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Incineration of doped sludges in fluidized bed. Fate and partitioning of six targeted heavy metals. I. Pilot plant used and results

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Abstract

Incineration of sewage sludge doped with several heavy metals was studied at small pilot plant scale in a bubbling fluidized bed of 15 cm i.d. and 5.2 m height. Some ceramic and metallic filters were tested at a relatively high temperature (600–700°C) to check their usefulness for partitioning of heavy metals in the flue gas. The work was focused on the fate of six selected heavy metals (Cr, Cd, Ni, Zn, Cu, Pb). In this process, there were four exit flows or discharges for these metals: bottom ash, coarse fly ash, cake filter or fine fly ash and flue exit gas. The distribution or partitioning of each heavy metal (HM) among these four exit flows was studied. Only cadmium and sometimes lead showed any difference between the different HMs considered. All other HMs seems to have the same fate, distribution or partitioning. Such distribution is governed or ruled by the fluid dynamics in the incinerator, cyclone and ceramic filter. Most of the HMs do not have enough residence time in this incinerator type to diffuse out of the ash particle and so remain in the particle. The amount of each HM in each exit flow in this process is governed by fluid dynamics and kinetics and not at all by thermodynamics.

Keywords: Incineration; Sewage sludge; Industrial sludge; Heavy metals; Fluidized bed; Combustion; Environment

1. Introduction

Fluidized bed incinerators have some advantages over rotary kilns, grates type and 'multiple hearths'... They are very well suited to incinerate solids of small size such as dried sewage and industrial sludges and refuse derived fuels. Incineration temperatures in fluidized beds are usually lower than in rotary kilns and grates, generating less NO_x emissions.

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Incineration of sewage sludge in fluidized beds is gaining popularity and replacing incineration in 'multiple hearth' incinerators. In the past, sewage sludge has been used as a fertiliser but this type of disposal is going to be restricted or forbidden because heavy metals and other components (such as microorganisms) in the sludge may contaminate soils and underground water, arriving then, by the trofic cycle, to humans. This potential problem is responsible for the increase of sludge incineration in developed countries.

Emissions of gaseous pollutants from incinerators is a very important problem. Perhaps, the most well known contaminants are dioxins and furans. There are also emissions of CO, NO_x, SO_2, and of volatile organic compounds (VOCs, PAHs, PICs). Emissions of heavy metals (HM) are not so well known by the public but they are, for these authors, as important and as dangerous as dioxins. This paper is concerned with only emissions of heavy metals and the extent they can be removed by using hot filters downstream from the incinerator.

Heavy metals are usually called 'trace metals' in the incineration process but this name will not be used in this work because, using doped (with metallic salts) sludge, the heavy metal content can be very high both in the feedstock and in the exit flows, and as such are not trace contents.

Several papers and reports have been published on the design and operation of industrial (commercial) fluidized bed incinerators for sludges [1–6,38]. All of them contain valuable information for this work. Concerning emissions from fluidized bed incinerators of sludge and waste, some papers are mainly focused on CO and NO_x emissions [7,8]. Other works are concentrated on VOC and PIC emissions [9,31], on technologies for cleaning the flue exit (stack) gas [10,34], or on other different scientific and technical aspects. There is a broad spectrum of work on subjects somewhat related to this paper.

This work is focused on the HMs behavior in fluidized bed incineration (FBI) of sludges, a field in which there is already an abundant literature. For instance, Lighty and co-workers [11,12] have developed new techniques to examine HMs distributions within cross-sections of ash particles which are of great interest for this work. Some other previous work focused on the fate, behavior or partitioning of different heavy metals in the incineration of sludges in fluidized beds (both bubbling and circulating ones) is indicated in Table 1.

In spite of such abundant and excellent previous work in this field, the authors and the European Commission (DGXII), among others, thought there were still details which needed further research. One of them is the usefulness of ceramic filters for hot gas cleaning of heavy metals. For this reason, EC (DGXII), under its BRITE-EURAM II Programme, financed a project to study the fate and possible partitioning of HMs in the incineration of sludges and of industrial waste. The authors of this paper were targeted (because of their experience of more of 30 years in fluidization technology) with evaluation of sludge incineration in bubbling fluidized bed. This type of incinerator will be the only one used in this study, although results obtained with it will be further compared with the ones from other types of incinerators.

Two characteristics make this work somewhat different with respect to those indicated in Table 1. First, sewage sludge contains a relatively small amount of HM. There can be contamination from the stainless-steel equipment. In fact in some introductory tests it was observed (from mass balances) how the overall amount of some HM increased after the incineration test. For this reason, to decrease the relative importance of contamination from the stainless steel, sludge was doped with some metallic salts. Second, hot (600–750°C)

Table 1
Compilating some previous papers on the fate of heavy metals in sludge incineration in fluidized bed

Ref.	Authors	Institution	Work
[13,14]	Latva-Somppi et al., 1996, 1998	VTT, Finland	HM distribution in combustion ashes
[15]	Lundberg et al., 1997	Kvaerner, Sweden	Analysis of gaseous emissions and of ashes
[16,17]	Lee et al., 1993, 1996	3 Universities, Korea	Behaviour of selected HMs
[18]	Louhimo et al., 1994	Tampella, USA and Finland	Results in some tests, commercial scale
[19,20]	Ho et al., 1994, 1996	Lamar University, USA	In-bed metal retention by sorbents
[21]	Kocinski et al., 1995	The University of Utah, USA	Cross-section analyses of fly ash particles
[22]	Rink et al., 1995		
[23]	Mininni et al., 1998	IRSA, Italy	Guideline for a safe incineration of sewage sludges
[1]	Barton et al., 1991	U.S. EPA	Compilation base of data
[9]	Parrish et al., 1991		
[24]	Ruth, 1998	U.S. DOE	Review on trace elements in coal and MSW incinerators
[25]	Wang and Lin, 1998	Inst. Techn., Taiwan	The use of solid sorbents to capture trace metals
[26]	Chen et al., 1997	University, Taiwan	In-bed metal retention in various Cl-containing feedstocks
[27–29]	Wey et al., 1996, 1997	University, Taiwan	Effect of PVC and HMs on emissions of organics
[30]	Chang et al., 1999	University, Taiwan	Effect of temperature on emissions of lead

filters were used in all incineration tests to study their usefulness regarding HMs partitioning. Hot filters have not been used very much to date as hot gas cleaning devices in waste incineration plants. To conclude, this work was focussed on three key aspects: (1) incineration in fluidized bed only; (2) sewage and industrial sludges as feedstocks; and (3) emissions of some targeted heavy metals (Cd, Cr, Cu, Ni, Zn and Pb). This paper is devoted to describing the full facility and methods used, together with important results. In a forthcoming paper, these results will be interpreted and modeled.

2. Experimental

2.1. Description of the pilot plant used

Incineration tests started at University ‘Complutense’ of Madrid (UCM) in 1992 in a small bubbling fluidized bed (FB) incinerator continuously fed near the bed bottom. The first facility had two in-series catalytic reactors located downstream from the incinerator for hot gas cleaning [31,32]. Due to the small height and/or size of the incinerator, the gas residence time in its upper part was quite low (<2 s) and relatively high amounts of CO and PICs were obtained in the flue exit gas, indicating poor incineration.

Since incineration tests had to be carried out at conditions close to those of industrial fluidized bed incinerators, CO and PICs in the flue gas had to be below the legal limits. Thus, a secondary air flow and a gas residence time (in the incinerator freeboard) higher

than 2 s had to be used and/or achieved. So, work was stopped in the first facility in 1995 and a new fluidized bed incinerator (FBI) was designed and constructed. The new incinerator was 70 mm inside diameter and 5.3 m total height. It was operated for 2 years (1996 and 1997) and 18 full incineration tests were carried out with it. This second incinerator still had some problems proving that it had some weak points/parts/devices which should be improved. For this reason, work with the second incinerator was stopped and modifications/improvements/revampings were made to it. The new incinerator (the third) is shown in Fig. 1. Results presented in this paper come from this unit only. Results and information from previous incinerators and plants are not reported on in this article.

