Background informal technical document on techniques to reduce emissions from aluminium production

TFTEI informal background technical document

December 2020

Prepared by KIT DFIU - TFTEI Techno-Scientific Secretariat

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List of Abbreviations / Acronyms

BAT	Best Available Technique
BREF	Best Available Technique Reference Document
BaP	Benzo(a)pyrene
ESP	Electrostatic Precipitator
HF	Hydrofluoride
LCA	Life-Cycle-Assessment
NaOH	Sodium Hydroxide
PAH	Polycyclic aromatic hydrocarbon
PFC	Polyfluorocarbons
PFC	Polyfluorocarbons
PM	Particulate Matter
TFTEI	Task Force on Techno-Economic Issues
UNECE	United Nations Economic Commission for Europe

Executive Summary

This informal background document was prepared by the TFTEI Technical Secretariat and provides a compact overview on state of the art reduction techniques of air pollutants from aluminium production. After a short general description of aluminium production processes, the report provides a systematic overview of relevant air pollutants and related abatement techniques mainly based on the European BREF on Non-Ferrous Metals Industries. This document was found to be up-to-date and in line with current emission limit values in the EU. Where available, beside the techical descriptions, this document offers input to economic aspects of emission reduction regarding capital and operational expenditure.

Due to the versatility of secondary aluminium processes (recycling), the current document mainly focuses on primary production while providing a general description of recycling processes. However, the inclusion of secondary processing focusing on air pollutants could be part of future TFTEI activities.

1 Introduction

This technical synthesis provides a rapid but comprehensive understanding about the main issues of air pollution from aluminium production and what measures have been applied or can be applied to reduce emissions. It is based on the latest information available from different scientific and industry sources as well as from public institutions such as environmental agencies.

TFTEI experts, among them relevant industry associations (European Aluminium and Eurometaux), were informed about this work during the 2018 TFTEI meetings in St. Petersburg and Brussels. With regard to the most relevant reference documents, the industry experts and further experts particularly from environmental agencies around Europe confirmed that the BREF on Non-Ferrous Metals Industries (Cusano et al., 2017) to be a vast and up-to-date body of knowledge, which serves as primary reference for the report. The draft aluminium report was circulated among the aforementioned experts through an adhoc working group to receive comments and agree on a final report in the course of 2019 and early 2020.

Even though emissions from aluminium industries are comparatively well controled in the western part of the UNECE region, this is not necessarily the case in the central and eastern part of the UNECE region. This document gives brief information about stat-of-the-art in emission abatement technologies, current emission levels and economic aspects of emission abatement as far as information were available.

2 Aluminium Production

The main producer of aluminium from a global perspective is China, followed by Russia, Canada and India as displayed in Figure 1. The list of countries reveals a concentration of aluminium production in countries with comparably cheap energy resources, such as oil in the United Arab Emirates and Bahrain or renewables (hydropower) in Norway and Iceland. Aluminium production can be generally separated into primary production based on bauxite from mining activities and secondary production from obsolete material or product flows (waste and scrap). Both processing routes are introduced briefly in the following in order to provide a

basis for the further assessment of emission control techniques. While primary aluminium production has reached over 50 million tonnes per year particularly driven by increasing production in China, aluminium production in the EU has been steadily decreasing in the previous decade and is currently at a level of around 2 million tonnes per year. The majority of aluminium production in the EU comes from secondary sources with a current level of around 4 million tonnes per year. The main cost driver of primary aluminium production is electricity. As a consequence, production has concentrated in places where low-cost electricity is available such as Norway with its high share of hydropower in overall electricity supply. Furthermore, the production of secondary aluminium from scrap has increased.
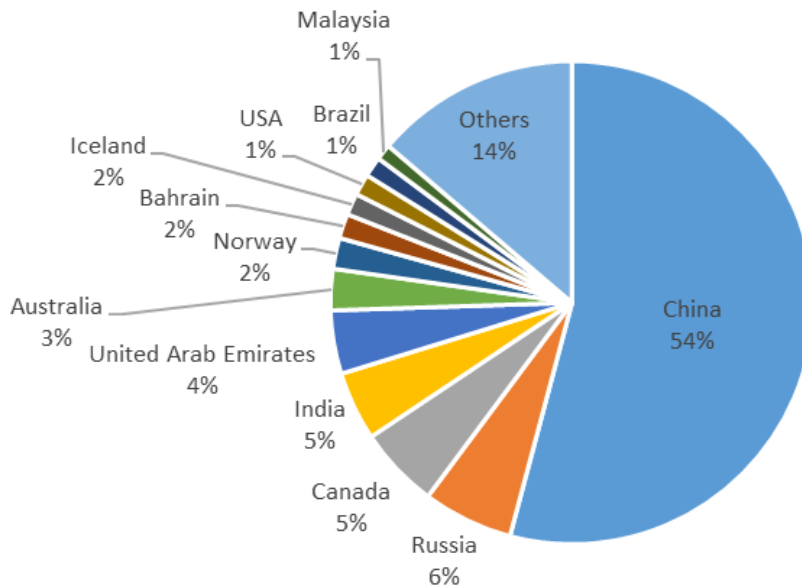
Figure 1: Main producers of aluminium worldwide in 2016 (US Geological Survey 2018a)

The European aluminium industry has made considerable efforts to reduce its electricity consumption, currently achieving an average use of 15 kWh per kg. The production and refining of secondary aluminium is much less demanding in terms of energy, accounting for a consumption per kg of about 5 % of the energy needed to produce primary aluminium (Cusano et al., 2017).

Table 1 displays a current overview of the input streams for aluminium production from primary sources (bauxite) which will be further assessed with regard to the production processes and the resulting emissions to air in section 3.

Table 1: Input streams for the production of 1 kg aluminium based on average values for Germany 2020 (Umweltbundesamt 2018)

Output:		
Aluminium	1	kg
Input:		
Aluminiumfluoride	0.018	kg
Anodes-C	0.43	kg
Alumina (Al_2O_3)	1.9	kg
Bauxite	4-7	kg
Electricity	48.2	MJ
Process heat	3.83	MJ

2.1 Primary Production

The primary production refers to the production of aluminium from natural resources. It starts with Bauxite mining, which is not considered in this report, as it is not a conventional industrial process. Bauxite reserves in the world are comparably large. The countries with the largest reserves are Guinea, Australia and Vietnam (US Geological Survey 2018b). In 2016, the highest production (mining) volumes were located in Australia and China (US Geological Survey 2018b). After the mining, the first step to be investigated is the alumina production from Bauxite via the Bayer process. This process will be briefly introduced in the following, as well as the reduction to aluminium, the anodes production and further steps of primary production.

