

The distribution of heavy metals during fluidized bed combustion of sludge (FBSC)

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Abstract

During combustion of wastewater treatment sludge, the inorganic constituents are converted into ash which contains the major fraction of the heavy metals present. The behaviour of heavy metals in combustion processes has been studied extensively for mostly coal combustion and waste incineration. For biomass and sludge, literature data are scarce and mostly limited to laboratory experiments. The present paper assesses the partitioning of eight heavy metals (Hg, As, Cd, Cu, Pb, Cr, Ni and Zn) in the different residues from a large-scale fluidized bed sludge combustor of 4.4 m i.d. The origin of the sludge is mostly from treating urban wastewaters (>90%), although some mixed sludge (urban + industrial, <10%) is also burnt. The different residues (bottom ash, fly ash, filter cake, scrubber effluent and stack emissions) were sampled and analysed during 33 weeks, spread over a period of 1 year. The mass balance of relevant heavy metals closes for 96.5%, inaccuracies being related to the unsteadiness of the process, the accuracy of the mass flow data monitored at the plant, and on collecting representative samples. It is also shown that all heavy metals under scrutiny, except Hg, are concentrated in the fly ash as collected in the electrostatic precipitator.

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1. Introduction

1.1. Waste activated sludge

WAS processes produce considerable amounts of sludge, about 70 g of dry solids (DS) per inhabitant equivalent (IE) [1]. The FBSC (fluidized bed sludge combustor) burns sludge, mostly from urban wastewater treatment (>90%) although some mixed urban and industrial sludge (<10%) is also dealt with. The implementation of the European Urban Waste Water Treatment Directive [2] and a more efficient treatment in general have caused a steady increase in the annual sludge production in the EU from 5.5 million tonnes of dry solids (DS) in 1992 to nearly 9 million tonnes in 2005 [3]. Sludge poses both an environmental and economical problem, the former due to the tonnage and to the variety of toxic and polluting compounds present, the

latter due to the high cost of sludge treatment and its disposal representing 35–50% of the total WWTP cost [4].

Sludge treatment involves various steps: thickening, anaerobic digestion, mechanical dewatering, possibly drying [1]. Anaerobic digestion produces an energy-rich biogas but increases the mineral fraction of the residual dry solids.

The problems associated with sludge disposal are complex due to its content of hazardous substances such as heavy metals (ranging from less than 1 ppm to over 1000 ppm) [5], PCBs, PAHs and dioxins [6,7], pesticides and endocrine disruptors [8], pathogens and other microbiological pollutants [1]. Beside these potentially hazardous materials, sludge also contains valuable materials such as nitrogen, phosphorus, and organic carbon as well as some inorganic compounds. The major routes for the disposal of sludge are landfilling, use in agriculture and (co-)incineration, each route with advantages and disadvantages.

Whereas, landfilling has for long been the major route for sludge disposal, the European Landfill Directive [9] forbids landfilling of biodegradable waste since 2003: this disposal route of sludge is hence excluded in the E.U.

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Table 1
Maximum acceptable total heavy metal concentration (in mg/kg DS) in sludge for agricultural use following the Flemish VLAREA [15] and European legislation [14]

	As	Cd	Cr	Cu	Hg	Pb	Ni	Zn
VLAREA	150	6	250	375	5	300	50	900
EU Sludge Directive	–	5	800	800	5	500	200	2000

Sewage sludge contains several components of agricultural significance (nitrogen, phosphorus, potassium, organic matter, and calcium) [10]. It can therefore be used as natural fertiliser and soil amendment [11]. This soil application could be the preferred environmental disposal route [12], if hazardous substances were not present.

Heavy metals in the sludge [13] often outweigh the soil's heavy metal content and the application of sludge could indeed increase the concentration of heavy metals in the agricultural soil, affect the crop production due to uptake, and lead to the transfer of heavy metals to humans through the consumption of plants and animals [11].

The use of sewage sludge for agricultural purposes is regulated by the Sewage Sludge Directive [14]. This Directive is currently under revision. In Flanders, the more stringent VLAREA [15] is already applicable and is illustrated in Table 1 towards the acceptable total concentration of heavy metals in sewage sludge for use in agriculture. The regulation does not specify a separate concentration in the leachate.

