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Long-term monitoring of dioxins and furans near a municipal solid waste incinerator: human health risks

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Abstract

Since 1996, a wide surveillance programme has been developed to get overall information on the impact of a municipal solid waste incinerator (MSWI) in Tarragona (Catalonia, Spain). The concentrations of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) have been periodically measured in soil and vegetation samples collected at locations in the incinerator surroundings. Furthermore, air PCDD/F levels have been also monitored by using active and passive sampling devices, generating a huge amount of information regarding the environmental status of the zone. In the last survey (2009–2010), mean PCDD/F levels in vegetation, soil and air were 0.06 ng I-TEQ kg^{-1}, 0.58 ng I-TEQ kg^{-1} and 10.5 fg WHO-TEQ m^{-3}, respectively. Both soil and herbage showed a notable reduction in the PCDD/F concentrations in comparison with the baseline study, with this decrease only being significant for soils. In contrast, PCDD/F values in air remained similar during the whole assessment period. Human exposure to PCDD/Fs was evaluated under different scenarios, and the associated non-carcinogenic and carcinogenic risks were assessed. The hazard quotient was below unity in all cases, while cancer risks were under 10^{-6}, which is lower than the maximum recommended guidelines. The current results clearly show that the MSWI of Tarragona does not produce additional health risks for the population living nearby.

Keywords

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), municipal solid waste incinerator, human health risk assessment, soil, vegetation, ambient air, monitoring programme

Introduction

In recent decades, the generation of municipal waste in Europe has continuously increased. According to Eurostat statistics (European Commission, 2011), the EU currently produces 3 billion tonnes of waste per year, corresponding to approximately 6 tonnes of solid waste per person. Under this scenario, it is evident that the safe treatment and disposal of municipal solid waste is a major concern. Due to legislative, environmental, social and economic constraints, the identification and use of sustainable ways of municipal solid waste (MSW) disposal must be enhanced (de Araújo et al., 2010). The European waste hierarchy, developed in the Waste Framework Directive (2008/98/EC), reports a priority order of what constitutes the best overall environmental option in waste legislation and policy: prevention, preparing for re-use, recycling, other recovery (e.g. energy recovery), and disposal. Although incineration is not the top preference method for waste treatment, it is clearly more advisable than landfilling when managing waste that cannot be reused or recycled. Furthermore, incineration has multiple advantages in comparison with disposal in landfills, such as energy recovery and volume minimization (Chung et al., 2010). In recent years, modern incinerators have become widely recognized as an efficient way of MSW treatment, while their contribution as pollutant releasers has decreased by introducing new legislative and technical regulation measures (Inoue et al., 2009). Despite the fact that MSW incineration has been traditionally affected by the 'not in my back yard' (NIMBY) syndrome, public controversy is inevitable at those locations where MSW incinerators (MSWIs) are operating, but especially prior to the construction of new facilities (Elliott, 1998). Among the chemicals generated during MSW incineration, polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), commonly known as dioxins, have received special attention (Chang and Chen, 2000; Vilavert et al., 2009). Dioxins are persistent organic pollutants (POPs), which are formed in the production of certain organochlorine chemicals, as well as in other industrial processes as byproducts (Weber et al.,

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2011). All those chemicals are associated with adverse health effects (Rovira et al., 2010). Among the different congeners, the contribution of the 17 2,3,7,8-substituted congeners, which are the most toxic, is generally assessed (Fiedler et al., 2000; Vassura et al., 2011). Among these, 2,3,7,8-tetrachlorodibenzodioxin (TCDD) is known to cause adverse health effects, such as lethal toxicity, reproductive effects, immunotoxicity and endocrine disruptions. According to the International Agency for the Research of Cancer (IARC), TCDD is catalogued as a Group 1 agent, meaning that it is carcinogenic to humans (Schwartz et al., 2010; Viel et al., 2011).

The MSWI of Tarragona (Catalonia, Spain) has been operating since 1991. During the last 15 years, the concentrations of PCDD/Fs were uninterruptedly and periodically measured in soil and vegetation samples, as long- and short-term environmental monitors, respectively, collected from the MSWI surroundings. These data were used to evaluate the human health risks associated with exposure to those pollutants (Domingo et al., 2001; Mari et al., 2007). Recently, active and passive sampling devices have also been used to measure PCDD/Fs in air (Vilavert et al., 2009).

Herein, updated information regarding the levels of PCDD/Fs in soil, vegetation, and air samples collected in the last period of the monitoring programme (2009–2010) is presented. The temporal trends of the PCDD/F environmental burdens were determined by comparing the current data with those from previous surveys. Finally, human health risks derived from the exposure to PCDD/Fs were evaluated for the population living within the zone that is potentially influenced by the facility.