2.1.1 Alumina Production (Bayer Process)

The aluminium production starts with the production of alumina from bauxite according to the so-called Bayer process. In the Bayer process, caustic soda (NaOH) is used to extract alumina from bauxite in digesters. High temperatures and pressures are required for this process (Kuenen et al. 2016). The International Aluminium Institute (2018b) mentions temperatures between 140°C and 280°C and a pressure of approximately 3,5 MPa (depending on the temperature). The resulting slurry contains dissolved sodium aluminate and a mixture of metal oxides, which are bauxite residues (red mud). The aluminate solution is cooled and seeded with alumina to crystallize hydrated alumina. These crystals are then calcined in rotary kilns or fluid bed/fluid flash calciners before being ready to be used for the further production of aluminium (Kuenen et al. 2016). A more detailed description of the Bayer Process is also provided by The International Aluminium Institute (2018b). Figure 2 provides an overview of the Bayer-Process as described before.
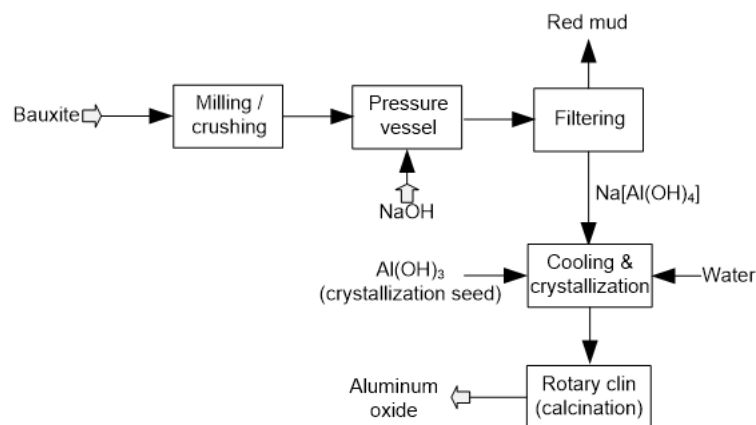
Figure 2: Basic flow-sheet of the Bayer Process which is commonly applied in industrial scale to produce alumina (Al_2O_3) from Bauxite (The International Aluminium Institute, 2018b)

According to The International Aluminium Institute (2018b), two to three tons of bauxite are required to produce one ton of alumina. The global alumina supply is around 90 million tons, 90% thereof is used in aluminium production. Alumina refineries are often located close to bauxite mines and/or ports in order to reduce transport efforts for raw materials and the final product (The International Aluminium Institute 2018b). The calcining of bauxite generates dust, SO_2, CO_2 and NO_X emissions. The abatement technique applied is a bag filter or ESP (see section 3.1).

2.1.2 Aluminium Reduction

Aluminium is produced from alumina (aluminium oxide – Al_2O_3) via the Hall–Héroult electrochemical smelting process (The International Aluminium Institute 2018b). The description in the following is primarily based on the information of The International Aluminium Institute (2018a), can, however, be complemented by further references as the Hall-Héroult process is a standard industrial process that hardly varies among different applications.

The Hall-Héroult process was developed in 1886 independently by Charles Martin Hall and Paul Héroult. It is the sole industrial method for the smelting of primary aluminium. A large electric current (direct current at an amperage of up to 600 kA) is passed through a molten mixture of cryolite, alumina and aluminium fluoride to obtain pure, liquid aluminium metal. The current is fed into a line of electrolytic cells connected in series (a "potline"). The cells and potlines vary in size and configuration from one plant to another, the fundamental electrochemical process, however, is identical. Each pot is a large carbon-lined metal container, which is used as negative electrode (cathode) in the cell. Typically a cathode lasts between 1000 and 2500 days before it needs to be replaced (The International Aluminium Institute 2018a).

The high electricity consumption is the main reason, why the primary production plants are often located at different sites than the alumina production. While alumina is produced close to the mining sites of Bauxite, aluminium is often produced at locations with a reliable and cheap electricity supply (Cusano et al. 2017).

In the cells, an electrolytic bath of molten cryolite (Na_3AlF_6) is maintained at a temperature of around $960 - 980°C$ and alumina powder is dissolved in it. Aluminium fluoride (AlF_3) is added to the solution to ensure optimal chemistry and lower the electrolyte's freezing point. Large Carbon blocks are used as positive electrodes (anodes). The electrical current, which is necessary to split alumina atoms because of the high chemical reactivity of aluminium, passes from the carbon anodes via the bath to the carbon cathode cell lining and then to the anode of the next pot in line. The dissolved alumina is split into molten aluminium (Al) and oxygen (O_2). The oxygen further reacts with the carbon in the anode blocks and forms carbon dioxide according to the following reaction:

$$2\,Al_2O_3 + 3\,C \rightarrow 4\,Al + 3\,CO_2.$$
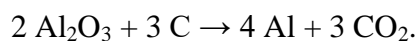
After having been split up, the molten aluminium sinks to the bottom of the cell, while the gaseous by-products evaporate to the top. The aluminium is then siphoned from the pot in a process called tapping, and transported to dedicated alloying and/or casting operations (The International Aluminium Institute 2018a). Figure 3 provides a schematic overview of the fused-salt electrolysis of aluminium.
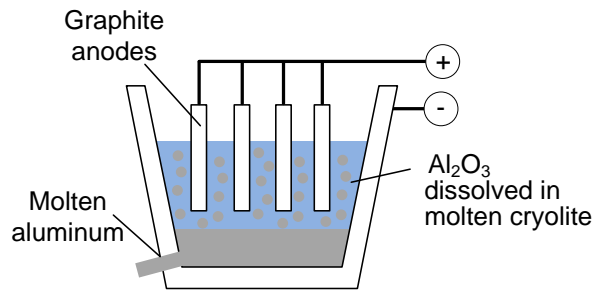
Figure 3: Basic concept of the Hall-Héroult process for fused-salt electrolysis of aluminium, which is the standard process for the reduction of alumina (Al_2O_3) to elementary aluminium metal

The main pollutants from the production of aluminium by fused-salt electrolysis are carbon dioxide and carbon monoxide (linked to anode consumption), polyfluorocarbons (PFC) in connection with the anode effect; fluorides, both gaseous and particulates; particulate matters; tars and PAHs in the case of Søderberg electrodes; SO_2; NOx and metal compounds. Fume and gas treatment installations are used to capture these pollutants and recycle fluorides for use in the smelting process. These flue gas treatment installations can also be used to remove other gaseous by-products such as sulphur dioxide and will be further introduced in section 4. A scheme of the overall primary aluminium production is displayed in Figure 4.
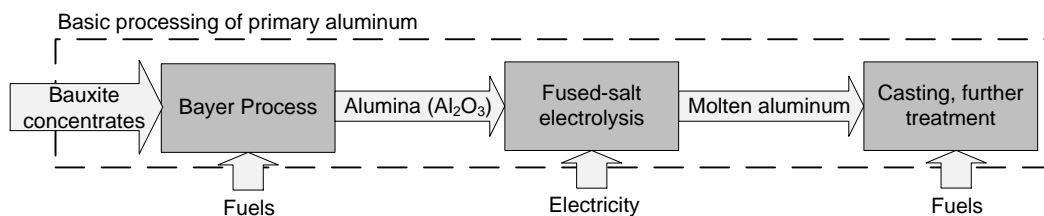
Figure 4: Process scheme for primary aluminium production (Kuenen et al. 2016)

2.1.3 Anodes Production and Smelting Techniques

The carbon anodes for the production of primary aluminium are often, but not necessarily, produced on the same site as the smelter itself (Cusano et al. 2017).