Some details of the third generation incineration plant are as follows:

1. *Feed system*: The feed system has two in-series screw feeders (Fig. 1). The first one is a dosifier; the second one runs at high speed to avoid pyrolysis of the sludge in it.
2. *Feeding*: Feed is injected near the bottom bed (not at the bed surface). The sludge really enters into the hot bed. The heating rate of the sludge fed is very high. The time for the pyrolysis/gasification of the sludge, first step in the overall incineration mechanism, is very low and the yield to intermediate products, such as VOCs, is lower than when feeding is at the bed surface [33].
3. *Bed geometry*: The incinerator has an internal diameter of 150 mm at its bed zone. At the top of this zone there is a V shaped baffle (see Fig. 1) to decrease the carry over of solids from the bed.
4. *Bed composition*: An 'inert' silica sand was used in all incineration tests as the fluidizing medium. Its size was 1.0–1.6 mm (u_{mf} at operating conditions=0.50 m/s).
5. *Temperatures*: In the incinerator, temperatures were measured at five different heights (T_1: pre-heated air; T_2: bed; T_3: just above the bed; T_4: just below secondary air injection; T_5: freeboard, upper part). Examples of temperature profiles along the incinerator and exit pipes are shown in Fig. 2 for a few representative incineration tests. Due to the relatively low feed rate, only a small amount of CO and PICs were generated in the bottom bed. So, the secondary air flow was not capable of increasing the temperature in the freeboard of the incinerator, as it usually does in industrial FB incinerators. Since the temperature in the freeboard was somewhat lower than in the bottom bed, as Fig. 2 shows, some condensation of HMs could occur there. To avoid this to the maximum extent possible, freeboard and exit pipe were externally heated with electrical wires. Temperature in the flue exit gas was thus never below 550°C (Fig. 2). Total electric power used in the plant for heating was up to 40 kW.
6. *Typical operating conditions* (in the incinerator) were:
   inlet superficial gas (air) velocity: 1.2–2.0 m/s
   waste fed: around 2 kg/h
   Some other main experimental conditions are indicated in Table 2.
7. *Cyclone*: To achieve some separation between coarse and fine (small) fly ash, a cyclone was used at the top of the incinerator, before the filter chamber, as Fig. 1 shows.

2.2. Hot filter

After the cyclone at the incinerator top there is a high temperature (HT) filter box (Fig. 1). The flow inlet is tangential to let the whole vessel act as a cyclone. Some coarse fly ash is still

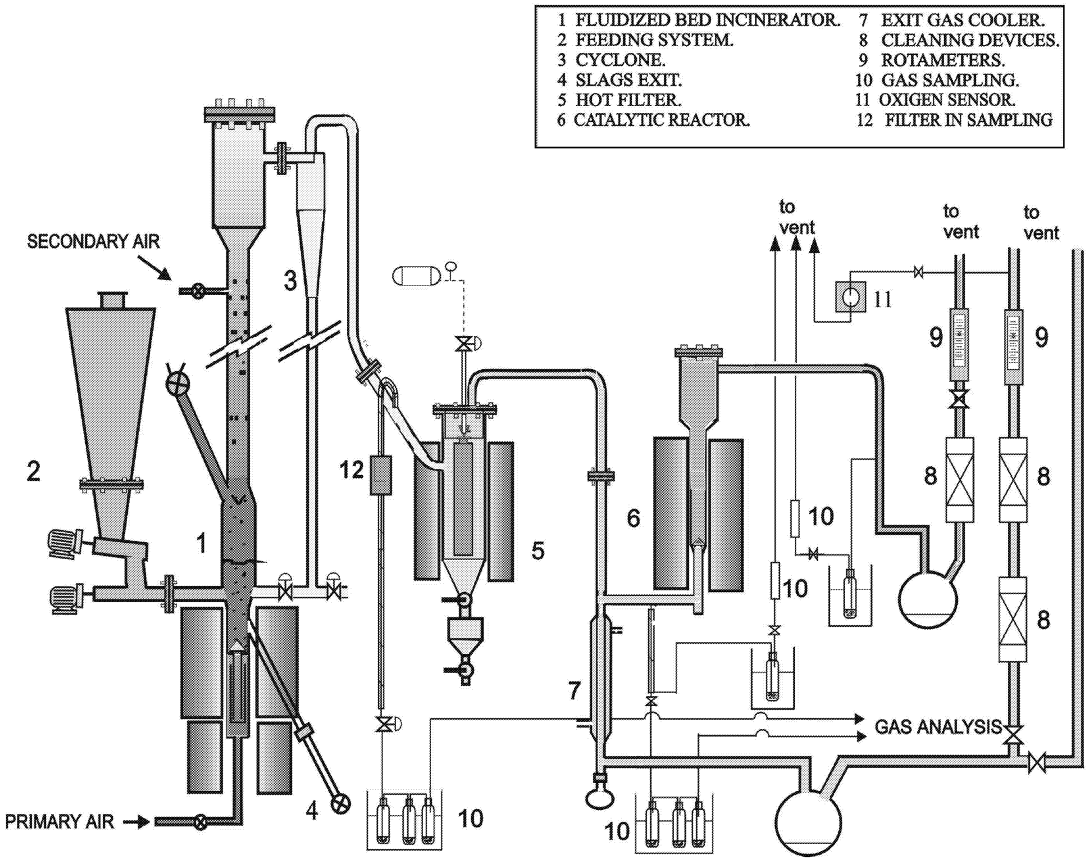

Fig. 1. Pilot plant at UCM for incineration of waste.

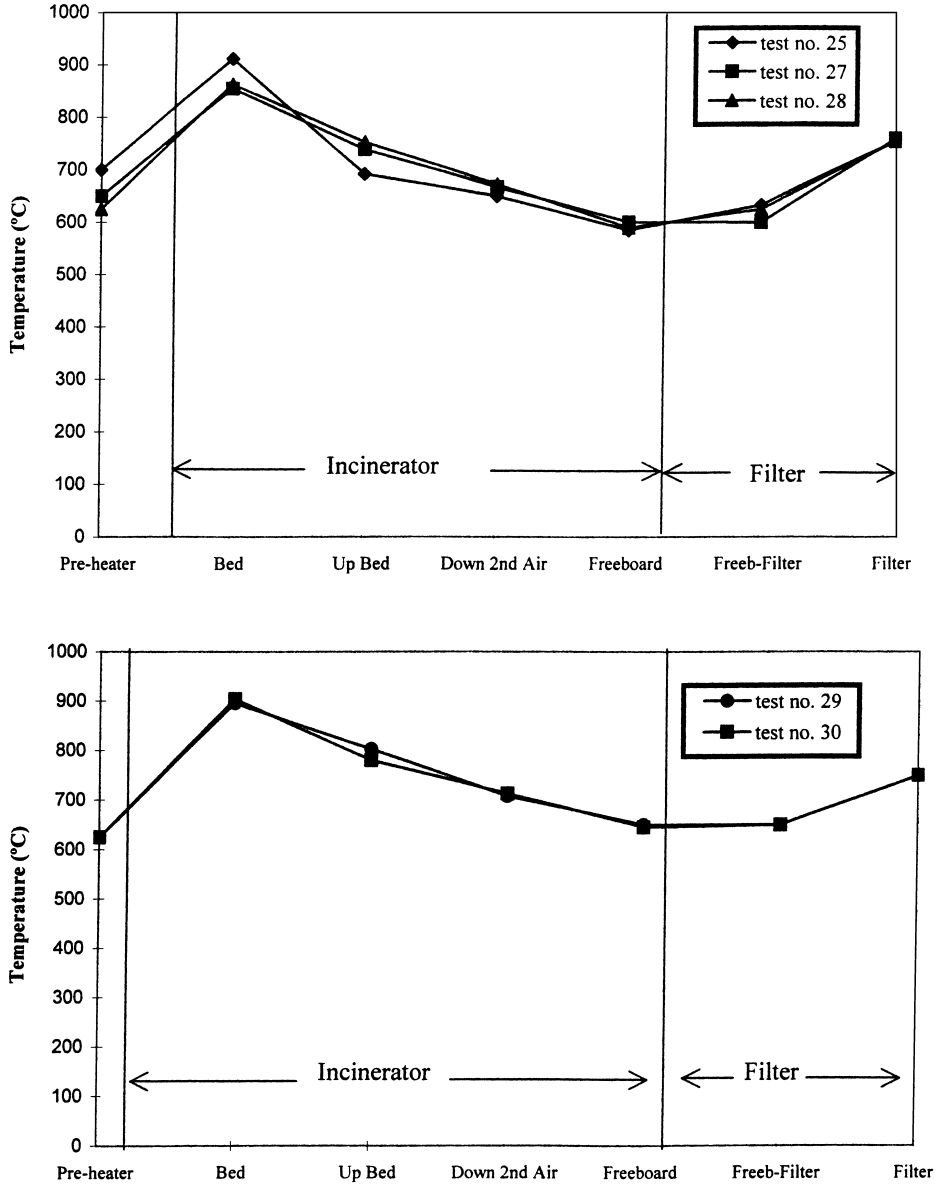

Fig. 2. Temperature profiles in some representative tests along the incineration facility.

separated from the main gas flow and collected at the bottom of this filter box (Fig. 1). This ash and the one collected in the cyclone (at the incinerator top) will be called 'coarse fly ash'.

