Two Decades of Environmental Surveillance in the Vicinity of a Waste Incinerator: Human Health Risks Associated with Metals and PCDD/Fs

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The concentrations of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), as well as the levels of a number of heavy metals, have been periodically measured in samples of soil and vegetation collected around a municipal solid waste incinerator (MSWI) in Tarragona (Catalonia, Spain), for approximately 20 years. Since 2007, the levels of the above pollutants have been also determined in air samples by means of either active or passive samplers. In the present study, data regarding the environmental impact of the MSWI, in terms of PCDD/Fs and heavy metals, are updated. The temporal trends of these pollutants were evaluated by comparison with data from previous surveys. In the current survey (2013-2014), mean concentrations of PCDD/Fs in soil, vegetation, and air were 0.63 ng I-TEQ/g, 0.07 ng I-TEQ/g, and 10.1 fg WHO-TEQ/m^3, respectively. Reductions of 47% and 35% of PCDD/Fs in soil and vegetation, respectively, were observed in relation to the background study (1999). Regarding air samples, a very slight temporal decrease of the PCDD/F levels was also found, remaining the concentrations nearly constant through time. With respect to metals, notable fluctuations in the concentrations were noted, being dependent on each specific environmental monitor. Overall, the current exposure to PCDD/Fs and metals does not mean any additional health risks for the population living near the facility. In conclusion, the results of the present study show that the environmental impact of the MSWI of Tarragona is not significant, in terms of PCDD/Fs and heavy metals, after more than 20 years of continuous operation.

According to data of the European Commission (EC), people use 16 tons of material per person per year, of which 6 tons are converted into waste (EC, 2015). Therefore, it is obvious that the safe treatment and disposal of waste in general, and municipal solid waste (MSW) in particular, is an issue of a great importance (Nadal et al. 2005, 2011). 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 recoveries, and disposal. Incineration is a well-known option for MSW treatment. In the Europe-27, about 20% (in weight) of MSW is being incinerated (Vassura et al. 2011). Although according to the European Union (EU), incineration is not the best option for waste treatment, it is better than landfilling when the MSW cannot be reused or recycled. Moreover, incineration has also some specific benefits such as volume reduction and energy recovery (Nadal et al. 2009). The EU sets a series of measures to prevent or reduce air, water, and soil pollution, caused by incineration and co-incineration of waste, as well as the human health risks derived from such operations. These measures include the obligation to obtain permission for incineration or co-incineration and setting emission limits for certain pollutants released into the air and water (Directive 2000/76/EC).

In recent decades, a considerable number of studies have demonstrated that incinerators emit environmental pollutants such as heavy metals and dioxins (Chen et al. 2014b; Margallo et al. 2014; Park et al. 2014; Peng et al. 2015; van Dijk et al. 2015). In 2000, a study revealed that at European scale, the largest annual emission of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) was released from MSW incinerators (MSWIs) (Quaß et al. 2000). However, operators of MSWIs have been working to find effective means of efficiently controlling the emissions of PCDD/Fs (Vilavert et al. 2010; Thacker et al. 2013). As it is well known, in recent years, PCDD/F and heavy metal levels around MSWIs have been decreasing due to the reduction of the emissions from the stacks (Domingo et al. 2001b; Ferré-Huguet et al. 2007; Schuhmacher et al. 1998; Takeda and Takaoka 2013; Venturini et al. 2013; Vilavert et al. 2010, 2012, 2014).

The MSWI of Tarragona (Catalonia, Spain) has been operating since 1991. Based on a wide surveillance program initiated approximately 20 years ago, the concentrations of PCDD/Fs and heavy metals are being periodically measured in soil and vegetation

samples collected in the vicinity of the facility (Domingo et al. 2001a,b; Llobet et al. 2002; Mari et al. 2007; Schuhmacher et al. 1996, 1997; Vilavert et al. 2009, 2012). Soils and herbage were chosen as indicators of long-term and short-term environmental pollution, respectively.

