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PCDDs AND PCDFs CONCENTRATION IN COMBUSTION GASES AND BOTTOM ASH FROM INCINERATION OF HOSPITAL WASTES IN POLAND

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Abstract

The result from the determination of polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in combustion gases and bottom ash from eighteen of Polish hospital waste incinerators is presented. Thirteen of the investigated plants were built between 1994 and 1997. Eight of them shown low PCDDs/Fs concentration in stack gases, below 0.1 ng-TEQ/m^3_n. For two plants concentrations of PCDDs/Fs in stack gas were above 20 ng-TEQ/m^3_n. In all of the samples of bottom ash taken from pyrolytic chambers, PCDDs and PCDFs were in the level of 8 - 45 μg TEQ/kg. A method used for sampling and analysis is described.

Keywords:

hospital waste incineration, bottom ash, pyrolysis, combustion gases, analysis, PCDDs, PCDFs

Introduction

Wastes which are produced in Polish hospitals have variable harmfulness. In conditions of hospital treatment two types of wastes are produced. The first group are wastes of structure and characteristics of municipal wastes, like glass, paper, kitchen wastes and others from hospital service. Due to the fact that till 1997 there has not been built any municipal wastes incineration plant in Poland, these wastes have been dumped on municipal dumping grounds. The second group includes wastes which are produced in laboratories, operating rooms, consulting rooms and hospital units. That group of wastes is incinerated because of thread of infection.

Table 1 presents characteristics and amounts of wastes produced in Polish hospitals monthly and actual means of treatment. Data are presented for 1996 [1].

Table 1.

Hospital beds (1996)	237 230
Total weight of hospital wastes monthly [kg]	**14 805 500**
Wastes landfiled	
Total	11 645 000
As infected	890 000
Wastes incinerated in power plants	1 578 000
in new incinerators	**1 580 000**
Wastes buried in graves	2 500

Average capacity of the new incinerator is about 100 000 kg monthly.

Incineration is commonly used method to be quite safe means of neutralization of infected wastes from hospitals provided that it is carried out under appropriate thermal conditions so that it does not lead to contamination of the environment by toxic chemical compounds, which are produced in reactions of secondary synthesis which takes place during high temperature processes.

Neutralization of wastes was realized in Polish hospitals until 1994 by burning them in hospital power stations fired with hard coal, which are not designed for waste incineration nor equipped with proper devices for purification of combustion gases [2]. Till May 1997, there have been built and commissioned 13 new, modern hospital waste incineration plants.

Based on actual needs analysis made by Ministry of Health and Social Welfare and on the base of guidelines of WHO [3], it was considered a list of 23 central incineration plants for hospital wastes. The Project has started in 1994 and is funded mainly by Polish National Foundation for Environmental Protection.

The operating areas of these incineration plants cover whole provinces. Some incinerators serve for hospitals which are more than 100 km from the plant.

Thermal utilization of wastes are differentiated in technology on the base of pre-treatment of wastes at higher temperature and later after-burning of combustion gases.

Most of the new installations for incineration of hospital wastes are equipped with systems which enable to undertake two-stage thermal process. Some of them apply only first step of thermal treatment , in others, these two stages take place in one technological operation, e.g. fluidized bed burning.

However, the solution of thermal treatment of wastes is applied, as a final result we obtain combustion gases and residue, which contain chemical compounds formed in thermal processes [4].

Because of complexity of chemical and physic-chemical processes of formation of highly toxic organic compounds during thermal processes and the fact that they remain in residue after burning, high exploitation and investment costs, it is necessary to find compromise between costs of the process of thermal utilization of wastes and its influence on the environment. In most cases in Poland, however, there were chosen technologies scanted with systems of stack gas cleaning. Economical aspects and actual neutralization needs of harmful and difficult to dispose wastes are considered as superior.

This policy, not without reason, has been realized in Poland for 5 years.