Although different filters have been tested, results presented here will refer only to a ceramic one, called *Pantel*®, from DIDIER Filtertechnik GmbH. Its dimensions are 60 mm o.d., 40 mm i.d. and 50 cm length. Its average pore size is 15 μm. Its chemical analysis is:

Table 2
Important experimental conditions in the fluidized bed incinerator

Test No.	18	19	20	21	25	27	28	29	30	31	32
Waste fed											
Code	SL-1	SL-1	SL-2	SL-2	SL-3a	SL-3b	SL-4	SL-5a	SL-5b	SL-6	SL-7
Flow rate (kg/h)	1.2	1.6	1.6	1.5	1.5	1.4	1.4	1.8	1.7	2.0	1.7
Size (mm)	−4	−4	−4	−3	−4	−4	−4	−4	−4	−4	−4
Incinerator											
Throughput (kg/h m^2)	381	493	511	473	476	454	445	574	551	636	532
Height (fixed) bed (m)	0.35	0.35	0.45	0.45	0.45	0.4	0.4	0.4	0.4	0.4	0.44
Superficial gas velocity at inlet (m/s)	1.34	1.45	1.29	1.22	1.25	1.11	1.20	1.77	1.93	0.92	1.37
Primary air flow rate (Nm^3/h)	4.4	5.3	5.9	5.6	4.2	3.9	3.9	6.0	6.3	3.0	4.8
Secondary air flow rate (Nm^3/h)	0.2	0.3	0.3	0.3	1.1	0.9	0.6	0.78	1.1	0.54	0.55
Excess air (vol.%)	70	60	72	75	50	50	50	50	50	50	50
Incineration temperature (bottom bed) (°C)	950	950	980	940	913	856	864	896	904	804	834
Pressure (atm)	1.17	1.33	1.59	1.50	1.28	1.24	1.18	1.27	1.24	1.12	1.24

SiO_2, 57.8%; Al_2O_3, 34.3%; TiO_2, 1.0%; Fe_2O_3, 2.7%; CaO, 0.6%; MgO, 0.7%; Na_2O, 1.2%; K_2O, 1.7%.

The filter chamber was externally heated to obtain the highest temperature possible. The temperature in it, measured in two different points, was always between 650 and 750°C. Face velocities used were between 2 and 5 cm/s. Some other experimental conditions in the filter chamber are shown in Table 3.

The pressure drop across the filter (and cake) increased somewhat with time-on-stream. For this reason, a periodic cleaning of the filter was carried out. The periodic (and relative) cleaning of the hot filter was carried out by pulses of N_2 (in a reverse mode) of 0.2 s every 5 min. Without the increase in pressure, the incinerator was able of operating several hours without problems. This time was enough to get the required samples of gas and ashes.

2.3. Exit pipes

For a simultaneous study on the catalytic total oxidation of VOCs and PICs in the flue gas [34,35], downstream from the incinerator there is a slip flow with a catalytic reactor in it (Fig. 1).

Table 3
Experimental conditions in the filter chamber

Test No.	18	19	20	21
Inlet flow gas $(m^3/h)_{T,P}$	16	12.3	17.4	15.9
Pressure average (atm)	1.1	1.2	1.5	1.4
Particle load (g/Nm^3)	25.2	28.5	39.9	45.2
Face velocity (m^3/m^2 h)	165	179	163	164
Face velocity (cm/s)	4.6	5.0	4.5	4.5
ΔP across the filter (mm Hg)	60	175	402	311

To avoid large fluctuations in the exit flow there were two empty vessels, of 40 l each, in the main and slip exit flows (before the rotameters). The exit flow was easy and continuously measured.

The third generation incineration facility performed very well. Nevertheless, it has still two main drawbacks: (1) the small diameter of the screw feeders does not allow feed waste with particle sizes bigger than 1 cm; and (2) there is no external cooling of the incinerator bed, resulting in high temperatures (above 900°C) in the incinerator when some dried feedstocks/sludges are used.

2.4. Control

A distributive control system was developed by J. Prieto and co-workers at the Spanish High Council for Research, CSIC (Cantoblanco, Madrid) for this specific incineration pilot plant using the latest technology in digital communication (RS-485). A computer is used for data acquisition with different control loops in the incinerator. The multitask software was built in Borland C++ 4.5 for Windows 95®. It includes a Visual PC monitoring of temperatures.

2.5. Waste (feedstock) used in the incineration tests

The 'basic' feedstock used was a dried sewage sludge (SL) from a wastewater treatment plant (La China) of Madrid city. This sludge was received at UCM with 20 wt.% of solids. It was then dried to about 95 wt.% solids and crushed to reduce its particle size. It was then sieved, and the size below 4 mm was used in the incineration tests. This size did not offer problems in the feeding system used (shown in Fig. 1). Size distribution of the feedstock(s) is described in Table 4.

The 'basic sludge' used had a typical homogeneous aspect and a grey color. Its bulk density was 0.50 g/cm^3. Its elemental and proximate analyses are indicated in Table 5. It is well known that SiO_2, CaO and Al_2O_3 present in the sludge, and in the bed, can capture or react with HMs having an influence on their partitioning [19,21,22,25,26].

The HMs content of these 'basic sludges' is shown in Table 6. The heavy metal concentrations are considered relatively low which was an important problem for the UCMs incineration tests. In the first incineration tests, the amount of some HMs (such as Ni, Cr

Table 4
Particle distribution (wt.%) of the basic sludge (once dried, crushed and sieved)

Size (mm)	SL-1	SL-2
−4+3	38.5	35.2
−3+2	24.0	22.1
−2+1.6	13.0	15.2
−1.6+1	9.5	10.3
−1+0.5	6.5	6.9
−0.5	8.5	10.3

Table 5
Analysis of the basic sewage sludge used in incineration tests

	SL-1	SL-2
Elemental analysis (wt.%, d.a.f.)		
Carbon	24.8	21.7
Hydrogen	3.13	2.67
Nitrogen	2.55	2.01
Oxygen	17.9	15.1
Sulphur	1.02	0.97
Chlorine	0.03	0.03
Proximate analysis (wt.%)		
Ash	46.5	53.0
SiO_2	64.0	64.0
Al_2O_3	4.0	4.0
CaO	30.0	30.0
Volatiles	42.5	36.6
Fixed carbon	5.8	5.3
Moisture	5.0	4.9
LHV (MJ/kg)	12.6	12.3

and Cd) collected at the end of the test was higher than the amount fed. In the case of Ni and Cr it was probably due to the amount of Ni and Cr lost (by corrosion and erosion) by the stainless-steel incinerator walls and pipes. The net result of this loss was like a generation or production of Ni and Cr in the incineration plant. The small content in Cd (1–2 ppm) in the 'basic sludge' also generated strange results. Results from the

Table 6
Content of metals of the basic and of doped sewage sludges used in the incineration tests

Metal content (ppm)							
Test No.	Basic sludge		Doped sludge				
	SL-1	SL-2	SL-3a	SL-3b	SL-4	SL-5a	SL-5b
			25	27	28	29	30
Al	210	195	200	200	210	205	205
Fe	180	190	195	195	190	180	180
Cr	50	60	150[a]	2530	n.m.[b]	n.m.	n.m.
Ni	25	20	40[a]	130[a]	7810	4120	3790
Zn	680	650	2670	8460	7230	4070	4410
Pb	315	300	1010	2510	n.a.	350	390
Cu	230	240	2570	5120	6500	3370	3920
Cd	1.5	2.0	3.3[a]	2600	7200	3670	3800
Hg	2.0	2.0	2.0	2.0	2.0	2.0	2.0

[a] There were not enough amounts of salts in the lab to get higher contents in the final doped sludge.
[b] Not measured.

first incineration tests (about the fate of these three heavy metals: Ni, Cr, Cd) were thus unreliable.

