Trace elements emission in iron ore sintering: A review

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ABSTRACT

Iron ore sintering is the main process of fines agglomeration for ironmaking with relevant particulate matter and trace element emissions of integrated steel plants. Due to the high environmental and human health impacts caused by trace elements, legislations are becoming more stringent for regulating emissions of the trace elements. This review consolidates several studies in the sector, focusing on the sources, behavior, and technologies employed to control emissions of trace elements during iron ore sintering. The emission of trace elements is strongly dependent on the characteristics of raw materials, operating parameters and technologies used for their abatement. Iron ore is the main responsible material for emissions of trace elements during sintering as it undergoes a series of physicochemical transformations throughout the process. In general, trace elements may remain in the solid-state, volatilize during combustion and recondense in the cleaning system, or remaining in the gaseous state during the flue gas treatment. Although most trace elements are retained in air pollution control devices in the form of particulates, volatile and semi-volatile elements (As, Se, Hg, Cd, etc) can remain as gases or concentrate in the finer particulates. Modern technologies have demonstrated high efficiency in removing these elements from the flue gases.

1. Introduction

Trace elements (TEs) are chemical elements found in very low concentrations, usually in the order of milligrams per kilogram (mg/kg) or parts per million (ppm) (Nimir, 2006). Although some elements are essential for human health and ecosystems, they are all considered to be toxic at some threshold concentration, including some being considered carcinogenic to humans (As, Cd, Cr (VI), Ni) (Van de Velden et al. 2008; Lin et al. 2010; Filizok and Görgün, 2019).

TEs can behave in different ways when submitted to high-temperature processes (like iron ore sintering and coal-fired power plant, that occurs typically up to 1000°C). Some of them tend to remain in the solid-state (V, Cr, Mn, Co, Ni), others partially volatilize, and may recondense again before being emitted to the atmosphere (Zn, Pb, Cd, As), and some volatilize remaining in vapor phase until emitted to the atmosphere (Hg, F, Cl, Br)(Xu et al., 2004; Sekine et al., 2008). The elements that volatilize can be emitted in the form of gas or fine particles, especially particulate matter smaller than 10 micrometers (PM$_{10}$) and smaller than 2.5 micrometers (PM$_{2.5}$), being able to travel great distances in the atmosphere before depositing, contaminating the soil and water, and entering the food chain through vegetables and fish (Senior, 2000; Tian et al., 2014; Boente et al., 2017; Zhao,S. et al., 2017a). Some elements are chemically stable and tend to accumulate in human tissues and the environment (Van de Velden et al. 2008). With increasing concerns for environmental preservation and control of the factors that impact human health, environmental legislations have also become increasingly strict. As a result, some regulations established limits to the emissions of some pollutants that were not previously controlled, such as TEs and fine particles. European legislation, for example, has limits for the concentration of PM$_{2.5}$, lead, arsenic, cadmium and nickel in the air (COTEU, 2008). In the United States, the Environmental Protection Agency (EPA) has air quality concentration standards for PM$_{2.5}$ and lead (USEPA, 2013, 2008). Following other countries and the World Health Organization (WHO), in 2018 Brazil also defined its concentration limits in the air for PM$_{2.5}$ and lead (Siciliano et al., 2020). The same occurred in China by setting its current limits for PM$_{2.5}$ and lead in 2012 (CMEP, 2012).

In 2005 WHO defined air quality guidelines for four pollutants,

including PM_{10} and $PM_{2.5}$ (WHO,2005). In 2015 WHO selected 32 pollutants, classifying them into 4 groups according to the need for a systematic review of standards. Among TE-related pollutants, PM was classified in group 1, cadmium, chromium and lead in group 2, arsenic, manganese, platinum and vanadium in group 3, and mercury in group 4, with group 1 being the highest priority while group 4 lowest priority (WHO, 2015).

Iron and steel production is considered to be one of the main industries in the world, generating an approximate value of $ 900 B per year, and practically all products used in the modern world either have some element of steel or need steel during their production process (Zhou et al., 2020). World steel production reached 1,869.9 Mt in 2019 (WSA,2020a), where around 70% of global production relies directly on inputs of coal via the BF/BOF (Blast Furnace and Basic Oxygen Furnace) route. China represents more than half of world production and it is mainly based with around 90% on the BF/BOF processes (Fernández-González et al., 2017a).

Sintering and pelletizing are iron ore agglomeration processes that allow the use of ores with a size lower than 10 mm in the blast furnace, without impairing the furnace's permeability (Fernández-González et al., 2017a). Sintering is the most widely used agglomeration process in the world for ferrous burden preparation to the blast furnace, as sinter is cheaper than pellet and has better reducibility characteristics than lump. Typically, the sinter corresponds to more than 60% of the ferrous load of blast furnaces in eastern Asia (Lu and Ishiyama, 2015).

Since the production of iron and steel is carried out at high temperatures and with a large quantity and variety of raw materials, this industry has a high polluting potential. Among all the processes involved in the production of steel, sintering is the main emitter of polluting gases and particles that contain TEs, being responsible for approximately 45% of the total emissions of an integrated steel plant (Menad et al., 2006; Chen et al., 2009; Remus et al., 2013; Abreu et al., 2015; Olmez et al., 2016; Ji et al., 2017a). The entire context presented, given the continuous growth in steel production in the world (WSA, 2020a), strongly justifies the study of emissions of TEs in the iron ore sintering process. Given the scarcity of specific research in the area, this work collects and consolidates the still insipient knowledge on the subject, complementing it with works on TEs in other high-temperature industrial processes, such as coal-fired power plants and combustion of sewage sludge.

2. Emissions of trace elements in high-temperature industrial processes

Although there are not many studies on the emission of trace elements in the iron ore sintering process, works regarding these emissions in other high-temperature processes are more common. Understanding the TEs behavior in other contexts can greatly assist to elucidate their behavior in the iron ore sintering, especially when they have common raw materials or process conditions.

2.1. Coal-fired power station

Coal-fired power plants are responsible for almost 40% of all electrical energy generated in the world and are considered as one of the main anthropogenic sources of TEs emissions, in addition to SO_2, NO_X and PM (Zhao et al., 2016; BP, 2019). A report published by the Environmental Protection Agency (EPA) points to the mercury emitted by coal-fired power plants as the hazardous air pollutant with the greatest potential to damage public health (Senior, 2000). Thus, several studies address the emissions of TEs in this process.

Similar to iron ore sintering, emissions of TEs in coal-fired power stations depend on the concentration of the element in the inputs (in this case coal), the chemical and physical properties of the element, process conditions and parameters (especially firing temperature and atmosphere) and the emission control equipment used (Meij and te Winkel, 2007; Pudasainee et al., 2010; Nalbandian, 2012; Zhou et al., 2020).

Several authors have characterized diverse samples of coal from different regions of the world. Table 1 presents a summary of the concentrations of TEs identified by some authors (Zhao et al., 2016; Zhu et al., 2016; Zheng et al., 2018, 2017).

In coal, lead is mainly associated with organic compounds. It volatilizes at approximately 850°C in the form of $PbO_{(g)}$. Between 730°C and 820°C, it can be transformed into $PbO_{(s)}$ and below 730°C, solid lead sulphate is formed. The presence of chlorine can result in the Pb remaining in the form of vapor (Zhao et al., 2018). On the other hand, most of the chromium in coal is associated with clay minerals content in the inorganic matter. Pb enriches in the ash due to crystallization of Cr_2O_3 and the interaction of chromium with hematite. In pulverized coal combustion, the high concentration of toxic Cr^{6+} in fly ash requires attention (Zhao et al., 2018).

Zinc and cadmium appear in coal mainly in the form of sphalerite. Zinc has low volatility, leading to concentration mainly in the bottom ash and fly ash, and can be found in the forms of $2ZnO \cdot SiO_2$, $ZnO \cdot Fe_2O_3$ and $ZnO \cdot Al_2O_3$. Cadmium, on the other hand, volatilizes during combustion. With the cooling of flue gas, the gaseous cadmium compounds, such as $CdCl_2$, condense below 580°C, forming particulates (Zhao et al., 2018).

Arsenic in coal is normally associated with pyrite or other sulphides (Zhao et al., 2018, 2016). During combustion, arsenic mostly volatilizes in the forms of $AsCl_3$ and AsO_2, depending on the chlorine content in the coal (Zhao et al., 2018). According to Zheng et al. (2018), the presence of Cl in coal contributes to emission of As, while the presence of S inhibits this emission. The presence of Fe or Ca results in the formation of $FeAsO_4$ or $Ca_3(AsO_4)_2$ on the surface of fly ash. Studies indicate that 0.2-52% of arsenic emitted as gas is in the form of As_2O_3. As^{3+} corresponds to 10% of arsenic in fly ashes (Tian et al., 2013; Zhao et al., 2018; Zheng et al., 2018).