Because of the lack, till May 1997, instructions which referred to the permissible level of emission and lack of laws imposing on the user the duty to control of the level (including polychlorinated dibenzodioxins and dibenzofurans), there have been built and exploited many incineration plants which have not been technically and technologically adequate to fulfill requirements posed on such installations in the West. There are, however, some of them which fulfill these requirements.

In Poland, it was accepted the project of utilization of wastes by thermal methods. Likewise, this problem is solved in other countries. In most cases, however, hospital wastes are utilized together with other wastes in installations of large capacity, at the level of tons per hour. In Poland, the installations which have been built during 1994-1997 have the capacity of 50 to 1200 kg/hour.

Determination of the concentration of toxic substances in stack gases and bottom ashes, emitted from incineration plants, was performed in 1994-1997. All of the new hospital waste incinerators in Poland were subjected to this determination.

The incinerators were tested from the moment they reached nominal exploitation parameters. In two cases, measurements were repeated after one year of exploitation.

To these measurements were subjected 13 new incineration plants and 5 older, which are equipped with ineffective devices for purifying of combustion gases or even do not have them.

From these 18 installations, only 5 are equipped with systems of purification of combustion gases which do not dispose liquid effluents. From 13 installations, 11 dispose the effluents to sewage-treatment plants whereas two of them dispose them to municipal collectors. In non-sewage solutions, wet systems of combustion gases purification are also applied but the excess of used, recirculating absorbing solution is evaporated, what leads to cooling the fumes.

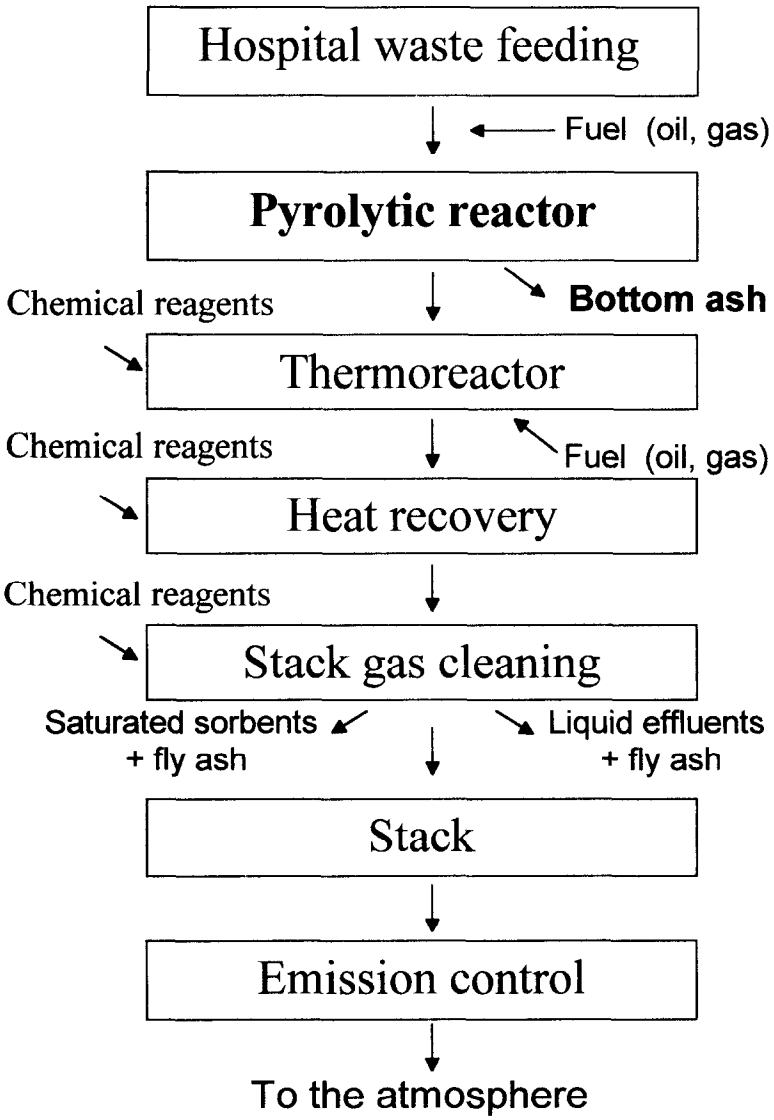
Ashes from pyrolysis chamber (7-10wt%/weight of load) and saturated sorbents are dumped to municipal dumping grounds in the form of grounded materials consolidated by cement mortar. This is the only and effective means of disposing toxic ashes in Poland so far.