Two main types of aluminium smelting technologies with different types of anodes can be identified, the Søderberg and the prebake technology. Green paste production is the starting point for the production of all carbon anodes. All green pastes are manufactured from a mixture of calcined petroleum coke, cleaned and recycled anode butts, and coal tar pitch which acts as a binder. The petroleum cokes are a residue from the distillation of crude oils and may therefore be contaminated with substances like metals (e.g. nickel) and sulphur compounds. Petroleum coke and anode butts are ground, preheated and then mixed with pitch in heated mixers.

The Søderberg technology uses a continuous anode, which is introduced into the cell (or pot) as a paste with addition of pitch and then bakes in the cell itself. The heat generated by the reduction process is used to bake the pitch into the carbon form required for reaction with alumina, thus recycling the waste energy in the pot.

The prebake technology, as the name implies, uses multiple anodes in each cell, which are baked in very large gas-fired ovens at high temperature before being lowered into the pot. At the end of their use, the spent anodes are replaced by new anodes, with the remaining anode butts being recycled (International Aluminium Institute 2018a). Prebake anodes are often

produced nearby the smelting facility and are made from petroleum coke, pitch and recycled anode butts returned from the smelting process. These materials are mixed together in heated containers and poured into moulds. Once formed, the anodes are transferred to a baking plant. Here, the anodes are placed in a furnace at a temperature of 1120°C for a period of up to two weeks. This bakes the pitch in the mix, forming a solid block of carbon, which is able to withstand the extreme conditions inside the smelting pots. The homogenous consistency of the anode blocks allows for efficient conduction of current and an even consumption of the anode in the electrolytic cell. Subsequently, the carbon blocks are fused to a steel rod (the means by which they will be lowered into the pot and through which the electrical current is passed) with molten cast iron. The rodded anodes are then transported to the smelter potroom to be placed in the reduction cells (Primary World Aluminium, 2019). The efficiency of the prebake technology compared to the Søderberg process, combined with its lower environmental impact (see chapter 2), means that prebake smelters predominate (see Table 2), while new facilities built today generally use this technology. Besides the types of anodes, there are different alumina feed configurations that can be distinguished.

Table 2: Aluminium smelting categories and their dissemination in Europe (Cusano et al. 2017)

Cell technology	Cell type	Anode configuration	Alumina feed configuration	Acronym	Breakdown in Europe
Prebake cell	Centre worked	Vertical	Bar broken centre feed	CWPB (*)	None
		Vertical	Point centre feed	PFPB	90 %
	Side-worked	Vertical	Manual side feed	SWPB (*)	None
Søderberg cell	Vertical stud	Vertical	Manual side feed	SWVSS (*)	None
			Point feed	PFVSS	10 %
	Horizontal stud	Horizontal	Manual side feed	HSS (*)	None
			Bar broken feed		
			Point feed		
(*): No longer in operation in Europe.					

In Europe, nowadays, only the point feed technology is used, and according to the TFTEI industry experts, this is also the case for most of the plants worldwide. Table 2 provides an overview of the different technological configurations and their dissemination in Europe (Cusano et al. 2017). The most common technique, the so-called point feed prebake technique, uses multiple in-cell feeders and other computerized controls for feeding alumina into the pot. This process improves energy efficiency while reducing emissions, dust and raw material use (The International Aluminium Institute 2018a). The computerized controls are also able to reduce the so-called anode effect and hence to avoid emissions of PFCs.

Emissions to air from the anode production are hydrocarbons and PAH, sulphur dioxide, dust, and fluorides (only if anode butts are used in the anode production). The formation of SO_2 is influenced by the fuel and raw materials used, whereas the formation of NO_X is determined by the combustion temperature (cf. Cusano et al, 2017 and descriptions in the next section of this report).

2.1.4 Further Steps

After the actual production of the aluminium, further production steps are usually required to form final or semi-finished products. Exemplary processes are casting, alloying, rolling, extruding, etc. Yet these processes have a comparably low influence on the direct emissions of

aluminium production and they are not directly caused by the production of aluminium itself. Instead, they aim at producing final goods and shall hence not be investigated in more detail in this report. More information is provided by e.g. UC RUSAL (2018).

2.2 Secondary Production

Secondary production of aluminium means the process of recycling aluminium scrap into reusable aluminium. According to The Aluminium Association (2018), this process that is about 92 % more energy efficient than primary production. Kuenen et al. (2016) mention that secondary production requires only 5 % of the energy demand for primary production. In North America, about 40% of the total aluminium supply stems from secondary production (The Aluminium Association 2018). In Germany, the share of secondary production is even higher. In 2017, 58 % of the total aluminium production was secondary aluminium (WirtschaftsVereinigungMetalle 2018). However, it may not be forgotten, that discarded aluminium has to be remelted several times before it ends up in a final product. The energy consumption of these remelting steps is often ignored when setting up energy balances or life cycle analyses (Harrison and Schéele 2009).

The process of secondary aluminium production begins with extracting used aluminium from waste. In this context, scrap segregated by chemical composition or alloy has a higher value than scrap with a mix of alloys and other materials. After collecting and sorting the scrap, it is molten in a furnace at temperatures ranging from 700°C to 750°C (The Aluminium Association 2018). The type of furnace depends on the quality of the scrap and other technical parameters, as there are several options (cf. Kuenen et al. 2016). The type of raw material and its pretreatment determine the most appropriate type of furnace, flux and abatement systems to be used for a particular type of scrap, based on its size, oxide content and degree of contamination, among other factors (Cusano et al. 2017).

Rotary or reverberatory furnaces (also known as closed well, chamber or hearth furnaces) are used for melting a wide range of secondary raw materials (mainly metal scrap). Rotary furnaces can incorporate a tilting mechanism. The tilting mechanism in some cases increases efficiency for melting scrap with low aluminium content including skimmings/dross, and the amount of flux needed for these materials can also be reduced Boin and Bertram (2005). Induction furnaces and shaft furnaces are used to melt cleaner aluminium grades. Reverberatory furnaces using different types of combustion fuels often include a side well. A pumping system (mechanical or electromagnetic) is sometimes used to convey molten metal through the side well and a charging well to assist the melting of small particles. Based on Cusano et al. (2017), Table 3 provides a general overview of various types of furnaces used for secondary aluminium production.

Table 3 Different types of furnaces used for secondary aluminium production (Cusano et al., 2017)

Parameter	Unit	Rotary drum furnace	Tilting rotary furnace	Closed well or Hearth furnace		Shaft furnace	Crucible furnace	Channel induction furnace
				Single chamber	Multiple-chamber with melting bridge			
Preferred application		Secondary aluminium	Secondary aluminium	Secondary aluminium / foundries		Moulding shops	Moulding shops	Secondary aluminium
Feedstock		Al scrap	Al scrap	Ingots, Al scrap		Ingots, new scrap	Ingots, new scrap	Ingots, Al scrap
Capacity	t	Up to 150	Up to 30	Up to 180		Up to 4	Up to 6	Up to 50

