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Long-term study of environmental levels of dioxins and furans in the vicinity of a municipal solid waste incinerator

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Abstract

From 1975 to 2004, a municipal solid waste incinerator (MSWI) was operating in Montcada (Barcelona, Catalonia, Spain). Because of the potential health risks derived from emission of pollutants by the facility, especially polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs), a long-term monitoring program focused on measuring the environmental levels of PCDD/Fs near the facility, and to assess the health risks for the population living in the neighborhood, was established between 1996 and 2002. A total number of 111 soil and 121 herbage samples were analyzed for PCDD/Fs during this period. Human health risks for the individuals living near the MSWI (500 and 1000 m) were also assessed before (1998) and after modernization (2000) of the facility. It included PCDD/F inhalation, dermal contact, soil and dust ingestion, and food intake. All these data are here summarized. The environmental levels of PCDD/Fs showed that the MSWI was not the main responsible of the atmospheric pollution by these compounds. In turn, human health risks for the population living in the vicinity of the facility after introduction of a modern technology were negligible in comparison with the dietary PCDD/F exposure.

Keywords: Municipal solid waste incinerator; Dioxins and furans; Environmental levels; Human health risks; Risk assessment

1. Introduction

Nowadays, environmental decision-making is facing with greater difficulties than ever before. Some of these difficulties are social and political and arise, at least in part, of controversial situations such as the sitting of municipal solid waste incinerators (MSWIs).

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 combustion processes, and as a result of their occurrence as unwanted byproducts in various chlorinated chemical formulations (Alcock and Jones, 1996; Jones and Sewart, 1997). Due to their highly toxic properties, PCDD/Fs have received prolonged attention by the scientific community and environmental regulators (Fiedler, 1996; UNEP, 1999; Alcock et al., 2001). Among the thermal sources, until recent years, MSWIs and hazardous waste incinerators (HWIs) have had a preponderant role as PCDD/Fs emitters (Fiedler, 1996; Schuhmacher et al., 2000; Domingo et al., 2002a; Park et al., 2004; Capuano et al., 2005; Kim et al., 2005). 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; Meneses et al., 2004).

From 1975 to 2004, a MSWI was operating in Montcada (Barcelona, Catalonia, Spain). The MSWI was located in an active industrial zone, in which there is also a heavy traffic. The annual emission rate of total PCDD/F equivalents (I-TEQs) had been, on average, approximately 32.3 mg. Some improvements

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in the control process (temperature and residential time) allowed to drop the emission of PCDD/Fs to about one-fourth of the previous rate. However, until March 1999 an electrostatic precipitator was still used as emission control device. At that time, measures were taken to reduce notably PCDD/F emissions and a modernization of the flue gas cleaning system was carried out. An acid gas (HCl/SO_2) and metal emission limit equipment was installed and an active-carbon adsorption filter was added to the fabric filter. As a consequence, PCDD/F emissions dropped, on average, to 0.086 ng I-TEQ/Nm^3 (Abad et al., 2003), which is below the legal limit of 0.1 ng I-TEQ/Nm^3. In spite of this notable reduction, because citizen groups living in the vicinity of the MSWI kept still maintaining serious concerns about the presence of the facility, the popular pressure on the local authorities did not disappear, and finally, the local government decided to close the MSWI.

From 1996 to 2002, and before making the final decision about the MSWI, local government raised some relevant questions: (1) which were the current environmental levels of PCDD/Fs and their spatial and temporal trends, (2) how proposed changes influenced in the environment, and (3) which were the PCDD/F health risks for the population living in the vicinity of the MSWI. In response to these questions, a two-step program was elaborated. As a first step, from 1996 to 2002 a monitoring program of the environmental levels of PCDD/Fs was developed, while in a second step the health risks for the population living in the neighborhood of the facility were assessed.

To establish the environmental levels in the area under potential influence of the MSWI, and to evaluate the improvement of the quality of the environment after the application of new measures (emission cleaning device), soil and herbage samples were analyzed for PCDD/Fs in 1996, 1997, and 1998 (before the introduction of the air-cleaning device), and in 2000 and 2002 (after the introduction of the air-cleaning device). The program was designed to reach the following objectives: (a) to establish the current concentrations of PCDD/Fs in environmental matrices collected in the vicinity of the MSWI, and to evaluate the principal cause of the contamination; (b) to determine the variations on the levels of these pollutants after introducing the new air cleaning device; and (c) to help in the decision-making process.

