting equipment, leading to more material losses. Post-collection processes usually generate an increase in entropy of input waste that may limit recycling due to inconvenient economics (Reck & Graedel 2012); however, crashing-to-recover practices reflect a lack in the design of new products, where *Design for Recycling* (DfR) and *Design for Disassembly* (DfD) procedures should be, instead, strongly encouraged.

Figure 20 – Distribution of elements among gas, metal and slag phase during the remelting of aluminium scrap. In yellow are alloying elements. Source: Hiraki et al. (2011) – modified.



Leaving out economic considerations that lie outside the goals and scopes of this research, but intending to give a contribution for enhancing the aluminium recycling industry in Italy, two case studies are following presented: they deal with application of MFA and LCA to the transportation and the containers and

packaging end-use markets, which were identified in Chapter 3 as the most relevant for scrap generation and as such, key levers to fulfil a closure of flows in the aluminium cycle. In this sense, collection of EOL products and efficiencies of recovery processes are the greatest forces to improve metal recycling (Reck & Graedel 2012). Anyway, as seen in Chapter 3, national collection rates differ between 50%, for containers and packaging and almost 100% for transport, and the low efficiency in recovering discarded containers and packaging is among the main reasons that determine relevant metal losses in the Italian aluminium cycle, charting the collection of obsolete products and scrap stage (CES) after the alumina refining step (AR) for absolute quantity of deposited losses. However, even in the case of transportation, considering the increasing use of aluminium and the metal stock potentially recoverable in next years, a systematic scheme to enhance the aluminium recycling industry need to be defined.
The case studies here presented shed also a light in terms of application of MFA and LCA methods for supporting decision-making at a sectorial and local level.

5.1 Aluminium recovery from the transportation sector

The use of aluminium in the transport sector is among the main end-use markets for this metal and it represents a domain in which the challenge towards a reduction in environmental burdens can be enhanced by its particular lightness to strength ratio and its high recyclability grade. Main environmental benefits rely, indeed, in decreasing energy requirements and fuels consumptions during the use phase, but as much relevant potentials as result from indirect substitution of virgin materials (Kim et al. 2011; Ungurenau et al. 2007; Boon et al. 2000).
Whether the use of aluminium for road applications covers the major transportation market, the search of lightweighting interests every conveyance sector and it constitutes the main driving force for innovative solutions based on the metal (EAA 2008). The design of Aluminium Intensive Vehicles (AIVs) is a current topic of interest and it has been discussed largely in literature (Boon et al. 2000; Field et al. 1994). Expectations for next years do indicate that aluminium will

increase at 140 kg per vehicle respect to 60-70 kg of nowadays (ATG 2009). Averagely, a 10% vehicle weight reduction is expected by enhancing aluminium application in the body structure as much as in secondary components (e.g. engine and transmission, suspension and chassis parts), and despite an increase in GHG emissions at the production stage can be expected, when compared to a conventional steel-based vehicle (Ungurenau et al. 2007), a 100 kg reduction in car weight leads to a saving of 10 g CO_2 per kilometre (EAA 2007).

Similarly, aluminium may constitute from 60% to 80% of structural weight in in commercial or short and mid-range aircrafts, constituting a core element in the aerospace industry (EAA 2012), even because the possibility to obtain high performing materials by alloying aluminium and technical metals such as magnesium and zinc (e.g. Aluminium 7075) to resist extreme environmental conditions. From an internal report by the *Center for Industrial Ecology* at Yale University (Eckelman et al. 2011), GHG emissions from scrap collection and processing of aluminium-based aerospace alloys were estimated in 98 kgCO_2eq and 90 kgCO_2eq per ton of Aluminium 7075 recovered, by mass and economical value respectively. However, the close-loop recycling of one ton of that alloy in aerospace engine components would result in a net-reduction of about 13 tCO_2eq, mainly as effect from substituting for virgin materials (Eckelman et al. 2011).

The efforts made by the transport sector to make vehicles more sustainable are not limited to the phases of design and use only, but cover all the life cycle stages, including waste treatments and management practices. Aluminium scrap recovered from EOL products is hence a key lever to act for really closing material cycles and to support the creation of circular economies in accordance with Industrial Ecology principles (Ayres & Ayres 2002).

Up to 75% aluminium recycled in Europe is used for applications in transportation, and total amount of aluminium in use stock embedded in the Italian transport sector was estimated in about 7.0 Mt or 115 kg per capita (**Table 16**); values are in line with the results reported in literature (Liu et al. 2011). Comparing variations over decades, the consumption of aluminium in transport sector shows an increase of 84% in the last 10 years, almost doubling the previous rate. The increase in stock in the same years was of 67%, while in waste generation of 59%.

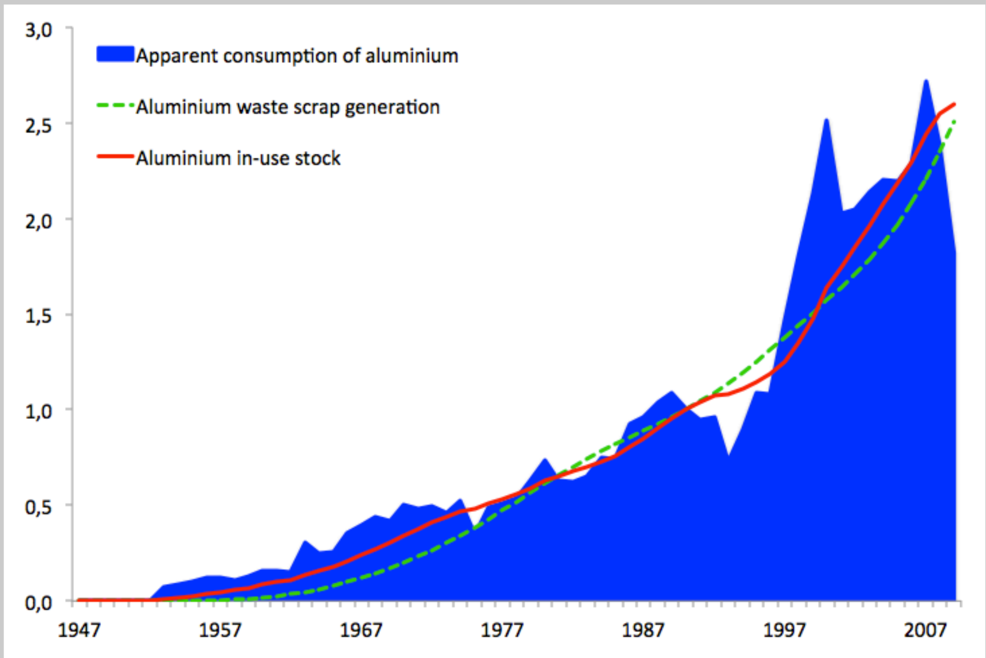
Considering the provisions for next years, it can be expected a stronger increase for each of the variables. Road vehicles seems to contain about 80% of all aluminium used in transportation, rail applications have about 2%, while air and marine ones almost equally shares the remaining fraction; annual averages trends are quite similar over the decades, and the portioning seems in accordance with Liu et al. (2011). Thus, focusing on End-of-Life Vehicles (ELVs), around 5.5 Mt of aluminium is potentially recoverable through the national ELVs management chain in the next years.

Figure 21 shows the evolution of annual change in apparent consumption of aluminium in the transportation, and the trends of both metal addition to in-use stock, and generation of waste and obsolete transport products. In the chart is evident the net increase occurred in the last decade as a consequence of the rising use of aluminium for transport purpose.

As pointed out, the in-use stock is increasing and considering the tendency toward the production of AIVs, the importance of achieving a full efficient separation and collection of the metal is of topical interest. Anyway, although the management chain of ELVs has an established procedure and the recovery of aluminium scrap is generally carried out in medium and big size Italian shredding plants, a considerable fraction of the metal particles ends up in the light fraction named car fluff. Since this residual waste is mainly disposed of in landfill, the aluminium unrecovered is entirely lost in ground, contributing to increase the set of resources named 'urban mines'.

The management chain for ELVs follows general dismantling and shredding practices commonly adopted worldwide, although differences in car material composition, legislative regulations or energy-related variables can sensitively influence environmental performances and material recovery (Passarini et al. 2012). Generally, 80-95% of ELVs enter the management chain as obsolete and discarded vehicles at the end of their life or even as new vehicles but damaged after road crashes. The drainage of fluids and the removal of batteries and similar hazardous components are the first processes an ELV undergoes before being sent to the dismantling phase. The latter includes the removal of profitable parts for reusing, remanufacturing, rebuilding or recycling processes: particularly, disassembly is usually extended to those parts whose market value exceeds removal costs, or for which the legislation claims for mandatory removing for safety reasons. Examples cover tires and glass, bumpers, engine and transmission parts or door skin (Passarini et al. 2012; Dalmijn & De Jong 2007).

Figure 21 – Evolution of trends in apparent consumption, waste scrap generation and in-use stock of aluminium from the transportation sector in Italy. Values are normalized to the year 1990 results.



The vehicle hulk resulting is compressed and processed in shredding plants to be reduced by a mill in pieces with dimensions and compositions suitable for material separation. Sorting and collecting processes commonly adopted by shredders include magnetic separators for ferrous scrap and eddy currents (ECs) equipment to concentrate non-ferrous metals, among which aluminium is the most abundant (about 80%), (Ecofer 2010). ECs are induced within the conductive scrap and they lead it to a magnet moment. When particles across the gradient field by magnets, they are subjected to acceleration, as response to the deflecting electrodynamic forces, and pushed over the belt. Ferrous metals scrap are quantitatively recovered and sent to steelmaking industries, while aluminium and coppers have less efficient separation grades due to the losses during aeraulic separation steps for light fluff fraction and the presence of alloying elements that affect efficiency in response to ECs magnets.

The high revenues from aluminium (about €/t 1500), (Gesing 2005) and copper scraps (up to €/t 3000), beside to the fulfilment of EC targets for ELVs (95% of overall recovery rate, with at least a minimum 85% for recycling and a maximum 5% for landfilling, Directive 2000/53/EC) they oriented the current interest towards solutions to improve the recovery of these fractions. Additional limits to aluminium scrap separation and recycling are represented by the need to improve the quality for secondary metal production through additional cleaning operations, e.g. the polyvinylchloride (PVC) coverage for copper wires that affect the yield of automatic collection systems.