Parameter	Unit	Rotary drum furnace	Tilting rotary furnace	Closed well or Hearth furnace		Shaft furnace	Crucible furnace	Channel induction furnace
				Single chamber	Multiple-chamber with melting bridge			
Melting efficiency	t/h	Up to 20	Up to 7	Up to 30		Up to 2.5	Up to 3	Up to 7
Preferred fuels		Natural gas, LPG, light fuel oil, medium/heavy fuel oil	Natural gas, LPG, extra-light fuel oil	Natural gas, LPG, extra-light fuel oil		Natural gas, LPG, extra-light fuel oil	Natural gas, LPG, extra-light fuel oil or electrically heated	Electrically heated
Ranges of energy use	GJ/t	2-5	2-2.5	2.1-4.3		2.1-3.3	0.9-7.4	around 3.6

Afterwards, the molten aluminium may be kept in its liquid state or cast into large slabs. Alloying elements may be added to the liquid aluminium in order to produce the desired metal for a specific application (The Aluminium Association 2018). The process of aluminium recycling is generally not limited to a certain number of cycles. Therefore, no loss of quality for recycled aluminium has to be expected, if impurities are sufficiently reduced during the secondary production process (The Aluminium Association 2018). Hence, it is important that impurities are reduced before the melting step. Otherwise, unwanted metal and impurities will remain in the final product. Unlike in the secondary copper production, where the final stage is an electrolysis process and primary and secondary copper are obtained in the same quality, this is not the case for secondary aluminium. Sometimes primary aluminium is used to "dilute" the secondary aluminium and thus reduce the concentration of impurities. Plant operators report different shares of primary aluminium input secondary production to reach the required quality depending on scrap composition und purity.

The schematic process is displayed in Figure 5.
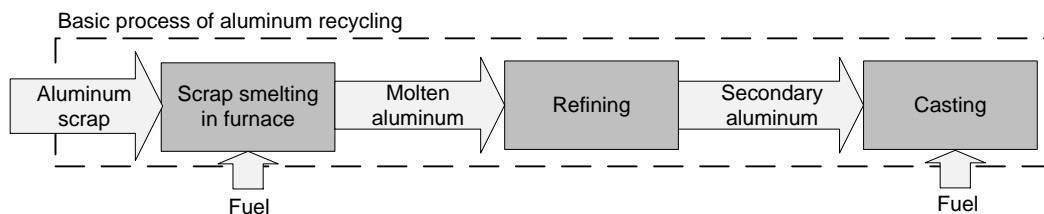
Figure 5: Process scheme for secondary aluminium production (Kuenen et al. 2016)

3 Emissions to Air

The emissions to air in secondary aluminium processing mainly depend of the type of fuel used to fire the furnace and in the impurities contained in the scrap fraction. For instance, secondary materials from the light metal fraction after conventional shredding processes of consumer products and machinery including end-of-life vehicles may contain certain amounts of plastics and further impurities which directly affects the emissions to air. Furthermore, secondary materials may contain impurities and are therefore in some cases diluted with high purity primary material. This may lead to several smelting processes, as the secondary material is usually converted into ingots or bars, which are then sold and introduced into casting processes that require a resmelting. Regarding the production of secondary aluminium, there are potential emissions to air of dust, metal compounds, Cl_2, HCl, NO_X, SO_2 and HF and products of poor combustion such as PCDD/F and other organic compounds from the melting and treatment furnaces (Cusano et al., 2017). As the overall emissions may strongly vary in secondary

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material processing, this document focuses on primary aluminium production for which air emissions can be better specified in general, while in the case of secondary production emissions strongly depend on scrap quality and impurities or contamination of input materials.

While the production of alumina from bauxite (Bayer process) mainly causes large amount of solid and liquid waste (particularly red mud which is usually landfilled), the anode smelting (e.g. in the prebake process, cf. Table 2) and subsequent electrolysis is the most energy intense step and causes significant air pollution. These emissions are determined in more detail in the following section.

3.1 Direct Emissions

In this section, we provide a general overview of air emissions occurring in the different steps of primary aluminium production, while a detailed discussion of abatement technologies is provided in section 3.

3.1.1 Calcining of bauxit

The calcining of bauxite generates dust, SO_2, CO_2 and NO_X emissions. The abatement technique applied is a bag filter or ESP (Cusano et al, 2017).

The calcining of bauxite during the Bayer Process generates dust, CO_2 and NO_X emissions also depending on the fuels used. Therefore, especially NOx emissions from existing plants show broad variations mainly depending on the type of burners (see section 3.1). Table 3 summarizes emissions from alumina production using the common abatement technologies (bag filters or ESP, see section 3 for description of technology).

Table 4: Major emissions from alumina calcining plants as average numbers per plant (Cusano et al., 2017)

Emission parameter	Emission value
Dust (kg/kg alumina)	$2\text{-}100*10^{-6}$
NO_x (kg/kg alumina)	$90\text{-}330*10^{-6}$
CO_2 (kg/kg alumina)	not reported

3.1.2 Anode Production

Emissions to air from anode production are hydrocarbons and PAH, sulphur dioxide, dust, and fluorides (only if anode butts are used in the anode production). The formation of SO_2 is influenced by the fuel and raw materials used, whereas the formation of NOX is determined by the combustion temperature (Cusano 2017).

When a new anode plant is associated with a primary aluminium smelter, the process gases from the plant can be combined with the electrolytic cell gases for scrubbing processes using alumina. In an existing plant, the gases are treated separately in a dry scrubber using alumina. A bag filter is used and the alumina collected is used in the electrolytic cells. Dust from handling and mechanical processes such as grinding, mixing and forming is collected in fabric filters. When pitch vapours are present in the gas, dry coke scrubbers, CTOs or RTOs can be used (Cusano 2017).

3.1.3 Overall emissions without emissions of Bayer process

Major air emissions in aluminium production mainly occur during the smelting and electrolysis process and the production of anodes from carbon materials. Nonetheless, aluminium industries have strongly decreased specific emissions to air in the previous decades: while primary aluminium production more than doubled over the period 1990 – 2010, total direct greenhouse gases from the production process increased by only 20% (Primary World Aluminium 2019).

Table 4 summarizes current direct air emissions from aluminium production for Germany. This does not include the aforementioned Bayer process to produce alumina (Al_2O_3) from bauxite but focusses on the subsequent smelting and fused-salt electrolysis process for the reduction of alumina to elementary aluminium metal. As emissions also depend on the smelting technologies (Søderberg and the Prebake technology, cf. section 1.1.3), the BREF document provides additional ranges of emissions for the respective smelting technology. Detailed information on emission reduction using different abatement technologies is provided in the following section. The figures in Table 4 also only refer to direct process emissions and do not include the high level of further emissions, mainly due to energy consumption which are often referred to in general Life-Cycle-Assessment (LCA) literature. Indirect emissions resulting from aluminium production are listed in the next section.

Table 5: Direct air emissions for the production of 1 kg of primary aluminium based on average values for Germany[1]

Pollutant	Direct emissions	Unit
CO	0.18	kg/kg Al
CO_2	1.4	kg/kg Al
HF	$40*10^{-6}$	kg/kg Al
Perfluoroethane	$25*10^{-6}$	kg/kg Al
Perfluormethane	$250*10^{-6}$	kg/kg Al
SO_2	0.007	kg/kg Al
PM_{10}	$706*10^{-6}$	kg/kg Al
$PM_{2.5}$	$581*10^{-6}$	kg/kg Al

In the past decades, fluoride air emissions were considered as most important pollutant from aluminium smelters. Fluorides may accumulate in vegetation and can cause damage to flora and fauna. They for instance accumulate in plants and from there enter the food chain of ruminants eating fluoride-contaminated forage. Hence, depending on local conditions, fluorides can have a serious environmental impact also to human health.