The main form that mercury is found in coal is linked to organic matter or sulphides. There are three ways in which Hg can be emitted from coal-fired power plants, elemental mercury (Hg^0), gaseous oxidized mercury (Hg^{2+}), and particle-bound mercury (Hg^P) (Pudasainee et al., 2010; Tian et al., 2014). During firing in the furnace, mercury is emitted in its elemental form (Hg^0). As the temperature of the gas reduces, mercury can react with several elements, such as O_2, HCl, Cl_2, SO_2, NO, NO_2, N_2O, NH_3 or H_2S, forming oxidized mercury (HgO, Hg_2Cl_2, $HgCl_2$). Oxides of transition metals, such as CuO and Fe_2O_3, as well as carbon, can also cause oxidation of mercury. This process results in formation of particulate mercury (Zhao et al., 2018; Zheng et al., 2018). According to Zheng et al. (2018), presence of Cl in coal results in the formation of $HgCl_{2(g)}$, resulting in the adsorption of Hg in fly ashes. The same occurs in the presence of Fe_2O_3, which due to its strong catalytic oxidation activity for Hg^0 in flue gas, contributes to the concentration of mercury in fly ashes.

Table 1

–Summary of the concentrations of TEs in diverse coal samples (Zhao et al., 2016; Zhu et al., 2016; Zheng et al., 2018, 2017) [1–4].

	Average (ppm)	Min (ppm)	Max (ppm)	Number of samples
Cr	33.645	6.7	394.0	26
Mn	148.1	116.2	180.0	2
Co	6.0	5.1	7.0	3
Ni	12.92	5.78	22.15	21
Cu	14.87	11.1	17.5	3
Zn	31.33	23.0	38.0	3
As	4.99	0.37	13.45	41
Mo	2.5	2.2	2.7	3
Ag	0.006	0.006	0.006	1
Cd	0.424	0.008	1.12	26
Sb	1.253	0.50	3.24	21
Ba	184.67	150.0	245.0	3
Pb	20.54	7.8	30.6	26
Hg	0.141	0.02	0.35	38
Se	2.88	0.87	5.43	38

TEs can be emitted as fly ash or volatiles, but emissions as fly ash are lower due to the high efficiency of removal with Electrostatic Precipitators (ESPs) or fabric filters (FF). On the other hand, gaseous emissions are more efficiently retained when Flue Gas Desulphurization (FGD) is present (Meij and te Winkel, 2007; Zhao et al., 2016). Flue gas from coal-fired power plants are generally cleaned by selective catalytic reduction (SCR) for De-NOx, low-temperature electrostatic precipitators (LLT-ESP)/electric fabric filter (EFF), wet flue gas desulphurization (WFGD) and wet electrostatic precipitators (WESP) (Meij and te Winkel, 2007; Zhu et al., 2016; Zheng et al., 2017). Each pollution control system shows different results in the retention of each specific element (Tian et al., 2014). ESP can retain 99.8% of the PM demonstrating high efficiency in retaining the TEs enriched in these particles. The elements emitted in gaseous form are retained mainly in the FGD. As an example, mercury was 49.6% removed in ESP and 80% in FGD, reaching a total removal of 89.92% (Meij and te Winkel, 2007). Other factors also affect the TEs emission in coal-fired power plants. Washing coal before combustion can remove 30 to 60% of TE (Tian et al., 2014), thereby reducing the overall emissions (Nalbandian, 2012; Zhu et al., 2016), while the presence of chlorine increases the formation of metal chlorides, favoring the emission of TEs (Pudasainee et al., 2010).

2.2. Combustion processes of sewage sludge

Emission of TEs has also been extensively studied in the processes of combustion of sewage sludge, municipal waste and industrial waste (Åmand and Leckner, 2004; Lin et al., 2010; Mu et al., 2012; Zhang et al., 2013; Yang et al., 2016; Wang, R. et al., 2017; Wang, X. et al., 2017; Wang, Z. et al., 2018; Fu et al., 2019). These products can be used as fuel alone or in a co-combustion with other fuels (Åmand and Leckner, 2004). Several studies have analyzed the concentration of trace elements in sewage sludge (Table 2). It can be seen that the concentration of trace elements in sewage sludge is high, especially for Cu and Zn, when compared with trace elements present in coal (Yang et al., 2016).

Yang et al. (2016) studied the co-combustion of sewage sludge with coal gangue and its environmental implications. Since both residues are available in abundance and are both potential energy sources, they were tested as alternative energy sources (Zhang et al., 2013; Yang et al., 2016). It was observed that As, Pb and Zn are the trace elements with the greatest volatilization potential during sewage sludge combustion. Since the coal gangue is composed mostly of kaolinite, its presence during the combustion of sewage sludge helped to retain trace elements, especially Pb and Zn (Yang et al., 2016).

Zhang et al. (2013) investigated the emission and distribution characteristics through a mass balance of Hg, Cd, Pb, Cr and Cu during co-combustion of sewage sludge in a pulverized coal fired power plant. They found that approximately 97% of Hg and 96% of Cd were distributed in fly ash and flue gas during the process. Most of the Cd was

detected in the fly ash, making it easier to remove from the raw gas, while the majority of the Hg was present in the flue gas. Pb was distributed with approximately 40% in fly ash and 20% in the flue gas. Cr and Cu showed only a small amount in the fly ash and almost nothing in the flue gas, with more than 90% concentrated in bottom ash. Additionally, by increasing the concentration of chlorine in the input, the distribution of the trace elements in fly ash and flue gas increased, with a consequent increment of emissions.

In another study, Van de Velden et al. (2008) analyzed the distribution of Hg, As, Cd, Cu, Pb, Cr, Ni and Zn during fluidized bed combustion of sewage sludge. They found among the elements studied only Hg does not concentrate on the fly ash collected by ESP and a remarkable amount of Hg, Cd and Pb was also identified in the stack emissions. In fixed bed municipal solid waste incineration, the effect of chlorine and calcium oxide on the volatility of Cd and Pb was studied by Wang, X. et al. (2017). During incineration without the presence of chlorine, the reducing atmosphere provided the transformation of Pb and Cd monoxides into elemental substances and sulphides causing the volatilization of these elements at temperatures between 600–700°C. CdO was more easily reduced than PbO, but the volatilization of PbO started before CdO when combusted in air. The addition of Cl in the mixture increased the volatilization of both elements and the addition of CaO had no impact on their volatility above 700°C. To minimize the emission of these elements, it was suggested the elimination of reducing atmosphere conditions in Cl-free incineration or the addition of CaO for combustion at temperatures below 700°C.

2.3. Ironmaking and Steelmaking

In the iron and steelmaking industry extensive amounts of gaseous and particle emissions are generated, especially fine particles where the main trace elements are enriched (Ji et al., 2017a; Yang et al., 2019). According to Li et al. (2017), the Iron and Steel Industry are responsible for 27% of airborne dust emission in China. In addition to iron ore sintering, discussed in more detail in the next section, other steps in the iron and steelmaking are relevant in emission of trace elements, such as coke-making, blast furnace and BOF converter processes.

Tsai et al. (2007) evaluated the emission of particulates and their composition in four processes of an integrated steel plant (coke-making, sintering, cold forming, and hot forming) in Taiwan, while Machemer (2004) characterized the airborne and bulk particulate material from a blast furnace (BF) and a basic oxygen furnace (BOF) in Maryland, United States. Table 3 presents a summary of the results found by these authors.

Regarding the TEs, chromium, titanium and vanadium showed higher concentration in BOF particulates. Aluminum, zinc, nickel and arsenic were detected in high amounts in particulates from cold forming. In the particles emitted during iron ore sintering, lead, cadmium and selenium were identified in higher content than the other processes, while the highest amount of copper was detected in the hot rolling

Table 2

– Summary of the concentrations of TEs in diverse sewage sludge samples in ppm of dry solid sewage sludge (Åmand and Leckner, 2004; Van de Velden et al., 2008; Zhang et al., 2013; Wang, R. et al., 2017).