In a few cases saturated sorbents like active coal and coke, which are used for adsorption of organic compounds and mercury were burnt in incineration chambers simultaneously with hospital wastes.

Description of hospital waste incineration with the use of pyrolytic chambers.

Typical process of incineration of hospital wastes in pyrolytic chamber, which has been realized in Poland since 1994 is given in Fig. 1.

Figure 1.



This is a two-stage process. In pre-chamber for pyrolysis, cartriges of wastes are degassed at temperatures 550-900^{0}C . Pyrolytic gases are burnt in the excess of oxygen at temperatures 1100-1200^{0}C during 2-3 sec. in thermoreactor. Hot combustion gases are cooled to tempeartures 250-300^{0}C in a heat exchanger and by water injection in Venturi nozzless (quenching). In most solutions, there are applied multi-step cleaning combustion gas technologies, in which the gas is purified by adsorption and chemisorption.

Experimental

Sampling procedure for determination of PCDDs/PCDFs in combustion gas and bottom ash.

Combustion gases were sucked by the stainless steel probe equipped with duct flow made of quartz tube. The probe construction allows gas stream to be heated or cooled inside the quartz tube. The tip of the probe was ended with replaceable stainless steel nozzles of different input diameters enabled to keep isokinetic conditions during sampling within the range 3-50 m/s of linear flow of the gas flux in the flume. After the probe, there were dust filters made of quartz wool set in a heating glass housing, which made it possible to sampling of the suspended dust and fly ash from the stream of combustion gases. Filtered, hot stack gas was cooled by water in glass condenser equipped with condensate receiver. Cooled to 20-25^{0}C, the gas was then passed through a cartridge filled with XAD-2 sorbent. The sampling method was adapted for combustion gases from incineration of hospital wastes, as a modification of procedures described by Williams, de Jong and Hutzinger [5,6,7].

Before the filters and XAD sorbent were taken to sampling, they were loaded with known amount of standard mixture of ^{13}C-PCDDs/PCDFs (recovery standard) in order to determine the PCDDs and PCDFs losses during sampling and later analytical operations.

Filters, condensate and cartridges with XAD-2 sorbent were analyzed in the analytical laboratory.

The sampling took 6 hours under full control of isokineticity of the sampling and measurement of the volume and vacuum of the combustion gas. The concentration of PCDDs and PCDFs in combustion gases was calculated and reported with reference to standardized cubic meter volume - m^3_n for dry gas with respect to 11 % of O_2.

For the determination of PCDDs and PCDFs in bottom ash, a portion of 20 g of sample was taken from about 1 kg of raw material after separation out of metal and glass parts and after homogenization. From periodic-type installations bottom ash samples were collected next day when the pyrolytic chamber was

unloaded. At continuous operating incinerators, samples were taken from pipeline carrying the bottom ash away to filling ground. Bottom ashes come from burning of non-segregated hospital wastes, which composition was as follows: cellulose-paper materials, bandages, protective clothing, packaging and plastics like syringes, latex gloves, expendable materials, etc, body fluids, organic tissue, glass packaging of medicines and other wastes. Concentration of organic chlorine in hospital wastes was in the range of 1-3 wt %, mainly from PVC content.

Sample preparation and analysis

Samples of bottom ash, exposed quartz wool filters, XAD-2 cartridges and condensed water were delivered to laboratory and extracted with toluene in Soxhlet apparatus. Aqueous samples were extracted with toluene in a separatory funnel.