Incineration has become the major disposal route for sludge in the European Union and it is expected that its role will increase in the future, since it results in a large reduction of sludge volume to a small quantity of ash, which accounts only for 10% of the volume of mechanically dewatered sludge; the thermal destruction of toxic organic constituents and pathogens, and the recovery of the flue gas energy [16].

Incineration includes dedicated sludge combustors, co-incineration of sludge with other wastes (e.g., municipal waste) and co-fuelling coal-fired power plants or cement kilns. The incineration of sewage sludge is governed by the European Directives on the Incineration of Waste [10,17].

1.2. Heavy metals in WAS

Heavy metals are present in urban wastewater and hence in sewage sludge as a result of domestic, industrial and road runoff inputs. Industrial contributions are the primary source of heavy metals in urban wastewater and despite increased control on discharges and changes in processes they still represent up to 50% of the total metal loads in sewage sludge [18]. Electroplating, plastics manufacturing, fertiliser, pigment, mining, and metallurgical processes are major sources [19]. Diffuse domestic sources are mainly Cu, associated with leaching from plumbing materials and the use of Zn in body care products [18]. Some and Lagerkvist [20] made a detailed study concerning the diffuse inputs, especially on Cu, Zn, Ni and Hg. The sources of Pb, Cr and Cd were less understood, although car washes were definitely the largest contributors for these elements. Aonghusa

and Gray [21] report a significant contribution of detergents and washing powders to the presence of Cd, Cu and Zn in wastewater. Jenkins and Russell [22] showed that household washing products account for about 13% of the wastewater arsenic content.

Heavy metals are found in considerable amounts in waste activated sludge. However, large variations in metal concentrations are observed in sludge produced by different WWTPs of comparable size, reflecting localised metal discharges to the sewer system. This will be further illustrated for the studied case of Brugge. This emphasises the importance of continued vigilance by the water industry in reducing metal discharges in industrial effluents.

Metals, in particular, heavy metals such as Pb, Hg, Cd and As constitute a significant potential threat to human health, both occupational and environmental. As trace elements, some metals are essential to normal metabolism, but most of them pose a high toxicity at higher levels of exposure; others (such as Pb and Hg) are xenobiotic and exert toxic effects at any level of exposure [23]. Several metals are chemically stable and tend to persist in human tissues and in the environment. The most important routes of exposure are inhalation of metals dust or fumes as well as involuntary ingestion. Most metals are excreted through the kidney and the gastrointestinal tract; another fraction is accumulated in some organs such as bone, liver kidney and brain [23].

Many studies have shown that activated sludge concentrates heavy metals from influent wastewater. Insoluble metals or metals adsorbed onto settling particles are removed during primary settling [24], finely suspended or dissolved metals are adsorbed on the sludge flocs during secondary treatment and thus removed during secondary settling [25–27].

Whereas, agricultural use of sludge needs to prevent heavy metal accumulation in the cultivated soils, co-incineration as treatment route should avoid releasing heavy metals to the environment. The formation and subsequent emission of particles enriched with toxic metals which can be inhaled as well as gaseous emissions during incineration processes have been studied [27]. Since sludge has a high mineral content (usually about 40–50% on a dry solid basis) a large quantity of ash is generated [28]. From the perspective of reutilisation, e.g. incorporation in cement [29–32] and towards landfill disposal as illustrated before, the environmental compatibility of these residues must be guaranteed, notably their heavy metal content and their resistance to leaching.

1.3. The combustion of WAS

Fluidized bed sludge combustors (FBSC) are widely used, and a strategy for their design has recently been updated [33]. Sludge combustion results in the formation of char and a mineral residue, mostly entrained from the FBSC as fly ash when at small enough particle size. Fly ash includes mainly fuel-originated ash, but may include also fragmented bed material particles. Flue gas is directed through the heat recovery to either preheat the fluidization air, or steam or thermal fluid (when sludge drying is added). Fly ash particles are then collected from the flue

gas, usually by an electrostatic precipitator (ESP) or a fabric filter, before the flue gas is directed through the stack to the atmosphere, possibly after scrubbing.

The possibility of using biomass ash, e.g. as soil conditioner or as an additive to concrete and asphalt, is affected by the properties of the ash since concentrations of harmful trace elements can limit its use [34] because of the solubility and/or the toxicity of the trace elements.