Today, due to effective gas collection from the electrolytic cells within the gas treatment center (GTC), fugitive emissions from point fed prebake cells, which produce almost 90% of the world's primary aluminium, are the main source of fluoride emissions. The balance of the emissions is collected inside the cell itself and carried away to efficient scrubbing systems, which remove particulates and gases.

Regarding the occurrence of perfluorcarbons (PFCs), computer technology controls the process down to the finest detail, which means that occurrence of the anode effect – the condition that

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[1] see https://iir-de.wikidot.com/2-c-3-aluminum-production

causes perfluorocarbons to be produced – can be minimised. Reduction in the number and duration of anode effects since the early 1990s, through effective controls of alumina and anodes, but perhaps more importantly through the growth in production using state of the art, low PFC emitting technology, has seen PFC emissions per tonne of aluminium production fall by almost 90%. A reduction in total PFC emissions of over 70%. The aluminium industry has a voluntary objective to further reduce its PFC emissions per tonne of aluminium by 50% between 2006 and 2020 (World Aluminium 2017).

This means that today, the sector's direct greenhouse gas emissions are predominantly from the consumption of carbon anodes and from fuel combustion in the refining, anode production and casting processes, both of which are a function of production, with only limited opportunity for reduction over and above incremental efficiency improvements. The industry has an objective to improve the energy efficiency of alumina refining by 10% between 2006 and 2020 at global level, a target which has almost already been met (Primary World Aluminium 2019).

Beside the electrolysis process, also the production of anodes from petroleum coke, coal tar bits or recycled anode butts may cause relevant air emissions. There are potential emissions to air of simple hydrocarbons or polycyclic aromatic hydrocarbons (PAH), SO_2, dust, and fluorides (only if anode butts, which is secondary anode material, are used in the anode production). The formation of SO_2 is influenced by the fuel and raw materials used, whereas the formation of NO_X is determined by the combustion temperature. Emissions can escape the process either as stack emissions (primary emissions) or as diffuse emissions (secondary emissions), the relative importance between the respective emission sources depends on the age of the plant and the technology used. Stack emissions are normally monitored continuously or periodically and reported by on-site staff or off-site consultants to the competent authorities. Provisions are made for fume treatment plants to be bypassed in the event of scrubber failure or during maintenance periods (Cusano et al., 2017). Table 5 summarizes the ranges of air emissions from existing anode baking plants.

Table 6: Ranges of major air emissions during anode production (Cusano et al., 2017)

Emission parameter	Emission value
Total fluoride (kg/kg anode)	$10\text{-}100*10^{-6}$
Dust (kg/kg anode)	$10\text{-}1000*10^{-6}$
SO_2 (kg/kg anode)	$100 - 6000*10^{-6}$
NO_x (kg/kg anode)	$100 - 400*10^{-6}$
BaP (kg/kg anode)	$0\text{-}3*10^{-6}$

3.2 Indirect Emissions

Indirect emissions caused by aluminium production mainly result from the large amount of energy consumption during smelting and reduction of alumina to elementary aluminium metal in the fused-salt electrolysis process. These indirect emissions are not in the focus of this document, however, as several reports do not explicitly distinguish between direct and indirect emissions, these figures are provided here as additional information in order to avoid misunderstanding. Naturally, these indirect emissions strongly depend on the energy mix, the electricity share from renewables (e.g. hydropower, wind or photovoltaics) and the fossil fuels used for power generation. Therefore, these indirect emissions can only be roughly quantified for specific countries, regions or production facilities. Table 6 provides indirect emissions

resulting from primary aluminium production in Germany. Note that only those emissions to air are listed, which are relevant for the TFTEI activities. An entire list of all emissions and waste streams caused by aluminium production has been published by the German Federal Environment Agency (Umweltbundesamt 2018).

Table 7: Indirect emissions of selected pollutants for the production of 1 kg aluminium based on average values for Germany 2020 (Umweltbundesamt 2018)

Pollutant	Indirect emissions	Unit
CO	0.011	kg/kg Al
CO_2	8.02	kg/kg Al
HF	$14.2*10^{-6}$	kg/kg Al
Perfluoroethane	$0.1*10^{-6}$	kg/kg Al
Perfluormethane	$1*10^{-6}$	kg/kg Al
SO_2	0.0184	kg/kg Al
Dust	0.02234	kg/kg Al
Hg (to air)	$85.1*10^{-9}$	kg/kg Al
N_2O	$558*10^{-6}$	kg/kg Al
NH_3	$612*10^{-6}$	kg/kg Al
NMVOC	0.0011	kg/kg Al
NO_X	0.0204	kg/kg Al

At a global level, it is reasonable that indirect emissions from electricity supply have increased as a function of increasing global aluminium production. Due to the fact that the industry's global power mix has shifted towards fossil thermal power sources, it can be assumed that the indirect emissions at global level are even higher than those listed in Table 6 even though the energy mix in some aluminium producing countries is dominated by renewable sources (e.g. hydropower in Norway). The industry has reduced the electrical energy required for the smelting and electrolysis process by 10% between 1990 and 2010 and has an objective to further improve energy efficiency by 5% by 2020 (Primary World Aluminium 2019). Nonetheless, aluminium production will remain highly energy intensive and the indirect emissions will mainly depend on the share of renewable power generation.

4 Emission Abatement Techniques (BAT)

This section mainly refers to the "Best Available Techniques (BAT) Reference Document for the Non-Ferrous Metals Industries" (Cusano et al. 2017) provided by the European Union. Additionally, information from the BAT conclusion document are summarized and presented (BAT Conclusions, 2016). We give a comprehensive overview on the state of the art of abatement technologies for air emissions from aluminium production and discuss open tasks in this context. The technology overview focusses on primary aluminium production via the processing steps described in section 1. As described before, secondary material production from recycling processes is more heterogeneous regarding impurities, emissions and process conception. Therefore, for secondary processing and emission abatement techniques, we refer to existing literature sources but do not go into detail in this document.

4.1 Emission abatement for alumina production

During the production of alumina [Al_2O_3] in the Bayer process, particularly the calcining of bauxite (see Figure 2) generates dust, SO_2, CO_2 and NO_X emissions. The abatement technique applied mainly to remove dust (particulate matter) is a bag filter or an electrostatic precipitator (ESP). As these are common techniques in process industries, they are only shortly specified in

this section, while a detailed description is provided by Cusano et al. (2017). Bag filters for treating process air emissions during alumina production via the Bayer Process are mainly used at the crushing and milling step (cf. Figure 2) where larger dust particles are produced. When using a bag filter for cleaning the process air from the milling and crushing step, the polluted air is lead through the fabric filter and the dust particles are separated. The dust is periodically removed from the filter and collected in a funnel (hopper) placed below the filtering installation. The incoming air usually does not stream directly into the filters but is lead through one or multiple dividing plates in order to achieve a good distribution of the pressure on the cloth. This also leads to a loss of kinetic energy of the process air, allowing for a pre-removal through gravity. A tapping mechanism is used to frequently remove the accumulating dust from the filter (Rijkswaterstaat Environment, 2017). The dust that falls off the cloth, is caught in the bottom of the filter and can be recycled into the process (see Figure 6).