Materials and methods

Sampling

In May 2009, vegetation and air samples were collected around the MSWI of Tarragona. The sampling sites were the same as those corresponding to the baseline and subsequent studies (Vilavert et al., 2009). Sampling points were selected in different wind directions and at various distances in order to consider all factors that might significantly influence the concentration of PCDD/Fs in the different environmental monitoring points. Eight samples, chosen among the 24 original sampling sites (Vilavert et al., 2009), were collected at six distances (250, 500, 750, 1000, 1250 and 1500 m) and wind directions (NE, NW, SE and SW) from the facility (Figure 1). These eight locations were considered to be the most representative of the zone. The choice of these locations was done after performing a detailed study of the temporal trends of PCDD/Fs in each one of those sampling sites, between 1996 and 2006. The representativeness was determined by rejecting points with important fluctuations of PCDD/Fs along time, which could denote the potential existence of specific pollution sources. Moreover, at least one sample per direction and/or distance was included in the set. This modification was proved to be successful (Vilavert et al., 2009), as more information was acquired without jeopardizing the scientific accuracy of the investigation. Approximately 150 g of vegetation (*Piptatherum paradoxum*) were obtained by cutting the plants at a height of approximately 5 cm from the ground. Samples were quickly packed in aluminium foils, and processed in the laboratory, where they were dried at room temperature and packed until analysis. On the other hand, air samples were collected by using a TE-1000-PUF high-volume active sampler (Tisch Environmental, Cleves, OH, USA). Sampling volumes were within the range 636–709 m^3. In June 2010, eight soil and air samples were collected at the same sampling points. Soils were taken from the upper 3 cm and stored in polyethylene bags. They were dried at room temperature and homogenized by sieving through a 2-mm mesh screen. In addition, passive air samplings were also performed. Polyurethane foam (PUF) passive samplers (PacWill Environmental, Stoney Creek, ON, Canada) were deployed for approximately 3 months. Applying a sampling rate of 2 $m^3\ day^{-1}$ (Mari et al., 2008), the total volume of sampled air was estimated to be 174 m^3. Once collected, the PUFs were kept in a glass airtight container with Teflon cap and amber colour to avoid photodegradation of the compounds. Once in the laboratory, the samples were kept at −20°C until analysis.

Analytical procedure

The concentrations of PCDD/Fs in soil and vegetation samples were determined by means of high resolution gas chromatography coupled to high resolution mass spectrometry (HRGC/HRMS), according to the US EPA 1613 method. In turn, the concentrations of PCDD/Fs in air were also determined by HRGC/HRMS, following the German VDI 3499 method. Appropriately labelled extraction standards ($^{13}C_{12}$-PCDD/Fs substituted congeners) were added in order to control the whole sample preparation process and to evaluate potential losses. An accelerated solvent extraction (ASE) was carried out with toluene. The extract was concentrated and divided into separate parts for determining the target compounds. The clean-up procedure was executed by using adsorption chromatography on a mixed silica column and adsorption/fractionation on alumina. The final obtained PCDD/F-extracts were injected and analysed separately on an Agilent 6890 capillary gas chromatograph equipped with a DB5-MS capillary column and coupled to a Waters Autospec Ultima high resolution mass spectrometer, with selected ion recording at resolution of 10,000. The ranges of recovery percentages were 8–85%, 36–116% and 51–170% for soil, vegetation and air samples, respectively.

Human health risk assessment

A human health risk assessment study was subsequently performed. Risk assessment is being largely used for estimating the environmental impacts of anthropogenic activities, in general, and those from incinerators, in particular (Morselli et al., 2011). PCDD/F concentrations in soil and air samples collected around the plant were used to estimate human exposure and to characterize health risks in the vicinity of the facility. Exposure of the local

