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LEVELS OF POLYCHLORINATED DIBENZO-P-DIOXINS AND DIBENZOFURANS IN DUST AND AIR FROM LUBLIN

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Abstract: Polychlorinated dibenzodioxins and dibenzofurans are present in the air either adsorbed on particulate matter or as vapor (minor part). They are most known carcinogens and mutagens. In this study dust samples from Lublin were being collected for 12 months and analyzed for the PCDD/Fs content. For the analysis of these samples classical extraction with Soxhlet apparatus, double step clean-up based on SEC and charge-transfer interactions chromatography, and finally analysis with GC-MS were applied. Results of the determinations were compared with results obtained by other authors.

Keywords: Polychlorinated dibenzodioxins, polychlorinated dibenzofurans, fine dust

INTRODUCTION

Industrialization, urbanization, economic growth and associated with them increase in energy demands have resulted in a profound determination of urban air quality. Modernization and enhanced industrial activities led to the increased use of fossil fuels and their derivatives, particularly in developing countries such as Poland. This increase in the in use of fossil fuel is sharply observed in the Lublin to which Lublin Hard Coal Mining belong.

Combustion processes are primary sources of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) in the environment (Rappe, 1994, 1996). Once released into the atmosphere, they are subject to atmospheric dispersion and deposition (Tysklind et al., 1993). Soils and watersheds are the sinks for these compounds (Hagenmaier and Krauß, 1993). Therefore, atmospheric deposition to an ecosystem is an important process in determining the environmental fate of PCDD/Fs (Dudzinka & Kozak 2001).

In recent decades, the environmental presence of polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs) has meant a serious public health problem. It is now well known that PCDDs and PCDFs, especially the 2,3,7,8-substituted congeners, belong to the most hazardous environmental contaminants. These organic pollutants are toxic in extremely tiny amounts and biaccumulate in humans (Boening, 1998; Sweetman et al., 2000; Kogevinas, 2001; Birnbaum and Cummings, 2002). PCDD/Fs are released into the environment in ultra trace amounts from various chlorinated chemical formulations. Due to their highly toxic properties, PCDD/Fs have received prolonged attention by the scientific community and environmental regulators (Fiedler, 1996; Alcock et al., 2001). Among the thermal sources, until recent years, MSWIs and hazardous waste incinerations (HWIs) have had a preponderant role as PCDD/Fs emitters. Although it has meant a potential source of exposure for subjects living near these facilities, it must be taken into account that for the general population, food intake is usually the primary source of exposure to PCDD/Fs, while inhalation and dermal contact are only minor routes (Karademir, 2004; Menseses et al., 2004).

The main objectives of this study are to investigate the characteristics of PCDD/Fs concentration and dry depositions in the Lublin area which is quite a clear part of Poland with no industry but the traffic and household heating system are becoming more and more important as a source of pollutants bounded with PM-10 dust.

Table 1 presents PCDD/F data in ambient air. In general, concentrations of the sum of the tetra- to octa-CDD.Fs homologues in ambient air are between 0.5 and 20 pg/m^3. As expected, there is a general gradient, increasing from remote to rural to urban/industrial centres. PCDD/Fs concentrations for the sum of the tetra- through octa-PCDD/Fs homolougues and the ΣTEQ are typically as follows: remote-0.5 pg/m^3 ΣTEQ-10 fg/m^3; rural 0.5÷4 pg/m^3 ΣTEQ; 20÷50 fg/m^3 and urban/industrial ; 10÷100 pg/m^3 ΣTEQ ; 100÷400 fg/m^3. This trend is consistent with expectations, given that combustion sources and chemical usage are believed to be the principal sources of PCDD/Fs release to the atmosphere.

Table 1. Ambient air levels of PCDD/Fs, together with information on other variables