TE	Sweden (ppm)	Netherlands (ppm)	Germany (ppm)	Italy (ppm)	Belgium (ppm)	Hangzhou China (ppm)	South China (ppm)
Cr	33.00	113.00	91.00	240–650	85	196.67	198.1
Mn	280.00	546.00	-	75–98	-	-	-
Co	6.20	8.70	-	-	-	-	-
Ni	20.00	83.00	39.00	36–53	38.7	48.33	-
Cu	390.00	406.00	330.00	243–599	300	-	181.5
Zn	550.00	1349.00	1318.00	2796–4231	1581	1083.33	-
As	4.70	8.40	-	-	10.5	-	-
Cd	1.20	3.80	3.80	14–23	4.5	2.67	2.80
Sb	2.40	4.10	-	-	-	-	-
Pb	33.00	260.00	159.00	104–137	162	138.33	3.80
Hg	1.10	3.30	2.70	-	1.1	-	1.48
Se	1.30	2.40	-	-	-	-	-
V	18.00	24.00	-	-	-	-	-

Table 3

– Elemental composition of particulates emitted by the stages of the steelmaking process (Machemer, 2004; Tsai et al., 2007).

TE (ppm)	Coke making[a]	Sintering[a]	BF[b]	BOF[b]	Cold forming[a,c]	Hot forming[a,d]
Al	2,837	4,276	1,000	6,500	8,778	5,290
As	125	75	NAc	NA	370	203
B	NA	NA	5	20	NA	NA
Ba	1,417	75	ND	61	1,603	160
Bi	NA	NA	35	NDd	NA	NA
Ca	6,793	15,819	700	56,050	32,222	13,592
Cd	97	1,336	8	25	1,178	73
Co	1	7	24	15	240	35
Cr	ND	211	182	846	523	182
Cu	119	1,154	73	155	620	7,953
Fe	17,657	54,573	635,667	257,500	130,722	52,833
K	9,806	157,207	8,333	ND	25,056	8,099
Li	NA	NA	5	5	NA	NA
Mg	956	1,387	447	25,500	3,889	1,611
Mn	130	483	4,207	12,850	4,352	685
Na	11,715	31,510	NA	NA	33,333	40,702
Ni	225	631	130	65	20,980	769
P	NA	NA	477	980	NA	NA
Pb	292	22,882	ND	150	814	1,452
S	51,943	42,120	7,733	1,300	146,944	60,036
Sb	4	3	40	ND	3	511
Se	68	398	NA	NA	157	ND
Si	NA	NA	5,907	17,650	NA	NA
Sn	NA	NA	4	20	NA	NA
Sr	32	56	ND	39	5	24
Te	NA	NA	200	ND	NA	NA
Ti	NA	NA	207	1,270	NA	NA
V	ND	ND	75	201	ND	20
Y	NA	NA	ND	2	NA	NA
Zn	1,892	4,631	2,377	3,695	9,427	5,684

[a] Composition of particulate emitted in processes of an integrated steel plant in Taiwan found by Tsai et al. (2007).
[b] Composition of the airborne and bulk particulate material from a BF and a BOF found by Machemer (2004).
[c] NA: Not analyzed
[d] ND: Not detected

particles (Machemer, 2004; Tsai et al., 2007).

Mercury is the most studied trace element of the iron and steel industry emissions due to its high volatility and insolubility in water. Between the raw materials, the iron concentrates are the major source of Hg (followed by coking coal), consequently, sinter and pellet plants are the major emitters of this element in the steel industry (Machemer, 2004; Tsai et al., 2007; Lau et al., 2016; Li,M. et al., 2017; Yang et al., 2019). In 2015, these processes were responsible for the emission of 15.9 t of Hg in China, followed by the blast furnace with 7.9 t. Coke ovens and roasting plants emitted together 7.0 t in the same year, while oxygen steelmaking generated less than 2.0 t (Yang et al., 2019).

Mercury can be emitted in a gaseous state, as Hg^0 or Hg^{2+}, or in a solid-state as particulate (Hg^p). According to Pacyna and Pacyna (2002) and Pirrone et al. (2001), the emission of Hg^0, Hg^{2+}, and Hg^p in the iron and steelmaking processes follow the approximate proportion of 80%, 15% and 5%, respectively. However, according to Xu et al. (2017) and Wang et al. (2016), the Hg^{2+} was found as the predominant specie in the iron and steelmaking emissions.

3. Iron ore sintering

Iron ore agglomeration is the first step of the steel production in an integrated steel mill and the sintering process represents the vast majority installed in these plants, while the pelletization process is commonly found in the iron ore mining companies. Sintering is a thermal agglomeration process (1300–1480 °C) (Eisele and Kawatra, 2003) of a mixture of iron ore mineral fines (0.5–8 mm), by-products of the iron and steelmaking industry, fluxes, slag-forming elements and fossil fuel (coke), aiming to produce a ferrous burden (12–35 mm) for the blast furnace with the suitable physico-chemical and mechanical properties (Fernández-González et al., 2017b). An important feature of the sintering process, from an environmental point of view, is the possibility of using steel residues as an input to the process (Oliveira and Bagatini, 2019).

The sintering raw materials are firstly mixed in a granulation process, which conducts their homogenization in a mixing drum with water addition (6–8%). Dwight-Lloyd is the most used iron ore sintering machine in the world. It operates continuously by a series of pallets where the granulated sinter mix is loaded forming a bed height of 500–600 mm, which is subjected to the ignition on the top by oil or gas burners. Following, an air suction system promotes the formation of a narrow combustion zone (flame front) moving downwards (10–30 mm/min) and heating the mixture to 1250–1350 °C (Fernández-González et al., 2017b). In a series of reactions, a semi-molten material is produced and then cooled into several mineral phases of different chemical and morphological compositions (hematite, magnetite, ferrites and gangue composed mostly of calcium silicates). Moreover, ahead of the combustion zone, water evaporates and volatile substances are driven off by the wind boxes below the strand, which are connected to a fan via a gas scrubbing system. Finally, the sintered material is discharged and subjected to a final cooling, crushing and screening (Cores et al., 2013). Fig. 1 shows a schematic flowchart of the process.

Fig. 2 shows the temperature profile and the atmosphere regime of the sintering bed. The heating time at the high-temperature zone (above 1100 °C) is around 1.5 min under a low partial oxygen pressure (pO_2) due to coke combustion and the cooling time (to 1100 °C) must vary from 3 to 5 min in a condition of high partial oxygen pressure (Cores et al., 2013).

3.1. Iron ore sintering air pollutants

The volume of gas emitted by a sintering plant ranges from 1500 to 2500 Nm^3/t of graded sinter, depending on the operational conditions and plant size (Remus et al., 2013). The off-gas contains carbon
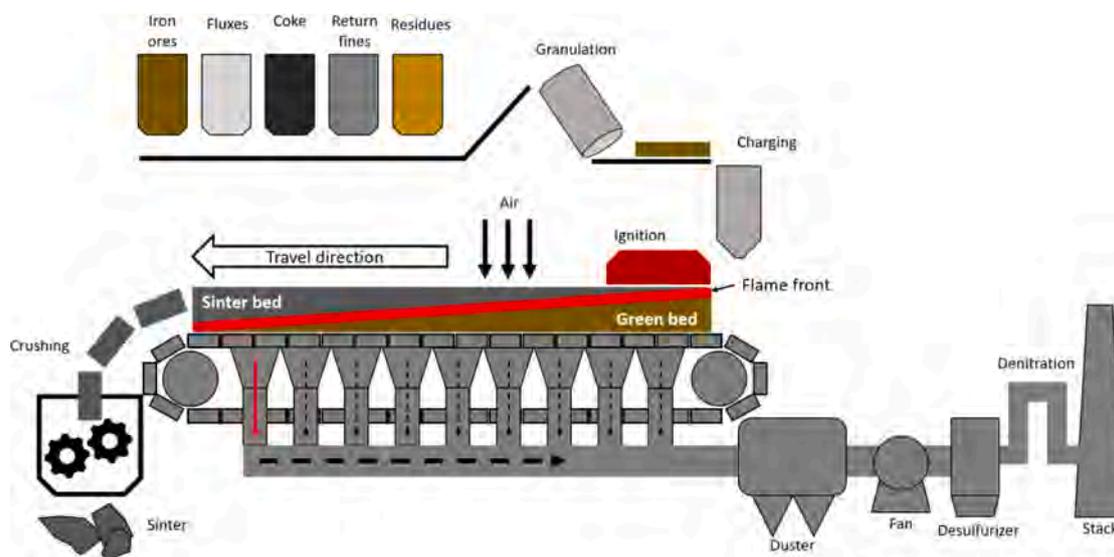
Fig. 1. –Schematic diagram showing the material flow in a sinter plant (adapted from Wielgosinski and Łechtańska, 2014).
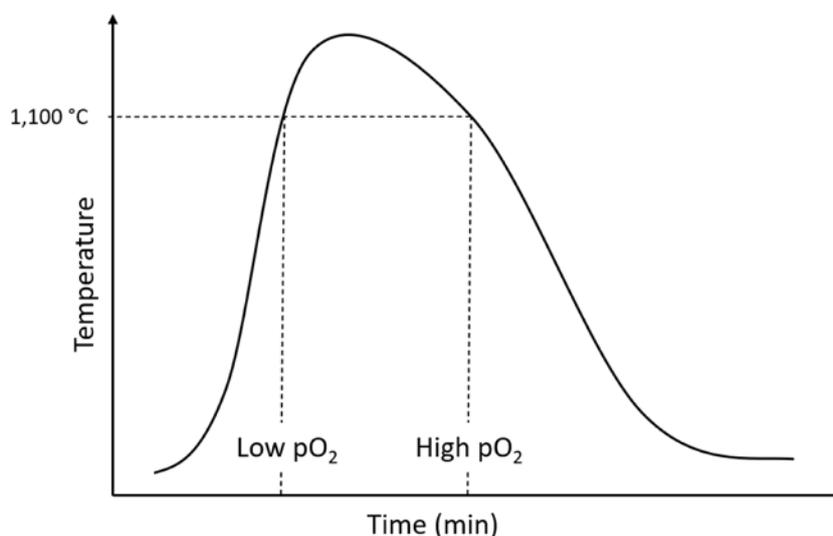