The mixture of unrecovered different fractions, in which thermoplastic and thermosetting polymers, natural or anthropogenic textiles, rubbers and inert materials are embedded, is the waste named Automotive Shredder Residue (ASR) or car fluff. Generally, ASR term refers to the whole heavy and light fractions of car fluff: the former is about 5% on an weight basis (compared to the initial ELV) and it is generally processed by means of ECs to separate aluminium scrap, while the latter is up to 25% and it is generated from aeraulic separator after the shredding phase. Due to its heterogeneous composition, a great volume and a scarcity of valuable materials embedded, the light fluff is generally untreated before disposal. Although the landfill disposal of waste with a LHV > 13 MJ/kg is prohibited by law, ASR is currently landfilled in most of the European countries. Of course, even some metal scrap unsorted remains in the car fluff, depending on the efficiency of separation techniques integrated to ECs in shredding plants (Staudinger & Koleain 2001). Since some projection to 2015 quantifies the amount of ELVs at around 14 Mt in Europe (COM Report 2007), of which about 1.3 Mt generated in Italy, at the current recovering rate for aluminium scrap, the metal content can be estimated to increase up to 10% in the remaining shredding waste ASR, with a relevant loss in terms of material and savings for energy requirements.

In Ciacci et al. (2010) a comparison among different ASR treatment processes was carried out with the aim to propose alternatives to the present landfilling in terms of benefits from resource conservation and reduction of waste disposal. The results of the LCA analysis showed the recovery of non-ferrous metals, including in prevalence aluminium, ensured a reduction of total environmental burdens related to resource depletion categories for all the scenarios investigated. Moreover, the option modelling Post Shredding Technologies (PSTs) with feedstock recycling, which resulted in the best environmental performance, it is even the treatment scenario that allowed an additional amount of metals recovered. Full paper has been presented in (Ciacci et al. 2010) and the final publication is available at www.springerlink.com.

In a following study (Passarini et al. 2012), LCA was applied to quantify potential implications of ASR composition evolution in EOL treatments. Particularly, increasing in light material content such as polymers and aluminium in new vehicle production, and improving in separation and recycling rates were modelled. Among the main outcomes, estimations regarding impacts avoided by recycling aluminium from ASR in year 2015 amounted at −29.1*10E+09 GJ and −2.2*10E+09 tCO_2eq for non-renewable energy consumption and contribution to climate change respectively.

Eventually, the research pointed out the need to adopt ecodesign and DfE approaches as tool for conceiving new generation vehicles and supporting decision makers in waste management strategies. Full paper has been presented in (Passarini et al. 2012) and the final publication is available at www.elsevier.com.

5.1.1 Characterization of aluminium in the light fluff

Aiming to support the improvement of aluminium recovery from light fluff, sampling and sorting procedures in a dedicated campaign of characterization have been performed in order to identify and quantify shape, dimensions and measures of metal scrap contained in that fraction. Indeed, a uniform distribution in size and shape enables efficient mechanical metal scrap concentration (Gesing 2005), although some shapes as wires and thin foils may fail to be separated because of an insufficient response to ECs magnets (Gaustad et al. 2012). The research can be intended as complementary to a pyrolysis trial previously carried out (Morselli et al.

2011). The two works, indeed, aim at investigating possible solutions for a quantitative exploitation of metallic and polymer fractions embodied in the light fluff. Full paper has been presented in (Morselli et al. 2011) and the final publication is available at www.springerlink.com.

The campaign has been carried out in a medium-size plant currently operating in Italy and representative of the national status quo. Fraction < 50 mm is generally rich in organic materials and it contains certain amount of residues from aluminium scrap (Morselli et al. 2010; Fiore et al. 2012). Thus, after screening a common output car fluff at 50 mm, a sample of 14.0 kg of aluminium scrap was handy sorted and collected: despite a strong difference in shape, metal scrap showed a high purity from contaminants, which has been estimated up to 90-95%. This level could even be improved by shredding collisions that cause the removal of paint layers covering up to 15% of the metal chips (Gesing 2005).

The sample of scrap was then classified according to the granulometric features of metal pieces in size and shape: the former included a selection among ranges 50-100 mm, 100-150 mm, and > 150 mm, while the latter a distinction as flat, cylindrical, irregular, cubic and multi-layers form, particularly relevant for separation purposes. Multi-layers refer mainly to pressed and shredded scrap from radiators. **Figure 22** simplifies the scheme of characterization followed. Fraction < 50 mm has small particles unsuitable for size classification: the quantification of aluminium was therefore accomplished only on a mass basis.

Hence, a second hand-separation step, aimed at selecting the most representative aluminium scrap from each sorting classes, was made in order to measure average weight and surface. The information gained from the classification was used to set ECs parameters and a scale up of the campaign waste, tested by dealing with a sample of about 3.3 tons of light fluff, using the normal process line adopted to treat the heavy fraction. The results are presented and discussed in the next section.

Figure 22 – Simplified scheme for the light fluff characterization adopted.

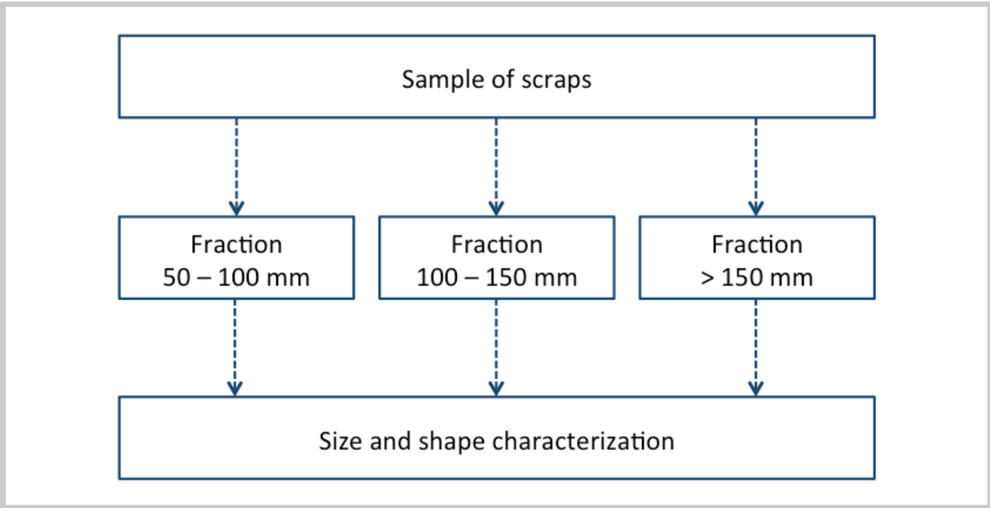


Table 16 – Cumulative amount of aluminium per decades: apparent consumption, waste scrap generated and per capita in-use stock in the transportation sector.

	Consumption	Waste generated	In-use stock
Years	*Mt*	*Mt*	*kg per capita*
1960-1969	0.7	0.1	15.1
1970-1979	1.3	0.5	28.0
1980-1989	2.1	1.1	45.1
1990-1999	3.1	1.7	69.3
2000-2009	5.7	2.7	115.4
Sum	*13.0*	*6.1*	*115.4*

Non-ferrous metals content in the ASR has been estimated in about 1% on mass basis, although some studies reported a maximum concentration up to 10% of the ASR weight (Gesing 2005). Aluminium scraps are the most abundant among non-ferrous metals, at 80% averagely (Ecofer 2010). **Figure 23** shows the distribution resulted from the characterisation campaign performed to partition aluminium in light fluff. ASR size distribution showed that about 60% aluminium present in light fluff is under 50 mm in size, and up to 87% is < 100 mm. Consequently, 10% is contained in fraction 100-150 mm and only 3% is bigger than 150 mm.

Figures 24a, 24b, 24c show size and shape distribution of aluminium scrap in fractions 50-100 mm, 100-150 mm, > 150 mm. Overall, the results present a median ratio weight to surface generally around 5 mg/mm^2. A higher uncertainty seems to affect the distribution of aluminium particles by shape: flat-shape metal scrap was present in each fraction considered, although a reduction in the interquartile difference, as index of dispersion, occurs when moving towards bigger fraction. Cylindrical-shape particles seems to concentrate in 50-100 mm and > 150 mm fractions. This class was the most variable, although the dispersion from median values remains similar. Lastly, irregular-shapes concentrate in fractions > 100 mm, while cubic and multi-layers shapes seem to be present in only one fraction: the formers concentrate between 100 and 150 mm, while multi-layers in 50-100 mm. The latter shapes show a narrow dispersion range since this class originates almost exclusively from one single source of scrap (i.e. radiators).

Figure 23 – Distribution of aluminium particles in untreated light fluff: the results are presented in percentage on a mass basis.

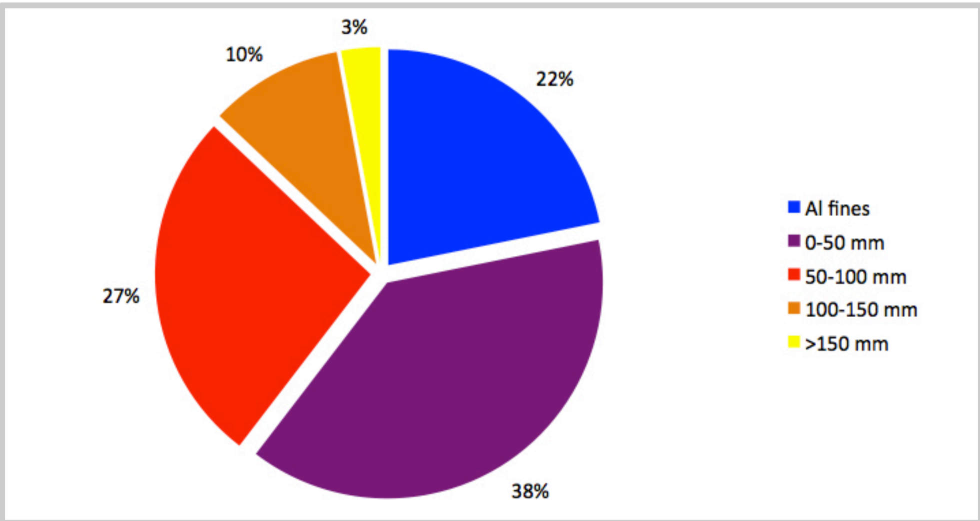


Figure 24a – Size and shape distribution of aluminium particles and scrap embedded within the light fluff, fraction 50-100 mm. On y-axis weight to surface ratio (mg/mm²) is showed.