Country	Location	Comment	$\sum P_{4-8}$CDD/F (pg/m^3)	OCDD (% of $\sum$)	PCDD/PCDF ratio	$\sum$TEQ[a] (fg/m^3)	Year	Reference
Austria	Brixlregg	Industrial	98 (73-130)	2	0.51 (0.39-0.65)	1200 (800-1600)	1988	Christmann et al., 1989
	South Graz (Urban)	Day + night	20	24	2.3	370	1994	Thanner and Moche, 1995
		Days	19	24	2.2	390		
Belgium	Flanders	Urban				110 (20-380)	1992	Wevers et al., 1993
Germany	Köln, Duisburg	Urban	12 (6.5 -17)	13	0.75	240	1987	Hiester et al. 1997
	Essen, Dortmund		6.7 (3.2 – 9.9)	21	1.2	90	1993	Buck and Krischmer 1986
	8 towns in NRW	Urban	8.6 (5.1 - 15)	11	0.61 (0.35-1.0)	140[e] (50-160)	1985	Kaupp, 1996
	Egge	Rural	2.3	27	0.78			König et al., 1993
	Bayeruth	Rural	0.81 (0.27 - 1.4)	23	1.1 (0.80 – 1.3)		1994	Lohmannet al., unpublished
	Hessen	Urban	6.6 (5.2-8.8)	32	1.6 (0.94 – 2.2)	100 (80-150)	1990	Benfenati et al., 1994
		Rural	3.3	32	1.5	50		
Ireland	Mace Head	Remote	0.45 (0.37-0.53)	18	1.1 (0.9-1.2)	4 (3-4)	1997	Lohmann et al., unpublished
Italy	Milan (urban)	Day	3.4 (2.8 - 4.1)		0.35 (0.15-0.93)		1991	Benfenati et al. 1994
		Night	7.7 (5.9-9.2)		0.38 (0.13-0.92)			
	Rome	Urban	1.5-2.95[b]	9[b]		85 (50-280)	1990-1991	Turrio-Baldassarii et al., 1994
Poland	Cracow	Traffic	60	17		950		Grochowalski et al., 1995
		Centre	1100	26		12000	1995	
Spain	Catalunya	Urban	18 (6.7-160)	7	0.38 (0.10-0.82)	250 (70-530)	1995	Abad et al., 1997
		Rural	4.2	31	1.5	50		
	Rörvik	Rural	1.9 (0.3-	32	1.8 (0.91-2.7)	21 (4-60)	1989-	Tysklind et al.,

Sweden			5.2)				1990	1993
	Gothenburg	Urban	1.3 (1.0-1.8)	21	1.2 (0.96-1.8)	22 (16-30)	1988	
	Coast	Remote	0.32	18	1.1	4	1987	Broman et al., 1991
	Stockholm		1.3	19	1.2	19		
UK	Manchester	Urban	17 (ND-62)	22[a]	1.4 (1.0-1.8)	410 (ND-1800)	1991-1993	Duarte – Davidson et al., 1994
	Cardiff	Urban	8.7	31	1.7	190	1992-1993	Jones and Davidson, 1997
	Bolsover [f]	Industrial	26	11	2.2	330		
	Hazelrigg	Rural	1.05(0.72-1.60)	23	1.3 (0.7-3.6)	11 (8-18)	1997	Lohmann et al., unpublished
	East coast	Remote	0.43 (0.24-0.69)	20	1.05 (0.8-1.4)	4 (2-6)	1997	Lohmann et al., unpublished
Japan	Tokio	Urban	0,133-0,158	39	nd	nd	2001	Sugita et al 2004
	Matsuyama[c]	Urban	14.5 (4.2-26.7)		0.61	160 (80-280)	1996-1997	Seike et al., 1997
Japan	Kanazawa	rural	0.0091-0.039				2003	Oka et al 2005
USA	Mississipi	Rural	0.45 (0.29-1.0)	47	4.7 (2.9 – 8.3)		1991	White and Hardy, 1994
	Ohio	Urban	6.2 (1.2-30)		2.9- 4.3[c]	81 (16-210)	1995	Riggs et al., 1996
	Phoenix, AZ	Urban	26.6 (9.9-52)	33	7.0 (0.45-21)	250 (90-450)	1996	Hunt et al., 1997
	Wisconsin	Rural	2.9 (1.5-4.4)	13	0.83 (0.63 -1.2)	58 (30 – 100)	1989	Harless et al., 1990
	Connecticut	Urban	6.9 (0.24-18.9)	31	1.7 (0.83-10)	110 (8 - 1900)	1987	Hunt and Maisel., 1990
	North Carolina	Rural	1.8 (0.72-4.6)	14	0.71 (0.60-0.89)		1991	Harless et al., 1992
	New York State	Urban	3.9	31	1.4		1986-1990	Smith et al., 1992
	Niagara	Rural	2.5 (0.48-53)	67	4.2	14 (ND-1300)	1986-1988	Smith et al., 1990
Australia	Sidney	Urban	3.7-15			16-62	1990	Taucher et al., 1992
	Brisbane	Rural	1.1 (0.60-1.2)	22	1.5 (0,86-2,9)	11 (4-17)	1996	Müller, 1997
Antarctica	Mc Murdo	Remote	0.005	7	0.65		1992	Lugar et al., 1996

Note. In general, mean values are given, with the range of concentrations in parentheses - values below detection limits taken as half the detection limit.