According to the results of the periods 1996-1999 and 2002-2005, it was concluded that the impact of the plant was not remarkable in comparison with other potential sources of metals and PCDD/Fs in the same area. Taking into account the notable reduction found in the levels of metals and PCDD/Fs in soil and vegetation samples, in the last campaign, initiated in 2006 and completed in 2011, we decided to reduce the number of sampling points from 24 to 8. However, since 2007 we also decided to collect air samples, as indicators of current environmental pollution, by means of active and passive sampling devices, determining PCDD/F and metal levels (Vilavert et al. 2009, 2012). Because high levels of metals in soils were found in the sampling of 2010 (especially for nickel), we considered to reestablish the same 24 sampling points, already included in our previous campaigns, for the next surveys of metals (not for PCDD/Fs) in vegetation and soil samples. The increase in the number of sampling points provides a better discerning of the real effect on metal concentrations evaluated around the facility.

In 2012, a new pluriannual campaign (2012-2014) was started to monitor environmental pollution, analyzing heavy metals in soil and vegetation samples. Table 1 shows a summary of the different sampling campaigns, environmental matrices and chemical pollutants whose levels have been periodically analyzed. Moreover, the method used to collect air samples, by means of either active or passive samplers, is also specified. The objectives of the survey were the following: a) to determine the current environmental status in the vicinity of the MSWI by analyzing metal and PCDD/F levels in soil, vegetation, and air samples collected in 2013 and 2014, b) to determine the temporal trends of metals and PCDD/Fs by comparing the data of the current study with those from previous surveys, and c) to assess the human exposure to these pollutants, as well as to characterize the associated non-carcinogenic and carcinogenic risks, for the population living in the neighborhood of the facility.

Materials and Methods

Sample Collection

In May/June of 2013, 24 vegetation samples and 8 air samples were collected for metal analyses in the vicinity of the MSWI of Tarragona, while 8 vegetation and 8 air samples were collected for PCDD/F analyses. Two hundred grams of herbage (*Piptatherum L.*) were obtained by cutting the plant at 4 cm above the ground. In turn, two high-volume active sampling devices (TE-1000-PUF and TE-6070-DV samplers, Tisch Environmental, Cleves, OH, USA, for the determination of PCDD/Fs and trace elements, respectively) were used to collect ambient air samples. Sampling volumes were within the range 402–622 $m^3$for PCDD/Fs, and 1827 $m^3$for metals. The sampling sites were the same as those corresponding to the baseline and our subsequent surveys (Schuhmacher et al. 1996, 1997, 1998; Vilavert et al. 2009, 2010, 2012). Sampling points were located at different wind directions (NE, NW, SE and SW), and at various distances (250, 500, 750, 1000, 1250, and 1500 m) from the facility (Fig. 1). In June of 2014, 24 soil samples were again collected for metal analyses at the same sampling points. Moreover, 8 soil and 8 air samples were also collected for PCDD/F analysis. Soil samples were taken from the upper 3 cm, stored in polyethylene bags and, once in the laboratory, sieved through a 2-mm mesch screen. In turn, PUF passive air samplers (PacWill Environmental, Stoney Creek, ON, Canada) were deployed for 3 months. Applying a sampling rate of 2 m^3/day (Mari et al. 2008), the total air volume was estimated in 182 m^3. Average temperatures during the sampling periods were 17.6°C and 21.1°C in May/June 2013 and in June 2014, respectively, while the mean relative humidity was 68%, in 2013, and 69%, in 2014. Finally, the accumulated rainfall during the sampling was 72.8 and 43.9 mm in 2013 and 2014, respectively.

Analytical Procedure

The concentrations of arsenic (As), beryllium (Be), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), antimony (Sb), thallium (Tl), and vanadium (V) in samples of soil, vegetation, and air filters were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS) (ICP-MS Perkin Elmer Elan 6000), using rhodium (Rh) as internal standard. Metal analytical methods were previously described (Rovira et al. 2011, 2014a). In brief, approximately