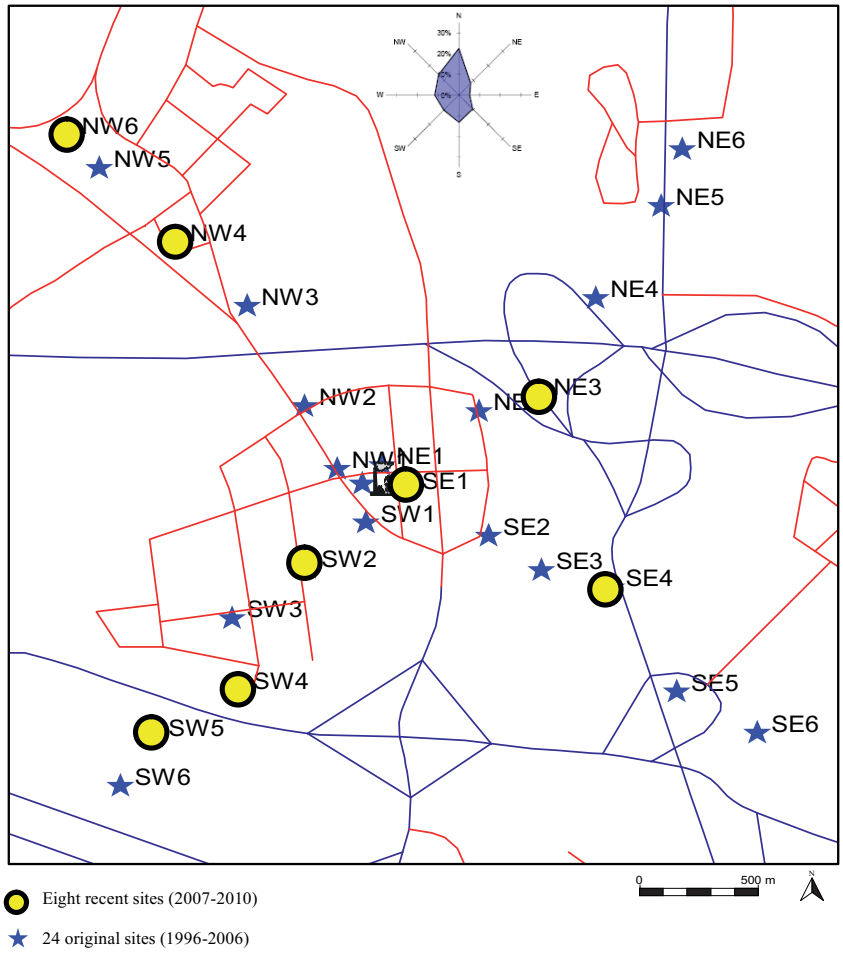

Figure 1. Sampling sites and wind rose.

population was calculated considering three different pathways: soil ingestion, dermal contact and air inhalation. The equations used to assess exposure through ingestion (Exp_{ing}), dermal contact (Exp_{derm}), and inhalation (Exp_{inh}) were taken from the US EPA methodology of human health risk assessment (US EPA, 1989), and largely applied in previous studies (Nadal et al., 2011). The specific equations and values of the parameters can be found elsewhere (Rovira et al., 2011a,b). The non-cancer risk [Hazard quotient (HQ)] was estimated by comparing the exposure concentration and the reference dose (RfD). In turn, carcinogenic risk was calculated by multiplying the predicted ingestion and dermal exposure by the oral slope factor. Furthermore, inhalation risks were determined based on the most updated US EPA RAGS methodology for this pathway (US EPA, 2009).

Statistics

Data were statistically analysed by using the SPSS 17.0 statistical software package. The Levene test was applied to analyse the equality of variances. Analysis of variance (ANOVA) or Mann Whitney U-tests were subsequently executed depending on whether data followed a normal or non-normal distribution, respectively. A probability lower than 0.05 ($p < 0.05$) was considered to be statistically significant.

Results and discussion

The levels of PCDD/Fs in vegetation, soil and air samples collected in 2009 and 2010 in the vicinity of MSWI of Tarragona are summarized in Table 1. Data for each individual sampling site, as well as the temporal trends, are also shown. In 2009, the mean PCDD/F concentration in vegetation was 0.06 ng International Toxic Equivalents (I-TEQ) kg^{-1} dry weight (dw) (0.06 ng WHO Toxic Equivalents (WHO-TEQ) kg^{-1} dw), ranging from 0.04 to 0.11 ng I-TEQ kg^{-1} dw. It meant a non-significant decrease in comparison to the previous (2007) and baseline (1999) surveys (reductions of 40 and 45%, respectively). It must be taken into account that the first campaign of the monitoring programme (1996) was not used as background, as it was performed prior to the installation of modern pollution control devices (Domingo et al., 2001). Specifically, between 1999 and 2009, herbage concentrations decreased in five of the eight sampling points, and increased in the remaining three points. Comparing with the previous survey (2007), the current concentrations decreased in seven of the eight sampling points and increased in only one. In 2010, the range of PCDD/F concentrations in soil was between 0.11 and 1.35 ng I-TEQ kg^{-1} dw (0.13 and 1.14 ng WHO-TEQ kg^{-1} dw, respectively), with a mean value of 0.58 ng I-TEQ kg^{-1} dw. A significant decrease was observed between the baseline

Table 1. Concentrations of PCDD/Fs in herbage, soil and air samples collected in the vicinity of the MSWI of Tarragona (Catalonia, Spain). Temporal variations.

