

## Sludge incineration tests on circulating fluidised bed furnace

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**Abstract** Results of sludge incineration tests on a demonstrative fluidised bed furnace are reported and discussed. They show that particulate, heavy metals and acidic compounds in the emissions can be easily controlled both when sludge is spiked with chlorinated hydrocarbons up to a chlorine concentration in the feed of 5%, and when the afterburner is switched off. As for organic micropollutants, polynuclear aromatic hydrocarbons (PAH) were much lower than the Italian limits of  $10 \mu\text{g}/\text{m}^3$  (no limits are at present considered in the European Directives). Dioxins (PCDDs) and furans (PCDFs) in some tests exceeded the limit of  $0.1 \text{ ng}/\text{m}^3$  (TE) but the concentrations in the fly ashes were much lower, thus evidencing a possible presence of contaminants in gas phase. PAHs and PCDD/PCDFs were not depending on the afterburning operation, the presence of organic chlorine in the feed sludge and the copper addition to sewage sludge.

**Keywords** Emissions; fluidised bed furnace; heavy metals; incineration; organic micropollutants; sewage sludge

### Introduction

Currently over 6.5 millions tonnes/year of dry sludge are produced in EU, and this amount will increase after the full implementation of the European Directive (91/271/EEC). In recent years many efforts have been made to improve sludge quality in order to increase the amount of sludge suitable for agricultural reuse, but in many cases this option still remains not applicable, due to insufficient sludge quality or to practical aspects. Incineration appears, therefore, in such cases the only practicable alternative, provided that landfilling is considered no longer sustainable as a disposal option.

Principal fears in sewage sludge incineration are still linked with organic micropollutants and metals emissions. Two main mechanisms are reported in literature for dioxins/furans formation (Altwicker *et al.*, 1990): a) PCDD/Fs are produced from related chlorinated precursors such as PCBs, chlorinated phenols, or chlorinated benzenes; b) they can be formed via *de novo* synthesis, i.e. they are formed from chemically unrelated compounds such as polyvinyl chloride, or other chlorocarbons, and/or the burning of non-chlorinated organic matter, such as polystyrene, cellulose, lignin, and coal in the presence of organic (or inorganic) chlorine donors. Both possibilities may include heterogeneous reactions. As far as *de novo* synthesis is concerned, a laboratory study by Stieglitz (1998) on fly ash from municipal waste incinerators found that particles of carbonaceous material from incomplete combustion were converted to a series of chlorinated aromatic compounds including PCDDs and PCDFs, through two basic reactions: a) the transfer of inorganic chlorine to the macromolecular carbonaceous structure with formation of carbon-chlorine bonds, and b) the oxidative degradation of the structure. In both processes transition metal ions, especially Cu(II), were involved as catalyst or reaction partners. Other important factors promoting dioxins/furans formation are fly ash composition, temperature (range  $300\text{--}350^\circ\text{C}$  appears critical), and the presence of oxygen in the gas phase. Addink and Altwicker (1998) found that aqueous extract from fly ash can produce small amounts of PCDD/PCDF when heated. The addition of CuCl and  $\text{CuCl}_2$  enhanced formation rates

drastically. Based on reaction order, they found that  $\text{CuCl}$  and  $\text{CuCl}_2$  are more important as a chlorine source than as a true catalyst. In his survey on the thermal formation of PCDD/PCDF, Fiedler (1998) summarised that: a) PCDD/PCDF can be formed in the gas phase as well in the heterogeneous phase; b) geometry of the combustion chamber, time, temperature, feeding rate, input chlorine, may have an influence on the formation; c) different processes might be dominating in formation of PCDD or PCDF; d) formation can be prevented by addition of inhibitors such as sulphur- or nitrogen-containing agents. However, a general review by ASME (1995) found no statistically significant relationship between chlorine input and PCDD/PCDF concentrations in the flue gas for 80% of the 90 facilities under observation. Only 11% displayed an increase; 9% of the facilities displayed decreasing PCDD/PCDF concentrations with increasing chlorine.