Fig. 2. –Temperature profile and the atmosphere regime of the sintering bed (adapted from Cores et al., 2013).

monoxide, carbon dioxide, sulphur and nitrogen oxides, alkali chlorides, hydrogen chloride, hydrogen fluoride, hydrocarbons, PAH (polycyclic aromatic hydrocarbons), PCDD-F (polychlorinated dibenzo-p-dioxins/dibenzofurans), PCBs (polychlorinated biphenyls) and heavy metals (Fernández-González et al., 2017c).

Greenhouse gases play an important role in the warming of the earth. The main greenhouse gas related to sintering is CO_2, with sintering accounting for approximately 7 to 9% of direct emissions from the use of fossil fuels (1.85 t CO_2 per ton of steel produced) (WSA, 2020b). SO_x and NO_x are other important pollutants emitted in the sintering process, which can cause acid rain and a series of respiratory and other diseases (Fernández-González et al., 2017c). Iron ore sintering is responsible for approximately 3% of the total emission of NO_x in China (Fan et al., 2015).

Iron ore sintering is known as an important source of organic pollutants, such as PCDD/Fs (polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans) and PAH (Polycyclic Aromatic Hydrocarbons) which presents a high health risk due to its high toxicity and carcinogenic risk (Quaß et al., 2004; Remus et al., 2013; Fernández-González, 2017c). According to Li, S. et al. (2017), sinter plants are responsible for about 30% of total PCDD/F to the atmosphere in stack

gases in China. They studied the emission factors for some organic compounds in stack emissions from six Chinese iron ore sintering plants and found 2.47, 0.61, 552, 0.32, and 107 $\mu g.t^{-1}$ for PCDD/F, PCB, PCN (polychlorinated naphthalenes), PBDD/F (polybrominated dibenzo-p-dioxins/dibenzofurans) and PBDE (polybrominated diphenyl ethers), respectively.

PM (particulate matter) is another important air pollutant emitted by sintering plants (Fernández-González et al., 2017c). Due to its small dimension and high surface area, it is also capable of directly depositing in the lung's alveoli, causing a series of respiratory diseases due to the concentration of toxic elements (Przybysz et al., 2014; Ji et al., 2017a). The sintering process is the main PM emitter within an integrated steel plant (Ji et al., 2016) corresponding to approximately 45% of the total steelmaking PM emission (Gan et al. 2015a).

Another pollutant emitted during iron ore sintering that deserves special attention are heavy metals since they have a long residence time in the atmosphere and are easily assimilated by natural organisms (Boente et al., 2017). In addition to heavy metals emitted in PM, some of them can also be emitted as gas, such as Pb and Hg (Remus et al., 2013; Yang et al., 2019).

3.1.1. Control Measures to mitigate the iron ore sintering polluting potential

Environmental requirements have become increasingly restrictive concerning anthropogenic emissions, especially greenhouse gas (GHG) emissions. To contain global warming and climate change, industrial sector is motivated to find environmentally correct and efficient solutions to reduce their emissions. In steelmaking, through the blast furnace route, a global effort and cooperation in the political, environmental, and economic sectors for the export and import of technologies are fundamental as a basic initiative to mitigate the emission of GHG, especially CO_2.

The GHG released into the environment at the steps of steelmaking, particularly CO_2 emissions, strongly depend on the types and quality of reducing agents. In this regard, replacing fossil fuels with cleaner and renewable energy sources is an important consideration. An alternative for iron ore sintering is biomass and biomass-derived fuels. Biomass is not only carbon neutral, but has a calorific value close to solid fossil fuels and contributes to the reduction of emission of other polluting gases. Previous sintering tests have shown that when biomass or charcoal partially replaces coke breeze, emissions of CO_2, SO_x, NO_x, dioxins, and furans reduce (Abreu et al., 2015).

Regarding the emission reduction of organic compounds in iron ore sintering, mainly include three aspects: source control, process control and end treatment. Source Control occurs when the content of sensitive elements or substances in sintering raw materials reduce. For example, the presence of copper and chlorine, and the carbon source are the main factors affecting the formation of dioxins in the sintering process and they must be carefully evaluated and controlled. To not have an additional investment cost, process control can be done using inhibitors (e.g. nitrogen-containing and alkaline substances, which will reduce dioxin emission by either combining with Cl or interrupting the catalytic activity of metal catalysts) or flue gas recycling technologies (i.e. the flue gas generated in the process is injected in the high temperature sintered layer to crack dioxins) and other routes to reduce the emission of dioxins in the sintering process based on existing sintering equipment. In particulate phase, dioxin can be removed by cyclone separator, bag filter, an electrostatic precipitator, while in the gas phase it is removed by activated carbon adsorption and selective catalytic reduction (SCR) (Qian et al., 2018).

PM removal techniques have been widely applied in the industry (including iron ore sintering process) as an important measure to protect the environment and recycle resources. Electrostatic precipitators can significantly reduce smoke emission, recycle useful materials in the smoke, and improve resource utilization. In addition, the purified gas in the precipitator can be recycled, saving resources indirectly. To further improve the reduction of PM released, Ji et al. (2017b) suggested the use of the separate granulation process. This process consists of identifying the key elements in the formation of PM to avoid the formation of chemical compounds that favor the conversion to the particle phase, restricting the migration of these elements from the raw material to the PM, controlling their emission. With the evaluation in a laboratory-scale sinter pot, this technique did not affect the sintering performance and delivered promising results for the removal of $PM_{2.5}$, being, therefore, promising for use in industrial-scale iron ore sintering plants.

Trace elements emission, including heavy metals, will be better discussed in a specific chapter following.

3.2. Source of trace elements in the iron ore sintering process

The sintering mixture is composed of a wide variety of raw materials with the most diverse properties and chemical composition, which vary according to the input source, processing, transport and storage stages that precedes sintering (Lau et al., 2016; Fernández-González et al., 2017a).

In addition to iron ore, the sintering mixture is also composed of solid fuel (coke), slag-forming elements and fluxes (especially limestone, quick-lime and dolomite), returned fines (sinter with granulometry < 5

mm), and steel-making residues (like blast furnace, sinter and coke plant dust) (Lau et al., 2016; Fernández-González et al., 2017a). In our previous study, divers iron ore sintering inputs were chemically characterized regarding the trace elements (Lau et al., 2016). Table 4 presents a summary of these data in comparison with the results obtained by other authors.

Iron ores contain the largest concentrations of As, V and Hg. Coke breeze have the highest concentrations of Ni, Cr and Ti. The other elements did not show any clear pattern of concentration in one of the components of the sintering mixture. Considering a typical sintering mixture (61% of iron ore, 12% of fluxes, 23% of return fines and 4% coke breeze) (Gan et al., 2015b) and the concentrations of trace elements presented in Table 4, it can be observed that iron ore is the main carrier of the trace elements. For example, Xu et al. (2017) evaluated the supply of mercury in three different Chinese sintering and found between 74.84 and 92.22% of the mercury originating in iron ore.