Samples Herbage[a]	Distance (m)	Year			Temporal variations (%)	
		1999	2007	2009	1999–2009	2007–2009
SE-1	250	0.21	0.16	0.05	–76	–71
SW-2	500	0.06	0.09	0.08	33	–27
NE-3	750	0.10	0.10	0.06	–40	–45
NW-4	1000	0.07	0.08	0.06	–14	–25
SE-4	1000	0.08	0.06	0.05	–38	–29
SW-4	1000	0.05	0.05	0.11	120	120
SW-5	1250	0.04	0.07	0.06	50	–25
NW-6	1500	0.07	0.12	0.04	–43	–64
Total TEQ		*0.11 ± 0.07*[c]	*0.10 ± 0.04*	*0.06 ± 0.02*	*–45*	*–40*
Soil[a]		1999	2008	2010	1999–2010	2008–2010
SE-1	250	0.65	0.63	1.16	78	84
SW-2	500	1.05	0.13	1.35	29	938
NE-3	750	4.89	0.28	0.43	–91	54
NW-4	1000	0.95	0.31	0.36	–62	16
SE-4	1000	1.62	0.13	0.11	–93	–15
SW-4	1000	1.45	0.34	0.44	–70	29
SW-5	1250	2.09	2.41	0.27	–87	–89
NW-6	1500	0.90	0.86	0.51	–43	–41
Total TEQ		*1.20 ± 1.01*[c]	*0.64 ± 0.76*	*0.58 ± 0.44*	*–52**	*–9*
Air[b]		2007	2009	2010	2007–2010	2009–2010
SE-1	250	32.5	3.61	7.67	–76	113
SW-2	500	5.49	15.1	22.3	306	47
NE-3	750	8.49	22.7	12.0	41	–47
NW-4	1000	12.3	4.95	6.95	–43	40
SE-4	1000	19.9	7.00	11.2	–44	60
SW-4	1000	5.15	5.98	7.47	45	25
SW-5	1250	8.48	3.70	7.50	–12	103
NW-6	1500	3.93	5.66	8.52	117	51
Total TEQ		*12.03 ± 9.70*	*8.59 ± 6.78*	*10.45 ± 5.14*	*–13*	*22*

[a]Results are expressed in ng I-TEQ kg^{-1} (dry weight).
[b]Results are expressed in fg WHO-TEQ m^{-3}.
[c]Mean value of 24 samples.
*Asterisks indicate significant differences at $p < 0.05$.

(1999) and last (2010) study (52%; $p < 0.05$), with important reductions in six of the eight sampling sites. In the 2008–2010 period, PCDD/Fs in soils showed a non-significant decrease (9%; $p > 0.05$). Air samples showed similar figures in the different campaigns. In 2010, airborne PCDD/F levels ranged between 6.95 and 22.3 fg WHO-TEQ m^{-3}, with a mean value of 10.5 fg WHO-TEQ m^{-3}. A slight increase of 22% was noted with respect to the previous survey (2009), whereas a 13% reduction of PCDD/F levels was observed with respect to the first campaign (2007) for which information regarding ambient air concentrations is available. Anyhow, no significant differences were noted over time.

The current results are in accordance with those found in similar areas, and very especially with those corresponding to soils and vegetation with the same characteristics as those here evaluated. Rovira et al. (2010) reported that PCDD/F concentrations in soils surrounding a MSWI in Mataró (Catalonia, Spain) were between 0.14 and 0.46 ng WHO-TEQ kg^{-1}, while PCDD/F levels in air samples ranged from 0.008 to 0.015 pg WHO-TEQ m^{-3}. In turn, Vilavert et al. (2010) reported a mean soil PCDD/F concentration of 0.75 ng I-TEQ kg^{-1} near a HWI (Constantí, Catalonia) after 10 years of continuous monitoring. On the other hand, levels ranging 0.16–14 ng I-TEQ kg^{-1} were observed in an industrial zone of Trondheim (Norway) (Andersson and Ottesen, 2008), while PCDD/F concentrations in agricultural soils surrounding two MSWIs in Shanghai (China) ranged from 0.64 to 61.15 pg I-TEQ g^{-1} (Deng et al., 2011). PCDD/F concentrations in soils near the MSWI of Tarragona are currently in the low part of the range, in comparison with literature data from industrial and urban zones (Nadal et al., 2011), and more specifically lower than results from other authors for corresponding soils near MSWIs (Kim et al., 2009; Wu et al., 2009). Furthermore, they are even lower than those typical of agricultural soils of the United States (US EPA, 2007). The comparison of data concerning the vegetal compartment is rather difficult because of the particularities of different species. In recent years, an important database on