The calcination in a rotary clin, which forms the final process step in alumina production (cf. Figure 2), causes most of the emissions to air occuring during alumina production from bauxite. No matter which calcination technology is used, fine alumina dust occurs and is generally collected by an electrostatic precipitator (ESP) or fabric bag filters. The applicability of bag filters also depends on the exhaust gas temperatures and ESP technology usually provides sufficient removal of particulate matter. However, it has to be taken into account that the performance of an ESP is proportional to the dust content in the flue gas (the higher the content, the higher the emissions), while a fabric filter enables constant levels of emission values and generally performs better than an ESP system (Cusano et al., 2017). Particularly during the shut-down and start-up process of the plant, dust emissions with an ESP treatment are significantly higher than with fabric filters. Furthermore, the fabric filter materials have been improved in previous years making their applicability at higher flue gas temperatures possible today. Table 8 gives an overview on the performance of ESP technology in comparison to bag filters for some selected plants based on the BREF document (Cusano et al. 2017). As indicated in Table 8, there are significant difference in the performance of ESP systems.

Table 8: Comparison of ESP and bag filter performance during alumina calcining in a rotary kiln for selected industrial plants in Europe (Cusano et al., 2017)

Average flue gas flow (Nm^3/h)	Abatement technology	Average emission value of dust	
		mg/Nm^3	(kg/t alumina)
220 000	ESP	68	0.1
300 000	ESP	23	0.01
107 000	Fabric filter	23	0.07
93 000	Fabric filter	23	0.05

Hence, the question whether to use ESP or bag filter technology needs to be evaluated individually for each case taking into account economic aspects described hereafter. The basic concept of an ESP is shown in Figure 7, while Figure 6 depicts the classical concept of a fabric bag filter.
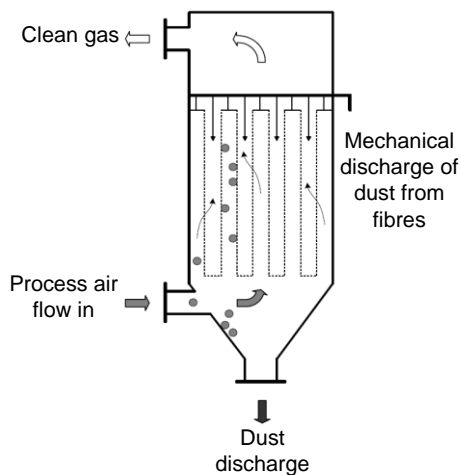
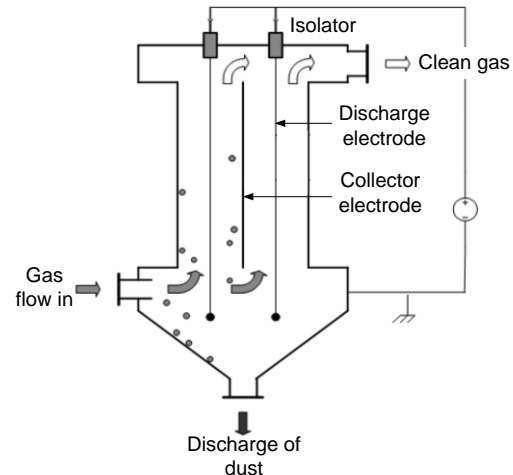**Figure 6: Principle of a bag filter (Rijkswaterstaat Environment, 2017)**

Figure 7: Concept of a dry electrostatic precipitator (ESP) (Rijkswaterstaat Environment, 2017)

SO_2 emissions are directly related to the sulphur content of the fuel used to fire the rotary clin. NO_X is also formed during the combustion process. NO_X is highly dependent on the type of fuel used, the design of the burner, the combustion chamber and temperature. The European BREF document (Cusano et al., 2017) therefore recommends the selection of low-sulphur fuels or natural gas where available and the use of low-NO_X burners. Additional process air treatment of the alumina plant is not mentioned in the BREF document (Cusano et al. 2017). The BAT conclusions published by the European Commission in 2016 the recommended techniques are as described above and no additional emission levels regarding air pollution are mentioned: *"In order to reduce dust and metal emissions from alumina calcination, BAT is to use a bag filter or an ESP"* (BAT Conclusions 2016).

From an economic perspective, the question whether to use fabric filter or ESP technology may also depend on the size of the calcination plant. While the investment costs for ESP systems increase constantly with the specific collection and, hence, with the volume flow of process air and flue gas, the investment for a fabric filter system usually starts at a higher level but does not so strongly increase with the size of the installation (Schneider 2016). However, as most alumina production sites are very large scale industrial plants, the effect of differences in investment volume of both technologies is comparatively low. It can be generally noted that the operation costs of bag filters also due to cleaning and maintenance are slightly higher as compared to those of ESPs, while the investment volume for both abatement technologies in the capacity range of common alumina plants is in the same range. However, the exact investment and processing costs strongly depend on site specific parameters and it is difficult to provide reasonable economic figures (Schneider 2016).

4.2 Emission abatement for anode production

During carbon anode production, which is an essential component of the alumina smelting and the fused salt electrolysis for gaining elementary aluminium metal, different forms of dedusting systems are applied during coke storage and handling, crushing, grinding and separation operations. Bag filters are the most commonly applied technique and achieve better dust removal efficiencies than ESPs. In cases in which ESPs are installed, an additional cyclone is installed in the upstream treatment. As the coke used for anode production contains various

hydrocarbon, additional VOC emissions, particularly polycyclic aromatic hydrocarbons (PAHs) occur in the flue gases. When pitch vapours are present in the gas, dry coke scrubbers, catalytic thermal oxidizers (CTOs) or regenerative thermal oxidizers (RTOs) can be used. The concept of dry coke scrubbers and thermal oxidizers will be shortly described in the following, while the concept of bag filters and ESPs are comparable to those presented before.

In dry scrubbing technology, powder is introduced and dispersed in the waste gas stream. The material reacts with gaseous species or acts as an adsorbent to form a solid, which has to be removed by filtration (bag filter or electrostatic precipitator). The use of a reaction tower improves the removal efficiency of the scrubbing system. For treatment of process gases in anode production, the scrubbing agent is usually coke powder. The concept of a dry scrubbing system is illustrated in Figure 8.
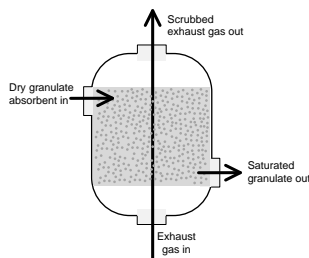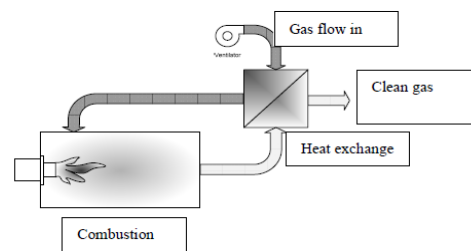

Figure 8: Principle of a dry scrubbing system

Figure 9: Concept of a recuperative incinerator (Rijkswaterstaat Environment 2017)

The recuperative incinerator is a thermal incinerator with an additional heat exchanger. With the help of the heat exchanger the untreated air is pre-heated with the combustion gasses causing up to 80% of the heat released to be available for use. In a regenerative incinerator two or more ceramic beds store the heat of the cleaned gas and preheat the untreated gas. The thermal efficiency can reach up to 97%.