0.5 g of soil and vegetation samples were digested with 5 mL of nitric acid (65% Suprapur, E Merck, Darmstadt, Germany) in hermetic Teflon bombs by means of a Milestone Start D microwave digestion system until raising 165ºC in 10 min, and keeping this temperature for 20 min. Once the solutions were cooled, they were filtered and made up to 25 mL with deionized water and kept frozen at -20ºC until the subsequent analysis. For air samples, one-eighth (50.2 cm^2) of the quartz fibber filter was treated with 2 mL of 65% nitric acid (Suprapur, E Merck, Darmstadt, Germany) and 3 mL of hydrofluoric acid (37% Panreac SA, Castellar del Vallès, Spain) in Teflon bombs for 8 h at room temperature followed by additional 8 h at 80ºC. The accuracy of the instrumental methods was done by analyzing duplicate and blank samples. Quality control was assessed by analyzing Loamy clay soil (National Institute of Standards and Technology, USA) for soils and air, and Trace element in spinach leaves (National Institute of Standards and Technology, USA) for vegetation.

The concentrations of PCDD/Fs in soil and vegetation samples were determined by means of high resolutions gas chromatography coupled to high resolution mass spectrometry (HRGC/HRMS), according to the US EPA 1613 method (US EPA 1994). In turn, PCDD/F levels in air were also determined by HRGC/HRMS, following the German VDI 3499 method (German VDI 2003). The ranges of the recovery percentages were 55-96%, 7-110%, and 47-96 for soil, vegetation, and air, respectively.

Human Health Risks

Human exposure to metals and PCDD/Fs was evaluated by applying the US EPA Risk Assessment Guidance for Superfund (RAGS) Methodology (US EPA 1989, 2012, 2014). The following three exposure routes were considered: soil ingestion, dermal contact, and air inhalation (Domingo et al. 2002). The numerical equations, as well as the values of the parameters used to calculate human exposure and to characterize health risks, were previously reported (RAIS 2013; Vilavert et al. 2012; 2014). The characterization of non-carcinogenic risks involves the calculation of the Hazard Quotient (HQ), which is defined as the relation between the predicted exposure and the reference dose (RfD). On the other hand, carcinogenic risks (CR) were estimated by multiplying the predicted exposure by the respective slope factor (SF). For dermal absorption estimations, the dermal RfD was calculated as the multiplication of the

respective oral reference dose by the gastrointestinal absorption factor, whereas dermal SFs were obtained by dividing the respective oral factor by the same gastrointestinal absorption factor (US EPA 1989).

Statistics

For undetected pollutants, concentrations were considered to be equal to one-half of the respective limit of detection (ND=1/2 LOD). Data analysis was carried out by means of the statistical software package SPSS 19.0. Significant differences ($p<0.05$) were analysed by applying the Levene test, being followed by ANOVA or Mann-Whitney's U-test, depending on whether data followed (or not) a parametric distribution.

Results and Discussion

Environmental Concentrations

Metal concentrations in soil, vegetation, and air samples collected in the vicinity of the MSWI of Tarragona are summarized in Table 2. Regarding the soil samples, higher levels were observed with respect to the background (1999) (Llobet et al. 2002) and with previous (2012) (data also presented here) surveys. It must be noted that we chose as initial (or background) study, the survey performed in 1999, just after the installation of modern pollution control devices (Domingo et al. 2001b). In comparison with that study (1999), only a decrease in As levels was noted. For other metals, there were increases of different degree, being significant for Be ($p<0.05$), Cr ($p<0.001$), Tl ($p<0.01$), and V ($p<0.001$). On the other hand, comparing the current results with those obtained in 2012 (data also presented here), an increase was noted for most metals, being significant for Ni and Tl ($p<0.01$), but not for the slightly high levels observed for Co, Cr, Cu, Mn, Pb, Sb and V. In contrast, As, Be and Cd presented non-significant slight decreases ($p>0.05$).

