

TEMPORAL TREND IN THE LEVELS OF PCDD/Fs IN PLASMA OF A NON-OCCUPATIONALLY EXPOSED POPULATION LIVING NEARBY A HAZARDOUS WASTE INCINERATOR

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Introduction

The notable public concern surrounding waste incineration is mainly due to the fact that incinerators have been considered as important emission sources of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)¹ among other toxic pollutants. However, there are currently other important industrial (cement kilns and power plants) and diffuse (vehicle emissions, domestic coal/wood combustion, natural fires, etc.) emission sources of PCDD/Fs. In humans, the most important exposure route to these pollutants comes from dietary intake².

In 1999, the first and up to now only hazardous waste incinerator (HWI) in Spain began to operate in Constantí (Tarragona County, Catalonia). Before the start-up (1998), a wide biological monitoring program (baseline) was initiated by analyzing PCDD/Fs in samples of plasma and other biological tissues from people living in the neighborhood of the HWI³. After 3 years of regular operations (2002), a new survey was carried out to determine the impact of the facility on that population⁴. A significant decrease of plasma levels was noted, which was due to the important reduction of PCDD/F intake through foodstuffs occurred in recent years⁵.

The present study was aimed at establishing the temporal trend in the levels of PCDD/F in plasma of subjects living in the surroundings of the HWI. The concentration of these pollutants in blood were determined and compared with those obtained in the baseline and 2002 survey.

Materials and methods

Samples of around 80 ml of plasma corresponding to 20 subjects aged 17-61 years (10 males and 10 females) were obtained in a blood bank. All individuals had been living near the HWI for at least the last 10 years. None of them had been occupationally exposed to PCDD/Fs. After collection, plasma samples were immediately stored at -20°C until analysis. PCDD/F levels were determined by following a derivation of the US EPA method 1625. Samples were firstly homogenized and subsequently extracted (liquid/liquid extraction). A mixture of ¹³C₁₂-PCDD/Fs standards was spiked to control any potential losses during the process. A multi-step clean-up using silica and alumina columns was carried out. The content of PCDD/Fs in the final extract was analyzed by high resolution gas chromatography coupled to high resolution mass spectrometry (HRGC/HRMS).

Statistical treatment of the data was done by using the SPSS 14.0 statistical software package. The Levene test, followed by ANOVA or Kruskal-Wallis test, was applied to calculate the statistical significance. A probability of 0.05 or less ($p < 0.05$) was considered as significant. When the concentration of a PCDD/F congener was below the limit of detection (LOD), that level was assumed to be one-half of the respective detection limit. Total concentrations in toxic equivalents were calculated using both NATO/CCMS- and WHO-TEFs.

Results and discussion

In this survey, the mean concentration of PCDD/Fs in plasma was 9.36 pg I-TEQ/g lipid (or 9.51 pg WHO-TEQ/g lipid), with minimum and maximum levels of 1.76 pg I-TEQ/g lipid (1.53 pg WHO-TEQ/g lipid) and 23.44 pg I-TEQ/g lipid (23.32 pg WHO-TEQ/g lipid), respectively. In the baseline survey (1998), the mean concentration of PCDD/Fs in plasma was 27.0 pg I-TEQ/g lipid, ranging from 14.79 to 48.95 pg I-TEQ/g lipid. In the 2002 study, the mean level of PCDD/Fs was 15.70 pg I-TEQ/g lipid (or 17.80 pg WHO-TEQ/g lipid), ranging 4.66-29.25 pg I-TEQ/g lipid (5.43-32.00 pg WHO-TEQ/g lipid). In comparison to both previous surveys

(baseline and 2002), a significant decrease of the current (2007) PCDD/F levels was observed (1998-2007, $p < 0.001$; 2002-2007, $p < 0.01$). The individual PCDD/F levels in plasma of the 20 participants in each study, together with the trend are depicted in Figure 1.

Diet has been clearly considered as the main route of entrance of PCDD/Fs to the human body. In fact, more than 90-95% of the current human exposure to these pollutants is estimated to be due to dietary intake⁶. Because of that, the concentrations of PCDD/Fs in foodstuffs highly consumed by the population and the dietary intake of PCDD/Fs were also complementarily studied to the biological monitoring of the non-occupationally exposed population^{5, 7, 8}. In the baseline survey, the dietary intake of PCDD/Fs for an adult was 210 pg I-TEQ/day, while in the 2002 study, that intake of PCDD/Fs was 59.6 pg I-TEQ/day (63.8 pg WHO-TEQ/day). The total market study performed in 2007 allowed calculating a total dietary intake of PCDD/Fs of 27.8 pg WHO-TEQ/g day. In consequence, the significant reduction of the PCDD/F levels in plasma of the local population is in total agreement with the notable decrease in the dietary intake of PCDD/Fs. In fact, the decline of PCDD/Fs in both the body burden and foodstuffs has also been observed in several studies around the world⁹⁻¹¹.

In addition to the biological monitoring of non-occupationally exposed people, the levels of PCDD/Fs in blood of the HWI workers have been annually determined since 1999. A continued and significant decrease has been observed since the incineration plant started to operate. In 2006, the mean concentration of PCDD/Fs in plasma of the HWI workers was 10.4 pg I-TEQ/g lipid, being significantly lower than the levels found in the first survey (26.7 pg I-TEQ/g lipid)¹².