1.4. Objectives of the research

Literature data concerning the partitioning of heavy metals in large-scale sludge combustors are scarce. The present paper reports findings for the 10,000 tonnes DS/year fluidized bed sludge combustor of Brugge (Belgium).

The report will detail the occurrence of eight target heavy metals at the various points of the plant (feed, bottom ash, fly ash, scrubber, and stack) in order to establish the distribution and the partitioning of heavy metals between the different waste streams.

2. Methods

2.1. Description of the Brugge sludge incineration plant

Sludge is mechanically dewatered by centrifuges, pre-dried to about 70% DS in a multiple hearth dryer (using thermal fluid at 230–260 °C). The FBSC has an internal diameter at the distributor level of 4.4 m, and a freeboard i.d. of 6.8 m. Primary (~80%) and secondary air (~20%) are used. The air pre-heater (fluidization air) is integrated in the freeboard. Flue gases are treated by electrostatic precipitation (ESP) and subsequent acid and alkaline scrubbing.

The plant schematics of Fig. 1 illustrate the FBSC operation at Brugge, and the various sources and sampling points of ash and residue. These points are indicated as: FBSC bottom ash and bed material (1), fly ash collected from the ESP (2), filter press cake after precipitation and filtration (3a) and the associated filtrate (3b). Emissions are measured in the stack (4). Filter

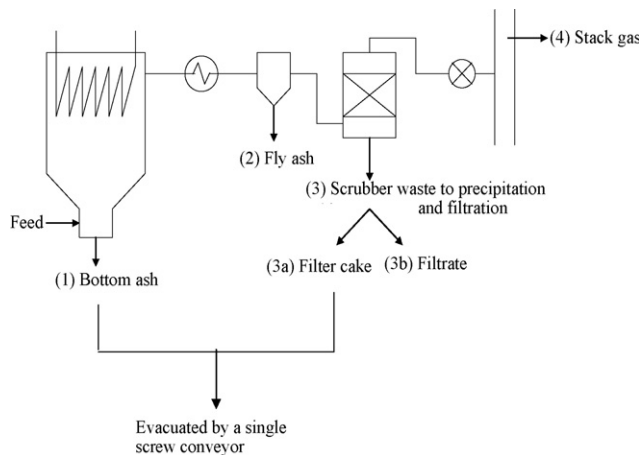


Fig. 1. The plant schematics and sources of heavy metals.

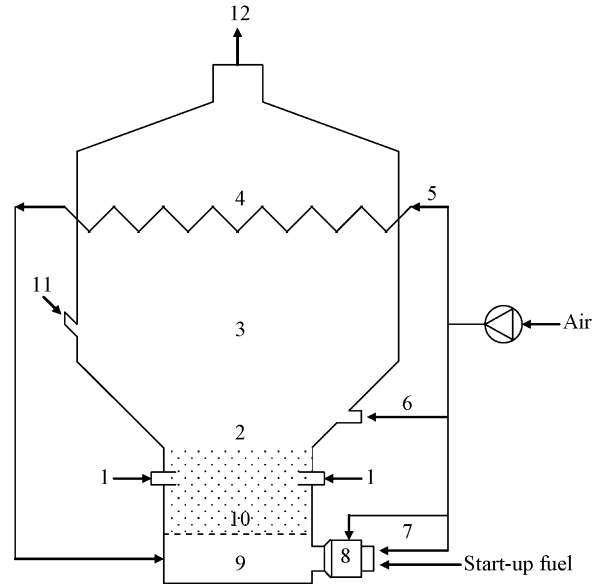


Fig. 2. The fluidized bed sludge combustor of Brugge with (1) sludge feed, (2) fluidized bed, (3) freeboard, (4) pre-heater of primary air (5), (6) secondary air, (7) air to start up burner (8), (9) windbox, (10) distributor, (11) make-up sand, and (12) exhaust to further heat recovery, ESP, pollutant abatement, stack. The distributor has a central hole feeding a water-cooled screw conveyor for bottom ash removal. This conveyor then joins the conveyor of the filter cake.