3.3. The behavior of trace elements

During the iron ore sintering process trace elements can have four different destinations: remain in the sinter product, be retained by the abatement methods in the form of particulates, be emitted into the atmosphere as particulate material, or be emitted into the atmosphere in the gaseous form (Xu et al., 2004). Fig. 3 presents a schematic diagram of these possible paths of trace elements in the iron ore sintering process.

Fig. 4 shows the distribution of trace elements according to their volatility. In general, the trace elements are classified into three different classes in the combustion process (Ratafia-Brown, 1994; Xu et al., 2004; Nalbandian, 2012):

- Class 1: Elements with low volatility index in the combustion processes, with a tendency to remain concentrated in the sinter or the coarse particles;
- Class 2: Elements that can volatilize and condense during the combustion process. They are mainly concentrated in the PM, being enriched in the finer particles;
- Class 3: Elements with low volatility temperature, being emitted mainly in their gaseous form.

Class 1 elements do not volatilize during the sintering process. In this way, these elements remain in the sinter cake or, the smaller particles can be dragged as particulate material, which is easily retained by the abatement systems (Nalbandian,2012). Once the gases move away from the combustion front, the course of the vaporized trace elements (Classes 2 and 3) depends on whether or not these elements are converted into solid and/or liquid forms by physico-chemical phenomena (Xu et al., 2004). These phenomena are:

- Heterogeneous condensation on surfaces or pre-existing particulate matter;
- Physical/chemical adsorption in pre-existing particulate material;
- Nucleation and coalescence as submicron particles in the presence of supersaturation;
- A chemical reaction between trace elements and constituents of the particulate material and flue gas;
- Remain in gaseous form for elements with a high vapor pressure at the exit temperatures of the gases.

According to Remus et al. (2013), during the sintering process lead reacts to form volatile compounds (PbO-$PbCl_2$, $PbCl_2$, and, probably, $PbCl_4$). Thus, the emission of lead is related to the presence of chlorine in the sinter feed. Deng et al. (2014) studied the correlation of lead emission and the presence of chlorine in coal in Chinese thermoelectric plants and found an *r*-value (Pearson correlation coefficient) of 0.7952, indicating that chlorine may have a significant effect on Pb_P (p indicates trace metals bound in particles) formation. Similarly, for iron ore

Table 4

– Trace element composition of iron ore sintering raw materials in ppm (Gan et al., 2015a; Lau et al., 2016; Ji et al., 2017a, 2016; Xu et al., 2017; Fan et al., 2018).

Raw material	TE	Lau et al. (2016)	Fan et al. (2018)	Gan et al. (2015a)	Ji et al. (2017a, 2016)	Xu et al. (2017)
Iron Ores (Sinter feed)	As	10.34	-	-	-	-
	Cd	0.11	-	-	-	-
	Cr	45.08	-	-	-	-
	Cu	16.88	-	-	-	-
	Ni	16.43	-	-	-	-
	Pb	7.47	60	62.0	120	-
	Cl	140.28	140	152.0	270	-
	Ti	-	-	-	564	-
	V	57.93	-	-	-	-
	Zn	20.08	-	-	350	-
	Hg	0.08	-	-	-	2.327×10^{-2}–4.997×10^{-2}
Fluxes	As	2.93	-	-	-	-
	Cd	0.16	-	-	-	-
	Cr	45.29	-	-	-	-
	Cu	15.13	-	-	-	-
	Ni	15.06	-	-	-	-
	Pb	5.04	30–60	25.0–62.0	0	-
	Cl	106.57	110–150	107.0–152.0	30–120	-
	Ti	-	-	-	0–66	-
	V	6.57	-	-	-	-
	Zn	14.29	-	-	20–40	-
	Hg	0.02	-	-	-	0–3.100×10^{-3}
Return fines	As	6.40	-	-	-	-
	Cd	0.16	-	-	-	-
	Cr	83.00	-	-	-	-
	Cu	11.30	-	-	-	-
	Ni	16.90	-	-	-	-
	Pb	12.80	60	55.0	80	-
	Cl	120.00	70	68.0	40	-
	Ti	-	-	-	528	-
	V	37.00	-	-	-	-
	Zn	113.00	-	-	240	-
	Hg	0.05	-	-	-	0–9.700×10^{-4}
Coke breeze	As	4.17	-	-	-	-
	Cd	0.04	-	-	-	-
	Cr	148.00	-	-	-	-
	Cu	35.57	-	-	-	-
	Ni	135.87	-	-	-	-
	Pb	273.47	60	62.0	10	-
	Cl	6.67	150	149.0	580	-
	Ti	-	-	-	1200	-
	V	6.33	-	-	-	-
	Zn	63.67	-	-	40	-
	Hg	0.03	-	-	-	1.950×10^{-2}

sintering Remus et al. (2013) suggest that the key factor in lead chlorine formation is not the lead in the sinter feed, but the chlorine concentration. Besides, depending on the fluorine content in iron ores, lead fluorides can also be formed, being even more volatile and contributing to the emission of lead. In the higher temperature zones, zinc volatilizes and then reacts to form zinc ferrites. These ferrites can remain in the sinter or in the form of particulates, which can be collected relatively easy by the sintering abatement systems (Remus et al., 2013).

Cadmium, in its elemental form, has a volatilization temperature of 767 °C and should volatilize during the sintering process (Haynes et al., 2016). On the other hand, if the formation of CdO occurs, volatilization should only occur at 1559 °C (Haynes et al., 2016). Furthermore, in the presence of Al and Si, CdO can form CdO.I$_2$O$_3$ and CdSiO$_3$, with both compounds being stable between 600 and 1100 °C, retaining most of the cadmium in the solid form (Zhu et al., 2019).

Arsenic, whether present in its elemental form or as As$_2$O$_3$, has a low boiling temperature and should volatilize during sintering (Haynes et al., 2016). On the other hand, some compounds stable at higher temperatures can be formed, such as FeAsO$_4$ (stable up to 1100 °C), and AlAsO$_4$ (stable up to 1400 °C). It is still possible, although unlikely, that Cd$_3$(AsO$_4$)$_2$, a stable compound up to approximately 1200 °C, is formed by the interaction of As with Cd (Contreras et al., 2009).

Regarding copper volatilization, the participation of chlorine is essential, since the boiling point of elemental copper is 2560 °C (Haynes

et al., 2016). In addition to allowing the volatilization of Cu due to the formation of CuCl$_2$, interactions of copper and chlorine have a major impact on the emissions of chlorinated aromatics. Copper acts as a catalyst in the formation of these compounds where the catalytic activity of CuCl$_2$•2H$_2$O is higher than elementary Cu for the *de novo* formation (one of the main formation pathways of chlorinated aromatics) (Sun et al., 2016). Concerning the formation of chlorobenzenes (CBzs) the catalytic activity is CuCl$_2$•2H$_2$O > Cu$_2$O > Cu > CuSO$_4$ > CuO and CuCl$_2$•2H$_2$O > Cu$_2$O > CuO > Cu > CuSO$_4$ for polychlorinated biphenyls (PCBs) (Liu et al., 2019).

Nickel and its main derivatives have a high boiling point, indicating low emissions of this element. If nickel is emitted, it occurs by volatilization through the NiCl$_2$ formation, which sublimates at a temperature of 985 °C (Haynes et al., 2016). Guangxu et al. (2017) analyzed the concentration of trace elements in the soil of an abandoned iron and steel factory in China and found that nickel concentration in soil was comparable to the background concentration of this element in the earth's crust. The low deposition of nickel in the soil is considered a consequence of a low air emission. These results agree with those obtained by Zhao,L. et al. (2017), who studied the PM emitted at different points in a sintering plant and found a low nickel concentration in the PM of the sintering strand.