PCDD/Fs in *Piptatherum* sp. has been created from a number of environmental surveys performed in Catalonia. Rovira et al. (2011a) found a concentration range of 0.17–0.32 ng WHO-TEQ kg^{-1} around a cement plant in Sant Feliu de Llobregat (Spain), and the same authors reported values from 0.08 to 0.31 ng WHO-TEQ kg^{-1} near another cement plant, located in Montcada i Reixac (Spain) (Rovira et al., 2011b). Recently, Vilavert et al. (2010) reported a wide range of values (0.05–3.60 ng I-TEQ kg^{-1}) in vegetation from the vicinity of a HWI.

The PCDD/F congener profiles in soil, vegetation and ambient air samples collected between 1999 and 2011 in the surroundings of the MSWI of Tarragona are depicted in Figure 2. In all cases, the most abundant congener was OCDD, followed by OCDF. In contrast, 2,3,7,8-TCDD and 1,2,3,7,8,9-HxCDF presented the minimum contribution to the total amount of PCDD/Fs. In soils, some predominance of the heaviest congeners was observed. However, an important variance in the values was observed irrespective of the evaluated matrix (Figure 2). No significant differences were noted according to the specific campaign for any of the monitors. However, a certain reduction of the OCDD levels in vegetation was observed concurrently to the slight increase in the 1,2,3,4,7,8,9-HpCDF concentrations. In order to establish the potential influence of the MSWI on its surroundings, the ambient air PCDD/F congener profile was particularly compared with that corresponding to emitted air (Figure 3). Both profiles were different. On one hand, 1,2,3,4,6,7,8-HpCDD was the predominant congener in emission samples. On the other hand, OCDF meant < 5% of the total PCDD/F burden in the released air, but > 25% in emission samples.

To identify potential changes in pollution sources, a principal components analysis (PCA) was applied to PCDD/F