ND- not detected.

[a] NATO-TEF values

[b] calculated as sum of 2,3,7,8-congeners only, average value form min – max data

[c] calculated form min – max data

[d] coelution with 1,2,3,6,9- PCDF

[e] 2,3,4,7,8-PCDF not always measured

[f] values below detection limits taken as detection limit

MATERIAL AND METHODS

1. Materials and equipment:
   - solvents: toluene, hexane and dichloromethane – "for residue analysis" (JT Baker – Germany),
   - anhydrous sodium sulphate (VI) *puriss-* (POCh – Gliwice, Poland),
   - standard solutions of 17 congeners of PCDD/Fs according to EN-1949, native and labeled (CIL – Germany),

- Soxhlet apparatus, 200ml capacity with reflexive/check cooling system and heating neck for 250ml flasks,
- rotary evaporator – (Büchi – Germany),
- vacuum pump with dephlegmator and vacuum control (Büchi – Germany),
- needle evaporator (Supelco – USA),
- reaction vessels – 3ml and 40ml capacity (Supelco – USA).

2. Methods:

Schematic diagram of analysis pathway is presented in figure 1.

- Dust samples were collected during the period of 12 months (from January 2005 till December 2005) on quartz fiber filters in the Botanical Gardens of Lublin in the vicinity of the most important route from Warsaw to the border areas of Poland and the residential district Slawinek.
- Filters were extracted in Soxhlet apparatus with 200ml of dichloromethane and toluene mixture (80/20 v/v) for 16 hours, providing at least four flows of solvent per hour.

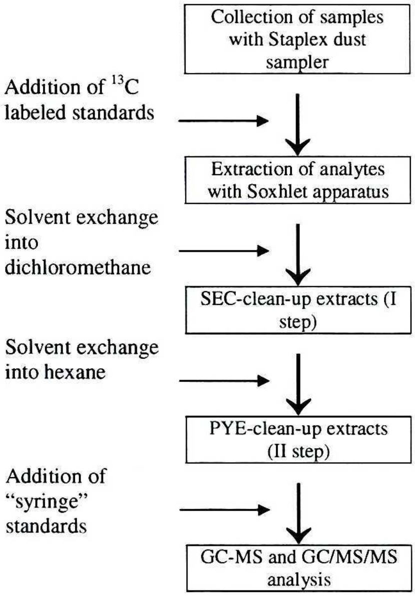

Fig. 1. Schematic diagram of analysis pathway of dust samples

Obtained extract was evaporated in vacuum evaporator at the temperature below 36 °C to the volume of about 3ml and then it was quantitatively transferred to 3ml reaction vessels, where it was evaporated to the volume of 2.5ml (including additional extracts from flask and Soxhlet apparatus flushing).

Purification of the extract:

- Purification was done with high-resolution elimination chromatography (HR-SEC) using liquid chromatograph Breeze 1525 (Waters – USA) built of the following elements:
    - Binary gradient pump – Waters 1525,
    - Injector – Rheodyne with 200µl sample loop,

- Columns: Envirogel GPC cleanup 19x150mm and Envirogel GPC cleanup 19x300mm,
- Two-wave, tunable detector UV-VIS Waters M2487,
- Fraction Collector – Waters FC III,
- Data acquisition and system control: – Breeze 3.30SPA software,
- Chromatographic syringes with nonstick pistons - 2500µl (Hamilton).

The system was standardized for standard solution containing:

- corn oil – lipid and waxes fraction marker,
- Bis(ethylcyclohexyl) phthalate – Phthalate fraction marker,
- Methoxychlor – PCDD/Fs fraction marker,
- perylene – PAH fraction marker,
- elementary sulphur – low-particle compounds fraction marker,

all compounds in dichloromethane.

Mobile phase (dichloromethane) flow rate – 5ml/min and wave length during chromatograms collection - λ=254 nm.

For eventual influence of column overloading protection but also for possible wide PAH fraction, collection was made for the following elution times – 14 – 21 minutes. Results of PAHs determination were described previously in Duda & Czerwiński.

