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Body burden monitoring of dioxins and other organic substances in workers at a hazardous waste incinerator

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ABSTRACT

The construction in Constantí (Tarragona County, Catalonia) of the first, and up till now the only hazardous waste incinerator (HWI) in Spain, finished in 1999. In this study, we measured the concentrations of a number of organic substances determined in blood and urine of the HWI workers. Samples of 18 men and 9 women, classified according to their respective workplaces, were collected in 2011, after approximately 12 years of regular operations in the facility. The current results were compared with those of the baseline survey, as well as with the most recent surveys performed in 2008, 2009 and 2010. Plasma analyses were carried out for hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs 28, 52, 101, 138, 153, and 180) and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), while the levels of 2,4- and 2,5-dichlorophenol (DCP), 2,4,5- and 2,4,6-trichlorophenol (TCP), pentachlorophenol (PCP) and 1-hydroxypyrene (1-HP) were measured in urine samples. In plasma, the mean concentrations were the following: 10.8 μg/kg lipid for HCB; 0.8 μg/kg lipid for PCB28; 0.3 μg/kg lipid for PCB52; 0.5 μg/kg lipid for PCB101; 42.2 μg/kg lipid for PCB138; 18.5 μg/kg lipid for PCB153, and 51.2 μg/kg lipid for PCB180. For PCDD/Fs, the mean concentration was 4.6 ng I-TEQ/kg lipid (4.7 ng WHO-TEQ/kg lipid). These levels, as well as those found in urine samples, are in agreement with the data of previous surveys performed in the same area. The current results in HWI workers do not show any evident sign of occupational exposure to PCDD/Fs and other organic substances. However, these results must be considered only as an indication of potential exposure, as the study presents notable limitations, such as the reduced number of participants and the lack of data relative to the air concentrations of chemicals. Consequently, general conclusions cannot be derived and the results should not be used as a basis for the implementation of industrial hygiene measures in other HWIs.

Introduction

The European Union (EU) generates annually about 3 billion tons of waste, being approximately 90 million tons of hazardous waste (HW). It is obvious that treating and disposing all this material without harming the environment has become a major concern. The EU's approach to waste management is currently based on these three principles: (1) waste prevention, (2) recycling and reuse, and (3) improving final disposal and monitoring. The last means that, whenever possible, waste that cannot be recycled or reused should be safely incinerated, with landfill only used as a last option. In recent years, incineration has been accepted as a strategic option for waste reduction and disposal (Assamoi and Lawryshyn, 2012; Monni, 2012; Narayana, 2009; Papageorgiou et al., 2009). With respect to HW, incineration is often selected as a disposal method, when the HW cannot be recycled (Ferré-Huguet et al., 2006; Salihoglu, 2010). Incineration of hazardous (and also harmless) waste may cause emissions of substances, which pollute the air, water and soil, having potential harmful effects on human health. Although most evaluations on stack emissions have been focused on three classes of pollutants (metals, semivolatile, and volatile compounds), polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs), which are characterized by their toxicity to humans and animals, their ability to bioaccumulate, and their persistence in the environment have raised the greatest concern (Kulkarni et al., 2008; Mari and Domingo, 2010). Once emitted to the atmosphere, PCDD/Fs are dispersed through the environment and accumulate in soils and vegetation, thereby easily reaching the food chain (Domingo et al., 1999a,b; Esposito et al., 2009; Lin et al., 2012). In order to limit these risks, the European Union (EU) has imposed strict operating conditions and technical requirements on waste incineration plants and waste co-incineration plants, according to the EU Directive Directive 2000/76/EC.

The construction in Constantí (Tarragona County, Catalonia) of the first, and up till now the only hazardous waste incinerator (HWI) in Spain, finished in 1999. A pre-operational monitoring program

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Table 1
Concentrations of organohalogenated compounds measured in plasma of workers at the HWI in 2011.

	Mean	Median	SD	Min	Max
HCB	10.8	10.2	8.1	2.1	25.0
2,4,4′-Tri-PCB28	0.8	0.8	0.2	0.5	1.1
2,2′,5,5′-Tetra-PCB52	0.3	0.3	0.2	0.1	0.6
2,2′,4,5,5′-Penta-PCB101	0.5	0.5	0.1	0.4	0.7
2,2′,3,4,4′,5′-Hexa-PCB138	42.2	37.5	15.8	26.0	62.0
2,2′,4,4′,5,5′-Hexa-PCB153	18.5	17.0	6.3	12.0	27.0
2,2′,3,4,4′,5,5′-Hepta-PCB180	51.2	53.0	16.1	32.0	68.0
PCDD/Fs	4.6 (4.7)	4.4 (4.4)	1.0 (1.1)	3.5 (3.7)	5.8 (6.3)

Plasma concentrations are given in μg/kg lipid, excepting those of PCDD/Fs which are expressed in ng I-TEQ/kg lipid (between parentheses, in ng WHO-TEQ/kg lipid). SD, standard deviation.

was established during the construction of the facility in order to assess its potential environmental and public health impact (Schuhmacher et al., 1997, 1999a,b,c). That program also included the assessment of the internal exposure of the HWI workers to various contaminants. Thus, baseline data from blood and urine of the workers were obtained. Since then, we have annually measured the concentrations of those chemicals that had been already determined in the baseline survey (Agramunt et al., 2003; Domingo et al., 2001; Mari et al., 2007, 2009). In occupational medicine, biological monitoring is an important tool to evaluate the levels of exposure to potentially harmful substances taken up from the occupational environment (Angerer et al., 2007, 2011; Bolt and Thier, 2006; Hardt and Angerer, 2003; Wilhelm et al., 2003). In this paper, we present the concentrations of a number of organic substances determined in blood and urine of the HWI workers. Samples were collected in 2011, after approximately 12 years of regular operations in the facility. The current results were compared with those of the baseline survey, as well as with the most recent surveys performed in 2008, 2009 and 2010.