Another concern regarding organic emissions from waste incinerators is the formation of products of incomplete combustion (PICs). A number of studies (Taylor and Dellinger, 1988; Brouwer *et al.*, 1994a, 1994b, Wendt, 1994) showed that the formation of PICs is highly dependent upon the local ratio of fuel and oxidant, and that their amount and composition are sensitive to both turbulent mixing and chemical kinetic constraints. Mascolo *et al.* (1997) confirmed the above results in laboratory tests carried out by the System for Thermal Diagnostic Studies (STDS). They also found that the PICs formed during combustion upset conditions are mainly constituted by PAHs, and suggested detecting these compounds to check that proper incineration conditions are maintained. Sacchi *et al.* (1996) showed that detailed chemistry models under the assumption of ideal mixing conditions can give qualitative indication on formation of PIC species; their use was found to be appropriate to determine which species and ratios should be monitored as faults indicators. Tirey *et al.* (1991) compared results from sewage sludge field tests (one fluidised bed and three multiple-hearth incinerators) to those obtained in laboratory by STDS. They found a good qualitative correlation between the destruction efficiencies of different hazardous constituents in field tests and their incinerability ranking obtained from STDS tests under pyrolytic conditions.

Metals are present in sewage sludge at noticeable concentrations and they can be transferred to the gaseous phase depending on their volatility, on the presence of other species, and conditions inside the furnace (temperature, time of exposure, oxygen concentration). The speciation of metals in a waste incinerator is influenced by chemical equilibrium and kinetics. Though equilibrium identifies the potential stable species, the stability of a particular species under incinerator operating conditions is not solely determined by equilibrium (Karimanal and Hall, 1996). Ho *et al.* (1994) in their analysis of data from five rotary kiln incinerators found that incinerator facilities, including the design of the air pollution system, have the dominant effect on metal emission rates. Chlorine was identified to have a significant effect on metal volatilisation, because most metal chlorides have a much higher vapour pressure than their oxides. Generally metal emission rates are relatively lower at a lower incineration temperature. Chang-Yu Wu *et al.* (1994) developed an algorithm to relate waste feed and operating parameters (characteristics, operating conditions of the incinerator, operating conditions of the pollution control devices, atmospheric dispersion) with the ultimate exposure to metallic emissions. They also validated the model by comparison with data reported in the literature. Hinshaw (1994) evaluated partitioning of metals among various effluent streams and determined the true air pollution control equipment removal efficiencies in an incineration facility for solid and hazardous waste. Karimanal and Hall (1996) found that Cd is more volatile than its oxide in a laboratory scale furnace and Cd particles are distributed predominantly in the sub-micron range. Pb is less volatile than Cd, but is highly volatile in its chloride forms, especially  $\text{PbCl}_2$ .

The Italian CNR-IRSA built a demonstrative plant including a fluidised bed furnace.

Tests were carried out to assess contaminants emissions in critical conditions. The operating conditions were stressed spiking sewage sludge with highly chlorinated compounds up to a chlorine concentration in the feed of 5% and using different operating conditions of the afterburning chamber including switching the burner off.

## Materials and methods

The demonstrative plant is located in Bari (Southern Italy) in the area of a municipal sewage treatment plant. The main units are an indirect dryer, two incineration furnaces (circulating fluidised bed and rotary kiln), an afterburning chamber, a heat recovery system and a flue gas cleaning system (bag filter and wet scrubber). The throughput of the plant is about 250 kg/h of sewage sludge at 20% solids concentration (60% volatile) or 160 kg/h of dried sludge at 75% solids. Hazardous compounds can be injected in the sludge pipe before the furnace. The two furnaces can operate alternatively. The pilot plant is equipped with process analytical instrumentation for gas monitoring in different sections: (a) oxygen (exhaust gas from the furnace), (b) oxygen, carbon monoxide and dioxide and total hydrocarbons (exhaust gas from the afterburning chamber), (c) oxygen, particulate,  $\text{NO}_x$ ,  $\text{SO}_2$  and HCl (exhaust gas in the stack). Exhaust gases can be sampled in five points of the plant. The main equipment characteristics of the plant are shown in Table 1.

27 tests were performed with the fluidised bed furnace, 10 of them as replicate, to verify reproducibility. Tetrachloroethylene ( $\text{C}_2\text{Cl}_4$ ) or a surrogate organic mixture (SOM) consisting of 55% of tetrachloroethylene ( $\text{C}_2\text{Cl}_4$ ), 25% of chlorobenzene ( $\text{C}_6\text{H}_5\text{Cl}$ ) and 20% of toluene ( $\text{C}_6\text{H}_5\text{CH}_3$ ) were used as spiking agents. In four tests also a solution of cuprum (II) nitrate was added (about 0,25% of copper on dry basis), to stress conditions favouring the *de novo* synthesis of PCDD/PCDF. A few tentative tests in starved air conditions failed due to high air inleakage. This problem was also evidenced by Ho *et al.* (1994) who found that in fluidised bed operation air inleakage can be accounted for 39 to 49% of flue gas flow rate. Heavy metal concentrations in the feed sludge are reported in Table 3.