After extracts analysis for PAH's, left part of it was evaporated to dry in gentle nitrogen stream and exchanged the solvent into hexane being a mobile phase in charge-transfer column fractionation. The volume of hexane extract was 550µl, where 500µl was injected to the previously described HPLC system, equipped with COSMOSIL PYE column (Phenomenex – Japan), working in the system of normal phases (hexane as the mobile phase @ 1 ml/min – 24 min and the last 5 minutes hexane/ CH_2Cl_2 80/20 v/v) to elution acceleration of PCDD/Fs mentioned. Initial 6ml of eluate was thrown out because of presence of non-planar compounds (for e.g. PCB and PBDE). Detailed time windows for PCDD/Fs determination are given in Duda & Czerwiński [2007].

25ml of eluate was collected and condensed to the 50µl volume (in gentle nitrogen stream). Purified extract was analyzed using GC-MS/MS system described and utilized before. Temperature program parameters and GC-MS/MS parameters were described before [Duda & Czerwiński, 2007].

RESULTS AND DISCUSSION

Results of PCDD/Fs determination are given in table 2. In current measurements absorption filters were protected against sunlight to prevent dioxin degradation. The concentration of PCDD/Fs in 2005 was higher than 2004 witch was caused by colder season which suggests influence of household a heating systems on the concentration of PCDD/Fs in the air and dust because of a vicinity of residential district Sławinek. We think that high concentration of PCDD/Fs in samples collected in Autumn is also connected with open burning of garden wastes in this area.

Accuracy of the determination was estimated to be about 30% (basing on error variance propagation law) and is not satisfactory, which may be explained by low PCDD/Fs levels and low weight of available sample. Optimal – 2g, compared to available maximum – 0.2g.

Temperature program used in the examination (over 60 minutes) is quite long and gives a possibility to trace the presence of other congeners for 17 typically determined according to EN-1949, which may indicate their source of origin.

Non-industrial emission sources will relatively become more important the near future and soon may dominate the overall annual emissions of PCDD/F in Europe. Besides educational efforts in order to inform the public about consequences when using inappropriate fuels also technical improvements, particularly with respect to solid fuel domestic heating appliances, may help to achieve further reductions.

Table 2. Levels of PCDD/Fs in samples collected in 2005

Congener	TEF	Spring		Summer		Autumn		Winter	
		ng/g_{dust}	pg/Nm^3	ng/g_{dust}	pg/Nm^3	ng/g_{dust}	pg/Nm^3	ng/g_{dust}	pg/Nm^3
2,3,7,8- TCDD	1	4.5	0.040	0.57	0.089	4.2	2.67	7.8	3.15
1,2,3,7,8-P5CDD	1	29.4	0.265	0.69	0.108	2.4	1.53	4.3	1.73
1,2,3,4,7,8- H6CDD	0.1	13.9	0.125	4.93	0.777	9.11	5.81	14.2	5.73
1,2,3,6,7,8-H6CDD	0.1	16.4	0.147	1.16	0.182	6.9	4.40	11.7	4.72
1,2,3,7,8,9-H6CDD	0.1	11.4	0.103	23.4	3.690	17.1	10.90	19.2	7.76
1,2,3,4,6,7,8-H7CDD	0.001	7.6	0.068	16.9	2.665	7.1	4.52	2.8	1.13
OCDD	0.0001 0.0003*	0	0	3.2	0.504	0	0	6.4	2.58
2,3,7,8-TCDF	0.1	71.4	0.644	12.7	2.00	46.2	29.47	78.4	31.61
2,3,7,8-P5CDF	0.5 0.3*	40.3	0.363	33.1	5.22	54.1	34.51	64.1	25.90
1,2,3,7,8-P5CDF	0.05 0.03*	32.6	0.294	12.8	2.018	46.5	29.66	56.2	22.71
1,2,3,4,7,8-H6CDF	0.1	5.74	0.051	3.4	0.533	11.8	7.52	19.4	7.84
1,2,3,6,7,8-H6CDF	0.1	9.11	0.082	5.1	0.804	8.4	5.35	27.1	10.95
1,2,3,7,8,9-H6CDF	0.1	5.45	0.049	14.9	2.349	9.2	5.86	31.1	12.57
2,3,4,6,7,8-H6CDF	0.1	27.6	0.249	8.9	1.403	11.4	7.27	8.7	3.51
1,2,3,4,6,7,8-H7CDF	0.01	36.9	0.332	6.3	0.993	26.8	17.09	39.3	15.88
1,2,3,4,7,8,9-H7CDF	0.01	40.1	0.361	14.8	2.33	31	19.77	47.1	19.03
OCDF	0.0001 0.0003*	11.45	0.103	22.1	3.485	44.9	28.64	59.6	24.09

*TEF 2005

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