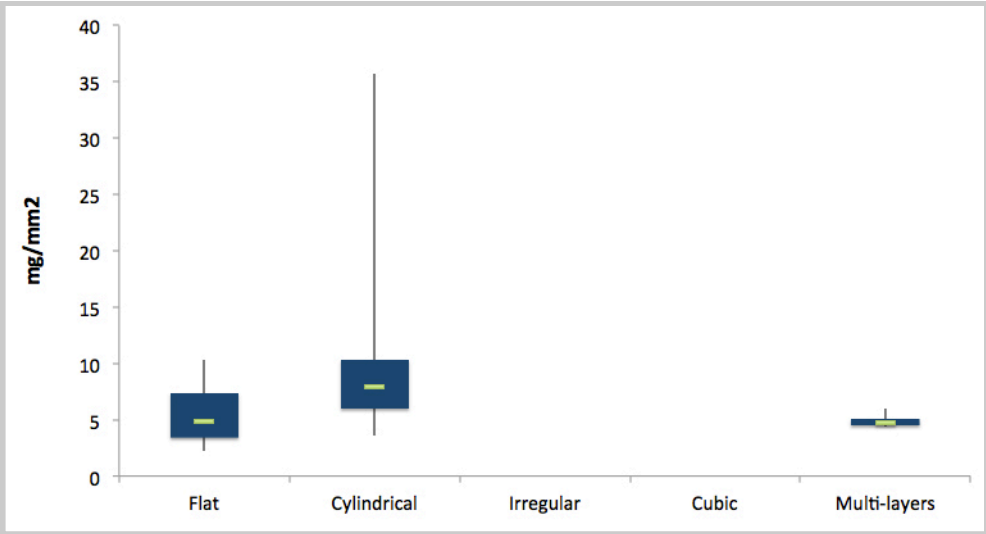


Figure 23b – Size and shape distribution of aluminium particles and scrap embedded within the light fluff, fraction 100-150 mm. On y-axis weight to surface ratio (mg/mm²) is showed.

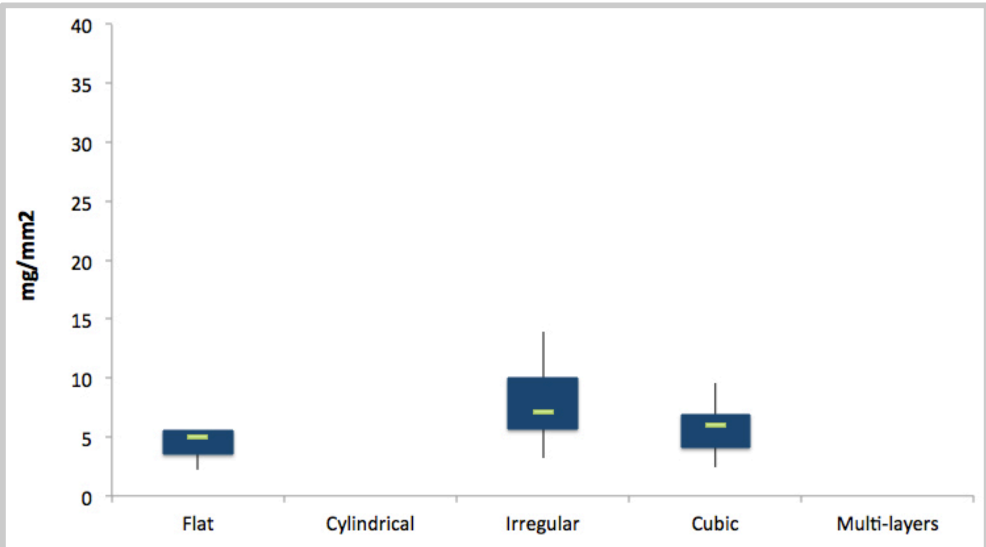
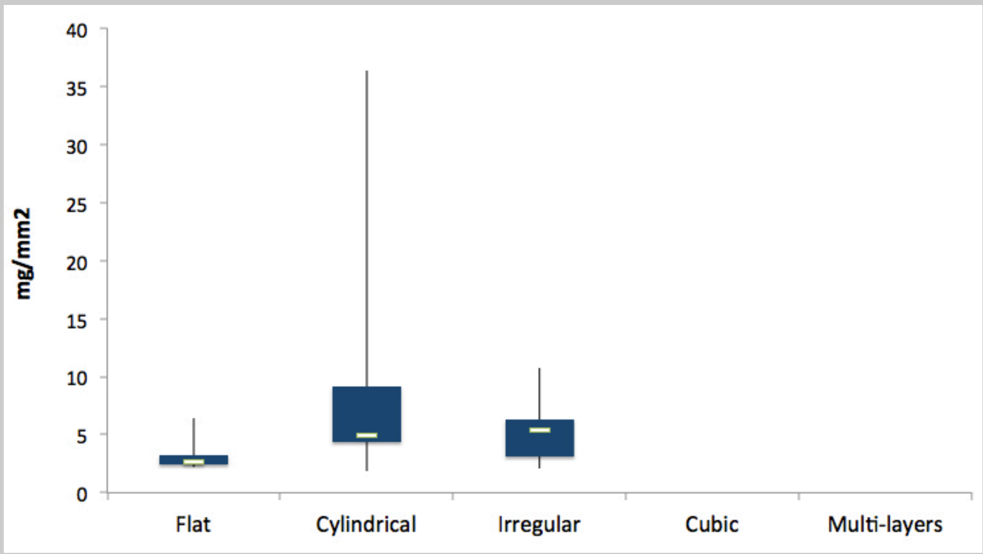


Figure 24c – Size and shape distribution of aluminium particles and scrap embedded within the light fluff, fraction > 150 mm. On y-axis weight to surface ratio (mg/mm²) is showed.



The campaign with 3.3 tons of light fluff resulted in 0.65% aluminium recovery on an input basis. Consequently, it follows that 6.5 kg of aluminium can be recovered from treating 1 ton of light fluff in normal operating conditions. Of course, that amount may change depending on shredding efficiencies, waste typologies treated by the shredder plants (since often they treat also discarded white goods and relative waste), the aluminium content, and parts removed in previous stages.

The results offer also perspective for improving aluminium scrap recovery from the management of the light fluff fraction in the country. Particle size and shape separation mechanism are included among the main criteria when implementing ECs (Zhang et al. 1999). Indeed, selective separation of aluminium scrap with different distribution in size and shape depends on magnetic deflecting forces as much as the gravitational and the centrifugal ones. Both deflecting and centrifugal forces are shape-dependent and influenced by length, width and thickness of particles (Zhang et al. 1999).

Critical particle size for ECs, which disables the deflecting forces, it seems to attest around 2-3 mm (Zhang et al. 1999), despite issues may occur even at 5 mm (Settimo et al. 2004). Since aluminium fines contributes for about 40% of the total metal weight in the light fluff, prior removal before EC separation seems to be preferable. Among the most innovative solutions, combining screening separation (e.g. trommels), (Coates and Rahimifard 2009) or adopting advance air-pulse separators (Lee and Rahimifard 2012) were discussed in literature. Particularly, since air-classifiers are low cost technologies, the latter seems to be a budget solution to improve removal efficiency up to 25% more than conventional air-based classifiers (Lee and Rahimifard 2012). Dense media separators are usually applied to heavy fluff (Coates and Rahimifard 2009) and they may increase rapidly in cost due to the higher volumes of light fluff. On the other hand, Settimo et al. (2004) discussed the chance to remove metal fines from waste streams by feeding them slightly wet to conventional eddy-currents machineries.

The remaining 60% has size > 50 mm, which is suitable to be processed by ECs: however, around 50% concentrate grade of metals from fluff leads to negligible losses in ECs separators (Gesing 2005). Even in this case, therefore, a concentration per dimensional classes through screening separators may help to enhance aluminium scrap separation for setting ECs depending on shape distribution. Besides, a further reduction in light fluff heterogeneity may derive from integrating advanced technologies for non-metal separation and recovery. Among others, the most interesting improvements include sorting sensors applying infrared and ultraviolet spectroscopy, visible and X-ray imaging, even coupled with 3-D size and shape systems (Gesing 2005; Gaustad et al. 2012).

Additional questions to be considered include the requirement, or not, of scrap grouping per type. The concentrations of different elements in aluminium scrap influence the possibility to produce aluminium-based alloys: for instance, mixing primary alloys, which generally contain low concentrations of impurities, with waste scrap, allows a dilution of the desired elements for secondary alloys production. Moreover, mainly due to the economical feasibility of the upgrading process, additives as nickel and tin must be diluted while, on the other hand, elements such as lithium can be segregated and refined but paying high cost (Gesing 2005).

Finally, designers and engineers as well as policymakers can powerfully help the recycling stage by limiting the choice of aluminium alloys, by reducing the number of joints and connections in composite assemblies, and possibly by consolidating standardized procedures in the production stages of globally sold new vehicles (Passarini et al. 2012; Gesing 2005; Morselli et al. 2011).

5.1.2 Estimations from the stock and flows model for the national transport sector

Assuming an average aluminium content of 6-7% ELV weight and 6.5 kg of metal scrap recoverable from 1 t of light fluff, it can be roughly estimated that for every ton of aluminium used in the transportation sector, about 102 ± 25 kg is recoverable by processing the light fluff. Extending the approximation to the total aluminium in-use stock embedded in that end-use market, an average of 560,000 tons of metal could be potentially recoverable from ELVs by treating the light fluff in the next years. This value is approximately 75% aluminium from national primary and secondary production in year 2009. Lastly, the recovery of the estimated amount would result in quantitative environmental benefits, mainly in terms of CO_2 emissions savings and in decreased energy consumption, from electrolysis to primary aluminium production.

5.1.3 Final considerations

The transportation sector constitutes the major end-use market for aluminium-containing products and expectations for next years do indicate that aluminium will increase at 140 kg per vehicle (ATG 2009). The exploitation of the current in-use stock is enhancing the metal recycling industry and will support the creation of circular economies. Thus, stocks and flows models may help to investigate the partitioning of aluminium within national borders and to detect chances of improvements.

In this sense, ELVs are a great source of aluminium scrap suitable to recycling; although the national management chain generally includes eddy currents machineries, a part of the metal residues is lost in the light fluff output. The results of this study contribute to shed a light on aluminium particles size and shape distribution in the light fluff fraction, and they may help Italian shredders and decision-makers to plan strategies for enhancing aluminium recovery from ELVs. Indeed, although the results of this preliminary screening campaign need to be further tested, the technical feasibility of processing light fluff fraction seems to require a selective separation of different scrap classes by size and shape in order to set ECs separators appropriately. Particularly, prior removal of fines and screening separation of particles > 50 mm by size and shape may increase efficiencies of recovering aluminium, and at supporting the closure of material flows in the national aluminium cycle.

5.2 Aluminium recovery from containers and packaging waste

Under the memorandum of understanding between the *Emilia-Romagna Region* and the *Consorzio Nazionale Imballaggi* (CONAI) to support the development of activities and the exchange of information about containers and packaging waste management aimed at increasing separate collection and recovery, a survey of present and future scenarios for aluminium waste at regional level was conducted (CiAl-CIRI Energia Ambiente-ARPA Emilia-Romagna 2012). Particularly, MFA and LCA were applied to detect and quantify partitioning of aluminium flows within the Emilia-Romagna region waste management system in years 2010 and 2020. Among key issues of the research were the capacity of operating incinerators and mechanical-biological treatment (MBT) plants, and potential for improving aluminium recovery from specific MSW flows and bottom ash (BA) output WtE sector; and, an estimation of environmental burdens associated to climate change and cumulative energy requirement from material recycling through a screening LCA.