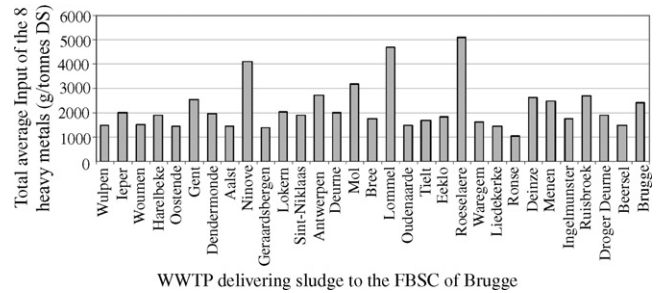


Fig. 3. Feed of heavy metals to the sludge FBSC of Brugge.

cake (3a) and bottom ash (1) are discharged by a single screw conveyor. All samples were collected once per week during 33 weeks, spread over 1 year. Grab samples were collected from the discharge of screw conveyors for flows (1), (2) and (3a). Effluent samples were collected at the effluent monitoring unit (3b). 2.5 tonnes DS/h is combusted at 800–830 °C, at a superficial gas velocity of 2.2–2.5 m/s. The temperature after the scrubber is 65 °C, resulting in an emission of approximately 35,000 m³/h. The fluidized bed furnace itself is illustrated in Fig. 2.

Table 2
Average sludge feed composition of heavy metals

HM (g/tonnes DS)	
As	10.5
Cd	4.5
Cr	85
Cu	300
Zn	1581
Ni	38.7
Hg	1.1
Pb	162

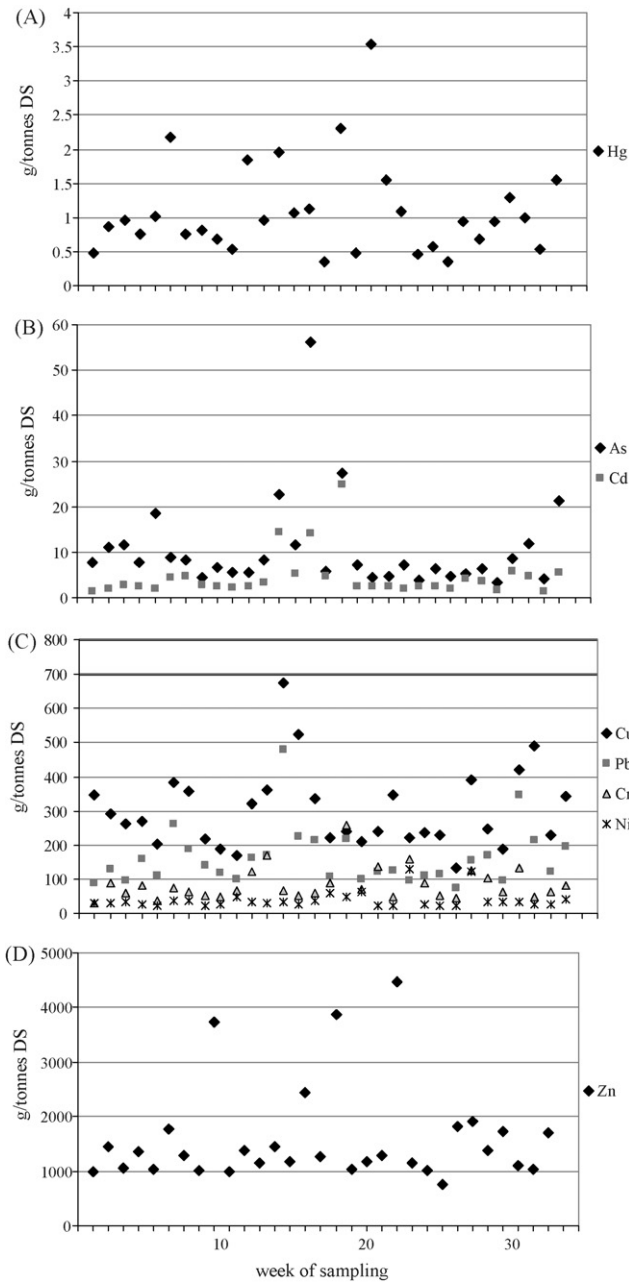


Fig. 4. Composition of sludge feed for the different suppliers in (A) Hg, (B) As and Cd, (C) Cu, Pb, Cr and Ni, and (D) Zn.

2.2. Samples and analysis

The operation was monitored throughout 2006, and this during 33 weeks, giving a good review of operating results. The sludge samples were analysed for heavy metals. Cr, Cu, Ni, Zn, As, Cd and Pb were measured by ICP-OES. Hg was determined by cold vapour AAS. Prior to the analysis, the sludge samples were submitted to a high-pressure microwave digestion method, as described in detail elsewhere [35,36].