Toluene extracts from quartz wool filters, XAD and condensed water were combined and analyzed together as combustion gas samples.

Afterwards extracts were concentrated and solvent was replaced with heptane. Then extracts were purified following standard analytical procedures [8] based on acid/base silica, carbon/celite, and basic alumina.

The last column eluates were evaporated up to 50 µl and analyzed by GC-MS technique.

Capillary column gas chromatograph HP 5890 series II equipped with quadrupol mass detector MSD 5971 was used for PCDD/F quantitative analysis and confirmation.

Two different columns were used for chromatografphic separation of PCDD/F congeners: SP-2331 for TCDFs separation and confirmation and RTX-5MS as routinely used capillary column.

Results and Discussion

Concentration of PCDDs and PCDFs in stack gases.

Table 2 presents the results of analyses of PCDDs and PCDFs in combustion gases and in bottom ash which were taken from 18 installations of incineration plants for hospital wastes in Poland in 1994-1997. There are also given temperatures of the combustion gas in sampling port and in the pyrolytic chamber.

Table 2:

1	2	3	4	5
Incinerator	PCDDs/PCDFs **in stack gas** [ng TEQ/m^3_n]	**Stack gas** temperature , average [^{0}C]	PCDDs/PCDFs **in bottom ash** [µg TEQ/kg]	**Pyrolytic chamber** temperature [^{0}C]
1	0.015	60	8.5	650 - 750
2	0.02	80	14.5	780 - 850
3	0.022	45	20	670 - 900
4	0.027	55	7.8	750 - 1000 !
5	0.047	75	12.1	500 - 600
6	0.055	40	12.5	650 - 850
7	0.075	90	15	550 - 780
8	0.09	105	22	600 - 700
9	0.13	65	19	575 - 800
10	0.215	140	29	550 - 700
11	0.32	40	9	780 - 900
12	0.42	60	19.5	550 - 700
13	3.9	120	9.5	650 - 800
14	9.7	80	18.4	600 - 650
15	12.1	200	22.5	580 - 650
16	18.5	170	43	750 - 900
17	26	270	35	600 - 700
18	32	250	30	500 - 850

According to recommendations of NATO CCMS [3], in order to calculate the Toxic Equivalency (TEQ) value, 17 PCDDs and PCDFs congeners having chlorine atoms in 2,3,7,8 positions were analyzed. In table 3 there are presented analytical results for all of the 17 PCDDs and PCDFs congeners analyzed in combustion gases taken from three hospital waste incinerators of low, medium and high emission, respectively. Results show that the distribution of congeners is very similar in all incineration plants. Some variations come from the difference in temperature of the analyzed gas. There were also differences in the humidity of the analyzed gas. In gases saturated with water vapour after chemisorption in alkali scrubbers, PCDDs and PCDFs concentration was much lower than in cases of combustion gas purified by dry methods. In spite of high content of tetrachlorodibenzodioxins (TCDDs) and tetrachlorodibenzofurans (TCDFs), the concentration of 2,3,7,8-TCDD and 2,3,7,8-TCDF in cool fumes was not high.

Table 3: PCDDs and PCDFs profile for combustion gases.