Main emissions of VOC and fluorides occur during the baking process in anode production (see also section 1.1.3). Since some anode plants accept recycled anode butts in their feed (containing significant amounts of fluorides), provision has to be made for fluorides recovery in a post-treatment phase. Regarding the treatment of flue gases, it has to be distinguished between stand alone plants in which only anodes are produced and plants in which alumina production and particularly the electrolysis of alumina to elementary aluminium are also conducted. When a new anode plant is associated with a primary aluminium smelter, the process gases from the plant can be combined with the electrolytic cell gases for scrubbing processes using alumina (see emission abatement for primary aluminium production in section 3.3). However, in most existing plants, the gases are treated separately in a dry scrubber using alumina. A bag filter collects the resulting alumina dust, which is reintroduced to the electrolytic cells in the fused salt electrolysis process.

The BAT conclusions provide additional information on BAT during anode production regarding abatement of emissions to air distinguishing between the paste plant, in which anode materials are formed (generally from petroleum coke, coal tar bits or recycled anode butts) and the baking plant (see process description in section 1.1.3). A summary of the recommended BAT and related emission levels reported in the BAT conclusions is provided in

Table 9 and 9. A discussed in the process description, anode baking is often integrated with a primary aluminium smelter, which affects the treatment of exhaust air and the related emission

level. While Table 10 only refers to a standalone plant, emission levels of an integrated smelter plant and the related BAT is provided in the following section in combination with the smelting and fused salt electrolysis process.

Table 9: BATs and related emission levels for anode production (paste plant) reported in the European BAT conclusions (BAT Conclusions 2016)

BATs for the paste plant (anode forming)		

• In order to reduce dust emissions to air from a paste plant (removing coke dust from operations such as coke storage and grinding), BAT is to use a bag filter.
• In order to reduce dust and PAH emissions to air from a paste plant (hot pitch storage, paste mixing, cooling and forming), BAT is to use one or a combination of the techniques given below:
   1. Dry scrubber using coke as the adsorbent agent, followed by a bag filter
   2. Regenerative or recuperative thermal oxidiser
   3. Catalytic thermal oxidiser		

Emission parameter	Process	Emission level mg/Nm^3
Dust	Hot pitch storage, paste mixing, cooling and forming	2-5, as a daily average
Benzo(a)pyrene (BaP as an indicator of PAH)	Removing coke dust from operations such as coke storage and grinding	0.001-0.01, as an average over the sampling period

Table 10: BATs and related emission levels for anode production (stand-alone baking plant) reported in the European BAT conclusions (BAT Conclusions 2016)

BATs for the baking plant (anode baking)		
In order to reduce dust, PAH and fluoride emissions to air from a baking plant in a stand-alone anode production plant, BAT is to use a pre-filtration unit and a regenerative thermal oxidiser followed by a dry scrubber (e.g. lime bed).		
Emission parameter	Process	Emission level mg/Nm^3
Dust	Introduction of anode paste in the baker and baking process	2-5, as a daily average
Benzo(a)pyrene (BaP as an indicator of PAH)	Baking at high temperature of around 1120°C for up to 2 weeks	0.001-0.01, as an average over the sampling period
HF	Baking at high temperature of around 1120°C for up to 2 weeks	< 3, as a daily average

Regarding economic aspects of air emission abatement for anode production, the BREF document (Cusano et al., 2017) provides several information on existing plants. A dry scrubbing unit with bag filter and a capacity of 50 000 m^3/h causes investments of around 1.4 million €. The investment costs for an RTO system with a capacity of 50 000 m^3/h would be around 4 million €, when using the RTO only for the higher pitch content gas and a dry scrubber for the lower concentration gas could reduce the overall investment to around 3 million € at the same capacity level. The yearly maintenance costs of such a system would be at around 100 k€. The overall investments for the abatement system in a production site with a capacity of 130 kt/a of baked anode using a dry scrubber with subsequent bag filter is reported at approximately 10

million € (Cusano et al., 2017) while the investments for an RTO/CTO system would be significantly higher.

4.3 Emission abatement for primary aluminium production

The smelting and subsequent electrolysis of alumina to elementary aluminium metal is the most energy consuming process step and causes the highest direct and indirect emissions as already quantified in section 2. Figure 10 summarises the major air emissions resulting from process air and pot room ventilation (electrolysis chambers), while the degassing and casting of molten aluminium in further processing causes comparatively little emissions.
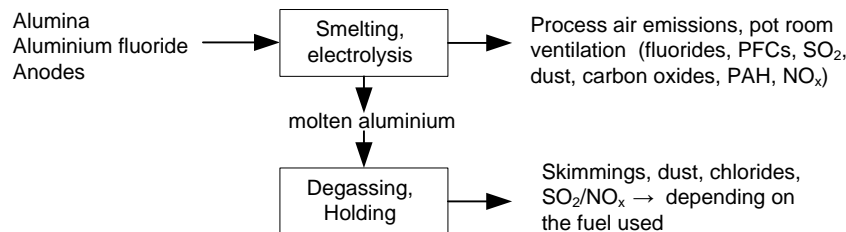
Figure 10: Overview of major air emissions in aluminium production (Cusano et al., 2017)

As discussed in section 2.1, the air emissions from primary aluminium production with highest environmental impact are perfluorocarbons. PFCs may occur in the primary aluminium reduction process, during so called anode effects. An anode effect is a specific process upset, in which an insufficient amount of alumina is dissolved in the electrolyte bath. This causes the voltage in the pot to be elevated above the normal operating range, resulting in the emission of gases containing the PFCs tetrafluoromethane (CF_4) and hexafluoroethane (C_2F_6). As already described in sections 1.1.3 and 2.1, the techniques of point feed prebaked cells (PFPB) and point feed vertical Søderberg cells (PFVSS) in conjunction with automatic multiple point feeding of alumina, computer control of the electrolytic process based on active cell databases and monitoring of cell operating parameters and automatic anode effect suppression, have strongly reduced the occurrence of anode effects and, hence, the generation of PFCs. In addition, scrubbing systems as described in the following are used to reduce flouride emissions.