In 2010, MSW produced within the Emilia-Romagna region amounted at 3.1 Mt or 698 kg per capita. Separate waste collection achieved 50.4%, and partitioned as reported in **Table 17**.

Compared to metal packaging entered in consumption, steel and aluminium C&P recovered were estimated averagely at 45%, although strong differences occurred between provinces depending on criteria applied for C&P classification and collection local systems. Whether aluminium C&P waste separately collected enters the management recycling chain, metal unsorted ends up in the residual MSW flow. Regional waste management system for residual waste accounts for incineration with energy recovery (i.e. WtE plants) at 42.5%, selection and MBT plants at 33.5%, and landfill disposal at 24.0%. Although the regional waste management system is able to fully deal with residual MSW flow, aluminium recovered is negligible as of today.

Table 17 – Shares of separate collection for different waste types. Source: ARPA Emilia-Romagna (2011).

Waste fraction	Quantity (t)	%	kg per capita
Organic	212725	13.7	48
Green	355983	22.8	80
Paper and cardboard	369443	23.7	83
Plastic	100455	6.4	23
Glass	143084	9.2	32
Ferrous and NF metals	42060	2.7	10
Wood	131087	8.4	30
WEEE	26387	1.7	30
Other	176810	11.3	39
Total	*1558033*	*100*	*352*

Indeed, only half of total BA produced from the eight regional incinerating plants is sent to treatment for recovery, and only two plants, outside region, do integrate ECs machineries for aluminium selection. Similarly, selection and MBT plants in Emilia-Romagna were 10 in year 2010, anyway only one plant recovered aluminium. It is interesting to point out that only 60% of total capacity is currently exploited in those plants, despite MSW directly landfilled (i.e. with no pre-treatment) corresponds at about 77% of the remaining capacity. Mass stock and flows model of aluminium in Emilia-Romagna region for the year 2010 is reported in **Figure 25**.

5.2.1 Description of the model created

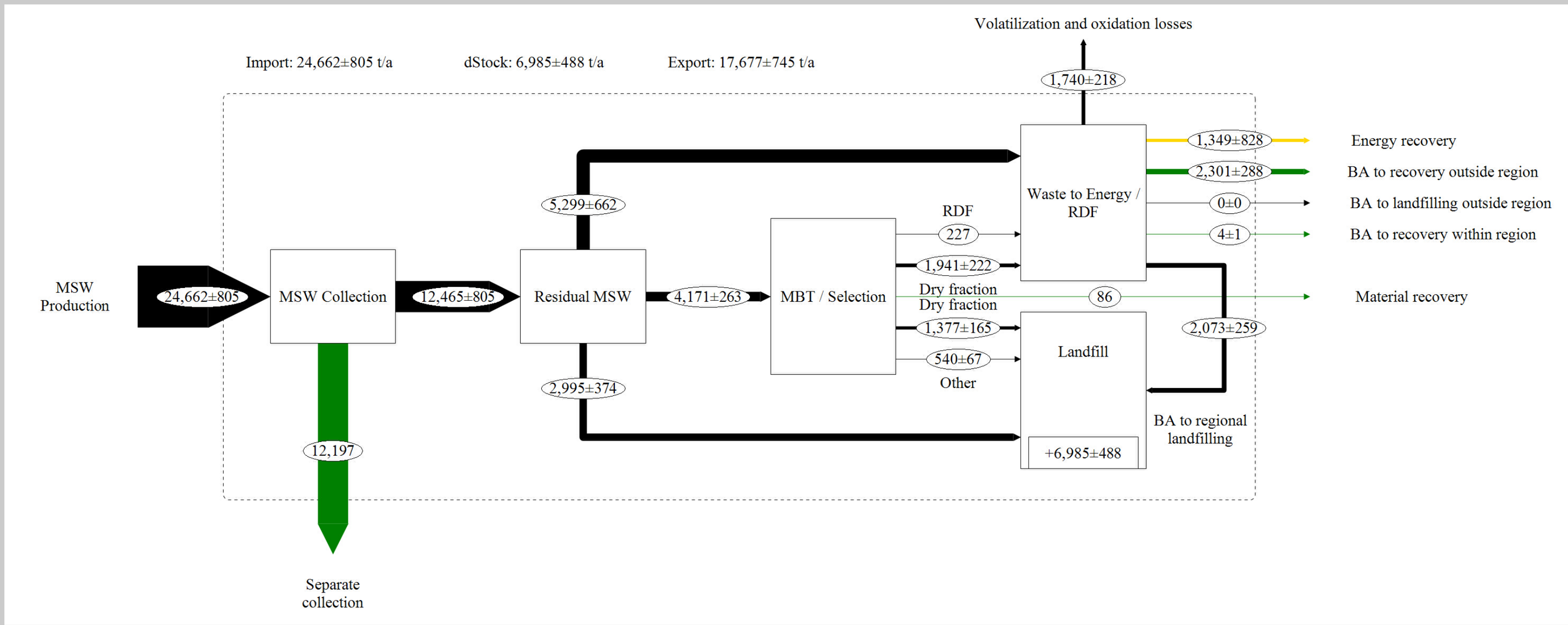
The partitioning of aluminium flows was modelled from the assumption of transfer coefficients (TCs) at each EOL process (Brunner & Rechberger 2004). More in detail, the data inventory required to set input and output TCs for collection of unsorted MSW, MBT and WtE plants, and PSTs technologies (e.g. BA recovery). The balance of mass flows was checked through a data reconciliation procedure in order to reduce uncertainties (TU Wien 2012). In **Table 18** the main TCs for the partitioning of aluminium in the model are listed.

Table 18 – Main TCs for the partitioning of aluminium assumed in the model created.

	Unit	Value
MSW management system		
Waste to Energy	%	43.0
MBT / Selection	%	34.0
Landfill	%	24.0
Main Waste to Energy output		
Losses	%	41.3
Bottom ash	%	58.7
Destination of bottom ash		
Recovery inside region	%	0.1
Landfilling inside region	%	47.3
Recovery outside region	%	52.5
Landfilling outside region	%	0.0
Main MBT / Selection output		
Dry fraction	%	79.5
RDF	%	0.6*
Aluminium recovered	kg	85.7**
Other	%	12.9
Destination of dry fraction		
Waste to Energy	%	58.5
Landfilling	%	41.5

* Estimation of aluminium content within RDF output. ** Reported in ARPA Emilia-Romagna (2011).

Figure 25– Flows and stock model for aluminium in Emilia-Romagna region in the year 2010. Quantities are expressed in tons of aluminium.



The main outcomes of the model resulted that potential of improvements should focus on (i) the recovery of about 3,000 t/y of aluminium directly landfilled; (ii) unrecovered aluminium from selection and MBT plants: of about more than 4,000 tons treated in 2010 only 86 tons of aluminium scrap were sorted and recovered. (iii) The recovery of about 2,000 t/y of metal from BA, currently disposed of; the model quantified the content of metallic aluminium in regional BA between 1.4% and 3.0%, with an average 2.2%. A similar score has been reported by the European Aluminium Association, which estimates a European average at 2.3% (Pruvost 2011). Finally, (iv) improving sorting and selection efficiencies of aluminium from BA sent to recovery plants. Overall, aluminium deposited and lost in landfill amounted at 6,985 ± 488 tons per year.

The study proposed an estimation of potential improvements in the regional waste management system at year 2020. Bearing in mind that long-time provisions are affected from several variables and they may result in erroneous considerations, the analysis aimed at discussing sources of improvements for the aluminium recovery at regional level. Therefore, the subjective choice of year 2020 should be intended as indicative span of time needed to adopt a virtuous waste management system rather than a mere temporal prediction. More in detail, such scenario models at year 2020 the full achievement of European target for avoiding the direct disposal of MSW in landfill.

The model developed assumes a conservative approach, in accordance with the European Aluminium Association provisions, regarding the content of aluminium in MSW for the next years. In first approximation, the reduction of MSW should be balanced by the increasing of aluminium containers and packaging consumed (Pruvost 2011). Then, partitioning of aluminium flows was determined by following parameters: MSW production and separate collection rate, population growth, hypothetical evolution of the regional waste management system, and wide endowment of technologies dedicated to quantitative recovery of aluminium in the light of the present operating capacity.

Figure 26 shows historical trends and estimates for MSW production and separate collection rates over years 2000-2020, resulting from the model created. Population growth was calculated as linear extrapolation of values over last ten years (**Figure**

27), while uncertainty ranges were determined for MSW production and separate collection rates. In the former case, a 5% of variation was applied to the annual quantities linearly extrapolated. In the latter case, instead, linear extrapolation was not suitable to estimate future rates since in the last three years separate collection increased significantly; furthermore, European targets claimed for years 2011 and 2012 result in 60% and 65% of separate collection. Thus, the lower limit was extrapolated by the best-fitting potential function over years 2006-2010, while the upper one the best-fitting potential function was calculated assuming to achieve the years 2011 and 2012 European targets.

Eventually, liner extrapolation was also used to simulate ideally the regional waste management system evolution to year 2020. Efficiencies of aluminium recovery treatments from the endowment of ECs in all the fleet of regional plants were, respectively, equal to 20% in selection and MBT plants, and varying between 30% and 70% for BA treatments, depending on the adoption of current or innovative separating technologies (CiAl-DIIAR 2010; Muchova & Rem 2007).

The results support the quantitative closure of aluminium cycle in the Emilia-Romagna region by reducing the deposited aluminium losses in landfill from 28% of total metal discarded in year 2010 to 4% in 2020. As showed in **Figure 28**, whether expectations will be confirmed, metal recovered from selection and MBT plants would seem to amount at an average of 1,408 t/y, while advanced technologies for BA may allow up to 1,900 t/y of aluminium suitable for recycling. Hence, up to about 4,000 tons of aluminium might be collected and recovered from residual MSW flows, to be added at more than 18.4 kt of aluminium separately collected.

Figure 26 – Historical trends and future estimates for MSW production and separate collection rates over years 2000-2020 according to the model created. Dashed lines individuate uncertainty ranges assumed.