1	2	3	4	5
No:	PCDD/F congener	Incinerator 7 [ng/m^3_n] **low**	Incinerator 13 [ng/m^3_n] **medium**	Incinerator 18 [ng/m^3_n] **high**
1	2,3,7,8-TCDF	0.051	0.89	6.9
2	2,3,7,8-TCDD	0.007	0.10	1.25
3	1,2,3,7,8-P_5CDF	0.003	1.77	10.2
4	2,3,4,7,8-P_5CDF	**0.054**	**4.00**	**27.4**
5	1,2,3,7,8-P_5CDD	0.022	0.30	5.9
6	1,2,3,4,7,8-H_6CDF	**0.095**	**5.34**	**49.7**
7	1,2,3,6,7,8-H_6CDF	0.036	1.66	27.1
8	1,2,3,7,8,9-H_6CDF	0.052	2.95	25.3
9	1,2,3,4,7,8-H_6CDD	0.007	0.56	4.2
10	1,2,3,6,7,8-H_6CDD	0.011	0.67	4.5
11	1,2,3,7,8,9-H_6CDD	**0.018**	**1.35**	**8.9**
12	2,3,4,6,7,8-H_6CDF	0.003	0.92	2.9
13	1,2,3,4,6,7,8-H_7CDF	**0.145**	**7.98**	**52.0**
14	1,2,3,4,6,7,8-H_7CDD	0.065	4.45	12.7
15	1,2,3,4,7,8,9-H_7CDF	0.02	1.52	7.8
16	OCDD	0.15	3.85	**21.8**
17	OCDF	0.07	4.20	**31.9**

Mainly PCDFs were detected in combustion gases. Among them, the main compounds were heptachloro-dibenzofuran - 1,2,3,4,6,7,8-H_7CDF, hexachlorodibenzofurans, e.g. 1,2,3,4,7,8-H_6CDF, 1,2,3,6,7,8-H_6CDF and 1,2,3,7,8,9-H_6CDF. In all samples, high concentrations of pentachloro-dibenzofuran 2,3,4,7,8-P_5CDF were detected. Among PCDDs, the highest concentration was detected for heptachlorodibenzodioxins H_7CDDs, mainly 1,2,3,4,6,7,8-H_7CDD. 1,2,3,7,8,9-H_6CDD was also detected in high concentration. For cool stack gases at temperatures below 70^0C the concentration of OCDD and OCDF were negligible whereas in installations, in which the temperature of the fumes was higher than 200^0C, concentrations of these compounds were very high. Even in those cases when the combustion gases were subjected to effective fly ash removal..

Among 18 incineration plants which were investigated, for 5 of them equipped with of low efficient systems of for purification of combustion gases, there were measured concentrations of PCDDs and PCDFs in gases emitted to atmosphere. Those concentrations were within the range 9.7-32 ng TEQ/m$^3_{n_2}$

From 13 new installations, 8 are equipped with very efficient systems of purification of combustion gases. For these installations, concentrations of PCDDs and PCDFs in combustion gases were between the range permissible in EU countries, i.e. 0.015-0.09 ng TEQ/m^3_n. In the remaining 5 installations, systems of purification of combustion gases are not advanced enough nor sufficient. For these installations, measured concentrations of PCDDs and PCDFs in combustion gases were between 0.13 and 3.9 ng TEQ/m^3_n.

Installations with low emission were usually equipped with multi-step systems of purification of combustion gases.

It was also revealed in those cases very low level of content of fly ash which was detected at levels of 0.2-1 mg/m^3_n. For installations of medium level of emission of PCDDs and PCDFs, concentrations of fly ash were within the 1 and 10 mg/m^3_n.

In the five incineration plants built in 60s and recently modernized, which are characterized by high emission of PCDDs and PCDFs, purification of combustion gases is undergoing only by dust removal to the content of fly ash levels below 50 mg/m^3_n. However, it was shown that the fly ash emission to the atmosphere for three of the investigated incinerators were higher than 100 mg/m^3_n (positions 16 to 18 in table 2). As a result, concentrations of PCDDs and PCDFs in combustion gases in these cases reached even 32 ng TEQ/m^3_n. It is clearly seen the dependence between high concentrations of CO (in some cases even 800 mg/m^3_n) as well as organic carbon (C_{org}) and concentrations of PCDDs and PCDFs. High levels of emission of HCl and heavy metals were also observed for these installations.

Concentration of PCDDs and PCDFs in bottom ash.

Samples of the ash, which is a result of incineration of hospital wastes in pyrolytic chambers, were taken from all 18 incineration installations. Chemical analysis revealed that there were not big differences in concentrations of PCDDs and PCDFs in those samples.