As in other high-temperature processes, mercury can be emitted in iron ore sintering as Hg^{2+}, Hg^{0}, or Hg^{P}. Xu et al. (2017) studied mercury
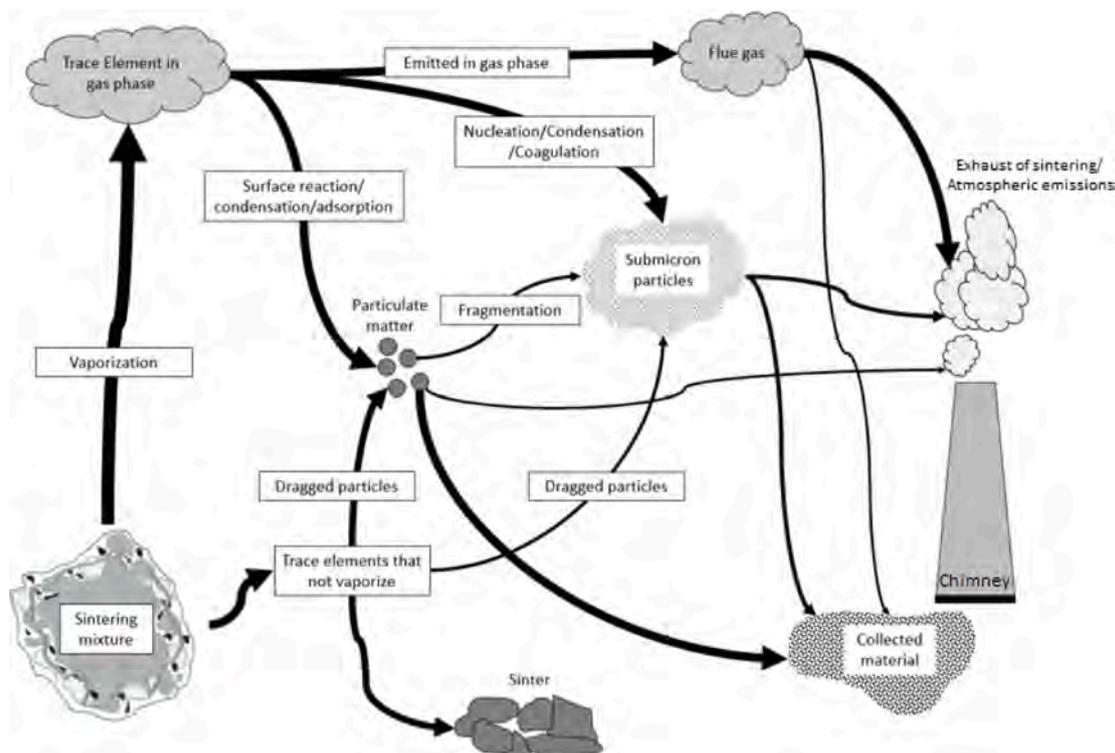
Fig. 3. –Paths that trace elements can follow in iron ore sintering process (adapted from Xu et al., 2004).
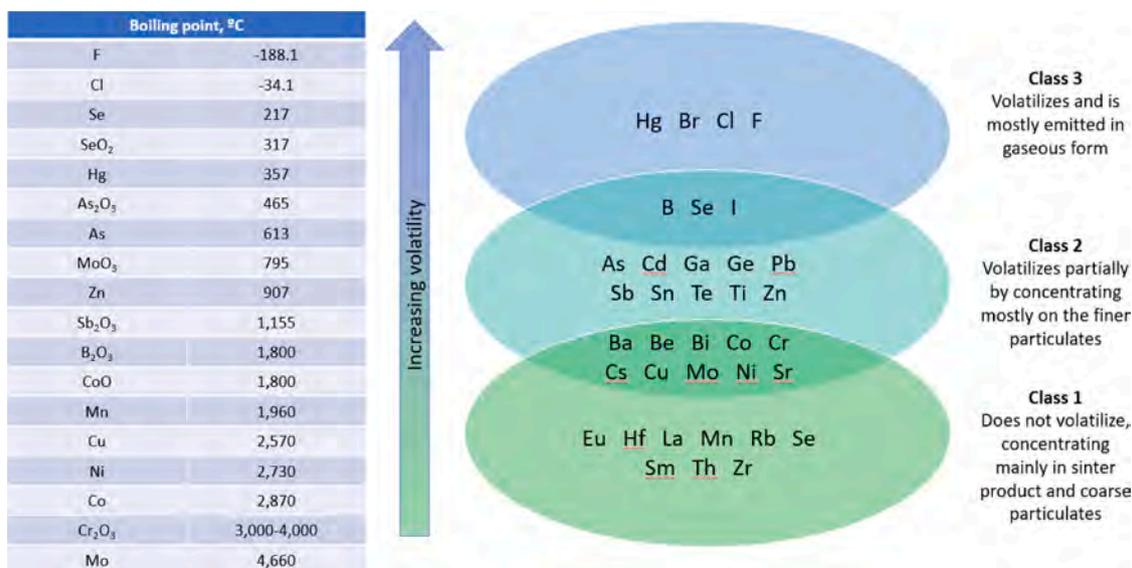

Fig. 4. –Classification of trace elements based on their volatilization (Ratafia-Brown, 1994; Xu et al., 2004; Nalbandian, 2012; Haynes et al., 2016).

emissions in three different Chinese sintering plants and found a high proportion of Hg^{2+} at the inlet of ESP (64.36–94.72%), followed by Hg^0 and, to a lesser extent, Hg^P. Wu et al. (2017) found similar results, with approximately 70% of Hg^{2+}, 30% of Hg^0, and no Hg^P in Chinese iron ore sintering emissions in 2015. The high proportion of Hg^{2+} can be explained since Fe_2O_3 has significant catalytic activity in the oxidation of Hg in the presence of Cl (Dunham et al., 2003; Ghorishi et al., 2005).

Fan et al. (2018) studied the concentration of Pb, K, Na, Cl, and S in $PM_{2.5}$ emitted in a laboratory-scale sinter pot test. It was identified that the concentration of these elements is significantly higher in the $PM_{2.5}$ collected after the increase of flue gas temperature (stage-2) than before (stage-1). The presence of these elements in Fe-rich and Fe-Si-Al-rich

particles was further detected, as well as the formation of $CaSO_4$ in both stages, while the $PbCl_2$-KCl, KCl, and NaCl in the stage-2.

3.4. Trace elements emissions factors

The emission factor is the average emission rate of a given source related to units of production or process. In atmospheric emissions from iron ore sintering, the emission factor corresponds to a mass of the pollutant emitted by mass of sinter produced.

Table 5 presents the results reported by Passant et al. (2002) and Remus et al. (2013) who evaluated the emission factors of trace elements in iron ore sintering. The values obtained show significant variation,

Table 5

–Trace elements emission factor in iron ore sintering (mg/t sinter) (Passant et al., 2002; Remus et al., 2013).

Element	Remus et al. (2013)	Passant et al. (2002)
As	0.6–15	6.2
Cd	0.2–276.7	32.0
Cr	3.6–125.1	5.5
Cu	1.9–600.5	183.4
Hg	0.1–207	18.3
Mn	3.4–539.4	110.0
Ni	1.3–175.6	8.2
Pb	26.1–5,661.2	3,136.4
Se	21.8–120.5	155.9
Tl	0.5–86.6	48.6
V	0.6–58.5	1.8
Zn	2.1–1931.3	255.8

which can be explained by the different properties of the raw materials employed and the different abatement systems of each sintering plant evaluated. Lead presents the highest emission factor, which can reach 5661.2 mg/t of sinter. However, great variability of emission factors was found for the same element in different sintering plants. The least variable was Se with a variation of 7.15 times between the highest and lowest emission factors.

3.5. Measurements used in studies/industry

A complete atmospheric study requires sample collection over long periods and covering a large area, making these studies expensive and complex to perform. A widely adopted alternative is the use of organisms that act as bio-accumulators (Abril et al., 2014). Rahayu et al. (2016) proposed the use of a wild plant (*Eichhornia crassipes*) as a bio-accumulation agent for Pb, Fe, Hg, Zn, Cu and Cd. According to Loppi, et al. (1994) some lichen species can be used for the same purpose. The latter used lichen *Parmelia Caperata (L.) Ach.* as a bio-accumulator to evaluate the atmospheric contamination of Cd, Cr, Cu, Hg, Ni, Pb and Zn. Holy et al. (2009) evaluated the mosses as bio-accumulators of Cd, Hg and Pb, finding strong correlations between the concentrations of Cd and Pb in mosses and sampling site-specific and regional characteristics. There are other studies that are also pointing out on several species that can be used as bio-accumulators for heavy metals (Freitas,1995; Gür and yaprak, 2011; Satya et al., 2012; Turkyilmaz et al., 2018; Bora et al., 2020).