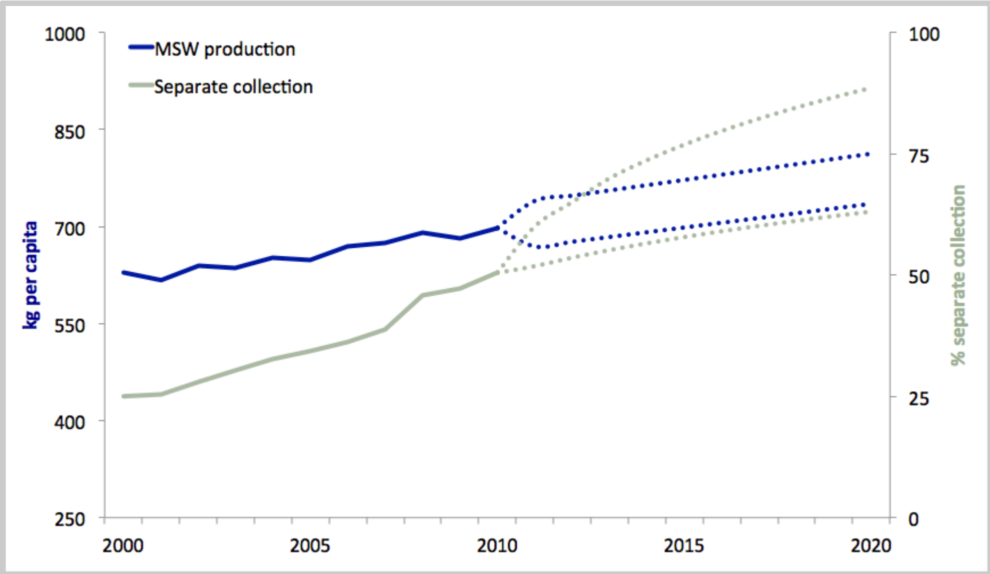
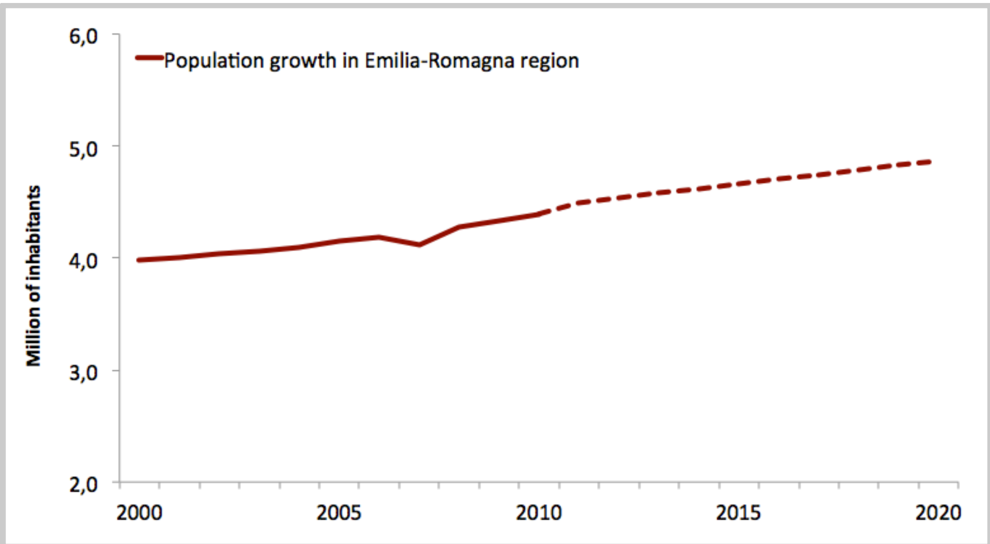


Figure 27 – Population growth in Emilia-Romagna region: historical and provisional trends. Values are in million of inhabitants.



5.2.2 Environmental impact assessment

Environmental impacts related to recovery of aluminium from the regional waste management system were then evaluated by screening LCA over MFA results for years 2010 and 2020. In accordance with goals and scopes of a screening LCA, data inventory was conducted investigating similar case studies in literature and processes embedded in the Ecoinvent 2.2 database. Impact assessment was performed in terms of *Cumulative Energy Demand* (CED), and contribution to global warming according to the method proposed by the International Panel on Climate Change (IPCC 2007).

Evaluation regarded two processes related with aluminium recovery, respectively in a MBT plant and from treating BA outputs an incinerator. A third process was also included to estimate benefits from recycling the inert fraction of BA for building purposes. In **Table 19** are summarised both the energy inputs to recover aluminium from existing MBT plants as reported in literature references consulted, and values assumed for the analysis. Specifically, the SATURN project refers to an innovative process defined as *Sensor-sorting Automated Technology for advanced Recovery of Non-ferrous metals* developed by RWTH-Aachen University (2012) and funded by the *EU Eco Innovation Funding Programme* in year 2008. Although the process is still on a pilot scale, high rates of efficiencies were here adopted as advanced technology for aluminium recovery.

Emission factors for processes modelling the recovery of aluminium from BA were assumed from (CiAl-DIIAR 2010).

Avoided impacts from the recycling of aluminium and inert fraction of BA were determined modelling as benefits from the substitution on virgin material. In the former case, the Ecoinvent process "*Aluminium, primary, at plant/RER*" and "*Aluminium, secondary, from old scrap, at plant/RER*" were used to estimate the environmental advantages from metal recycling; 78% as substituting rate was applied.

Benefits from recycling the inert matrix of BA was calculated as avoided impacts from concrete production for building appliances, as detailed in (CiAl-DIIAR 2010). In **Table 20** are reported the cumulative emission factors resulting from recovery and recycling processes investigated. In following tables and figures the final results in terms of avoided environmental impacts from the recovery and recycling of aluminium and inert, in years 2010 and 2020, are presented (**Tables 21-22**; **Figures 29-30**). These values were obtained multiplying theoretical quantities of aluminium and inert recovered from waste flows by emission factors in **Table 20**.

In general, if assumptions and trends assumed will be confirmed, the creation of a virtuous waste management system for MSW in Emilia-Romagna region would allow to cut 352 TJ of cumulative energy demand and about 21,000 tCO_2eq from a quantitative recovery of aluminium.

Table 19 – Energy-related consumptions from MBT plants as reported by the main literature references consulted, and values assumed for basic and advanced MBT technology in this study. Source: Abeliotis et al. (2012); Papageorgiou et al. (2009); RWTH-Aachen University (2012).

Author	Year	Unit	Quantity	Process
Abelotis et al.	2012	kg	0.4	Diesel
		kWh	42.4	Electrical energy
Papageourgiou et al.	2009	kg	0.8	Diesel
		kWh	80.0	Electrical energy
RWTH-Aachen	2012	kg	0.3	Diesel
University		kWh	98.5	Electrical energy
MBT basic	*2010*	*kg*	*0.8*	*Diesel*
		kWh	*80.0*	*Electrical energy*
MBT advanced	*2020*	*kg*	*1.1*	*Diesel*
		kWh	*178.5*	*Electrical energy*

Figure 28 - Flows and stock of aluminium in Emilia-Romagna region as resulting from the year-2020 model. Quantities are expressed in tons of aluminium.

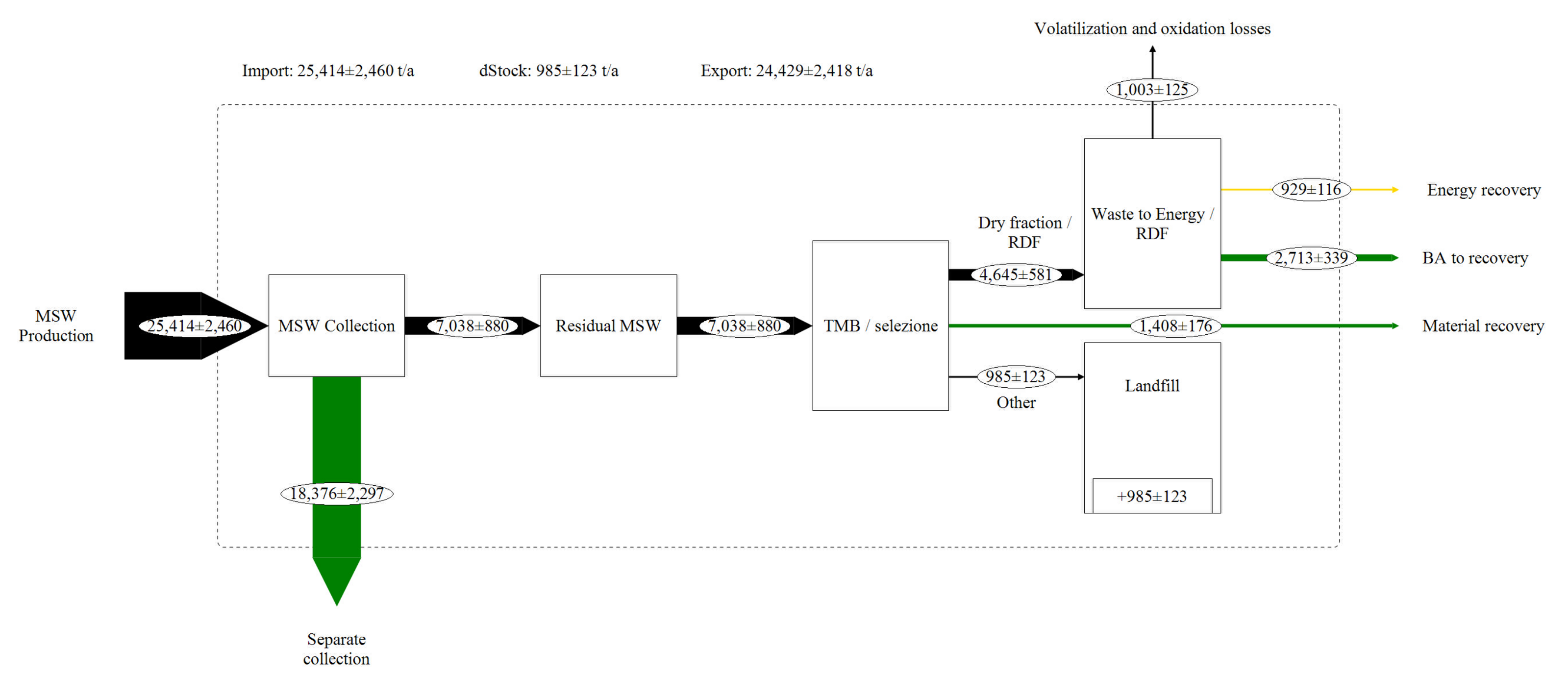


Table 20 – Cumulative emission factors from recovery and recycling processes modelled.

Process	Indicator	Unit	Emission factor
Aluminium from basic MBT	IPCC 2007	$kgCO_2eq/t$	– 10872
	CED	MJ/t	– 168544
Aluminium from advanced MBT	IPCC 2007	$kgCO_2eq/t$	– 10787
	CED	MJ/t	– 167140
Aluminium from WtE bottom ash	IPCC 2007	$kgCO_2eq/t$	– 7237
	CED	MJ/t	– 138769
Inert fraction from WtE bottom ash	IPCC 2007	$kgCO_2eq/t$	2
	CED	MJ/t	– 45

Table 21 – Quantities of aluminium and inert fraction of BA recovered according the scenarios 2010 and 2020. Values are in metric tons.