Table 2 gives total contents of PCDDs and PCDFs in μg TEQ/kg ash for 18 installations. There are also given temperatures of pyrolytic chambers in which hospital wastes were burnt.

Table 4 contains results of analyses of 17 congeners of 2,3,7,8- chlorosubstituted PCDDs and PCDFs for three samples of ashes with the reference to installations of high, medium and low emission of PCDDs and PCDFs respectively (see table 2).

There were detected PCDDs and PCDFs at high concentrations, within the range 8-45 μg TEQ/kg in these installations. Main substances were PCDFs, among which 1,2,3,4,6,7,8-H_7CDF covered 25 wt. % of 17 analyzed congeners of PCDDs and PCDFs. Mass content of OCDF was about 15 wt %. Mass

contents of 2,3,4,7,8-P_5CDF and 1,2,3,7,8,9-H_6CDF were about 10%. In all samples of ashes, the distribution of congeners was very similar.

Table 4: PCDDs and PCDFs profile for bottom ash from pyrolytic chambers

1	2	3	4	5
No:	PCDD/F congener	Incinerator 7 **low in stack gas** [μg/kg]	Incinerator 13 **medium in stack gas** [μg/kg]	Incinerator 18 **high in stack gas** [μg/kg]
1	2,3,7,8-TCDF	0.37	0.244	1.12
2	2,3,7,8-TCDD	0.29	0.235	0.84
3	1,2,3,7,8-P_5CDF	5.25	3.43	13.45
4	2,3,4,7,8-P_5CDF	**12.45**	**6.22**	**24.75**
5	1,2,3,7,8-P_5CDD	7.25	5.82	11.9
6	1,2,3,4,7,8-H_6CDF	9.45	6.35	22.5
7	1,2,3,6,7,8-H_6CDF	8.9	6.33	12.75
8	1,2,3,7,8,9-H_6CDF	**13.7**	**8.74**	**26.25**
9	1,2,3,4,7,8-H_6CDD	2.1	1.02	6.2
10	1,2,3,6,7,8-H_6CDD	3.1	1.36	7.85
11	1,2,3,7,8,9-H_6CDD	2.75	1.33	5.45
12	2,3,4,6,7,8-H_6CDF	3.5	2.05	10.35
13	1,2,3,4,6,7,8-H_7CDF	**52.6**	**34.75**	**106.8**
14	1,2,3,4,6,7,8-H_7CDD	11.4	6.47	37.9
15	1,2,3,4,7,8,9-H_7CDF	5.9	3.64	19.8
16	OCDD	12.7	7.27	45.7
17	OCDF	**21.3**	**13.84**	**65.4**

Those measurements and analysis resulted in information about the level of concentration of PCDDs and PCDFs in combustion gases and ashes from incineration of hospital wastes.

Most results were obtained in form of singular measurements which were undertaken on new installations or installations equipped with new exploitation materials. It makes possible for us to assess the level of emission of PCDDs and PCDFs from Polish incineration plants for hospital wastes.

For incinerators operated in West Europe countries the stack gases should not contain PCDDs and PCDFs at a level higher than 0.1 ng-TEQ/m^3_n with respect to the commonly accepted requirements [2]. The results presented in this paper were obtained for new or modernized installations. In some cases investments did not bring expected low emission level i.e. the PCDDs/Fs concentration in stack gas

exceeded 0.1 ng-TEQ/m^3_n. The concentration of PCDDs and PCDFs was determined by gas chromatography and mass spectrometry analysis.

Critical remarks do not refer to all installations. Almost half of the investigated installations emitted combustion gases with concentrations of PCDDs and PCDFs below 0.1 ng TEQ/m^3_n. As was already mentioned, there have not been established permissible amounts of substances emitted from incineration plants.