In general terms, metal levels in vegetation samples presented variations depending on the specific metal analysed. With respect to our initial study (1999) (Llobet et al. 2002), there has been a significant decrease in As and Pb concentrations ($p<0.001$). Furthermore, we also observed a significant increase in the levels of Cr ($p<0.01$). The remaining metals showed slight increases or decreases, none of them being statistically

significant ($p>0.05$). When comparing the current results with those obtained one year before (2013) (data also presented here), reductions of the levels of Cd, Pb, and Sb were observed. However, the differences were not significant. For other metals, heterogeneous increases were found, being significant for Ni ($p<0.05$). In both matrices, soils and vegetation, Mn was the metal presenting higher concentrations, with mean values of 236 and 40.0 µg/g, respectively. On the other hand, Hg was the element showing the lowest levels, although in the present study (2014), it was found above its limit of detection in both matrices. In the previous study, conducted in 2013, the different elements showed similar levels in air samples to those found in previous monitoring campaigns, although some variations could be observed. Comparing with the study performed in 2009 (data also presented here) a decrease was noted for Be, Mn, Ni, Sb, Tl and V, being only statistically significant for the latter element ($p<0.05$). In contrast, there has been a significant increase of Co values ($p<0.05$). The remaining elements (As, Cd, Cu, Cr, Hg and Pb) have shown no significant increases. Comparing to the 2007 study (Vilavert et al. 2009), there has been a reduction in the concentration of most metals, being significant for Ni and V ($p<0.01$). However, the levels of Mn and Pb in air increased significantly between 2007 and 2013 ($p<0.05$). In air, the element showing the highest concentration was Cu, followed by Mn, while Hg presented the lowest levels and Tl was found below its detection limit in all samples.

The mean concentrations of PCDD/Fs in soil, vegetation and air samples collected in 2013 and 2014 in the vicinity of the MSWI are summarized in Table 3. The mean levels determined in our initial/baseline (1999) and in our previous studies are also shown. The concentrations of PCDD/Fs measured in 8 soil samples varied between 0.16 and 1.69 ng I-TEQ/kg dry weight (dw), with a mean value of 0.63 ng I-TEQ/kg (dw). This means a non-significant decrease ($p>0.05$) of 47% with respect to the baseline study (1999) and a slight increase of 9% in relation to our previous study (2010) (Vilavert et al. 2012). There was a certain relationship between the distance to the MSWI and the levels of PCDD/Fs in soils, as the two points closest to the facility presented the highest levels. Nonetheless, both are very close to sampling points strongly impacted by the heavy traffic in the area. Furthermore, a similar increase was not observed in the other two monitors, vegetation and air.

The concentrations of PCDD/Fs in samples of vegetation collected in 2013, ranged from 0.04 to 0.10 ng I-TEQ/kg (dw), with a mean value of 0.07 ng I-TEQ/kg (dw). This

means a slight reduction compared with the average value obtained in our background study (1999), and a slight growth compared with the mean concentration obtained in the earlier study (2009) (Vilavert et al. 2012). However, the difference was not statistically significant. In contrast to soils, there was no significant correlation between PCDD/F concentrations in vegetation and the distance to the MSWI.

Finally, the levels of PCDD/Fs in air, obtained by means passive samplers, were in 2014 between 6.10 and 16.8 fg WHO-TEQ/m^3, with a mean value of 10.1 fg WHO-TEQ/m^3. In the last study (2010) in which air samples were analysed using passive samplers (Vilavert et al. 2012), PCDD/F concentrations ranged between 6.95 and 22.3 fg WHO-TEQ/m^3, showing an average level of 10.5 fg WHO-TEQ/m^3. In the 2013 study (data also presented here), when the air samples to determine the concentrations of PCDD/Fs were collected by active sampling, a range between 1.88 fg WHO-TEQ/m^3 and 5.29 fg WHO-TEQ/m^3, with an arithmetic mean of 3.56 fg WHO-TEQ/m^3 was found. Although passive samplers are a good system for assessing PCDD/Fs and other persistent organic pollutants, the outcomes are not entirely comparable to those obtained by active sampling devices.