Fluoride "scrubbing systems" use alumina to extract gaseous fluoride from pot gases. This "activated" alumina, which contains the residual fluoride, is then used as a feed for the reduction process. Thus the fluoride is recycled through in a closed loop system. A significant share of smelters worldwide employ such scrubbing systems, however, due to the increasing number of production facilities in China and a lack of knowledge about their exact technology, it is difficult to quantify the overall usage of fluoride scrubbing systems at global level. Nonetheless, this technology is common standard in Europe (Cusano et al. 2017) and is defined as BAT in the European conclusions document (BAT Conclusions 2016). In European aluminium production, optimum gas collection from the electrolytic cells and specific workplace training on minimisation of fugitive fluoride emissions, as well as improvements in cell technology have driven a 50% reduction in total fluoride emissions per tonne of aluminium production between 1990 and 2010 (around 80% since the 1960s). The industry has committed to a further reduction of at least 35% between 2006 and 2020 (World Aluminium 2017). An alternative to capture fluorides is the scrubbing with crushed limestone or a fresh water scrubbing in which water runs in a closed loop and regular purges are conducted to maintain the required operational conditions. An advantage of wet scrubbers is the additional removal of SO_2 from flue gases. However, wet scrubbers are mainly used in Scandinavian aluminium industries as they require

large amounts of water and related wastewater treatment as well as cross-media effects. Hence, the applicability of wet scrubbing systems may be limited in case of very high off-gas flow rates (due to the significant amounts of waste and waste water generated) and in arid areas (due to the large volume of water necessary). In order mitigate SO_2 emissions, the use of low level sulphur materials for anode production is recommended[2]. Table 7 shows the currently highest potential for emission reduction in primary aluminium production using wet scrubbing systems for the Søderberg process and boosted suction systems (BSS) to increase the amount of treated process air in the Prebake (PFPB) process in combination with a dry scrubber and a bag filter.

Table 11: Total emissions to air from primary aluminium production (smelting and electrolysis) with the currently highest level of abatement technologies which is wet scrubbing for the Søderberg process and BSS in combination with dry alumina scrubbing for the Prebake process (Cusano et al., 2017)

Emission parameter	Prebake (PFPB)	Søderberg
Total fluorides (kg/kg Al)	$250\text{-}450*10^{-6}$	$300\text{-}600*10^{-6}$
Dust (kg/kg Al)	$200\text{-}600*10^{-6}$	$800\text{-}1400*10^{-6}$
SO_2 (kg/kg Al)	0.01-0.025	not reported

In addition to the BREF document (Cusano et al., 2017), the BAT conclusions provide relevant information on emission levels (BAT Conclusions, 2016), particularly regarding regulations for new plants and the comparison to existing production facilities. Even though the Søderberg process is of minor importance for new plants (see technical description in section 1.1.3), the BAT conclusions provide technical information and related emission levels to air for both technologies. The BATs regarding air emissions are summarized in Table 12, while Table 13 gives an overview of related emission levels for existing and new plants.

Table 12: List of BATs for air emission abatement provided in the European BAT conclusions (BAT Conclusions, 2016) for both the Prebake (PFPB) and the Søderberg process. Associated emission levels are summarised in Table 13

Prebake (PFPB)	Søderberg
• Automatic multiple point feeding of alumina • Complete hood coverage of the cell and adequate off-gas extraction rates (to lead the off-gas to the scrubber followed by a bag filter) taking into account fluoride generation from bath and carbon anode consumption • Boosted suction system connected to the scrubbing system • Minimization of the time for changing anodes and other activities that require cell hoods to be removed • Efficient process control system avoiding process deviations that might otherwise lead to increased cell evolution and emissions (automatic anode effect suppression) • Use of a programmed system for cell operations and maintenance • Use of established efficient cleaning methods in the rodding plant to recover fluorides and carbon	• Use of paste with a pitch content between 25 % and 28 % (dry paste) • Upgrade the manifold design to allow closed point feeding operations and improved off-gas collection efficiency • Alumina point feeding technique. Point feeding of alumina avoids the regular crust-breaking (such as during manual side feed or bar broken feed), and thus reduces the associated fluoride and dust emissions. • Increased anode height combined with the treatment in a scrubber using alumina as absorbent followed by a bag filter. An increased anode height helps to achieve lower temperatures in the anode top, resulting in lower emissions to air.

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[2] Anodes containing less than 1,5 % sulphur as a yearly average can be produced by an appropriate combination of the raw materials used. A minimum sulphur content of 0,9 % as a yearly average is required for the viability of the electrolysis process (BAT Conclusions, 2016).

Prebake (PFPB)	Søderberg
• Storage of removed anodes in a compartment near the cell, connected to the treatment in the scrubber system, or storage of the butts in confined boxes (not applicable to existing plants, only for new facilities)	• Anode top hooding when high current density anodes are used, connected to the scrubber system

As illustrated in Table 13, the emission levels for the approval and permission of new production facilities in the European BAT conclusions (BAT Conclusions, 2016) are in the upper range of the emission levels reported for Point Centre Feed Prebake (PFPB) processes in the BREF document (cf. Table 11) regarding dust and fluorides. This underlines the up-to-dateness of the BREF document, which was also confirmed by industry experts during the survey work. Only in the case of SO_2 emissions for which wet scrubbing systems are the most efficient abatement technology (while dry scrubbing systems are commonly applied), emission levels reported in the BREF document for existing plants in some cases exceed the levels appointed in the BAT conclusions.

Table 13: BATs and associated emission levels for aluminium smelting and electrolysis collected from the electrolytic cells and roof vents as reported in the BAT conclusions (BAT Conclusions 2016). Wet scrubbers are only used in case of very high off-gas flow rates, due to cross-media effects and in case of the availability of large volumes of water.

Emission parameter	BAT defined in the European conclusions	Emission level kg/kg Al	
		existing	new plant
Dust	Dry scrubber using alumina as the adsorbent agent followed by a bag filter (and potentially an additional wet scrubber if applicable)	$1200*10^{-6}$	$600*10^{-6}$
Total fluorides, mainly HF		$600*10^{-6}$	$350*10^{-6}$
SO_2	Use of low-sulphur anodes and wet scrubbing system if applicable	0.0025-0.015	0.0025-0.015
Perfluorocarbons, mainly Perfluoroethane and Perfluormethane	• Automatic multiple point feeding of alumina • Automatic anode effect suppression • Computer control of the electrolysis process based on active cell databases and monitoring of cell operating parameters	not reported	not reported

Regarding economic aspects of air emission abatement in the smelting and electrolysis process, Cusano et al. (2017) give the example of a possible new gas treatment centre under consideration in France in 2012. For a modern PFPB smelter with a capacity of 260 000 tonnes of aluminium per year, the total investment costs for the emission abatement technology using common scrubbing systems would be at around 120 million € (two units costing 60 million € each). The cost for retrofitting an existing plant would be higher. If a BSS is included (see reference figures in Table 7), an extra investment of 12 million € would be needed to add a second network and set of valves, filters and fans in a new installation; and twice that amount or more for a retrofitting configuration (Cusano et al. 2017).

5 Conclusions

The goal of this document was to provide a brief overview of state of the art in primary aluminium production and related air emission levels as well as corresponding emission abatement technologies based on the current European BREF document for nonferrous metals (Cusano et al. 2017) and additional various literature sources and expert judgments. Comparison of emission levels to the European BAT conclusions showed that the BREF document is up-to-date and provides vast information on respective abatement technologies. This was also confirmed by industry staff and additional experts from environmental agencies. Only in the case of SO_2, dry scrubbing systems for aluminium smelters as described in the BREF document might exceed current emission levels stated in the BAT conclusions. However, SO_2 emission levels strongly depend on the sulphur content of the anodes and may therefore also be regulated by the use of low-sulphur anodes.

Regarding costs for abatement technologies, this report provides several investment figures, mainly from the BREF document. However, as costs or investment volume are generally reported for the plant as a hole and not for specific abatement technologies, it is difficult to provide resilient data and information in this issue.

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