Despite the fact that the maximal concentrations of compounds emitted from incineration plants differ in EU countries, we suggest for Polish hospital waste incinerators the maximum acceptable concentrations of: CO - 50 mg/m^3_n, NO_x - 200 mg/m^3_n, C_{org} - 20 mg/m^3_n, metals - 0.5 mg/m^3_n (zinc not included), total dust - 10 mg/m^3_n, PCDDs/PCDFs - 0.1 ng TEQ/m^3_n

As far as CO, NO_x, organic carbon and metals are concerned, there is no problem with reaching these levels in combustion gases from installations which are commonly used in Poland provided that the basic process parameters like installation load, temperature, oxygen content in gasification or burning chambers, after-burning and using of sorption media are properly set. Reaching values of 0.1 ng TEQ/m^3_n for PCDDs and PCDFs in combustion gases still remains problem. It is thought in Poland that this requirement is too severe and it is considered as technically and economically acceptable the level of 0.5 ng TEQ/m^3_n. Installations which are built in Poland are characterized by low unit capacity and the emission of combustion gases is no more than 3000 m^3_n /hour. Nevertheless, the level of 0.1 ng TEQ/m^3_n should be taken into consideration to assess the danger.

The amount of contaminants that are emitted to the environment from processes of thermal utilization of hospital wastes mainly depends on proper after-burning of combustion gases and removing of harmful substances, unable to destroy by thermal methods like heavy metals, hydrogen chloride, hydrogen fluoride, NO_x, sulfur, ashes and already mentioned chloro-organic compounds. Substances which can be thermally destroyed, i.e. oxidized to CO_2 and H_2O like aromatics, phenols, esters, carboxylic acids, etc. should be detected in combustion gases, sewage and ashes. Unfortunately, organic substances in concentrations of 1g / 1kg sample were detected in samples of ashes from pyrolytic chambers of more than half of investigated installations, together with PCDDs and PCDFs.

It proves that the process of after-burning of wastes in installations, which were commissioned in 1994-97, was incomplete. Incomplete after-burning of wastes leads to formation of organic compounds which contain many condensed rings in one molecule, mainly heterocyclic and their chloro-substituted derivatives which have high toxicity.

Simultaneously, carbon undergoes only partial oxidation to CO. The presence of CO in combustion gases in concentrations higher than 20 ppm (oxygen content 11%) testifies that the after-burning process is

ineffective. As a result, it was observed simultaneous increase in concentrations of aromatics and PCDDs/PCDFs. Concentrations of CO and oxygen in combustion gases is the basic indicator whether the process of burning and after-burning of gases is carried out properly or not. On this base, installations are adjusted. This adjustment refers to the system of loading of wastes to combustion chambers as well as the amount of the excessive air and temperature.

Conclusion

The measurements of concentrations of PCDDs and PCDFs in combustion gases and ashes from thermal utilization of hospital wastes by pyrolytic method showed variable levels of the emission of these compounds to atmosphere. There are incineration plants in Poland for burning of hospital wastes which emit exhaust fumes containing PCDDs and PCDFs at levels below 0.1 ng TEQ/m^3_n but there are also plants that emit these compounds at levels higher than 30 ng TEQ/m^3_n. GC-MS analysis showed that the distribution of congeners of PCDDs and PCDFs in combustion gases was very similar for all 18 installations.

Slight differences in the content of congeners might be caused by differences in temperatures of combustion gases directed to stacks and different methods of purification. Large differences in levels of the emission show that in older installations systems of purification of combustion gases are ineffective. It might also show that they are not technologically properly designed.

Concentrations of PCDDs and PCDFs in samples of the ash taken from pyrolytic chambers of 18 incineration installations were very high and were between 8-45 μg TEQ/kg.

Despite differences in the construction of pyrolytic chambers and gaseous or oil fuel, the distribution of congeners of PCDDs and PCDFs for ash samples was very similar.

There is still unsolved problem of the utilization of toxic ash, which is produced as a result of thermal utilization of hospital wastes in Poland. It is solidified into concrete compositions and deposited at dumping grounds for hazardous wastes or even municipal dumping grounds.

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