The PCDD/F congener profiles in soil, vegetation and air samples collected in the vicinity of the MSWI between 1999 and 2014 are depicted in Fig. 2. In the three environmental matrices, the most predominant congeners were OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,4,6,7,8-HpCDF. In contrast, 2,3,7,8-TCDD was the congener with the lowest contribution to the total amount of PCDD/Fs. In all cases, a predominance of the heaviest congeners could be noted. In both soil and vegetation, PCDD/F congener profiles were similar irrespective of the sampling year: 1999, 2013 and 2014, in soils, and 1999, 2009 and 2013, in vegetation (Vilavert et al. 2012). In general terms, levels of the PCDD/F congeners in vegetation have decreased compared to previous studies, although some specific congeners have increased with respect to the survey carried out in 2009. Regarding air samples, PCDD/F profiles obtained in the campaigns of 2010 and 2014, both based on passive samplers, are very similar. It indicates that not only the concentrations of PCDD/Fs are very similar, but also that no changes in terms of congener profiles occurred.

The results of some recent studies on PCDD/F levels in air and soil samples are summarized in Tables 4 and 5, respectively. In general terms, the current levels of PCDD/Fs in soils were lower than those found in studies conducted near other MSWIs.

Thus, Chen et al. (2011) reported a range of concentrations between 0.85 and 4.50 pg TEQ/g in the vicinity of a MSWI in Taiwan, while in Shanghai (China), the levels ranged between 0.64 and 64.15 pg I-TEQ/g in the surroundings of another MSWI (Deng et al. 2011). In a survey performed in the vicinity of an Italian MSWI situated in a suburban area near the Adriatic coast, Vassura et al. (2011) found PCDD/F concentrations ranging from 58 to 164 ng TEQ/kg. The levels of our current study were also lower than those reported by Colombo et al. (2014) in the Maldives, where soil samples from two areas with different levels of urbanization (one developed area on Malé Atoll and one undeveloped area on Faafu Atoll) were collected. The authors found concentrations ranging from 0.01 to 49.3 pg WHO_{2005}-TEQ/g. In Turkey, Turgut et al. (2012) reported PCDD/F levels between 0.19 and 1.05 pg WHO-TEQ/g in a forested zone.

With respect to PCDD/Fs in air, the current concentrations were similar, or even lower, than those found in the scientific literature in sampling sites presenting similar characteristics. For example, Rovira et al. (2010) reported a range of concentrations from 0.008 to 0.015 pg WHO-TEQ/m^3, in the neighborhood of the MSWI of Mataró (Catalonia, Spain), being the levels found very similar to those obtained in the current study. In another survey conducted in Sant Vicens dels Horts and Pallejà (Catalonia, Spain), we evaluated the use of traditional and alternative fuel in a cement plant. Mean levels of 0.009 and 0.018 pg WHO-TEQ/m^3, respectively, were found (Rovira et al. 2014b). Bogdal et al. (2014) carried out two sampling campaigns in Zurich (Switzerland), located in a large courtyard, in the city center. A median level of 62 fg WHO_{98}-TEQ/m^3 was obtained, which is higher than the levels found in the present survey. The current concentrations in air were even lower than those observed in remote and rural areas of USA, where Lorber et al. (2013) reported an average value of 10.4 ± 33.2 fg WHO-TEQ/m^3.

The comparison of concentrations of PCDD/Fs in different kinds of vegetation is more complex due to the intrinsic characteristics of each species. However, the current levels are similar, or lower, than those reported in other studies carried out in Catalonia and other countries, where the same species were analysed. Investigations conducted around various cement plants located in Catalonia (Spain), in which the same kind of vegetation (*Piptatherum paradoxum* L.) was analysed, reported levels of PCDD/Fs within the same order of magnitude as those here obtained (Rovira et al. 2014a,b). On

the other hand, Nadal et al. (2009) studied the concentrations of PCDD/Fs in one of the most important chemical/petrochemical complex in Southern Europe, which is located in Tarragona County (Catalonia, Spain). The results showed mean concentrations between 0.23 and 0.58 ng WHO-TEQ/kg. In turn, Åberg et al. (2010) analysed grass samples near a former saw mill of northern Sweden, ranging the concentrations between 0.26 and 0.61 pg WHO-TEQ/m^3. In the Setubal Peninsula, which is one of the most industrialized and densely populated areas of Portugal, Augusto et al. (2007) determined PCDD/F levels in lichens, obtaining a mean value of 8.8 ng I-TEQ/g, while in an industrial zone located around Dongting lake (China), Fang et al. (2008) reported mean concentrations of 0.64 pg I-TEQ/g in *Artemisia selengensis* and *Polygonum orientale L*. species.