The main operating conditions are shown in Table 2. Either dewatered sludge (10.6-16.9% cake concentration) or dried sludge (94.6 and 99.2%) were fed to the furnaces. The higher heating value resulted 22,100 kJ/kg loss on ignition.

**Table 1** Equipment: main characteristics of the incineration plant

Operating unit	Parameter
Toxic wastes storage	15 m <sup>3</sup> (volume)
Dewatered sludge storage	15 m <sup>3</sup> (volume)
Sludge indirect dryer	30 m <sup>2</sup> (exchange surface) 440 kg/h (evaporation capacity)
Fluidised bed furnace	590 mm (internal diameter) 9.6 m (height) 800,000 kcal/h (heat capacity)
Cyclone	1,000 mm (internal diameter)
Rotary kiln furnace	1,200 mm (internal diameter) 6 m (length)
Afterburning chamber	1,200 mm (internal diameter) 5.3 m (height)
Heat recovery section	360,000 kcal/h (exchange capacity) 21,000 kg/h (oil flow rate)
Air preheater	1,500 Nm <sup>3</sup> /h air flow rate 60,000 kcal/h (air exchanger capacity)
Bag filter	90 m <sup>2</sup> (filtration surface) 4,000 Nm <sup>3</sup> /h (gas flow rate)
Gas washing column	1,100 mm (diameter) 12 m (height) 4,000 Nm <sup>3</sup> /h (flue gas flow rate)

**Table 2** Main operating parameters

Test No	Feed sludge (kg/h)	Total solids (%)	Loss on ignition (% of total solids)	Additives <sup>a</sup> (kg/h)	Cl content in the feed (%)	Total CH <sub>4</sub> (m <sup>3</sup> /h)	Furnace Exhaust gas temperature (°C)	Afterburner (°C)
1	250	15.83	67.26	–	–	91.99	840	833
3	233	10.56	63.66	–	–	50.62	844	566
3r	250	15.52	67.51	–	–	57.72	851	560
4	242	15.04	62.99	2.25 <sup>a</sup>	0.79	106.52	838	948
4r	245	14.29	65.38	5 <sup>a</sup>	1.71	99.86	865	899
6	200	15.29	66.32	5.09 <sup>a</sup>	2.12	53.10	830	466
6r	245	14.24	65.50	5 <sup>a</sup>	1.71	62.94	879	606
7	237	15.68	65.80	11.33 <sup>a</sup>	3.91	104.64	813	1151
9r	230	16.04	63.51	12.45 <sup>a</sup>	4.39	100.61	819	971
9	240	16.90	65.92	11.59 <sup>a</sup>	3.94	48.99	823	580
7r	230	15.12	65.80	12.50 <sup>a</sup>	4.41	73.06	821	534
10	240	16.77	64.69	12.00 <sup>a</sup>	4.07	104.51	819	996
10r	230	16.90	62.62	12.30 <sup>a</sup>	4.34	134.23	810	902
10A	230	15.48	62.93	12.00 <sup>a</sup>	4.24	110.73	826	797
11	227	15.18	65.03	23.00 <sup>b</sup>	5.05	134.97	831	1115
11r	227	15.53	62.88	23.60 <sup>b</sup>	5.17	115.77	849	971
13	217	15.99	64.35	22.55 <sup>b</sup>	5.15	50.22	848	543
13r	227	15.79	62.99	21.85 <sup>b</sup>	4.82	53.55	847	559
14	227	15.14	63.77	23.00 <sup>b</sup>	5.05	132.13	831	1000
14r	227	15.04	61.83	21.93 <sup>b</sup>	4.84	129.64	836	919
14A	227	15.84	62.62	23.06 <sup>b</sup>	5.07	112.84	850	856
15	241	14.05	61.62	22.87 <sup>c</sup>	4.76	121.23	845	1031
17	241	14.64	59.25	22.00 <sup>c</sup>	4.59	66.96	877	531
18	241	14.45	59.45	21.26 <sup>c</sup>	4.45	92.28	903	945
18A	241	13.92	58.73	21.80 <sup>c</sup>	4.81	89.73	841	743
44	120	99.18	60.68	–	–	64.70	859	902
45	127	94.64	58.77	–	–	62.87	991	949

<sup>a</sup> C<sub>2</sub>Cl<sub>4</sub>; <sup>b</sup> Surrogate Organic Mixture (SOM); <sup>c</sup> SOM + Cu(NO<sub>3</sub>)<sub>2</sub>·3H<sub>2</sub>O

In conclusion, the pilot plant was run with different kinds of feed (dewatered or dried sludge, dewatered sludge + C<sub>2</sub>Cl<sub>4</sub>, dewatered sludge + SOM, dewatered sludge + SOM + Cu(NO<sub>3</sub>)<sub>2</sub>·3H<sub>2</sub>O) and different operating conditions (afterburning chamber temperature).