For this reason, it was decided to dope the basic sewage sludge with some metallic salts to increase the HM contents in the feedstock. The HM salts added were: chromium nitrate; cadmium sulphate; cadmium acetate; nickel acetate; copper acetate; zinc acetate; zinc carbonate and lead acetate.

Doping of the basic sludge was performed by mixing the wet sludge (before its drying) with the metallic salts dissolved in water. Once a homogeneous mixture was achieved, the sludge dried. A homogeneous feedstock was thus attained. The amount of each metallic salt used in the last tests was to raise the HM in the doped sludge to an average of around 4000 ppm. HM contents of the resulting doped sludge is shown in the last columns of Table 6. These contents are clearly much higher than the ones in the former sludge as shown in first columns of Table 6. The error produced by the Ni and Cr lost by the walls and pipes (and transferred to the flue gas) using this doped sludge is now much lower. Results obtained with the doped sludge are much more reliable and reasonable. They will be the only ones presented in this paper.

The possibility of cross-contamination between test runs has been taken into account in two ways: (1) by cleaning as much as possible all the plant between test runs; and (2) by using a similar (as far as possible too) doped sludge in all tests.

Heavy metals are known to react with chlorine during the incineration process and produce various metal chlorides. The chlorine content in the feedstock affects metal partitioning [27,29]. Chlorine content in the basic sewage sludge (Table 5) is low (0.03 wt.%) for some studies. So, it was decided to increase the chlorine-content in the feedstock. For this reason some organic chlorine was added using PVC to the sludge.

The (Cl/HM) ratio in the feedstock is considered to be very important. Regarding the reactions of Cl with HMs indicated in Table 7, the chlorine in the feedstock can be sub-stoichiometric or higher than stoichiometric. For this reason, stoichiometric chlorine was carefully calculated for each test run. This value and the experimental chlorine fed are

Table 7
Chlorine content in some incineration tests

Test No.	25	27	28	29	30
(a) Heavy metals fed (at-g)	1.88	13.8	3.09	0.96	1.01
Cl amount					
(b) Content in feedstock (%)	0.03	0.03	0.03	3.00	3.00
(c) Fed (g)	39.7	1.52	1.31	131.16	130.45
(d) Experimental amount fed (at-g)	1.12	0.043	0.037	3.7	3.68
(e) Stoichiometric amount[a] (g)	126.9	949.4	219.2	68.02	70.34
(f) Stoichiometric amount (at-g)	3.58	26.78	6.18	1.92	1.99
(g) Stoichiometric (%)	3.30	18.30	5.84	1.55	1.62
[at-g Cl/at-g Metals]					
Theoretical [f)/a)]	1.90	1.94	2.00	2.00	1.97
Experimental [d)/a)]	0.60	0.0031	0.012	3.85	3.64

[a]Reactions considered: $Cu+2Cl \rightarrow CuCl_2$; $Cd+2Cl \rightarrow CdCl_2$; $Zn+2Cl \rightarrow ZnCl_2$; $Pb+Cl \rightarrow PbCl$; $Ni+2Cl \rightarrow NiCl_2$; $CrO_2+2Cl \rightarrow Cl_2CrO_2$.

shown in Table 7. According to these values:
test nos. 27 and 28 were made with a very low relative amount of chlorine.
test nos. 29 and 30 were made with some 'excess' of chlorine, and
test No. 25 was made with a 'modest' amount of chlorine (not-small-not-big).

3. Sampling and analyses

3.1. Gas sampling

There are three different sampling and/or analysis of the flue gas:

1. Gas samples were taken periodically before and after the HT filter. Gas sampling system used was an adaptation of US EPA Reference Method 5 and it is shown in Fig. 3. Gas samples were representative of the flue gas due to the high velocity of the flue gas at the point of sampling. These samples were then analyzed by gas chromatography. Typical flue gas composition is 8–9 vol.% O_2, 11–12 vol.% CO_2, 10–11 vol.% H_2O and 50–200 ppm CO. Condensates were also analyzed and their TOC corresponded to <50 mg organics/m^3_n flue gas.
2. Another type of gas sampling, for detailed VOCs determination, was made at the inlet and exit of the catalytic reactor located in the slip flow downstream from the incinerator. The effectiveness of some catalysts for total oxidation of VOCs and Cl-VOCs were measured. Standard resins (Tenax) were used in this sampling, and VOCs or Cl-VOCs were later analysed by GC and mass spectrometry. These analyses correspond to a simultaneous work and are shown in other papers [34,35].

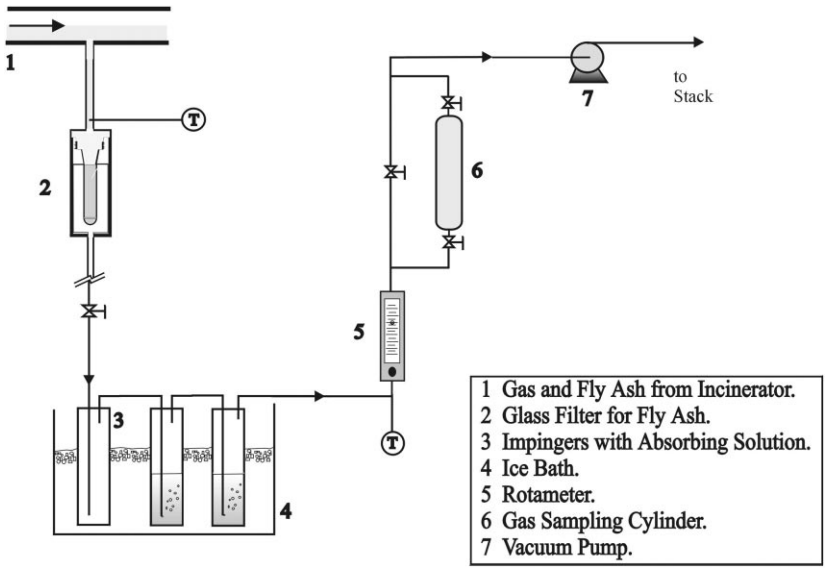

Fig. 3. Fly ash and gas (main components) sampling train system for measurement of the heavy metals content in the flue gas.

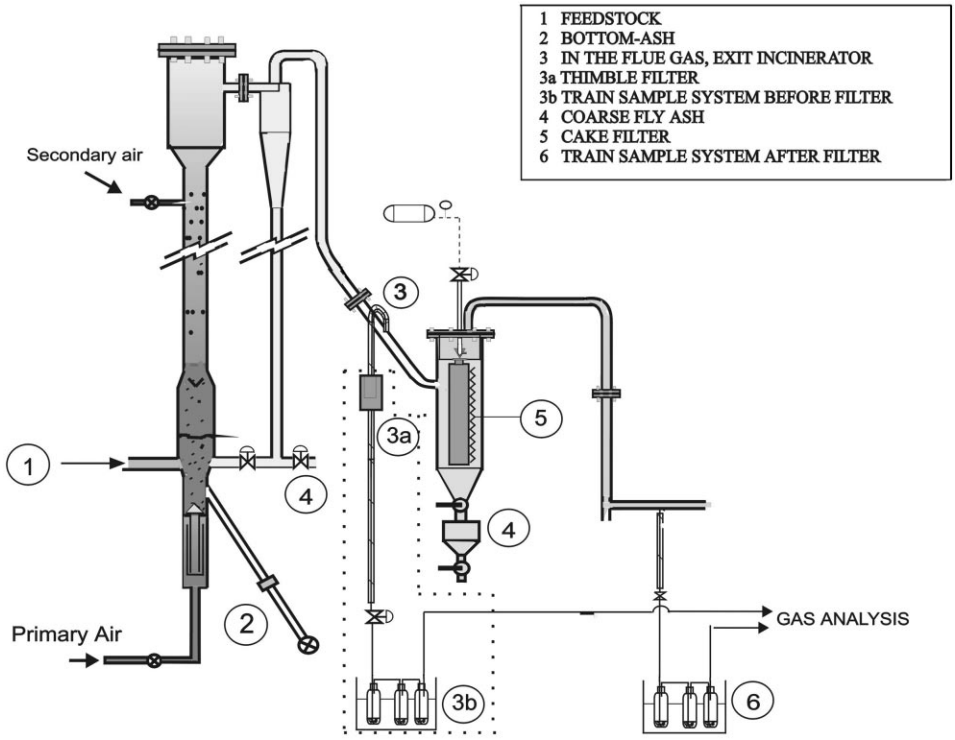

Fig. 4. Location of the sampling points for heavy metals.