Soil and herbage were chosen as indicators of the environmental PCDD/F pollution. While soils are mainly used to describe long-term exposure to PCDD/Fs, vegetation can bring information on the short-term exposure to these pollutants. On the other hand, health risks of PCDD/F exposure for the population living in the vicinity were also assessed before and after the pronounced decreases in the air emissions of PCDD/Fs from the facility were observed.

2. Materials and methods

2.1. Sample collection

The MSWI was placed in a residential area with an important industrial activity. There are some small mountains at the south and southeast of the facility. A motorway with a heavy traffic was near to the incinerator. A close meteorological station provided data on the wind regimens. The impact area under the influence of the facility was defined by the US EPA dispersion model Industrial Source Complex Long-Tem (ISC-LT).

For sampling, 24 monitoring sites were selected in a radius of 3 km taking into account considerations on the prediction of the time averaged emission plume obtained from the Gaussian model (ISC-LT) (Schuhmacher et al., 1998; Domingo et al., 1999a). These sites were located at 100, 250, 500, 750, 1000, 1500, 2000 and 3000 m from the stack in each of the three main historical directions of the wind rose in the area: S, NW and NE. During the surveys carried out in 1996, 1997, 1998 and 2000, 24 samples of soil were taken, while in 2002 only 15 soil samples corresponding to 100, 250, 500, 1500 and 3000 m from the stack, at the main wind directions were collected.

With respect to vegetation, *Pipatherum paradoxum* L. was selected as an important representative species of the vegetation of the area. In 1996, 24 samples were collected at the same sites than soils. In 1997, 1998 and 2000, due to difficulties in finding herbage in some cases, or for technical analytical problems in other cases, only 23 samples were taken. In 2001, only 12 samples corresponding to 100, 250, 500 and 1500 m from the MSWI at the same 3 wind directions were collected, while in 2002, 15 herbage samples were collected. Therefore, a total of 111 soil and 121 herbage samples were measured for PCDD/F levels during the total monitoring program (1996–2002).

Each soil and vegetation sample was taken from an area of approximately 50×50 m. Soils were clayey and sandy. The sample amounts consisted of a minimum of 500 g. Soil samples were sieved through a 2-mm mesh screen to obtain a more homogenous grain distribution. Determination of dry matter content was achieved by drying subsamples (1–3 g) at 130 °C overnight (Schuhmacher et al., 1997a). Vegetation samples of 120–150 g (dry weight) were obtained by cutting vegetation at a height of approximately 4 cm from the soil.

2.2. PCDD/F analyses

The extraction and clean-up procedures, as well as the analyses of PCDD/Fs were performed by HRGC/HRMS as widely detailed in previous reports (Schuhmacher et al., 1997a,b, 1998). For the determination of groups of PCDD/F congeners with the same degree of chlorination and for the hepta- and octa-PCDD/F congeners substituted at the 2,3,7,8-positions, a Hewlett Packard 5890 series II gas chromatograph provided with an on-column injector was applied in combination with a Hewlett-Packard 5972 low resolution mass spectrometer. For the determination of the tetra- to hexa-PCDD/F congeners substituted at the 2,3,7,8-positions, a Varian 3300 gas chromatograph provided with a septum-equipped programmable injector was applied in combination with a Finnigan MAT 95 high-resolution mass spectrometer. Good conditions of the instruments were regularly maintained by mass calibration and proved by standard runs. Analytical procedures included the use of ^{13}C-labeled internal standards. To achieve the best possible quality with the analytical process, internal standards were calibrated against references at regular intervals to ensure long-time accuracy. In turn, long-time reproduction of the analytical methods was ensured by analysis of internal and external reference materials. Recoveries of added surrogates were determined and evaluated according to the European norm EN-1948 part 1/2/3 for PCDD/F analysis in emissions (Domingo et al., 1999a,b; Nadal et al., 2002). For I-TEQ calculations, in the case of values under the detection limit, the congener was assumed to be present at one-half of that limit of detection.

2.3. Human health risks

To assess the health risks of PCDD/F exposure for the general population living in the area under potential influence of the MSWI, the following routes should be evaluated: (1) direct contact from inhalation of air and particles, as well as ingestion and dermal contact of soil and dust, and (2) exposure derived from the consumption of locally grown vegetables, local meats, and local dairy products, which could be contaminated by PCDD/Fs from exposure to polluted soils and herbage. However, according to the urban location of the facility, there were neither pasture grounds nor crops of vegetables, grains, or fruits. Consequently, additional exposure from ingestion of local products that might be potentially contaminated by PCDD/Fs was excluded. Inhalation exposure was calculated by assuming that individuals were exposed to polluted air 24 h/