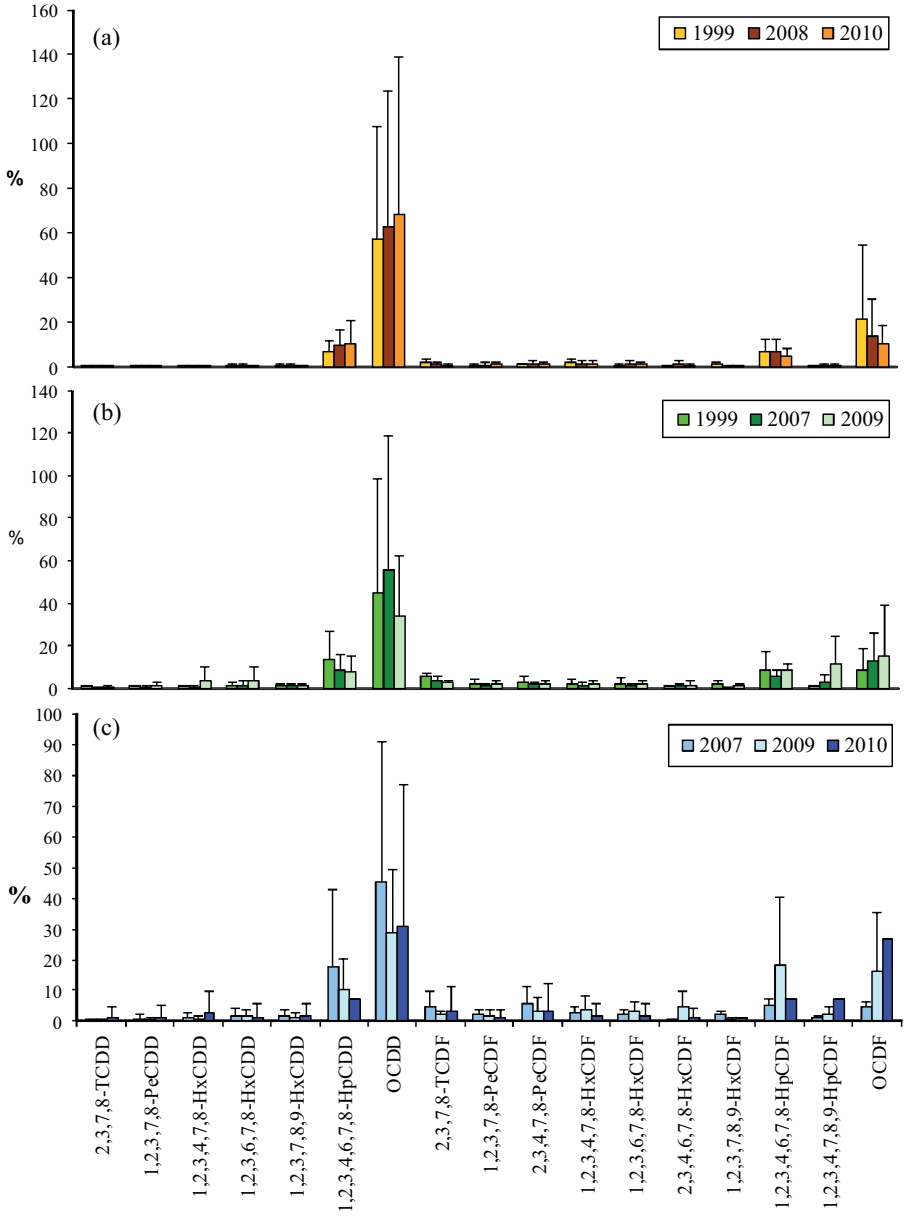

Figure 2. PCDD/F congener profiles in (a) soil, (b) herbage, and (c) air samples collected around the MSWI of Tarragona (Catalonia, Spain).

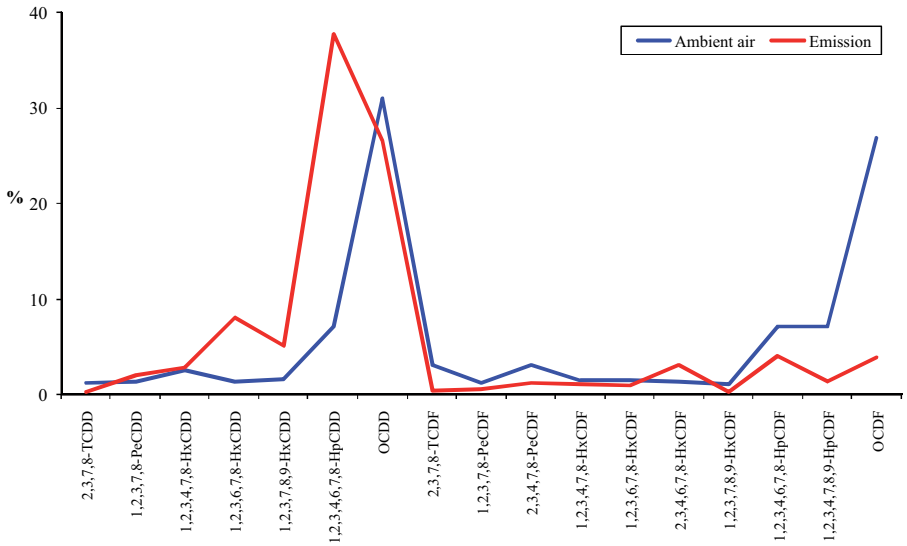

Figure 3. Comparison of PCDD/F congener profiles in air in 2010: Ambient air and emission gas.