The emissions were sampled in three points of the plant: before and after the bag house filter and at the stack. Organic micropollutants were sampled only in the first and last point.

**Table 3** Heavy metals concentration in the feed sludge (dry solid basis)

Metal	Concentration (mg/kg)		
	Minimum	Maximum	Average
Cd	3.1	23.3	10.6
Co	1.3	3.8	2.2
Cr	94.7	534.9	299.7
Cu	247.5	473.9	334.6
Hg	1.0	2.7	1.9
Mn	80.3	123.2	95.4
Ni	19.2	57.4	41.4
Pb	95.0	172.6	124.1
Sn	43.4	87.7	60.5
Ti	1028.4	1888.1	1448.3
V	2.0	22.0	18.3
Zn	1229.6	4213.0	2409.0

## Results and discussion

Pollutants concentrations at the stack are reported in Table 4 where the limit values of the European Directive 94/67 on hazardous waste incineration and of the Italian Decree 503/97 on special waste incineration are also shown.

Performance of the plant was very satisfactory for particulate,  $\text{NO}_x$ ,  $\text{SO}_2$ , metals and PAHs emissions. Also Zn concentration appears quite low in spite of its abundant presence in feed sludge and high volatility of chlorine related compounds. Conversion of organic nitrogen in the sludge to  $\text{NO}_x$  was found to be lower than 10%.

The CO concentration was normally below the limit of the European Directive 94/67 of  $100 \text{ mg/Nm}^3$  (half hourly average). Only 4 values were found to be higher, due to unsteady state conditions following a power black out or non-homogeneous mixing of additives and sludge with formation of spiking agents pockets of very high heat content. Two values of HCl exceeded the half hourly limit of  $60 \text{ mg/m}^3$  and some others the mean daily limit ( $10 \text{ mg/m}^3$ ), due to operating problems in the NaOH feeding system. It must be pointed out that the nominal capacity of the washing column was exceeded in some tests for over 50%.

PCDD/PCDFs concentrations (TE) were in some cases higher than limits of  $0.1 \text{ ng/Nm}^3$ . Some discrepancies were evidenced between such results and those relevant to the fly ash concentration. The expected concentration at the stack, on the basis of the particulate emissions, would be of some orders (up to three) of magnitude lower than the measured values, thus evidencing the presence of such contaminants especially in gaseous phase. Such aspects are still under investigation. Figures 1 and 2 show that no correlation exists between dioxins + furans and PAHs and between the above contaminants and the afterburner temperature. No remarkable *de novo* synthesis of PCDD/PCDFs was evidenced in the tests with high Cu dosage. Some peaks of dioxins and PAHs appear with no spiked sludge, thus evidencing a poor influence on the emissions of organic chlorine.

As far as the operating conditions are concerned, only the indirect dryer presented problems, requiring continuous monitoring due to the sticky nature of the sludge in the range 40–60% DS. It was necessary to recycle dried material to correct the input moisture to the dryer, which has not to overcome the value of 40%.

## Conclusions

Sludge incineration tests were performed on a demonstrative plant in different operating conditions to assess the influence of organic chlorine addition and of the afterburning chamber operating modes on the emissions. 27 tests were carried out with fluidized bed furnaces spiking sewage sludge with tetrachloroethylene ( $\text{C}_2\text{Cl}_4$ ) or with a mixture of tetrachloroethylene, chlorobenzene ( $\text{C}_6\text{H}_5\text{Cl}$ ) and toluene ( $\text{C}_6\text{H}_5\text{CH}_3$ ) up to a chlorine