3. There was an on-line and continuous measurement of the O_2 content in the exit flue gas (see Fig. 1).

3.2. Sampling and analyses of solids

3.2.1. Sampling

Besides the feedstock, samples of solids for heavy metals measurements were taken at the following locations (see Fig. 4):

1. incinerator bed (or bottom ash);
2. bottom of the cyclone and of the HT filter chamber (coarse fly ash);
3. cake on the filter surface (small or fine fly ash);
4. flue exit gas. This gaseous emission is collected in a serie of chilled impingers (see Fig. 3) containing a diluted (5%) nitric acid solution.

The samples are called:

1. bottom-ash (incinerator bed);
2. coarse fly-ash (collected at the bottom of the cyclone and of the filter chamber);
3. cake filter (collected on the filter surface, at the end of the test);
4. flue exit gas (after the hot filter).

Samples were also taken in the flue gas after the incinerator, before the hot filter, as Fig. 4 indicates. In this sampling point a thimble filter was used. The solid collected in the thimble filter was analyzed too.

3.2.2. Analysis

Six metals (Cr, Ni, Zn, Pb, Cu and Cd) were analysed in the above mentioned samples. The samples of ashes, once dissolved, were analysed by inductively coupled plasma-atomic emission spectrometry (ICP-AES) at the UCMs 'Atomic Spectrometry Center' (CEA). These analyses were not conducted by the UCMs Dept. of Chem. Eng. but were made by experts in inorganic chemical analysis at the CEA laboratories. The most advanced and standardised techniques were used. For instance, the solid samples (ashes) were first attacked (digested) with acid solutions (of FH, ClO_4H,...) following the procedure described in the DIN 38414/7 norm. The solid residue (fraction which was not totally dissolved by acids attack) was melted (microwave digestion) with lithium tetraborate at 950°C.

3.3. Main experimental conditions in the incineration tests

The best incineration tests were achieved after 1 year of using plant III. Such tests (25, 27, 28, 29, 30 and following ones) were carried out without major problems (such as like pipe blockages, filter damage, good filtration, flow rates and temperatures without fluctuations, ...). We have confidence in these tests and are the only ones reported. Other tests experience some trouble and are omitted from this paper.

Each incineration test required ≈2 man-months for cleaning, preparing and running the incineration facility, 2 man-months for chemical analysis and 1 man-month for managing, analysis of results, reporting, etc. ... Thus, one full incineration test in the facility needed 5 man-months of effort.

Each incineration test required much information, mass balances, feedstock preparation, etc. Only the main operating parameters (Table 2) are reported here. Among all operating parameters, the throughput (kg dry waste/h m^2 cross-sectional area) merits one comment: throughput in these tests was between 240 and 580 kg/h m^2 which is of the same order of magnitude than the throughputs of commercial fluidized bed incinerators.

A mass balance for the sludge fed and for the produced ashes was made in all test runs. Statistics or significance testing was always taken into account [36,37]. In the last test runs reported, error due to the ash lost in different points of the pilot plant was $<5\%$.

4. Results

In this facility and process, HM have four different exit flows, three as solids (ashes) and one as submicron particles diluted in the flue exit gas:

- bottom ash
- coarse fly ash
- cake filter or small-size fly ash
- flue gas after the hot filter.

Table 8
Fate of the heavy metals; partition coefficients (%) of the different heavy metals considered in test nos. 27 and 28

Incineration test No. 27						Incineration test No. 28				
Metal	Bottom-ash	Coarse fly-ash	Thimble filter	Cake filter	Flue gas	Bottom-ash	Coarse fly-ash	Thimble filter	Cake filter	Flue gas
Ni	24.1	39.5	0.30	35.6	0.5	50.4	29.6	0.20	19.6	0.2
Zn	21.9	42.2	0.20	35.4	0.3	50.1	27.6	0.20	21.8	0.3
Pb	22.2	43.8	0.20	33.7	0.2	–	–	–	–	–
Cu	26.6	42.1	0.20	32	0.8	55.2	28.6	0.20	15.8	0.2
Cd	22.3	35.6	0.20	40.2	1.8	48.5	20.7	0.30	29.9	0.5
Cr	24.4	41.2	0.20	34.1	0.1	–	–	–	–	–

There is also a minor amount of ash which is collected in the thimble filter, in the sampling point No. 3a in Fig. 4. This amount is small and can easily be overlooked.

The chemical analysis of each one of the four main exit flows together with a mass balance allows one to calculate the fate/distribution/partition of each heavy metal in each incineration test. It is shown in Tables 8 and 9 for four representative tests (27–30). This distribution or partitioning of each metal among the four exit flows is shown in Figs. 5 and 6 for four selected tests.

For a better understanding of the fate of the heavy metals, their content (percentage) in the three main exit flows of ashes are presented

bottom ash (Fig. 7)
coarse fly ash (Fig. 8)
cake filter (Fig. 9)

One very important fact appears in these figures: for a given incineration test, in each exit flow of solids (bottom ash, coarse fly ash, cake filter) the percentage or distribution of the different HMs is similar (between the interval of error). All HMs (except Cd) have the same partitioning ratio. No definitive enrichment of any HM (excepting Cd) in certain ash type was detected. For example, test nos. 27 and 30, all HMs (excepting Cd) have the following partitioning ratios:

Table 9
Fate of the heavy metals; partition coefficients (%) of the different heavy metals considered in test nos. 29 and 30

Incineration test No. 27						Incineration test No. 28				
Metal	Bottom-ash	Coarse fly-ash	Thimble filter	Cake filter	Flue gas	Bottom-ash	Coarse fly-ash	Thimble filter	Cake filter	Flue gas
Ni	24.1	54.4	0.70	20.7	0.04	44.2	34.9	0.5	20.5	0.03
Zn	27.4	50.1	0.80	21.6	0.1	52.9	27.0	0.6	19.4	0.14
Pb	19.6	55.2	1.8	23.0	0.4	34.0	37.5	1.0	27.0	0.59
Cu	19.3	57.9	0.90	21.8	0.1	37.9	39.4	1.0	21.1	0.67
Cd	18.1	48.3	2.5	30.7	0.4	18.3	34.3	2.4	43.1	1.83