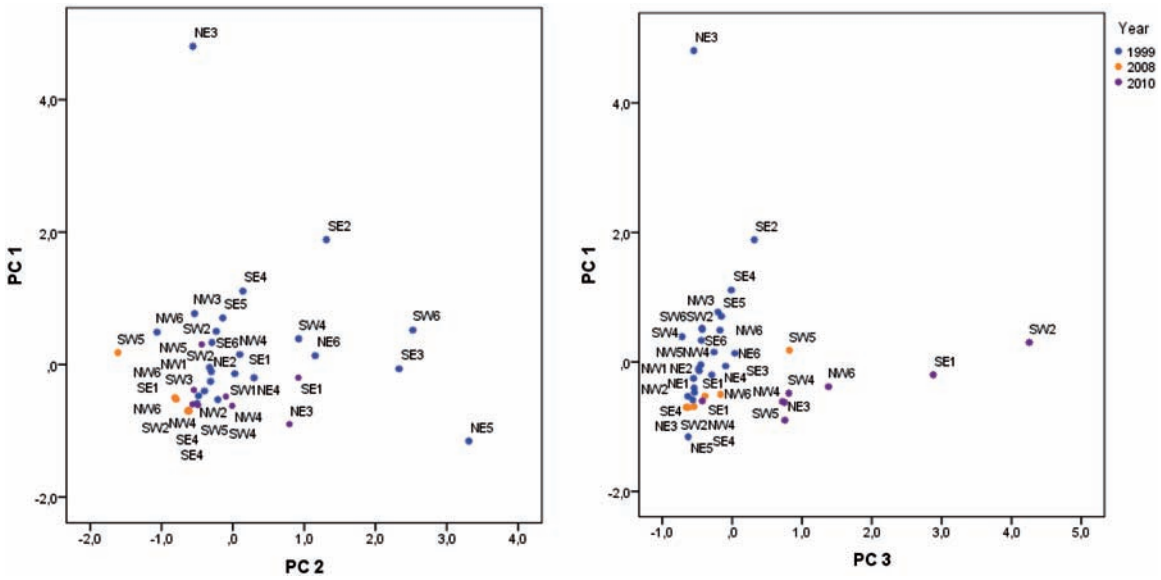

Figure 4. Principal component analysis of PCDD/Fs in soil.

concentrations in soils (Figure 4) and vegetation (Figure 5) samples collected in the vicinity of the MSWI. PCA, which has been widely used in environmental analysis (Chen Liu and Wu, 2011), is a very effective method to identify different sources of PCDD/Fs (Liu et al., 2012). The PCA in soil showed a three-dimensional model that explains 83.6% of the variance. The first principal component (PC1), which explained 64.3% of the variance, was correlated with penta- and hexa-dioxins, as well as with hexa-furans (1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF and 1,2,3,7,8,9-HxCDF). In turn, PC2 (11.5% of the variance) correlated with the heaviest congeners (1,2,3,4,6,7,8-HpCDD, OCDD and OCDF). Finally, PC3 (7.8% of the variance) correlated with 1,2,3,7,8-PeCDD and 1,2,3,7,8,9-HxCDF. Most samples collected in 2008 and 2010 formed a single cluster, and outliers were only detectable for some samples of the 1999 survey. Moreover, the PCA applied to vegetation samples provided a fourth-dimensional model that explained 82.2% of the variance. PC1, which would explain 31.2% of variance, was correlated with 1,2,3,4,6,7,8-HpCDD, as well as with penta-furans, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, and 1,2,3,4,6,7,8-HpCDF. PC2 (25.9% of the variance) was associated with 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, and 1,2,3,4,7,8,9-HpCDF. PC3 (13.7% of the variance) was positively related with 2,3,7,8-TCDD and 1,2,3,4,7,8-HxCDD, and negatively with 2,3,4,6,7,8-HxCDF, OCDF and OCDD. Finally, the fourth PC (11.4% of the variance) was positively correlated with 1,2,3,7,8,9-HxCDD and negatively with OCDF and 1,2,3,6,7,8-HxCDF. In general terms, most samples presented a similar profile during the whole monitoring period. Although certain particular samples showed high specific PCs, no spatial variation was observed when assessing

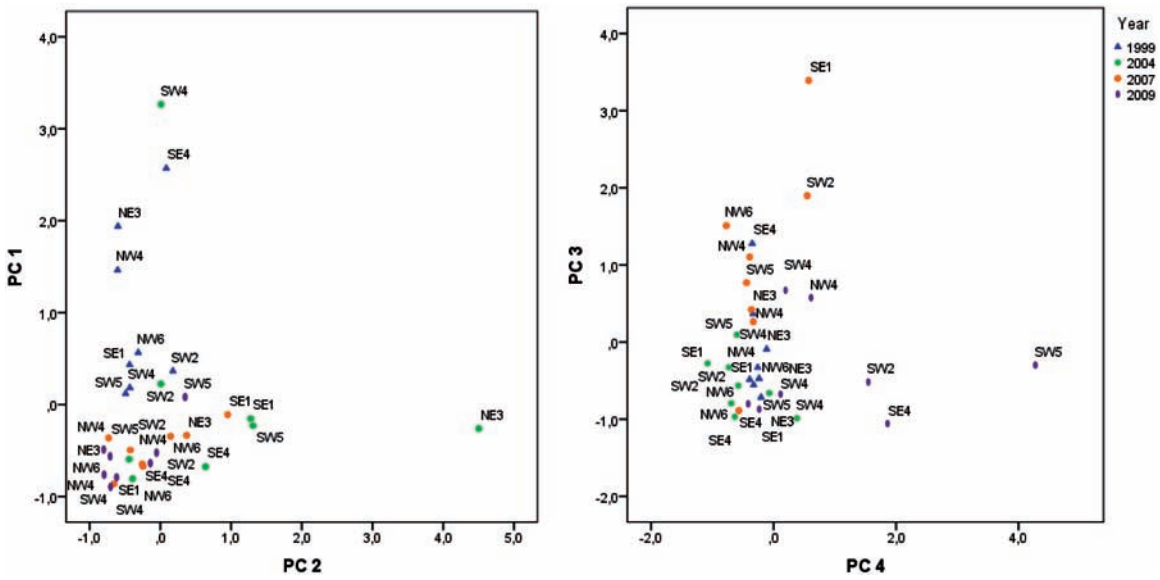

Figure 5. Principal component analysis of PCDD/Fs in vegetation.

the PCA results. There were no correlations between the levels of specific PCDD/F congeners and the distance/direction to the incineration plant.