	2010			2020		
Recovery process	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**
Aluminium from MBT	85.7	85.7	85.7	1408	1232	1584
Aluminium from BA	691	605	778	814	712	916
Inert fraction from BA	77961	77961	77961	92612	81035	104188

Table 22 – Total environmental impacts for CED and IPCC 2007 indicators resulting from the recovery and recycling of aluminium and the inert fraction of BA from the models 2010 and 2020.

		2010			2020		
Recovery process	**Unit**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**
Aluminium from MBT	TJ	14.4	– 14.4	– 14.4	– 235	– 206	– 265
Aluminium from BA	TJ	– 95.9	– 84.0	– 108	– 113	– 98.8	– 127
Inert fraction from BA	TJ	– 3.5	– 3.5	– 3.5	– 4.1	– 3.6	– 4.7
Total	*TJ*	*– 114*	*– 102*	*– 126*	*– 352*	– 308	*– 396*
Aluminium from MBT	tCO_2eq	– 932	– 932	– 932	– 15200	– 13300	– 17100
Aluminium from BA	tCO_2eq	– 5000	– 4380	– 5630	– 5890	– 5150	– 6630
Inert fraction from BA	tCO_2eq	181	181	181	215	188	242
Total	tCO_2eq	*– 5750*	*– 5130*	*– 6380*	*– 20900*	*– 18200*	*– 23500*

Figure 29 – Cumulative Energy Demand results for the two models created. Values are in TJ.

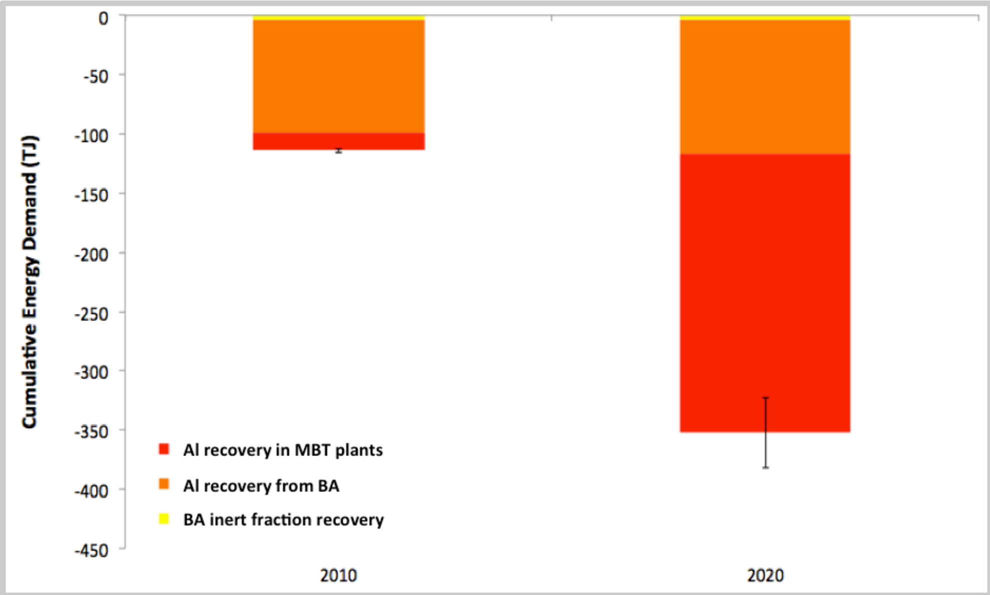
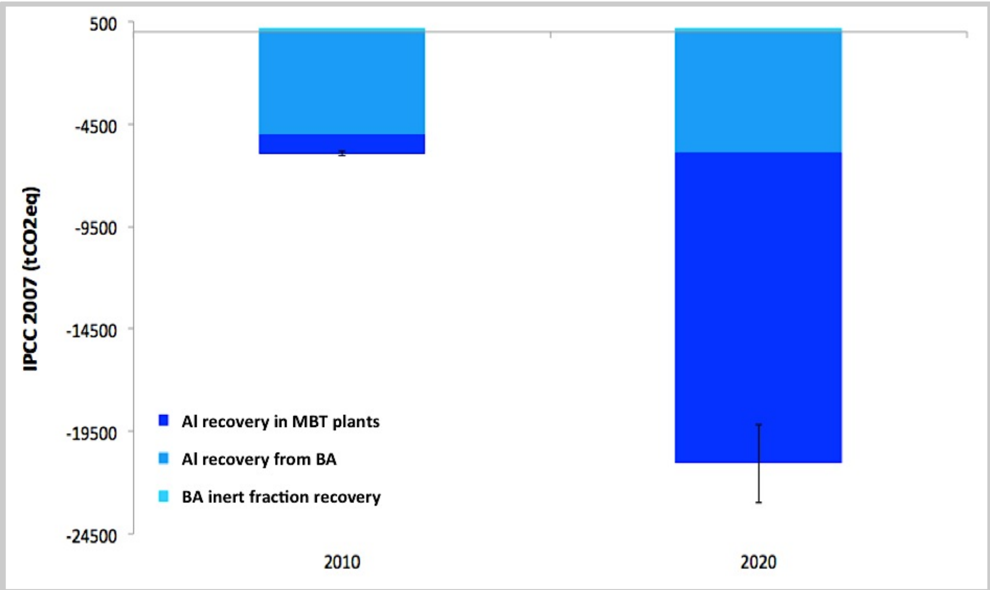


Figure 30 – IPCC 2007 results for the two models created. Values are in tCO_2eq.



5.2.3 Final considerations

The survey confirmed the regional waste management system is inadequate at present as about one third of MSW is disposed directly in landfill. Moreover, despite the operating plant capacity be able to treat all the residual waste produced, satisfying the requirement for self-sufficiency and proximity, the recovery of aluminium scrap from containers and packaging is undersized. According to the model created, the results indicated forthcoming strategies should focus on implementing aluminium-dedicated sorting and separating technologies, and for improving collection efficiencies in both MBT plants and in treatment plants for WtE bottom ash. The former would imply the widest margins for improvement, which may be even enhanced whether a regional waste management system oriented toward the EU waste hierarchy would take place, while the latter would allow achieving additional environmental advantages resulting from (i) destining aluminium forms < 50 micron to energy recovery, (ii) the possibility to recycle the inert fraction of BA in the building and construction sector, and eventually (iii) to reduce the total amount of residual waste landfilled.

It should be reminded that the research considered the impact categories for climate change and cumulative energy demand only: in this sense, further study might focus on complementary environmental effects for complementary categories of impact. Specifically, indicators for assessing rebounds to the human health should be carefully considered when evaluating the recycle of fractions from BA in order to avoid potential sources of environmental risk.

6.

Conclusions and personal considerations to the study

6.1 Overall highlights

The analysis through an integrated MFA – LCA model allowed to detect historical flows and stocks of aluminium and the resulting carbon profile from production. The results quantified in about 20 Mt the current in-use stock of aluminium in Italy, to which a reduction around 140 MtCO_2eq can be potentially associated with its exploitation in the next years. Beside, the quantification revealed trends over about the last sixty years, helped with the understanding of the national aluminium industry development. Thus, applying MFA – LCA approach over historical time spans allowed shedding a light on present and future trends, providing guidance for decision-making around the use of domestic secondary resources and for orienting industrial policy toward the goal of a low carbon society.

Today, much attention is paid to CO_2 emissions reduction from the production of primary aluminium since the Hall-Héroult process for electrolysis of alumina is the most energy-intensive step in the metal cycle. The current best technologies allow decreasing carbon intensity from the use of electricity by 5%, but up to 12% less could be achieved by adopting innovative BATs. Among the most promising options are wetted drained cathodes, which would permit the drainage of molten aluminium in continuous from the cell and thus leading to a cut of indirect emissions, or other similar solutions for reducing the energy consumption. Furthermore, carbothermic reduction processes are indicated as alternative technologies to reduce carbon footprint significantly by using low-carbon sources such as bio-coke. However, technologies based upon wetted drained cathodes as much as inert anodes, which would eliminate PFCs and direct CO_2 emission, despite of an increase in electricity consumption in same cases, they do likely seem to need five to ten years, at least, requiring for short-term interventions (Carbon Trust 2011).

In this sense, decarbonizing electrical energy sources implies maybe the strongest contribute to reduce GHG emissions from primary production of aluminium. Electricity supply is the key lever in GHG emissions from production of aluminium, as the process requires a continuous supply of electricity; inhibiting smelters' shift towards off-grid power plants based on renewable sources (e.g. wind or solar), deep reductions in the carbon intensity seems to be achievable by a connection to the national electricity grid when moving to renewable energy sources (Carbon Trust 2011).

However, in spite of European environment energy policies claims for enhancing initiatives to contrast climate change and progress have been showed, at national level implementation measures fall behind (EC 2011). Specifically, Italy still relies on thermoelectric sources for the main part in power production, with photovoltaic and wind ones underdeveloped. Hence, relying on a decarbonizing of energy sources to strongly reduce carbon leakage from the Italian aluminium production means, even in this case, to wait very likely for mid-long term rebounds.

Short-term effects could be achieved, instead, by enhancing the metal recycling industry. And, in the case of Italy, this is particularly true when considering the present capacity of secondary aluminium production. Specifically, collection of scrap is a key-issue in which Italy could act to decrease metal losses and consequently to increase the old scrap pool suitable for secondary production. Metal losses are among the highest along the aluminium cycle, following only the alumina refining stage for cumulative magnitude. Process losses for aluminium have been quantified in about 41% worldwide, mainly occurring during mechanical treatments as the removal of surface oxides. Carbon emissions cut from eliminating process scrap seems to reduce CO_2 emissions till 20 Mt on a global level, while the adoption of BATs would induce up to −12 Mt CO_2 (IEA 2008). The effect depends strictly with the share of secondary aluminium in the metal production mix (Milford et al. 2011). Considering the high ratio characterizing the Italian aluminium industry, an improvement in process efficiencies would have hence significant rebounds.

The MFA results revealed about 90% of the anthropogenic aluminium reservoir is embedded in the transportation sector, building and construction, and machinery and equipment; recovery and recycling initiatives should hence focus on these markets. Indeed, considering the expectations for an increase in the aluminium content within these end-use appliances, a wide-scale adoption of separating technologies for aluminium is necessary to avoid the loss of non-negligible amounts of aluminium scrap. Anyway, long lifetime of aluminium embedded in the transportation, and building and construction sectors affect scrap availability and so aluminium supply to the recycling industry.

On the other hand, containers and packaging is the largest supplier sector for secondary aluminium flows, although, as seen in the Chapter 5, the current recovery chain from MSW is still far from achieving a net closure of flows. The survey carried on for the Emilia-Romagna region, which is one of the most developed regions in Italy, showed a great potential for improving the waste management system that would imply relevant benefits from a carbon emissions reduction whether the European waste hierarchy was applied.