Materials and methods

Twenty-seven workers, 18 men and 9 women, participated in this study. The study design was limited by the reduced number of available personnel working in the plant, whose current staff is 30. As in our previous surveys (Agramunt et al., 2003; Domingo et al., 2001; Mari et al., 2007, 2009), subjects were classified according to their respective workplaces in the facility. Group 1 was composed of plant workers (sixteen individuals: incinerator operators, boiler maintenance, control panel, furnace maintenance, and waste-gas-washing operators), group 2 contained laboratory workers (six individuals: analytical jobs), and group 3 was formed by five workers (management and administrative tasks). The two latter were considered as control groups, as occupational activities basically take place in closed environments. Therefore, laboratory and administration workers do not have direct contact with the potential emissions of the HWI. Despite the notable percentage of workers participating voluntarily in the investigation (90%), the reduced number of total subjects, as well as the fact of being classified according to the workplace, means an important limitation for deriving conclusive recommendations. To evaluate exposure to organic substances in plasma and urine, as in previous studies, the individual samples were pooled into six "composite" samples corresponding to plant (four samples), laboratory (one sample) and management/administration (one sample) workers. Samples were mixed by equal volume per subject. In each group, the main criteria used for pooling was the specific workplace, as well as gender and age of the subjects, when possible (group 1).

The air concentration of target pollutants was not evaluated in each one of the working areas. Therefore, the exposure through inhalation could not be properly assessed. This is another important limitation of the investigation, as data of environmental monitoring are necessary to validate/corroborate the results of body burdens. Future campaigns of the monitoring study must consider the implementation of complementary analyses of PCDD/Fs and other organic pollutants in air. In recent years, a number of investigations have reported the ambient air levels of PCDD/Fs in the area where the HWI of Constantí is located (Nadal et al., 2009; Vilavert et al., 2009, 2012), providing estimative values of the exposure of PCDD/Fs by non-occupationally exposed individuals through air inhalation. In general terms, the environmental concentrations of PCDD/Fs have been found to be low, in comparison with those usually reported in urban and industrial areas of similar characteristics.

In October–November 2011, blood samples were drawn from each participant in thoroughly cleaned glass bottles. Approximately 50 ml were centrifuged for getting plasma. Moreover, urine samples were also collected and centrifuged. Plasma analyses were carried out for hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs 28, 52, 101, 138, 153, and 180), and PCDD/Fs. For PCDD/Fs, total tetra-, penta-, hexa-, hepta-, and octa-chlorinated dibenzodioxins and dibenzofurans, as well as the specific congeners containing chlorine in the 2,3,7,8-positions, were measured. In turn, the levels of 2,4- and 2,5-dichlorophenol (DCP), 2,4,5- and 2,4,6-trichlorophenol (TCP), pentachlorophenol (PCP), and 1-hydroxypyrene (1-HP) were measured in urine samples.

Plasma analysis of PCDD/Fs, PCBs and HCB was performed in accordance with the US EPA method 1625. The extraction and the clean-up procedures, as well as the analytical determination (HRGC/HRMS) were carried out as previously reported (Mari et al., 2007, 2009). Results were expressed according to the international NATO/CCMS and WHO systems. The urinary analysis of DCPs, TCPs and PCP were performed by HRGC/HRMS following the NIOSH method No. 8001, while 1-HP levels were determined by HPLC/fluorescence using the appropriate method from the DFG method collection analyses of hazardous substances in biological materials (Mari et al., 2007, 2009).

For calculations, when a result was below the limit of detection (LOD), the value was assumed to be half of that limit (ND = 1/2 LOD). When possible, the Levene test was used to compare the homogeneity of the variances. Significance of the data was computed by the Kruskal–Wallis and the Mann-Whitney U-test. The software SPSS (version IBM Statistics 19) was used for the statistical analyses. A probability of 0.05 or less was considered as significant.

Results

A summary of the current concentrations of HCB, PCBs and PCDD/Fs in plasma of the HWI workers is presented in Table 1. The mean concentrations were the following: 10.8 μg/kg lipid for HCB; 0.8 μg/kg lipid for 2,4,4′-tri-PCB28; 0.3 μg/kg lipid for 2,2′,5,5′-tetra-PCB52; 0.5 μg/kg lipid for 2,2′,4,5,5′-penta-PCB101; 42.2 μg/kg lipid for 2,2′,3,4,4′,5′-hexa-PCB138; 18.5 μg/kg lipid for