The annual average quantities of heavy metals of the Brugge-associated WWTPs are given in Fig. 3. These quantities are the total concentration of the eight metals under scrutiny in the present paper. The concentration of relevant

Table 3
Quantities of waste streams

	Quantity
Bottom ash	41 tonnes DS/yr
Fly ash	8302 tonnes DS/yr
Scrubber cake	40 tonnes DS/yr
Stack gas	35,000 m ³ /h

heavy metals in the sludge feed is illustrated in Table 2 and Fig. 4.

2.3. Partitioning of the heavy metals

Fig. 1 presented the FBSC and the sources of samples. Annual average quantities of the various waste streams

Table 4
Composition of waste streams

Bottom ash (1) and scrubber dewatering cake (3a), discharged simultaneously and mixed in the same screw conveyor	
Basic elements	39 wt% SiO ₂ 16 wt% Al ₂ O ₃ 18 wt% CaO 22 wt% Fe ₂ O ₃
Trace elements	0.3 mg Hg/kg DS 35 mg As/kg DS 112 mg Cr/kg DS 96 mg Cu/kg DS 2130 mg Zn/kg DS 195 mg Ni/kg DS 250 mg Pb/kg DS 12 mg Cd/kg DS
Fly ash (2)	
	39.1 mg As/kg DS 19.5 mg Cd/kg DS 181 mg Cr/kg DS 951 mg Cu/kg DS 0.67 mg Hg/kg DS 485 mg Pb/kg DS 105 mg Ni/kg DS 4176 mg Zn/kg DS
Effluent water (3b)	
	0.31 µg Cd/l 33.14 µg Cr/l 43.14 µg Cu/l 30.00 µg Ni/l 7.86 µg Pb/l 95.00 µg Zn/l 16.14 µg As/l 54.24 µg Hg/l
Stack gas (4) (emissions expressed in g/h)	
	0.104 g Cd/h 0.118 g Cr/h 0.060 g Cu/h 0.055 g Ni/h 0.248 g Pb/h 0.165 g As/h 0.753 g Hg/h 0.156 g Zn/h

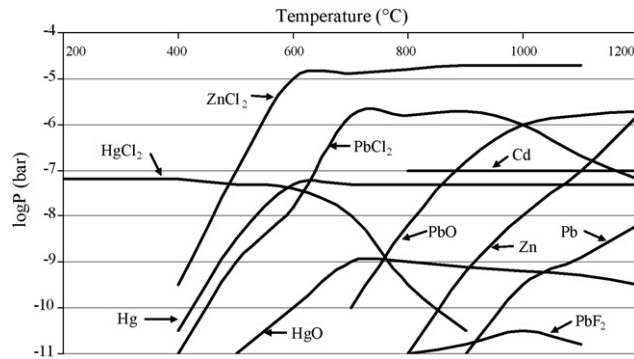


Fig. 5. Vapour pressure of relevant heavy metals and their salts.

and their composition are given in Tables 3 and 4, respectively.

The quantities of heavy metals in effluent water (3b) are extremely low, even considering a scrubber water effluent discharge of 40 m³/h.

Since stack gas emission standards are very stringent [37], with an allowed emission of less than 5 g/h as far as total mass flow of the eight heavy metals are concerned, this contribution is not significant in the overall balance. Stack gases were hence only fully analysed twice during 2006, each time over a period of 8 h as prescribed by the Flemish legislation [37].

The monitoring of the stack gas (Table 4, stream (4)) revealed that concentrations of heavy metals are indeed in compliance with the legal standards (total \ll 5 g/h).

It is also evident that more volatile metals such as Hg, Pb and Cd are significantly present in the stack emission, despite the scrubber stages, but in line with the volatility of the heavy metals and their salts as illustrated in Fig. 5.

3. Discussion

3.1. Mass balance of heavy metals

Accounting for the quantities of Table 3 and individual concentrations (Table 4), the mass balance for the heavy metals under scrutiny in this paper is presented in Table 5. The bottom ash of the FBSC contains bed material, agglomerated ash and residual coarse additives (CaCO₃ for SO₂ abatement). An average quantity of approximately 120 kg/day is withdrawn.