The PCDD/F congener profiles obtained in the 3 studies (1998, 2002 and 2007) are shown in Figure 2. When assessing the temporal trend, a general decrease of the individual concentration of all PCDD/F congeners was observed. Some high-chlorinated PCDDs (OCDD, 1,2,3,6,7,8-HxCDD and 1,2,3,4,6,7,8-HpCDD) were the predominant congeners, while the lowest contribution to the total concentration corresponded to 1,2,3,7,8,9-HxCDF and 1,2,3,4,7,8,9-HpCDF. The levels of 2,3,7,8-TCDD, the most toxic congener, were specially low (from <LOD to 1.20 pg/g lipid), being only detected in 9 of the 20 current samples.

The PCDD/F levels in plasma according to gender and age are summarized in Table 1. A significant reduction of the PCDD/F concentration was observed (for males and females) between the current and the previous studies (1998-2007, $p < 0.001$; 2002-2007, $p < 0.05$). PCDD/F levels in plasma of middle-aged (31-45 and 46-55 years) individuals also decreased significantly between the baseline and the current study ($p < 0.001$), as well as between the 2002 and the present survey ($p < 0.05$). A direct correlation between PCDD/F concentration in plasma and age has been previously reported, with higher values in older people¹³. However, in the present study no significant correlation between both variables was observed. It might be probably due to the unexpected relatively higher PCDD/F levels in 3 young subjects.

Because of the concern that waste incinerators mean for local populations and public authorities, in recent years a number of investigations have focused on determining the exact influence of some specific facilities, mainly derived from exposure to PCDD/Fs. The concentrations of PCDD/Fs in non-occupationally exposed people living near the HWI of Constantí seem to be in the lower part of the range, in comparison to those found in other locations of similar characteristics¹³⁻¹⁶.

In conclusion, the current results show that the influence of the HWI of Constantí (Tarragona, Catalonia, Spain) on exposure to PCDD/Fs is not significant for the subjects living in the vicinity of the facility. The significant reduction of PCDD/F concentrations in plasma is in agreement with the general decrease observed in the same biological monitor reported in recent investigations of other sites impacted by waste incinerators. Moreover, the agreement between the reduction of PCDD/F levels in plasma and the dietary exposure to PCDD/Fs corroborates that food consumption is the main human exposure pathway to these pollutants.

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Table 1: Total concentrations of PCDD/PCDFs (pg I-TEQ/g lipid) in plasma from subjects living in the vicinity of the HWI of Constantí according to gender and age

	Samples			Mean			Standard deviation			% of variation	
	1998	2002	2007	1998	2002	2007	1998	2002	2007	2002-07	1998-07
Total	20	20	20	27.0	15.7	9.4	8.2	5.9	5.4	-40**	-65***
Gender											
Male	13	10	10	26.7	17.0	9.9	2.5	6.1	6.2	-42*	-63***
Female	7	10	10	27.8	14.3	8.9	2.7	5.7	4.6	-38*	-68***
Age											
18-30	4	4	4	17.2	10.4	14.1	3.8	5.0	7.1	36	-18
31-45	4	8	6	25.2	15.7	7.4	5.0	3.7	4.4	-53**	-71***
46-55	8	4	7	32.0	17.6	9.5	8.4	1.9	3.4	-46*	-70***
>56	4	4	3	28.8	19.0	6.5	5.1	10.3	6.8	-66	-77**

Significant differences at: *p<0.05; **p<0.01; ***p<0.001.

Figure 1: Individual levels and temporal trend of PCDD/Fs in plasma of a non-occupationally exposed population living near the HWI of Constantí (Catalonia, Spain)

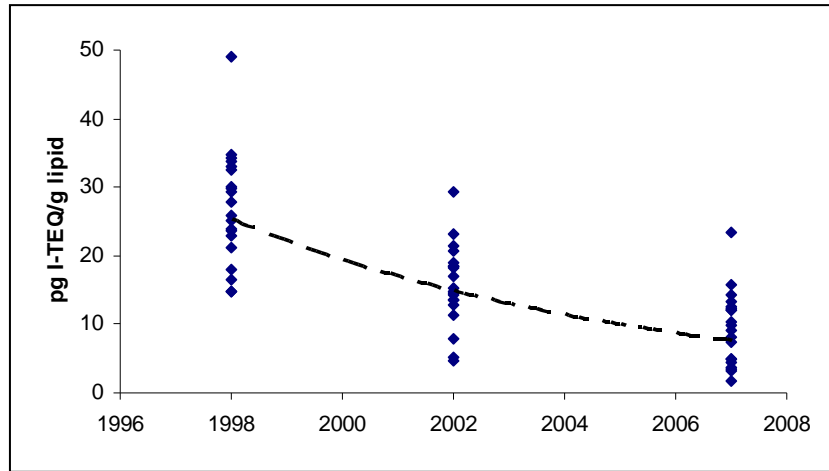


Figure 2: PCDD/F congener profiles in plasma of people living in the vicinity of the HWI of Constantí (Tarragona, Catalonia, Spain) in the baseline (1998), 2002 and current (2007) surveys