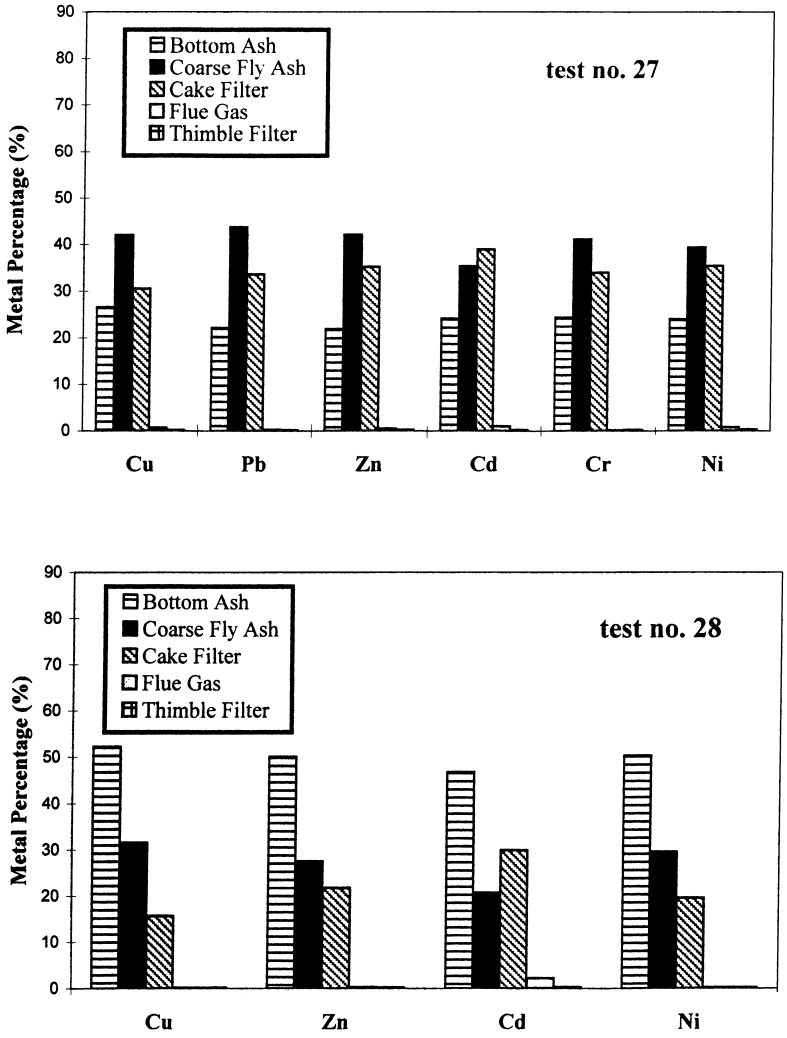

Fig. 5. Distribution/partition of each metal among the different exit flows in incineration test nos. 27 and 28.

Test 27	*Test 30*	
23–25 wt.%	36±4 wt.%	of each HM is in the bottom ash
39–42 wt.%	37±5 wt.%	of each HM is in the coarse fly ash
32–34 wt.%	21±8 wt.%	of each HM is in the cake filter
0.8±0.5 wt.%	1±0.5 wt.%	in the flue gas

The partitioning in each test can be different because of the somewhat different fluid dynamic conditions, such as superficial gas velocities, used in each incineration test.

The above results are a clear example that, concerning the fate or partitioning of the HMs, *there are no important differences between the different HMs; all HMs (except Cd) seems*

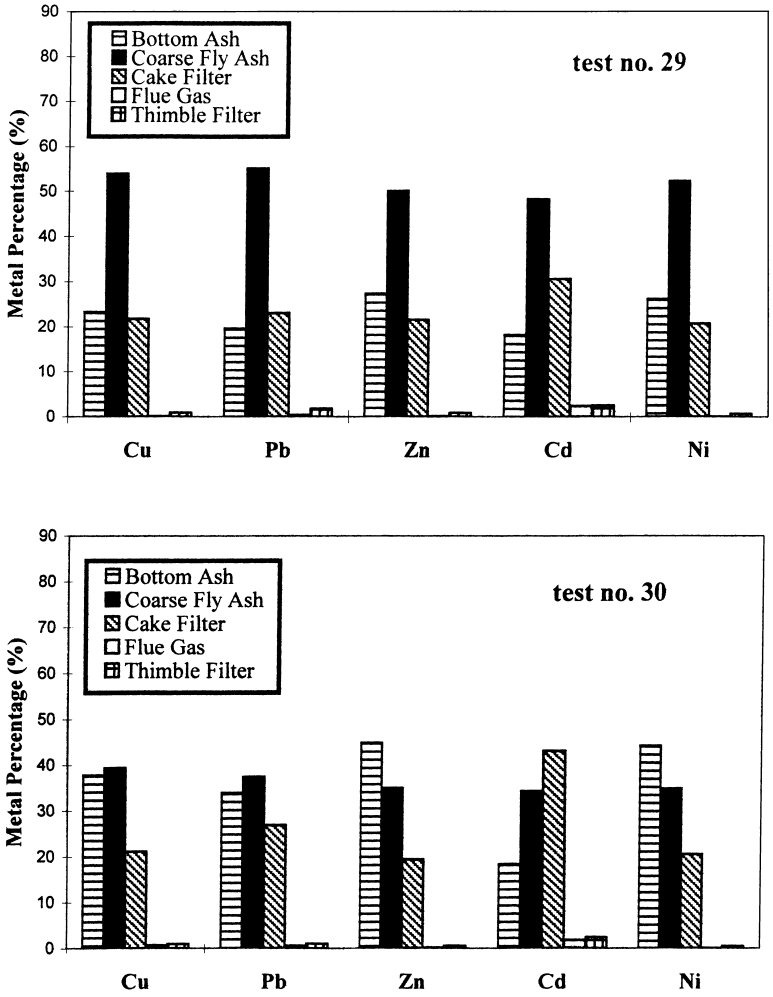

Fig. 6. Distribution/partition of each metal among the different exit flows in incineration test nos. 29 and 30.

to have the same fate, distribution or partitioning. These results agree for instance, with the previous findings of Latva-Somppi et al. [13] in VTT-Finland and of Lundberg et al. [15] in Kvaerner Pulping AB in Sweden. If the differences between the incinerator used by Lee et al. [16,17] in Korea and the one used in this work are taken into account, these results agree with Lee's work also. Partition coefficients shown in Tables 8 and 9 of the present work and those of Lee et al. [16,17] are similar, the small differences being due to the different FB incinerator and superficial gas velocities used in both works.

A detailed analysis of the results obtained in this work indicates that Cd shows a small difference with respect to the other HMs considered: *Cd-content in the bottom ash* is somewhat lower than the other HMs and Cd-content in *the flue exit gas* is higher than the other HMs. This fact can be easily understood remembering that Cd is the most volatile of the

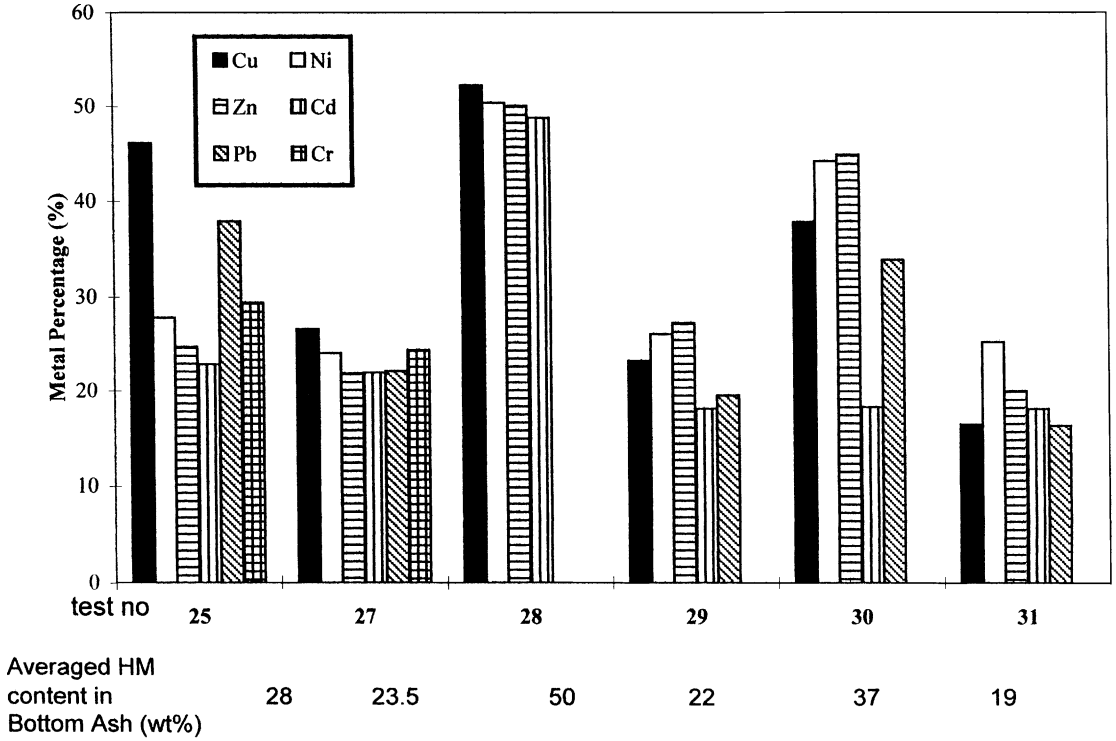

Fig. 7. Contents in heavy metals of the bottom ash got in six different incineration tests.