The concentrations of PCDD/Fs in soil and air samples collected around the facility were used to determine human exposure and health risks for the population living in the vicinity of the MSWI. Three scenarios were considered according to the location of receptors: (A) all sampling points, (B) sites closest to the facility (< 750 m), and (C) sites furthest to the facility (> 1000 m). Exposure to these pollutants is summarized in Table 2. Although human exposure to PCDD/Fs was somewhat higher in the closest area, the difference was not statistically significant. Furthermore, the percentage of each route was very similar in the three scenarios, with inhalation being the main exposure pathway. In turn, dermal contact and soil ingestion presented similar contributions (15–20%). The non-carcinogenic and cancer risks derived from environmental exposure to PCDD/Fs are shown in Table 3. The HQ was notably lower than the reference value, which is set at the unity (Morselli et al., 2011). The dermal route presented the greatest risk, although it was at least three orders of magnitude lower than the threshold value. All carcinogenic risk values were clearly below 10^{-5}, which is the acceptable excess of cancer risk (1/100 000 for lifetime-exposed individuals), according to the Spanish legislation. These results indicate that MSWI of Tarragona should not represent any additional risk due to the exposure to PCDD/Fs for the population living in the surroundings.

In this study, updated data on the environmental levels of PCDD/Fs near a MSWI have been shown. The length of the monitoring programme must be highlighted, as the surveillance programme has been performed for 15 years, with an uninterrupted monitoring of the PCDD/F levels in different environmental compartments. As previously stated (Nadal et al., 2011), these results reinforce the need to consider long time periods through multi-annual campaigns, instead of performing single campaign investigations, as these may lead to an important under- or overestimation of the results, and therefore of the human health risks. Secondly, the complementarity of using diverse monitors, such as soil, vegetation and air, is important, not only because they offer complementary information, but also to minimize the possible biases derived of analysing a single monitor. More specifically, the results of the current study are new evidences that the environmental impact of the MSWI of Tarragona, regarding the emissions of PCDD/Fs, is rather low. Consequently, the facility

Table 2. Environmental exposure to PCDD/Fs (in ng WHO-TEQ kg^{-1} day^{-1}) through different pathways for the population living in the vicinity of the MSWI of Tarragona (Catalonia, Spain).

Pathway	A	%A	B	%B	C	%C
Inhalation	2.86 E–06	64	3.83 E–06	59	2.28 E–06	70
Dermal contact	8.33 E–07	19	1.37 E–06	21	5.11 E–07	15
Soil ingestion	7.82 E–07	17	1.28 E–06	20	4.80 E–07	15
Total exposure	4.48 E–06	100	6.48 E–06	100	3.27 E–06	100

A, all sampling points.
B, points closer to the facility (< 750 m).
C, farthest points to the facility (> 1000 m).

Table 3. Non-carcinogenic and carcinogenic risks derived from exposure to PCDD/Fs in the surroundings of the MSWI of Tarragona.

	Pathway	A	B	C
Non-cancer risk	Soil ingestion	7.82 E–04	1.28 E–03	4.80 E–04
	Dermal contact	8.33 E–04	1.37 E–03	5.11 E–04
	Air inhalation	2.50 E–04	3.35 E–04	2.00 E–04
	TOTAL	1.86 E–03	2.99 E–03	1.19 E–03
Cancer risk	Soil ingestion	4.35 E–08	7.16 E–08	2.67 E–08
	Dermal contact	4.64 E–08	7.63 E–08	2.85 E–08
	Air inhalation	1.63 E–07	2.18 E–07	1.30 E–07
	TOTAL	2.53 E–07	3.66 E–07	1.85 E–07

A, all sampling points.
B, points closer to the facility (< 750 m).
C, farthest points to the facility (> 1000 m).

does not represent any notable risk to the health of the population living nearby. However, as the MSWI has been already operating for more than 20 years, the extension of the monitoring programme is now even more desirable to ensure a good environmental status of the zone for the near future.

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