Table 6
Average annual partitioning of heavy metals from feed to waste streams

Heavy metal	Feed (kg/yr)	Bottom ash (1) and scrubber cake (3a) (kg/year)	Effluent water (3b) (kg/year)	Fly ash (2) (kg/year)	Stack gas (4) (kg/year)
As	348	1	1.33	325	1.4
Cd	108	1	0.03	162	0.9
Cr	1,807	9	2.74	1,503	1.0
Cu	7,155	8	3.56	7,895	0.5
Hg	30	3	4.48	6	6.2
Pb	3,947	20	0.65	4,026	2.0
Ni	856	16	2.48	872	0.5
Zn	37,215	170	7.85	34,669	1.3

Table 5
Average annual tonnages in the FBSC of Brugge

	Source (Fig. 1)	Tonnes/year	%Removal on total residue
Average in sludge feed		51.5	–
Bottom ash and scrubber cake	(1) + (3a)	0.224	0.45
Effluent water	(3b)	0.024	0.05
Fly ash	(2)	49.46	99.49
Stack gas	(4)	0.008	0.02
Total recovered		49.716	
Balance (feed—recovered)		1.784	

The balance is closed for 96.5%. This is considered fair and acceptable, since the evaluation involves several sources of inaccuracy, which are difficult to quantify. These are related to the unsteadiness of the process, accuracy of the mass flow rate values that are based on process data monitored at the plant, and on collecting a representative fly ash sample which constitutes the major fraction of all samples collected (Table 3).

The unsteady process conditions cause inaccuracy in the mass balance mainly because of different response times of ash flows to changes in process conditions. Fly ash concentration reacts quickly, whereas, the composition of the bottom ash shifts slowly with time, due to the large inventory of bed material compared to the feeding of ash-forming constituents into the furnace. The bed-material feed and removal rates may vary remarkably from day-to-day, according to the assessments by the plant operating staff. However, the elemental concentrations in the bed material do not vary accordingly, since only a fraction of the total inventory in the bed is changed in a day (approximately 120 kg/day is extracted). The fly ash samples were moreover collected from the ESP discharge by grab sampling.

Although the overall mass balance closes in an acceptable manner, the balances for some individual heavy metals show larger discrepancies, as illustrated in Table 6 and discussed below. The overall trend confirms earlier findings in a laboratory scale FBSC [38].

3.2. The partitioning of heavy metals

The partitioning of the individual heavy metals between the fly ash, stack emission and scrubber effluent indicates an efficient capture of the heavy metals in the fly ash, except for Hg (Table 6). The figures are obtained by multiplying the tonnage of each

stream with its average concentration of each of the considered heavy metals.

It can be clearly seen that individual quantities differ between feed and residues, although the inaccuracy is only significant for Cd and Hg. It should however be stressed that the stack gas was only sampled twice during the year, thus leading to expected significant errors for these volatile metals, but the emission quantities are only of minor influence on the overall mass balance.

3.3. Methods of reducing the heavy metals content of WAS

Contrary to allowing heavy metals to reach the FBSC, pre-treatment methods have recently been described [39]. It was shown that thermochemical methods and/or peroxidation can reduce the heavy metals input by over 50% in the case of Cd, Cr, Ni, Zn and Cu, whereas, results for Hg and Pb are less outspoken.

This will however further impact on the cost of the sludge disposal and an economic viability study is needed.

4. Conclusions

During combustion of wastewater treatment sludge, the inorganic constituents are converted into ash which contains the major fraction of the heavy metals present. The present paper assessed the partitioning of heavy metals in the different residues from a real scale fluidized bed sludge combustor of 4.4 m i.d. The different residues (bottom ash, fly ash, filter cake, scrubber effluent and stack emissions) were sampled and analysed over a period of 1 year. The mass balance of relevant heavy metals closes for 96.5%. All heavy metals under scrutiny, except Hg, are concentrated in the fly ash collected by the electrostatic precipitator. The quantities of heavy metals in effluent water of the scrubber are extremely low. The monitoring of the stack gas revealed that concentrations of heavy metals are in compliance with the legal standards (total < 5 g/h). It is also evident that more volatile metals such as Hg, Pb and Cd are significantly present in the stack emission, despite the scrubber stages, but in line with the volatility of the heavy metals.

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