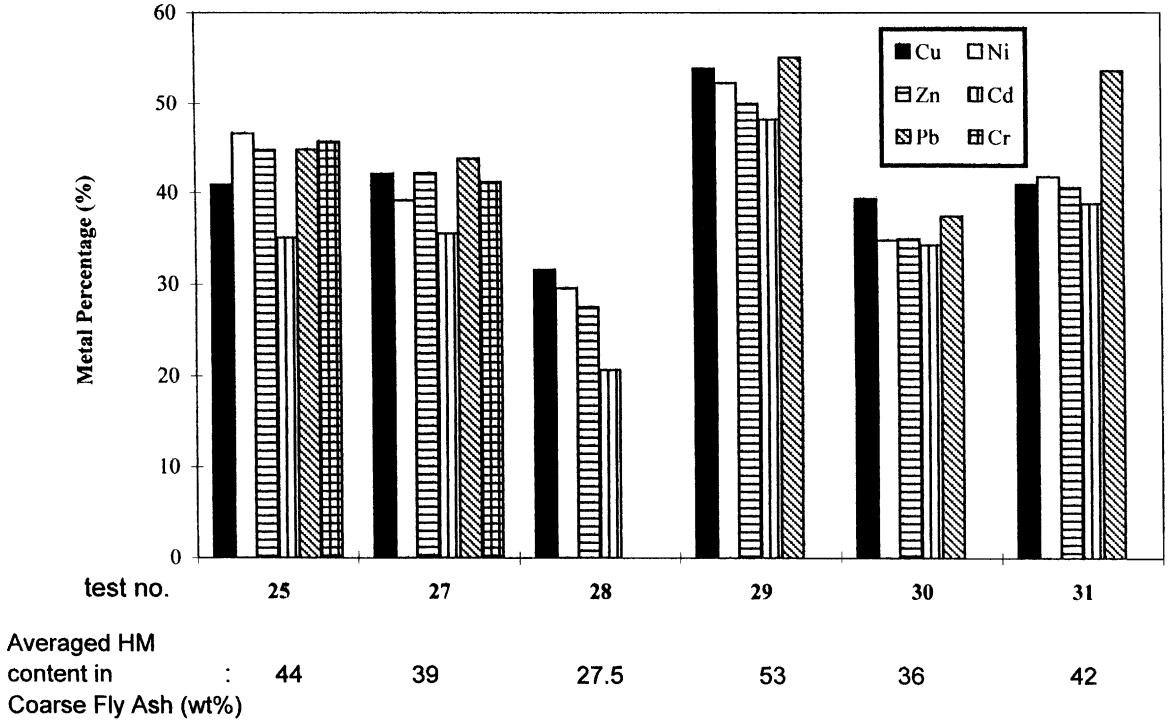

Fig. 8. Contents in heavy metals of the coarse fly ash got in six different incineration tests.

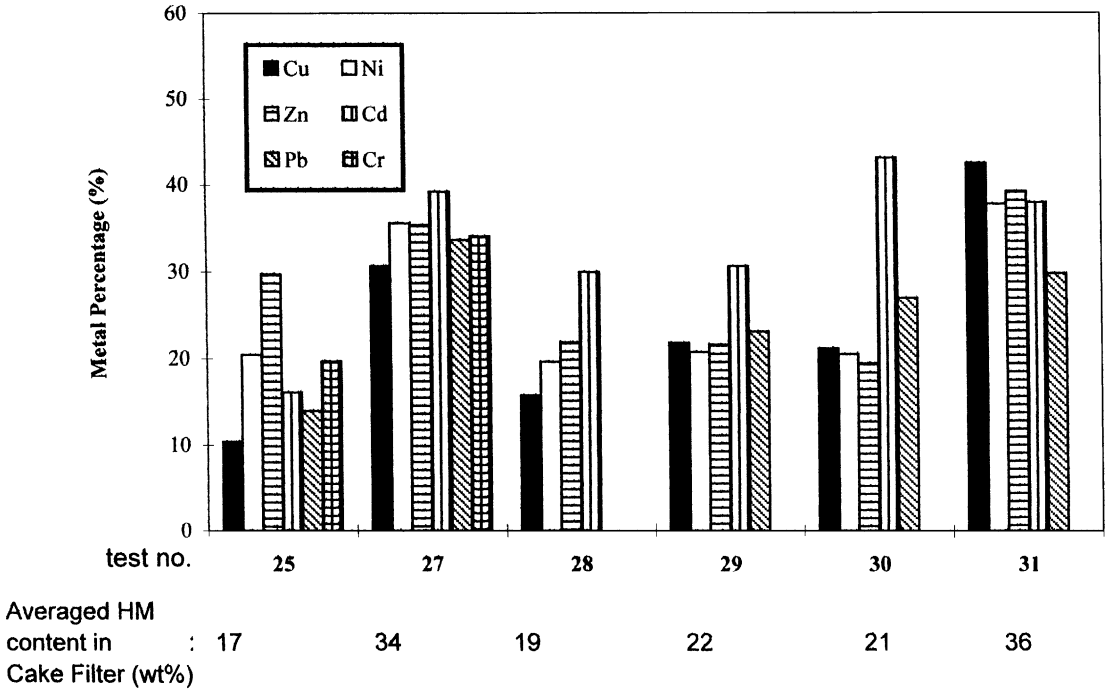

Fig. 9. Contents in heavy metals of the cake filter got in different incineration tests.

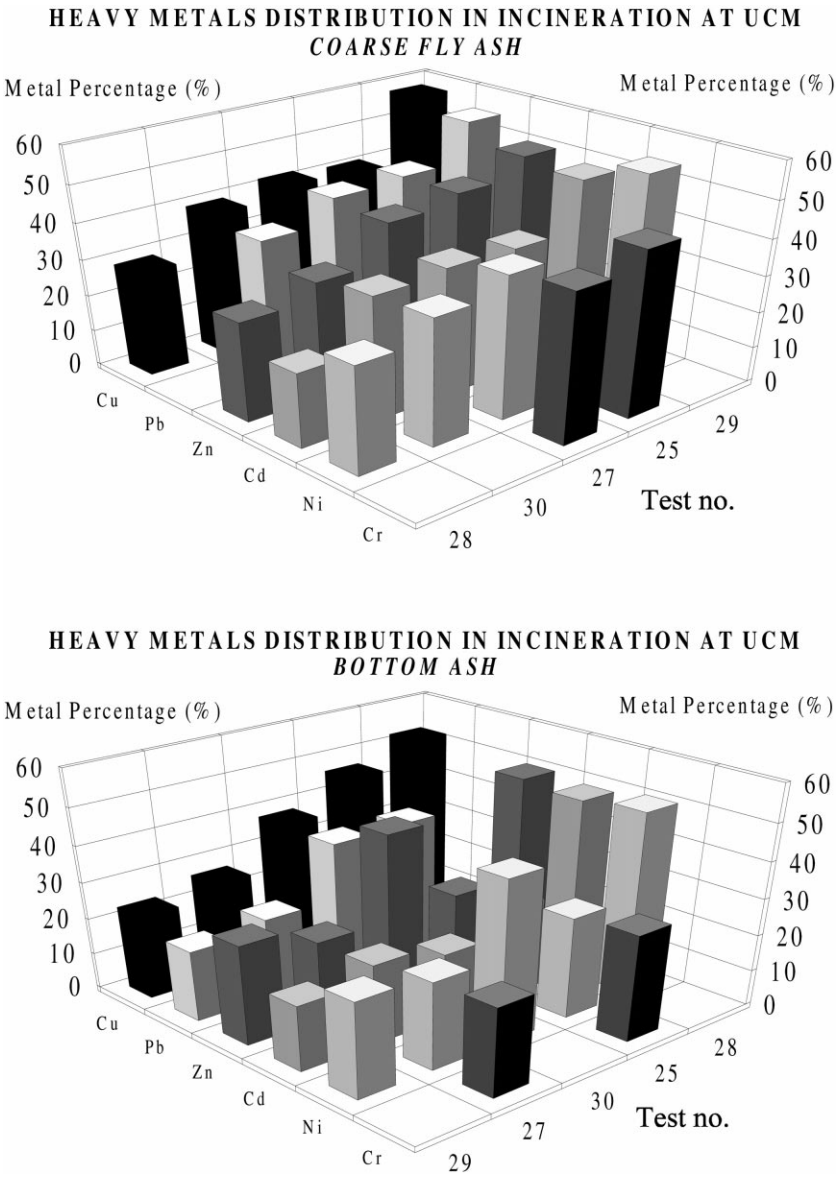

Fig. 10. Heavy metals behavior in incineration tests at UCM.