Focusing on short-term effects from enhancing the secondary aluminium industry may have in addition the advantage to decrease the country's dependence from net-imports, or in other words, to shift the attention in terms of GHG emissions from the production to the real consumption of aluminium. At global level, about 1% of anthropogenic carbon emissions is driven by the world's consumption of aluminium, whereof more than half are embodied in international trade of commodity or final products containing aluminium. Emissions embodied in European net-imports of aluminium are influenced both by carbon leakage and the consumption increasing. However, whilst the former seems to be potentially responsible only for 2-3% of all EU-ETS emissions by 2020, the latter might determine an increase by 72% in total imported CO_2 emissions. Indeed, whether aluminium production in Europe seems to continue at current levels (no new smelters are intended to be built in the continent), consumption of the metal will likely increased three or four times. The resulting disproportion between production and consumption has been discussed in Chapter 4, and here is reminded the need for standardized accounting procedures within international carbon markets.

Finally, beside measures for energy efficiencies improvement and the enhancing of recycling industry, further strategies should even take into consideration prevention principles and procedures in the production stage, such as ecodesign or Design for Environment practices. Among the most discussed today, an extension of lifetimes, dematerialization and lightweighting initiatives for present and future products seem to imply the best way for decreasing the total input for liquid metal production (Milford et al. 2011).

6.2 Recommendations

The doctoral thesis *Integration of MFA and LCA methodologies: the anthropogenic aluminium cycle in Italy* proposed a first attempt to integrate dynamic MFA and LCA methodologies to the identification and quantification of flows and stocks along the Italian aluminium cycle, and its environmental rebounds on climate change.

The intended goals of the study were ambitious and challenging, but data inventories and modelling procedures offered the chance to deal with the main obstacles resulting when approaching a long-term perspective. Despite of assumptions and uncertainties were checked and validated through sensitivity analyses, subjective choices may be arguable and affecting the results. For instance, in the MFA model, a higher level of detail for lifetimes of sub-categories for final products and end-use markets might help to increase awareness of scrap generation and hence the accuracy for the in-use stock estimation, rather than adopting different statistical distributions (Chen & Shi 2012; Müller et al. 2011; Spatari et al.

2005); similarly, in the LCA model, future considerations should be extended to the whole aluminium life cycle, or the environmental dimension should include as much impact categories as possible, and not just limited to the climate change only. Particularly, the last aspect should be carefully explicated when dealing with the recycle of particular waste types after the recovery of aluminium scrap in order to avert potential risks to the environment. As seen in Chapter 5, environmental benefits resulting from aluminium recovery and recycling, albeit relevant, cannot be enough to decide for the recycle of the inert fraction for WtE bottom ash, but need to fully comply requirements for human health protection.

However, reasons behind those choices were discussed and clearly described along the Thesis, including among others the data availability for historical inventories, the current interest in climate change profiles, or accuracy in estimating aluminium-content in a globalized way of production. Anyhow, create and apply dynamic MFA and LCA models fitting system boundaries at national level for a specific substance it requires consolidating procedures that have time as a limiting factor. In this sense, the Thesis sought to balance continuously completeness of study, time efficiency, and quality of the results, under the goals and scopes prefixed.

Lastly, keeping in mind the feature of on going improvements for MFA and LCA studies, a credit of the model is its opening toward iterative consolidation and update: improving the data quality and extending temporal and spatial boundaries investigated may be done anytime, making such model a versatile and functional tool.

In this sense, beyond the results, the model constitutes a general framework potentially applicable to analyse other materials or substances within industrial or urban metabolisms. Particularly, criticality minerals or metals for their topic importance today related with the deployment of low carbon strategic energy technologies (EC-JRC 2011) in front of a rapid growth in demand, an uncertain and non-uniform geographical distribution worldwide, and shortage of supplying, they do constitute an application field of great potential.

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Appendix A

In this section, supporting material to the analysis for stocks and flows of the anthropogenic aluminium cycle in Italy, described in the Chapter 3, is reported.

Table A1 – Decadal MFA results for the Bauxite Mining process. Values are in kt of aluminium.

Years	Bauxite Mining				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	83	12	95	0	35
1950 - 1959	260	39	299	0	381
1960 – 1969	221	33	254	1040	1558
1970 – 1979	62	9	71	3930	2810
1980 – 1989	14	2	16	4117	4062
1990 – 1999	20	3	23	5254	5324
2000 – 2009	0	0	0	5780	6388
Sum	*661*	*99*	*759*	*20121*	*20559*

Table A2 – Decadal MFA results for the Alumina Refining process. Values are in kt of aluminium.

Years	Alumina Refining				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	29	6	35	0	29
1950 - 1959	316	65	381	0	316
1960 – 1969	1293	265	1558	41	1334
1970 – 1979	2332	478	2810	– 503	1830
1980 – 1989	3372	691	4062	– 466	2906
1990 – 1999	4419	905	5324	– 1002	3417
2000 – 2009	5302	1086	6388	– 2016	3286
Sum	*17064*	*3495*	*20559*	*– 3945*	*13118*

Table A3 – Decadal MFA results for the Primary Aluminium Smelting process. Values are in kt of aluminium.

Years	Primary Aluminium Smelting				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	83	1	84	-	83
1950 - 1959	583	7	589	-	583
1960 – 1969	1121	13	1134	-	1121
1970 – 1979	2028	24	2051	-	2028
1980 – 1989	2351	27	2379	-	2351
1990 – 1999	1868	22	1889	-	1868
2000 – 2009	1875	22	1897	-	1875
Sum	*9908*	*115*	*10023*	-	*9908*

Table A4 – Decadal MFA results for the Ingot Casting process. Values are in kt of aluminium.

Years	Ingot Casting				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	82	0	83	*	82
1950 - 1959	579	3	583	*	579
1960 – 1969	1114	7	1121	*	1114
1970 – 1979	2015	12	2028	*	2015
1980 – 1989	2337	14	2351	*	2337
1990 – 1999	1856	11	1868	*	1856
2000 – 2009	1864	11	1875	*	1864

Sum	9849	59	9908	*	9849

Remark:

Net-import has been included in the Aluminium Production (Table A14).

Table A5 – Decadal MFA results for the Foundry Casting process. Values are in kt of aluminium.

Years	Foundry Casting				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	0	0	0	0	0
1950 - 1959	65	4	69	0	65
1960 – 1969	650	42	696	0	650
1970 – 1979	1816	118	1942	0	1816
1980 – 1989	2712	176	2900	0	2712
1990 – 1999	4006	260	4285	0	4006
2000 – 2009	7188	467	7688	0	7188
Sum	*16437*	*1068*	*17580*	*0*	*16437*

Remark:

Net-import was assumed negligible in first approximation as no data were found.

Table A6 – Decadal MFA results for the Rolling process. Values are in kt of aluminium.

Years	Rolling				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	0	0	0	0	0
1950 - 1959	257	12	270	– 5	252
1960 – 1969	932	45	977	– 54	877
1970 – 1979	1931	94	2024	– 122	1809
1980 – 1989	2921	141	3062	88	3009
1990 – 1999	4363	211	4575	988	5351
2000 – 2009	4970	185	5155	1521	6492
Sum	*15373*	*688*	*16062*	*2417*	*17790*

Table A7 – Decadal MFA results for the Extrusion process. Values are in kt of aluminium.

Years	Extrusion				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	0	0	0	0	0
1950 - 1959	78	2	80	– 1	77
1960 – 1969	344	9	354	9	353
1970 – 1979	1162	32	1193	– 53	1109
1980 – 1989	2275	62	2338	– 6	2269
1990 – 1999	3793	104	3897	– 281	3513
2000 – 2009	4757	116	4872	– 793	3964
Sum	*12409*	*326*	*12735*	*– 1125*	*11284*

Table A8 – Decadal MFA results for the Other fabrication process. Values are in kt of aluminium.

Years	Other				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	0	0	0	0	0
1950 - 1959	45	2	47	– 2	43
1960 – 1969	94	4	97	– 1	93
1970 – 1979	163	6	170	– 10	153
1980 – 1989	251	9	261	0	251
1990 – 1999	308	12	320	1	309
2000 – 2009	274	8	283	17	292
Sum	*1136*	*41*	*1177*	*4*	*1140*

Table A9 – Decadal MFA results for the Manufacturing process. Values are in kt of aluminium.

Years	Manufacturing				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	0	0	0	0	0
1950 - 1959	437	0	437	0	437
1960 – 1969	1973	0	1973	– 308	1666
1970 – 1979	4887	0	4887	– 1078	3809
1980 – 1989	8241	0	8241	– 1586	6654
1990 – 1999	13179	0	13179	– 3369	9810
2000 – 2009	17935	0	17935	– 4532	13403

Sum	*1136*	*41*	*1177*	*4*	*1140*

Table A10 – Decadal MFA results for the Use process. Values are in kt of aluminium.

Years	Use				
	Production	***Loss***	***Input***	***Stock***	***Stock change***
1947 - 1949	0	11	0	– 11	– 11
1950 - 1959	29	87	437	321	321
1960 – 1969	270	222	1666	1174	1174
1970 – 1979	1047	390	3809	2372	2372
1980 – 1989	2416	432	6654	3807	3807
1990 – 1999	4089	460	9810	5262	5262
2000 – 2009	6712	500	13403	6190	6190
Sum	*14563*	*2101*	*35779*	*19116*	*19116*

Table A11 – Decadal MFA results for the Collection of EOL Products and Scrap process. Values are in kt of aluminium.

Years	Collection of EOL Products and Scrap				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	0	0	0	0	0
1950 - 1959	23	6	29	0	23
1960 – 1969	216	54	270	316	532
1970 – 1979	838	209	1047	580	1418
1980 – 1989	1933	483	2416	1161	3094
1990 – 1999	3271	818	4089	2065	5336
2000 – 2009	5370	1342	6712	2719	8089
Sum	*11650*	*2913*	*14563*	*6841*	*18491*

Table A12 – Decadal MFA results for the Treatment of Scrap process. Values are in kt of aluminium.

Years	Treatment of Scrap				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	0	0	0	0	0
1950 - 1959	22	1	23	0	22
1960 – 1969	506	27	532	0	506
1970 – 1979	1347	71	1418	0	1347
1980 – 1989	2939	155	3094	0	2939
1990 – 1999	5069	267	5336	0	5069
2000 – 2009	7684	404	8089	0	7684
Sum	*17567*	*925*	*18491*	*0*	*17567*

Table A13 – Decadal MFA results for the Melting of Scrap process. Values are in kt of aluminium.