Continuous emission monitoring systems can be used for some pollutants, but they are not yet commercially viable for trace elements, therefore measurements are performed manually (Nalbandian,2012). Trace element sampling is performed using isokinetic methods. A probe/pitot is used in the duct orifice with a vacuum pump to adjust the sampling flow to isokinetic conditions. The sampled gas passes through a hot box and a cyclone and/or filter where the particles are collected. The gas then flows through impingers with a series of oxidizing agents to collect the trace elements present in the gas. This method has several limitations and is an expensive procedure, which requires a long time for collection to obtain significant results. Moreover, it does not present punctual results over time, only the average of emissions over the collection period, and, in general, needs approximately one day for analysis and results (Nalbandian,2012). Technologies towards the online measurement of trace element emissions were not developed yet. According to Nalbandian (2012), the solution seems to be based on spectroscopy, where the most promising technologies are: Atomic Fluorescence Spectroscopy, Atomic Absorption Spectroscopy, and Tunable Diode Laser Absorption Spectroscopy.

3.6. Air Pollution Control Devices (APCD) employed/available

The atmospheric emissions abatement system installed in iron ore sintering has diverse configurations and equipment, each having its advantages and disadvantages. Among the various equipment that can be used alone or in combination are the Electrostatic Precipitators (ESP), Fabric Filters (FF), Cyclones, Flue Gas Desulphurization (FGD) and Selective Catalytic Reduction (SCR), although no equipment is specific for the removal of trace elements (Remus et al., 2013; Yang et al., 2019). Each of these equipment are described in detail.

3.6.1. Electrostatic Precipitator (ESP)

The most common equipment for treating flue gas from sintering plants is ESP. This equipment works by generating an electrostatic field in the dust path. A voltage of thousands of volts is applied ionizing the gas around the electrodes. The negative ions flow to the plates charging the particles in the gas stream, causing them to be attracted to the grounded plates. In dry ESPs the plates go through a mechanical stirring process, causing the particles to fall into the collectors. In the wet ESPs, a constant flow of water removes the particles, and the liquid obtained is treated later (Guerriero et al., 2008; Nalbandian,2012; Remus et al., 2013).

ESPs reduce dust emissions by more than 95%, in some cases reaching reductions of more than 99% (Nalbandian,2012; Remus et al., 2013). They have good efficiency for particles with specific resistivity is in the range of 10^4 to 10^9 Ωm. In general, the particles in the sintering flue gas fall in this range, but in some compounds, such as alkali chlorides, heavy metal chlorides and calcium oxides, the specific resistivity may be greater than the appropriate range, making their removal difficult (Remus et al., 2013). Also, since they are more difficult to charge, ESPs are less efficient with fine and ultrafine particles, where the semi-volatile trace elements tend to concentrate (Nalbandian,2012).

Several technologies can increase the efficiency of ESPs. Remus et al. (2013) highlighted three of them:

- Use of energy pulse superimposition: the pulse system provides high voltage pulses at a frequency of up to 200 pulses per second. This technique helps to energize particles with high resistivity, providing a better charging of particles and distribution of current in the precipitator;
- Moving Electrode Electrostatic Precipitator (MEEP): in this equipment, several electrode plates move during operation, being cleaned continuously by rotating brushes. This prevents the formation of insulating dust layers that adversely affect the operation of the ESP;
- Electrostatic Space Cleaner Super (ESCS): this equipment works with high voltages and greater distance between the electrodes.

3.6.2. Fabric filter (FF)

In FF the particulate material is removed from the flue gas by passing through a system of filter bags. The collected dust forms a porous cake on the surface of the filters where it also acts as an additional filtering layer (Nalbandian, 2012). During the filtration process, a series of re-agents can be used to increase the removal of other pollutants besides dust, such as HCl, HF, SO_x, PCDD/F, PCB, HCB and PAH. The particulates accumulated on the filter are removed, being partially recirculated in the waste gas to increase the efficiency of the adsorbents. The remaining dust can be disposed and/or recirculated in the sintering strand (Remus et al., 2013).

In general, the filtering process takes place with the following steps: injection of adsorbents (lignite coke, active carbon and zeolites) to reduce the emission of hazardous compounds; injection of $NaHCO_3$ or slacked lime to reduce acidic emissions; capture of dust in the filter element; dust removal from the filter element; recirculation of part of the collected dust; use of an exhaust fan to extract the filtered gas (Remus et al., 2013).

Although they can be used in isolation, in iron ore sintering the FFs are usually used in combination with the cyclone (Nalbandian, 2012; Remus et al., 2013; Yang et al., 2019). The use of FF with ESP increases the retention of particulate material and ultrafine particles, directly impacting the retention of trace elements, especially semi-volatile

elements (Zhao,S. et al., 2017b; Zheng et al., 2017). According to Nalbandian (2012), fabric filters can achieve an efficiency of 99.95% in the collection of particles of all sizes and more than 99% efficiency for fine particles.

3.6.3. Cyclones

Cyclones are used mainly as a pre-treatment of gases. Since they are based on inertia, they are efficient only for the retention of coarse particles. More than one cyclone can be used in series, increasing efficiency for particles of different sizes. Its main function is to protect the following equipment from the abrasion generated by the larger particles (Remus et al., 2013).

3.6.4. Wet fine scrubber

Scrubbers are devices that remove particulate material from the flue gas using a liquid. Since the particles emitted by iron ore sintering are relatively fine and contain hydrocarbons, a special wet fine scrubber is required (Remus et al., 2013). AIRFINE is one model of wet fine scrubber, which consists of two steps of work. The first step is responsible for cooling the waste gas by spraying a solution of NaOH in water on the surface of the scrubber. In the next stage, the fines are washed by injecting a mist of water and compressed air into the gas flow, removing fine particles, including heavy metal chlorides. The collected liquid is treated and recirculated in the process (Remus et al., 2013).

Other models of wet fine scrubbers are electrostatic or ionizing advanced wet scrubbers or condensing wet scrubbers (Nalbandian, 2012). Electrostatically enhanced wet scrubbers apply a charge to the particulate material, collecting it through neutral or negatively charged electrodes. In a condensing wet scrubber, the flue gas is cooled below its adiabatic saturation temperature, causing the condensation of a large fraction of elements that are in the vapor phase. Besides, cooling induces condensation of water vapor, forming droplets that assist in the collection of fine particulate matter (Xu et al., 2004).

Some trace elements of greater volatility and, consequently, more difficult to remove by ESP and FF, can be removed in the wet fine scrubber due to water solubility. This is the case for mercury in the form of Hg^{2+} (Zhao,S. et al., 2017b). According to Zheng et al. (2017), other examples of trace elements that can be removed in wet fine scrubbers are As, Cr and Pb.

3.6.5. Flue gas desulphurization (FGD)

Desulphurization can be achieved wet or dry. Wet desulphurization is carried out through the wet fine scrubber with the addition of Ca or Mg as reagents, forming $CaSO_4$ or $MgSO_4$. The residues from this process, such as gypsum, magnesium sulphate and ammonium hydrogen sulphite (NH_4HSO_3), are treated and recirculated or reused in other processes, such as cement production. This process also helps to remove HCl, HF and dust from the flue gas, but does not affect emissions of NO_x (Remus et al., 2013).

Dry desulphurization is achieved through the adsorption of SO_2 on activated carbon. The waste gas flow is directed to an activated carbon bed, where several pollutants are adsorbed. In addition to SO_2, this process removes HCl, HF, Hg^{2+}, dust, PCDD/F and, if ammonia is injected into the gas before the catalytic bed, NO_x. As a co-product, sulphuric acid (H_2SO_4) can be obtained (Remus et al., 2013).

3.6.6. Selective catalytic reduction (SCR)

A catalytic reduction process can be used for the control of NO_x emissions. In this process vanadium pentoxide (V_2O_5) or tungsten oxide (WO_3) on titanium oxide (TiO_2) carrier can be applied as catalysts, where NO_x is reduced by ammonia or urea to N_2 and H_2O. The proper operating temperature is between 300 and 400 °C, making it necessary to reheat the gas before the process (Remus et al., 2013).

SCR is beneficial for Hg^0 oxidation, in which hydrogen chloride (HCl) and sulphur dioxide (SO_2) have positive effects. On the other hand, the literature does not offer much information about the efficiency

of the SCR in controlling the emission of other TEs (Zhao et al., 2017a).

3.6.7. APCDs configurations and efficiency in retaining trace elements

An iron ore sintering plant may have several combinations of control equipment described above as APCD. According to Xu et al. (2017), a typical configuration is the use of ESP, FF and FGD. These authors evaluated the efficiency of mercury removal in three sintering plants with the following APCD configurations:

(1) ESP, circulating fluidized-bed flue gas desulphurization (CFB-FGD) and FF;
(2) ESP and dense flow absorber flue gas desulphurization (DFA-FGD);
(3) ESP and ammonia flue gas desulphurization (AFGD).