six HMs considered. Again, this finding agrees with previous results from other fluidized bed incinerators [13,15].

Results are shown together in Figs. 10 and 11. In these figures, the above conclusion is confirmed:

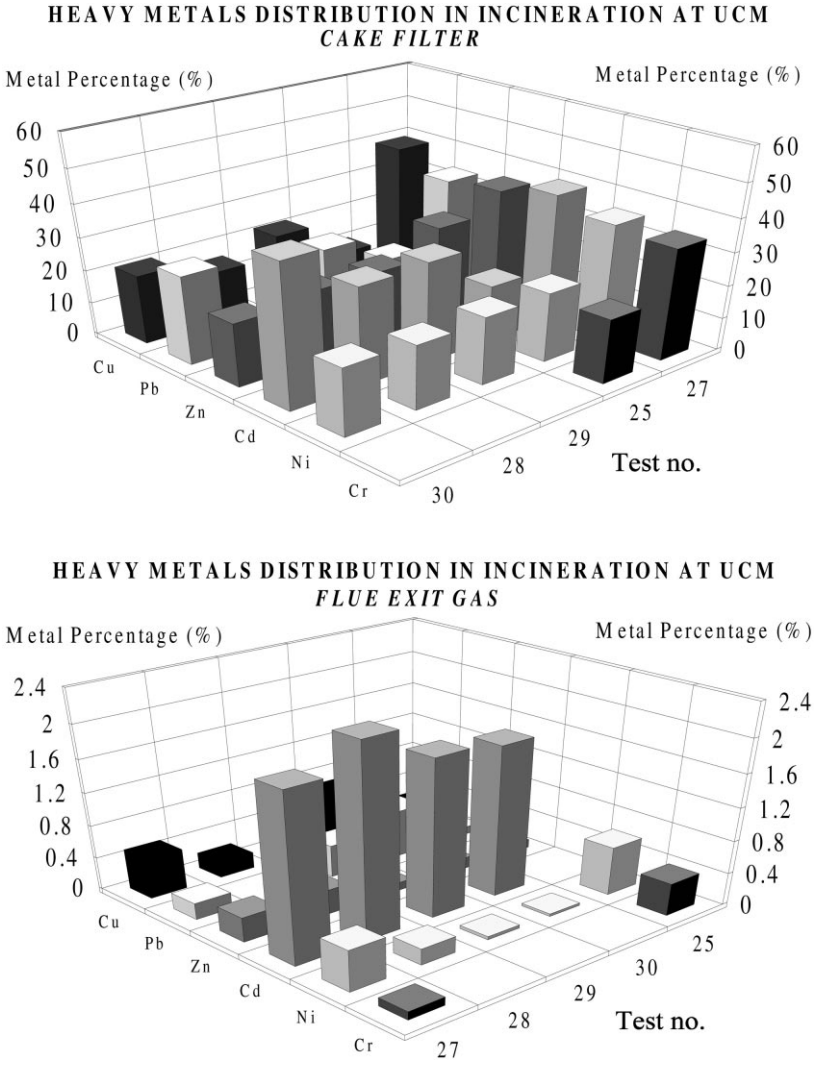

Fig. 11. Heavy metals behavior in incineration tests at UCM.

All the HMs have the same fate/distribution, excepting for the Cd which is more 'volatile'. (Cd-content in bottom ash is a little bit less than the other HMs. In the flue exit gas, and maybe also in the cake filter or fine fly-ash, the Cd is found in a slightly higher content than the other HMs).

There is some indication that Pb also behaves in a manner between Cd and the others HMs, although Figs. 7–9 and Figs. 10 and 11 do not offer a clear, statistical or conclusive trend. It seems that the Pb content in the fly ashes is a little higher than in the bottom ash, but such a difference is not important. More tests are needed to check this trend.

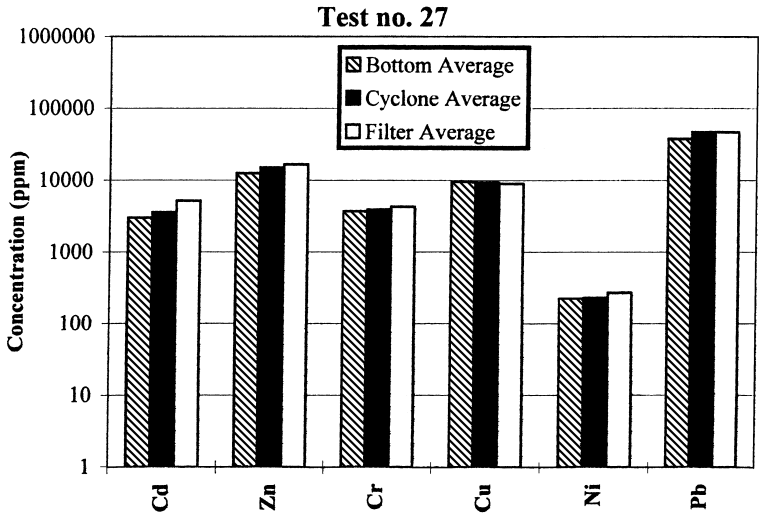

Fig. 12. Heavy metals content in the three flows of ashes in a test (No. 27) at UCM.

The fate of the HM can be presented in still another way. For a given incineration test, No. 27 for instance, concentrations of Cd, Zn, Cr, Ni and Pb in the three flows of ashes (bottom bed, cyclone and cake filter) are presented in Fig. 12. If these results are compared with the ones by Lundberg et al. (Ref. [15], see their Fig. 3) from an industrial fluidized bed incinerator from Kvaerner Pulping AB in Sweden, it can be determined how in both cases: (1) the concentration of each HM (excepting Cd) is near the same in all exit flows of ash; and (2) Cd-content is higher in the filter ash in both facilities/plants. In this comparison, the Zn content in ashes, for instance, is higher in results obtained in this work than in the ones from Kvaerner AB because the Zn content in both feedstocks was quite different: much higher in the feedstock at UCM than in the one used by Kvaerner AB.

All experimental results presented can be interpreted and modeled taking into account that numerous simultaneous phenomena occur in the fluidized bed incinerator used in this work (such as fluid dynamic aspects, residence time of the different sizes of the sludge and ashes, temperatures, volatility of the different HMs and of the their salts and oxides, etc....). This analysis and modeling will be presented in part II of this work.

List of symbols

BFB	Bubbling fluidized bed
FBI	Fluidized bed incineration test
HM	Heavy metal(s)
PAH	Polyaromatic compounds
PIC	Products of incomplete combustion
SL	Sewage sludge
T_1	Temperature of the pre-heated air
T_2	Temperature of the bed at the bottom of the incinerator

T_3	Temperature just above the bed
T_4	Temperature just below secondary air injection
T_5	Temperature of the freeboard, upper part
u_t	Terminal velocity of a particle, m/s
u_{mf}	Minimum fluidization superficial gas velocity, m/s
UCM	University 'Complutense' of Madrid
VOC	Volatile organic compounds

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