Years	Melting of Scrap				
	Production	***Loss***	***Input***	***Net-import***	***Consumption***
1947 - 1949	0	0	0	*	0
1950 - 1959	21	1	22	*	21
1960 – 1969	480	25	506	*	480
1970 – 1979	1280	67	1347	*	1280
1980 – 1989	2792	147	2939	*	2792
1990 – 1999	4815	253	5069	*	4815
2000 – 2009	7300	384	7684	*	7300
Sum	*16688*	*878*	*17567*	*	*16688*

Remark:

Net-import has been included in the Aluminium Production (Table A14).

Table A14 – Decadal MFA results for the Aluminium Production process. Values are in kt of aluminium.

	Aluminium Production			
Years	***Production***	***Input***	***Net-import***	***Consumption***
1947 - 1949	82	83	-	82
1950 - 1959	600	604	-	600
1960 – 1969	1594	1626	401	1996
1970 – 1979	3295	3375	1550	4846
1980 – 1989	5129	5290	2691	7820
1990 – 1999	6672	6936	4973	11645
2000 – 2009	9164	9559	7360	16524
Sum	*26537*	*27475*	*16975*	*43512*

Remark:

Net-import is calculated as sum of Ingot Casting (Table A4) and Melting of Scrap (Table A13) values to consider imports of unwrought aluminium for both primary and secondary metal production.

Table A15 – Metallic aluminium loss, and detail for average dissipated and deposited losses over decades.

	Metallic aluminium	***Dissipated losses***	***Deposited losses***
Years	*kt*	%	%
1947 - 1949	11	39.6	60.4
1950 - 1959	119	52.2	47.8
1960 – 1969	435	48.8	51.2
1970 – 1979	999	49.2	50.8
1980 – 1989	1620	43.3	56.7
1990 – 1999	2396	40.1	59.9
2000 – 2009	3419	37.4	62.6
Sum	*8999*	*n.a.*	*n.a.*

Remark:

Metallic aluminium column considers losses from processes IC, FC, FW, U, CES, TS, and MS only.

N.a. – not applicable.

Table A16 – Trade of main commodities for aluminium-containing products. Values are in kt of aluminium.

Years	Bauxite & alumina	Unwrought	Semis	Final products	EOL & scrap	Life cycle total
1947 - 1949	-	-	-	-	-	-
1950 - 1959	-	-	-	-	-	-
1960 – 1969	1080	316	401	– 47	– 308	1444
1970 – 1979	3428	580	1550	– 185	– 1078	4295
1980 – 1989	3652	1161	2691	82	– 1586	5999
1990 – 1999	4252	2065	4973	708	– 3369	8629
2000 – 2009	3764	2719	7360	746	– 4532	10056
Sum	*16175*	*6841*	*16975*	*1304*	*– 10873*	*30423*

Table A17 – Shares of aluminium used in each end use market for some selected years. Values are in percentage.

Year	Trans	B&C	M&E	ConDur	EE	C&P	Other
1990	28.2	30.3	9.8	9.7	7.6	11.5	2.9
2000	46.9	11.6	13.5	6.4	1.1	16.1	4.3
2005	42.4	21.3	11.7	6.4	0.5	13.5	4.1
2009	42.0	20.7	11.7	6.7	0.6	13.4	4.9

Table A18 – Amounts of aluminium from primary and secondary production, and the total unwrought metal. Values are in kt. Last column lists average shares of secondary aluminium in total production over decades.

Years	Primary	Secondary	Unwrought	Share of secondary
1947 - 1949	82	0	82	0.0
1950 - 1959	579	21	600	3.0
1960 – 1969	1114	480	1594	27.9
1970 – 1979	2015	1280	3295	38.9
1980 – 1989	2337	2792	5129	53.9
1990 – 1999	1856	4815	6672	71.6
2000 – 2009	1864	7300	9164	79.6
Sum	*9849*	*16688*	*26537*	*n.a.*

Remark:

N.a. – not applicable

Appendix B

In this section, supporting material to the analysis for GHG emissions embodied in the Italian aluminium, described in the Chapter 4, is reported.

Table B1 – Primary energy emission factors for NF metals production in Italy, some selected years. Source: ISPRA (2012)

	Primary energy								
	CO_2			CH_4			N_2O		
	Liq.	***Sol.***	***Gas.***	***Liq.***	***Sol.***	***Gas.***	***Liq.***	***Sol.***	***Gas.***
Year		*t/TJ*			*kg/TJ*			*kg/TJ*	
1990	62.39	95.09	55.33	1.00	1.50	1.00	2.00	1.50	1.00
1995	78.46	95.41	55.42	1.22	1.50	1.00	2.00	1.50	1.00
2000	72.06	94.93	55.47	2.55	1.50	1.00	2.00	1.50	1.00
2005	70.88	104.78	55.59	2.41	1.50	1.00	2.00	1.50	1.00
2006	71.36	104.41	55.67	2.47	1.50	1.00	2.00	1.50	1.00
2007	71.48	102.57	55.64	2.38	1.50	1.00	2.00	1.50	1.00
2008	72.08	98.96	56.91	2.46	1.50	1.00	2.00	1.50	1.00
2009	71.77	95.87	57.13	2.41	1.50	1.00	2.00	1.50	1.00

Remark:

Liq. – liquid fuel; Sol. – solid fuel; Gas. – gaseous fuel.

Table B2 – CO_2eq emission factors per fuel type from primary energy source; some selected years. Values are in kgCO_2eq per kg of fuel combusted.

	Primary energy				
	kgCO_2eq/kg fuel				
Year	***Coal***	***Oil***	***Natural gas***	***Diesel***	***Propane***
1990	2.29	2.55	2.55	2.69	2.58
1995	2.30	3.20	2.55	3.38	2.58
2000	2.29	2.95	2.55	3.11	2.58
2005	2.53	2.90	2.56	3.06	2.59
2006	2.52	2.92	2.56	3.08	2.59
2007	2.47	2.92	2.56	3.08	2.59
2008	2.39	2.95	2.62	3.11	2.65
2009	2.31	2.93	2.63	3.09	2.66

Table B3 – GHG emissions per thermal fuel type from power production in Italy, some selected years.

	Electrical energy								
	CO_2			CH_4			N_2O		
	Liq.	***Sol.***	***Gas.***	***Liq.***	***Sol.***	***Gas.***	***Liq.***	***Sol.***	***Gas.***
Year		*Mt*			*kt*			*kt*	
1990	63.05	28.15	15.79	2.45	0.45	0.43	0.49	0.45	0.03
1995	73.61	20.68	14.81	2.87	0.33	0.40	0.58	0.33	0.03
2000	52.95	23.16	38.73	2.05	0.38	1.05	0.42	0.38	0.07
2005	22.56	39.32	57.03	0.89	0.64	1.54	0.18	0.64	0.10
2006	22.00	39.27	58.88	0.87	0.64	1.59	0.18	0.64	0.11
2007	14.48	40.28	64.88	0.56	0.65	1.75	0.12	0.65	0.12
2008	9.74	39.57	63.46	0.38	0.65	1.67	0.08	0.65	0.11
2009	7.60	35.44	53.54	0.29	0.58	1.41	0.06	0.58	0.09

Remark:

Liq. – liquid fuel; Sol. – solid fuel; Gas. – gaseous fuel.

Table B4 – CO_2eq emission factors per sources from electrical energy production in Italy; some selected years. Values are in kgCO_2eq per MJ of power produced.

	Electrical energy			
	kgCO_2eq/MJ			
Year	***Thermal***	***Hydro***	***Nuclear***	***Renewable***
1990	0.176	0.003	0.007	0.019
1995	0.162	0.003	0.007	0.019
2000	0.153	0.003	0.007	0.019
2005	0.145	0.003	0.007	0.019
2006	0.142	0.003	0.007	0.019
2007	0.139	0.003	0.007	0.019
2008	0.134	0.003	0.007	0.019
2009	0.133	0.003	0.007	0.019

Table B5 – PFCs emissions from the primary aluminium smelting process (electrolysis and anode production), and the cumulative CO_2eq emission factor; some selected years.

	Process		
	CF_4	C_2F_6	CO_2eq
Year	*kg/t aluminium*	*kg/t aluminium*	*kgCO_2eq/t aluminium*
1990	0.86	0.18	7246
1995	0.2	0.04	1668
2000	0.14	0.02	1094
2005	0.12	0.01	872
2006	0.11	0.01	807
2007	0.14	0.02	1094
2008	0.08	0.01	612
2009	0.12	0.01	872

Table B6 – CO_2eq emissions from the transportation process per freight type; some selected years. Values are in $gCO_2eq/t*km$. Source: Eurostat (2012).

	Transportation		
	$gCO_2eq/t*km$		
Year	***Ship***	***Rail***	***Lorry***
1990	13.68	42.70	98.35
1995	13.71	35.10	92.83
2000	13.87	26.49	86.85
2005	13.92	22.95	81.95
2006	13.93	21.92	81.03
2007	13.95	21.74	79.64
2008	13.97	21.56	78.37
2009	13.98	21.37	77.20

Table B7 – CO_2eq emission factor per energy source from aluminium recycling in Italy; some selected years.

	Aluminium recycling		
	Oil	**Natural gas**	**Electricity**
Year	*$kgCO_2eq/kg$ fuel*	*$kgCO_2eq/kg$ fuel*	*$kgCO_2eq/kWh$*
1990	2.55	2.55	0.52
1995	3.20	2.55	0.48
2000	2.95	2.55	0.44
2005	2.90	2.56	0.44
2006	2.92	2.56	0.43
2007	2.92	2.56	0.43
2008	2.95	2.62	0.40
2009	2.93	2.63	0.38

Table B8 – CO_2eq emission factor per ton of aluminium recycled in Italy; some selected years.

Year	Aluminium recycling kgCO$_2$eq/t aluminium		
	Oil	***Natural gas***	***Electricity***
1990	43.65	516.41	185.39
1995	54.78	517.29	168.89
2000	39.34	430.87	119.11
2005	11.59	214.14	26.62
2006	11.67	214.43	26.10
2007	11.69	214.32	25.98
2008	11.79	219.19	24.20
2009	11.74	220.04	22.77

Table B8 – Sensitivity analysis for the cumulative results; some selected years.

Year	MtCO$_2$eq		
	Mean	***Min***	***Max***
1960	1.71	1.39	1.88
1965	1.89	1.58	2.14
1970	5.72	4.81	6.51
1975	3.30	2.73	3.68
1980	6.94	5.79	7.82
1985	9.30	7.99	10.81
1990	12.51	10.83	14.65
1995	10.12	9.33	11.28
2000	7.77	6.79	9.18
2005	10.59	9.20	12.45
2006	10.61	9.11	12.33
2007	11.39	9.85	13.32
2008	10.44	8.99	12.15
2009	7.73	6.86	9.28