Human Health Risks

Data regarding environmental exposure to metals and PCDD/Fs for the population living near the MSWI are shown in Table 6. We considered three different scenarios: (a) all sampling points, (b) sampling points closer to the facility from 250 to 750 m, and (c) sampling points located at 1000-1500 m from the plant. Manganese presented the maximum exposure for the three scenarios established, followed by Pb in scenarios A and C, and for Cu in the scenario B. For most metals, the exposure pathway with the highest percentage of contribution was soil ingestion, followed by dermal contact. In contrast, the minority via was air inhalation, with the only exception of Sb. Arsenic was the only element for which the dermal contact was the main route of exposure, followed closely by soil ingestion. The PCDD/F profile was found to be quite different with respect to those for metals, being inhalation the predominant route, with 60% of total exposure. The other two routes, soil ingestion and dermal contact, contributed almost equally, with approximately 20% each one.

Health risks associated to environmental exposure to metals and PCDD/Fs were also characterized (Figs. 3 and 4). Non-carcinogenic risks were assessed by comparing metals and PCDD/F exposure with the RfD. The Hazard Quotient (HQ) for all metals and PCDD/Fs did not exceed (in any case) the unity, which is considered the safety threshold. Manganese, followed by As, were the elements showing the maximum HQ, while PCDD/Fs had a HQ value of $2.15 \cdot 10^{-3}$. With respect to cancer risks, Cr was the

metal showing the highest value, with a risk index of $1.73 \cdot 10^{-5}$, followed by As with a value of $1.14 \cdot 10^{-5}$. Both, Cr and As, exceeded the limit value set by the Spanish legislation at 10^{-5}. Notwithstanding, this could be a consequence of a risk overestimation related to metal speciation. We considered that 1/6 of the total Cr was Cr-VI, which is the carcinogenic form (US EPA, 1998). Thus, we could have overestimated the exposure and the associated health risks. Similarly, we only analysed the levels of As total, assuming to be present as inorganic As, which is the carcinogenic species.

Conclusions

The main interest of the current study is probably the comparison of results belonging to a very long (20 years, approximately) environmental monitoring program, involving analyses of PCDD/Fs and heavy metals in environmental samples collected near the MSWI of Tarragona. The introduction of air samples since 2007 allows an integrated diagnosis of the impact of the facility, considering different environmental compartments.

Despite the slight increase in the concentration of metals and PCDD/Fs in soil near the MSWI, the lack of a similar pattern in vegetation and air samples (used as short-term and immediate environmental monitors, respectively), reflects the low impact of the facility on the neighborhood. Human exposure to PCDD/Fs and metals and the associated non-carcinogenic and carcinogenic risks have shown that the emissions of these pollutants by the MSWI do not generate additional health risks for the populations living nearby.

Based on the above results, it can be concluded that, to date, the environmental impact of the MSWI of Tarragona is not significant in terms of PCDD/Fs and heavy metals. Furthermore, the current environmental levels of these pollutants are comparatively low compared to those observed in other areas impacted by various urban and industrial sources.

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Table 1 Structure of the environmental monitoring program of PCDD/Fs and metals in the period 1996-2014.

Metals			PCDD/Fs		
Year	**Matrix**	**Method used to collect air samples**	**Year**	**Matrix**	**Method used to collect air samples**
1996	Vegetation and Soil		1996	Vegetation and Soil	
1997	Vegetation and Soil		1997	Vegetation and Soil	
1999	Vegetation and Soil		1999	Vegetation and Soil	
2002	Vegetation		2002	Vegetation	
2003	Soil		2003	Soil	
2004	Vegetation		2004	Vegetation	
2005	Soil		2005	Soil	
2006	Soil		2006	Soil	
2007	Vegetation and Air	Active samplers	2007	Vegetation and Air	Active samplers
2008	Soil		2008	Soil and Air	Passive samplers
2009	Vegetation and Air	Active samplers	2009	Vegetation and Air	Active samplers
2010	Soil		2010	Soil and Air	Passive samplers
2011	Soil		-	-	
2012	Vegetation and Soil		-	-	
2013	Vegetation and Air	Active samplers	2013	Vegetation and Air	Active samplers
2014	Vegetation and Soil		2014	Vegetation and Soil	Passive samplers