The results obtained showed removal of mercury between 97.5 and 98.3%, which is the best result for the configuration (2) (Xu et al., 2017).

Although there are not many studies evaluating the efficiency of APCDs for the removal of trace elements in sintering, there are many of that evaluated the efficiency of APCDs in coal-fired power plants (Attalla et al., 2004; Zhu et al., 2016; Zhao,S. et al., 2017a). Attalla et al. (2004) accessed the efficiency of ESP, FF, and FGD in controlling trace element emissions of a conventional pulverized coal plant with results shown in Table 6.

In general, a high retention capacity of most trace elements can be observed in the evaluated equipment. The elements that presented the greatest difficulty in retention were B, Cd, Cu and, especially, the most volatile Hg and Se. ESP, FF and FGD showed efficiency greater than 95% for all other elements.

Comparing the results found by Attalla et al. (2004) and Xu et al. (2017) it is possible to observe a much higher mercury retention capacity in the sintering process than in coal-fired power plants. This can be explained by the higher concentration of Hg^{2+} in the sintering gas. According to Ghorishi and Sedman (1998), Hg^{2+} tends to be adsorbed on particulates and is partially removed by the ESP. Besides, FGD has a greater capacity for removing Hg^{2+}, since it is more reactive and water-soluble (Xu et al., 2017).

Zhu et al. (2016) evaluated several combinations of APCD concerning the ability to remove trace elements from emissions of coal-fired power plants in China. The following range of technologies were proposed, individually or in combination: ESP, FF, WFGD, SCR, SNCR (selective non-catalytic reduction) and SMC (a special mercury control technology, like activated carbon injection (ACI), bromide injection into the furnace (BIF), oxidation catalysts, low-temperature mercury capture, the thief carbon process, etc.). From the results obtained, as presented in Table 7, the best configurations are SCR + SMC + ESP + WFGD

Table 6

–Trace element control efficiencies in different APCD technologies in % of retention (Attalla et al., 2004).

Trace element	ESP	FF	FGD
Sb	96	97	99
As	98.5	98.6	98.7
Ba	99.5	98	99.5
Be	98.5	98.6	98.7
B	68	97	63
Cd	83	94	96
Cr	97.5	99.5	99.5
Co	98	99	99
Cu	89	99.5	99
Pb	98	98.5	99
Mn	97	99.5	99.5
Hg	30	60	20
Mo	96	100	99
Ni	96	99	99
Se	21	65	60
V	98	100	99.5

Table 7

–Trace elements removal efficiency of 8 different APCD combinations in coal-fired power plants in China (Zhu et al., 2016).

APCD combination	Co-benefit removal efficiency (%)							
	Hg	As	Se	Pb	Cd	Cr	Ni	Sb
ESP	33.17	86.20	73.78	97.16	96.46	98.53	93.52	83.5
ESP+WFGD	71.41	97.29	93.41	99.39	99.31	99.79	98.70	97.1
SCR+ESP+WFGD	74.82	97.29	93.41	99.39	99.31	99.79	98.70	97.1
SNCR+ESP+WFGD	71.41	97.29	93.41	99.39	99.31	99.79	98.70	97.1
SCR+SMC+ESP+WFGD	97.48	97.29	93.41	99.39	99.31	99.79	98.70	97.1
FF	67.92	99.00	65.00	99.00	97.63	95.13	94.83	94.3
FF + WFGD	86.28	99.80	91.20	99.78	99.54	99.32	98.97	99
SCR + FF + WFGD	87.91	99.80	91.20	99.78	99.54	99.32	98.97	99

and SCR + FF + WFGD.

In another study, Zhao et al. (2017a) evaluated the emissions of Zn, Sb, Pb, Cd, As, Cr, Mn and Ba in a Chinese coal-fired power plant equipped with SCR, ESP, and WFGD. The efficiency of removing the eight elements until the ESP was between 99.43 and 99.95%. The WFGD increased the efficiency to between 99.78 and 99.96%, retaining 18.10% of Sb, 22.25% of Ba, 23.16% of Cr, 28.39% of Mn, 31.15% of As, 53.17% of Pb, 61.26% of Zn, and 68.47% of Cd.

Font et al. (2009) evaluated the retention capacity of trace elements by the APCDs of two coal-fired power plants in Spain. High efficiency of ESP was found in the retention of most of the elements, but Hg, Cl, F, As, Se and B showed volatile behavior, making retention by ESP difficult. For these elements, the FGD acted with high retention efficiency for gaseous Cl, F, and B ($> 95\%$), As (78-90%), Se (84-94%), and relatively high for Hg (64 -74%).

As can be seen, the removal of elements that are not in the vapor phase (Classes 1 and 2) is carried out mainly by dust collectors and the biggest challenge is the removal of elements with high volatility. An alternative to remove these elements would be to force their condensation through the gas cooling, collecting them as particles. However, in this case, there is a need to reheat the gas before the stack makes the process less economically viable (Xu et al., 2004). The use of sorbent injection to capture these elements is also an interesting alternative, which has led to some studies (Biswas and Wu, 1998; Ghorishi and Sedman, 1998; Lineberry et al., 2009; Yao and Naruse, 2009).

Yao and Naruse (2009) proposed the use of compounds based on silica and calcium as sorbents to capture Pb, Cd and Cr during solid fuel combustion. The tested sorbents based on their order of efficiency in retaining Pb and Cd were kaolin, zeolite, limestone, scallop, mullite, apatite, bauxite and lastly silica.

For the retention of mercury, activated carbon is widely used and can be efficient in adsorbing other trace elements, such as As (Pandey et al., 2011). Ghorishi and Sedman (1998) suggest Ca-based sorbents to control total mercury emissions and found that the most effective sorbents are those with significant surface area and pore volume. Moreover, Lineberry et al. (2009) proposed the use of palladium-decorated carbon as a substrate for mercury capture and found promising capabilities, with total efficiencies greater than 90% in laboratory-scale tests and in the order of ~60% in a real industrial flue gas.

4. Conclusions

This paper reviews the emission of trace elements in the iron ore sintering process, focusing on the source of the elements, their behavior under high-temperature industrial conditions, and the devices employed to control emissions. Moreover, processes with similar characteristics, such as coal-fired power plants, were explored to assist in understanding the behavior of the trace elements in sintering plants. The mechanisms of emission of trace elements proved to be complex and their control can be performed through the appropriate choice of raw materials.

Iron ore is the main source of trace elements input in the sintering process. However, some new trends in the sintermaking can also impact the emission of trace elements and must be carefully evaluated. The use

of alternative fuels such as biomass and charcoal, even though they are viable to mitigate CO_2 emissions in the iron ore sintering process, they present harmful levels of toxic metals and trace elements (Vassilev et al., 2014). The use of iron and steelmaking solid waste as raw materials for sintering can support this industry to increase its potential in the circular economy concept. However, the trace elements content in iron and steelmaking solid wastes (Oliveira and Bagatini, 2019) must become another barrier to recycling them, and the understanding of trace elements emissions in the sintering process to be even more complex. Finally, the use of iron ore ultrafine in iron ore sintering makes the granulation step increasingly relevant, not only to make it feasible to use ultrafine particles and to achieve a high-quality sinter, but also to reduce the emission of particulate matter carrying trace elements. In addition, the use of new binders (including organics or inorganics), which improves the granulation performance, should be carefully evaluated to verify their impact on the trace elements emission (Linhares et al., 2019; Junior et al., 2021).

In general, trace elements undergo a series of physicochemical transformations throughout the process and may remain in the solid-state, volatilize during burning and recondense in APCD or remain in the gaseous state during the flue gas treatment. Most trace elements are retained in APCD in the form of particulates, but volatile and semi-volatile elements (As, Se, Hg, Cd, etc) can remain as gases or concentrate in the finer particulates, making them difficult to remove by conventional methods of controlling particulate emissions. Modern technologies (e.g the use of activated carbon or other sorbents) have demonstrated high efficiency in removing the volatile and semi-volatile elements from the flue gas. However, the atmospheric emission abatement systems currently installed in iron ore sintering do not focus on the removal of trace elements. Although there are alternatives for the online measurement of emissions (e.g. those based on spectroscopy), chemical knowledge of the raw materials to identify the source of trace elements, as well as the evaluation of the partition of these elements, between solid product and gaseous product, during the sintering process can effectively contribute to control the emission of trace elements.

The risks associated with the emission of these elements and the still incipient knowledge about their source and behavior during iron ore sintering make evident the need for further studies.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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