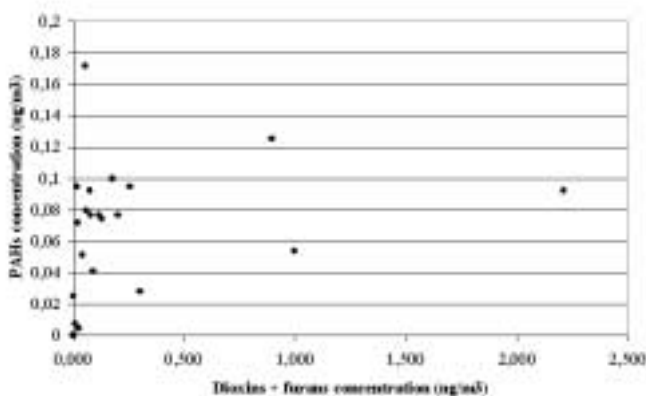


Figure 1 PAHs vs PCDD+PCDF

**Table 4** Main pollutants concentration at the stack (reference conditions: O<sub>2</sub> concentration 11%, 101,3 kpa, 273°k)

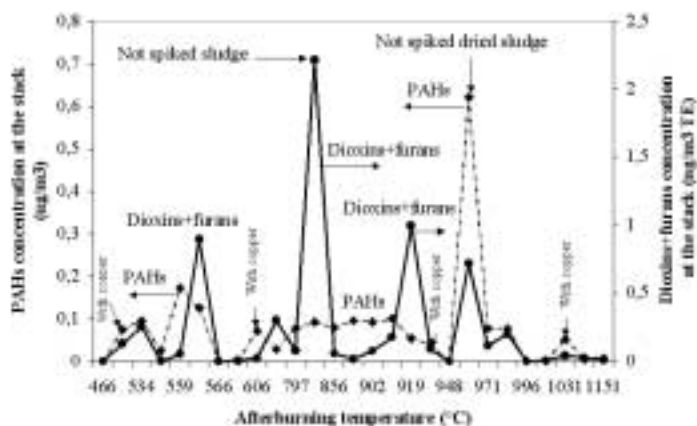
Test No	Particulate (mg/m <sup>3</sup> )	NO <sub>x</sub> (mg/m <sup>3</sup> )	SO <sub>x</sub> as SO <sub>2</sub> (mg/m <sup>3</sup> )	HCl (mg/m <sup>3</sup> )	CO (mg/m <sup>3</sup> )	Zn (mg/m <sup>3</sup> )	Σ 10 metals <sup>a</sup> (mg/m <sup>3</sup> )	PAHs (mg/m <sup>3</sup> )	PCDD+PCDFs (ng/m <sup>3</sup> TE)
1	0.75	21.10	30.00	74.34	0.60	0.050	0.042	9.231E-02	2.211E+00
3	0.42	10.60	6.20	2.60	3.12	<0.001		3.967E-04	1.411E-05
3r	0.38	64.03	25.10	68.20	62.50	0.042	0.018	1.256E-01	8.952E-01
4	0.25	65.50	14.00	10.50	115.00	<0.001		4.597E-05	1.110E-04
4r	0.22	75.95	120.00	17.38	72.70	<0.001	0.023	9.487E-02	1.644E-02
6	<0.10	94.00	<1.00	28.00	12.90	<0.001		6.164E-05	1.058E-04
6r	0.32	69.50	34.30	9.95	9.72	0.065	0.023	7.179E-02	2.125E-02
7	11.45	67.50	14.50	<0.10	8.50	0.012		7.751E-03	9.023E-03
7r	0.10	79.56	13.10	27.85	29.90	<0.001	0.086	9.487E-02	2.546E-01
9	1.27	10.10	16.00	4.90	7.15	0.040		2.893E-04	3.013E-03
9r	<0.10	148.60	1.39	1.06	10.35	<0.001	0.057	7.692E-02	1.143E-01
10	1.05	65.00	<1.00	17.75	2.66	0.052		3.975E-04	8.769E-04
10r	0.10	92.11	2.33	14.00	4.62	<0.001	<0.001	9.231E-02	7.313E-02
10A	0.16	57.36	154.50	29.20	123.40	0.084	0.017	7.692E-02	7.960E-02
11	3.40	3.15	<1.00	2.05	0.70	0.021		4.821E-03	2.438E-02
11r	<0.10	84.40	35.70	13.95	6.05	0.095	<0.001	7.692E-02	2.011E-01
13	0.65	80.50	50.00	3.15	9.30	0.100		2.503E-02	9.907E-04
13r	<0.10	105.10	10.20	21.00	38.50	0.042	0.236	1.718E-01	5.285E-02
14	2.60	110.00	6.60	17.50	17.00	0.050		2.718E-04	3.802E-03
14r	1.73	273.00	16.00	10.39	164.70	0.054	0.261	5.385E-02	9.970E-01
14A	0.74	171.20	16.60	39.67	12.90	<0.001	0.061	7.949E-02	5.656E-02
15	0.07	65.55	40.20	26.57	25.76	<0.001	<0.001	5.128E-02	4.082E-02
17	2.38	120.69	3.76	55.00	1550.0	0.003	0.052	7.436E-02	1.274E-01
18	0.13	67.40	34.40	20.13	13.60	<0.001	<0.001	4.103E-02	8.872E-02
18A	0.10	94.54	15.55	38.38	37.24	0.045	<0.001	2.821E-02	3.009E-01
44	0.71	2.55	15.00	15.50	8.40	0.004		1.000E-01	1.761E-01
45	0.09	9.21	4.27	15.00	0.83	0.013		6.205E-01	7.203E-01

<sup>a</sup> Sb+As+Pb+Cr+Co+Cu+Mn+Ni+V+Sn

	Particulate	NO <sub>x</sub>	SO <sub>2</sub>	HCl	CO	Zn	Σ 10 metals	PAHs	PCDD+PCDFs
Range	<0.10–11.45	2.55–273	1.39–154.5	<0.10–74.34	0.6–1550.0	0.003–0.1	0.017–0.261	4.597E-05-0.6205	1.411E-05-2.211
Mean value	1.264	78.081	28.321	22.462	30.696	0.045	0.080	0.076	0.240
EU Directive 94/67	10 <sup>a</sup> /30 <sup>c</sup> /10 <sup>d</sup>	–	50 <sup>a</sup> /200 <sup>c</sup> /50 <sup>d</sup>	10 <sup>a</sup> /60 <sup>c</sup> /10 <sup>d</sup>	50 <sup>a</sup> /100 <sup>c</sup> /150 <sup>e</sup>	–	0.5 <sup>f</sup>	–	0.1 <sup>g</sup>
Italian DM 503/97	10 <sup>a</sup> /30 <sup>b</sup>	200 <sup>a</sup> /400 <sup>b</sup>	100 <sup>a</sup> /200 <sup>b</sup>	20 <sup>a</sup> /40 <sup>b</sup>	50 <sup>a</sup> /100 <sup>b</sup>	–	0.5 <sup>h</sup>	10 <sup>i</sup>	0.1 <sup>i</sup>

<sup>a</sup>daily mean value; <sup>b</sup>hourly mean value; <sup>c</sup>half hourly mean value; <sup>d</sup>97° percentile half hourly mean values; <sup>e</sup>95° percetile 10 minutes mean values; <sup>f</sup>mean value over a sampling period of 0.5–8 h;

<sup>g</sup>mean value over a sampling period of 6–8 h; <sup>h</sup>mean value over a sampling period of 1 h; <sup>i</sup>mean value over a sampling period of 8 h



**Figure 2** PAHs and dioxins + furans concentration vs. afterburning temperature

concentration in the feed of 5%. Besides chlorinated hydrocarbons, in four tests also a solution of copper nitrate ( $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ ) was injected before the furnace to investigate the catalytic effect of copper on the *de novo* synthesis of dioxins/furans.

The performance of the plant resulted very satisfactorily for particulate, metals, sulphur dioxide, nitrogen oxides and polynuclear aromatic hydrocarbons (PAHs), which fulfilled the standards of the European Directive 94/67 in all the tests. The values of nitrogen oxides showed a low conversion of organic nitrogen to  $\text{NO}_x$  (around 10% of the total nitrogen). Some problems were evidenced with HCl and CO due to overloading of the washing column and to a poor mixing of spiking agents with sludge before the furnace with formation of pockets of products with a very high calorific value.

Organic micropollutants were very low in most of the tests and only in some cases (sludge without any spiking agent) PCDD/PCDFs have overcome the limit of  $0.1 \text{ ng/m}^3$ . The concentration in fly ashes was always very low thus evidencing an unexpected presence of such contaminants in the gaseous phase. No correlation was found between organic micropollutant emissions and chlorine and copper addition to sludge as well as the afterburning operation.

In conclusion, co-incineration of sewage sludge and hazardous compounds up to a chlorine concentration in the feed of 5%, if properly operated, appeared a safe and feasible outlet option, and afterburning of exhaust gas does not seem to have a remarkable effect on final emissions.

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