Table 2 Metal concentrations in soil, vegetation and air samples collected near the MSWI of Tarragona (Catalonia, Spain). Temporal trends

	Years				
Soil[a]	**1999**	**2012**	**2014**	**%1999-2014**	**%2012-2014**
As	5.56 ± 3.45	5.90 ± 2.05	5.14 ± 1.54	-7	-13
Be	0.33 ± 0.12	0.43 ± 0.15	0.42 ± 0.17	27*	-1
Cd	0.15 ± 0.06	0.19 ± 0.11	0.18 ± 0.06	17	-9
Co	NA	3.88 ± 1.19	4.10 ± 1.42	-	6
Cr	11.3 ± 4.25	15.5 ± 4.75	17.8 ± 6.12	58***	15
Cu	NA	24.0 ± 15.8	28.2 ± 24.2	-	18
Hg	0.06 ± 0.02	ND	0.06 ± 0.04	0	-
Mn	224 ± 72.1	229 ± 59.0	236 ± 122	5	3
Ni	8.75 ± 2.87	7.39 ± 2.74	9.16 ± 2.93	5	24**
Pb	25.7 ± 21.4	34.2 ± 24.9	39.5 ± 30.0	54	16
Sb	NA	0.12 ± 0.13	0.16 ± 0.13	-	40
Tl	0.06 ± 0.02	0.16 ± 0.04	0.43 ± 0.17	625**	165**
V	16.0 ± 4.20	24.6 ± 6.41	24.7 ± 10.7	54***	1
Vegetation[a]	**1999**	**2013**	**2014**	**%1999-2014**	**%2013-2014**
As	0.13 ± 0.01	0.05 ± 0.02	0.06 ± 0.03	-56***	7
Be	ND	ND	ND	-	-
Cd	0.03 ± 0.02	0.03 ± 0.06	0.03 ± 0.03	-4	-15
Co	NA	0.08 ± 0.05	0.07 ± 0.04	-	30
Cr	0.27 ± 0.15	0.59 ± 0.24	0.78 ± 0.58	187**	31
Cu	NA	7.84 ± 2.59	9.27 ± 2.57	-	18
Hg	ND	ND	0.01 ± 0.006	-	-
Mn	36.2 ± 1.32	35.2 ± 12.9	40.0 ± 24.0	12	15
Ni	1.02 ± 0.65	0.45 ± 0.67	1.25 ± 1.10	22	177**
Pb	1.22 ± 0.92	0.23 ± 0.16	0.20 ± 0.19	-83***	-9
Sb	NA	0.06 ± 0.03	0.05 ± 0.05	-	-20
Tl	ND	ND	ND	-	-
V	0.60 ± 0.34	0.14 ± 0.09	0.15 ± 0.07	-74	10
Air[b]	**2007**	**2009**	**2013**	**%2007-2013**	**%2009-2013**
As	0.49 ± 0.22	0.36 ± 0.18	0.42 ± 0.13	-15	17
Be	0.07 ± 0.04	0.06 ± 0.04	0.02 ± 0.02	-67	-63
Cd	0.11 ± 0.07	0.08 ± 0.02	0.11 ± 0.07	-4	39
Co	0.26 ± 0.12	0.06 ± 0.03	0.22 ± 0.08	-17	262***
Cr	5.62 ± 4.35	ND	2.83 ± 5.32	-50	-
Cu	69.6 ± 25.3	46.3 ± 27.6	56.6 ± 25.4	-19	22
Hg	ND	ND	0.03 ± 0.04	-	-
Mn	4.18 ± 1.17	10.9 ± 5.12	9.00 ± 4.39	115**	-17
Ni	6.63 ± 2.67	4.14 ± 1.71	3.51 ± 2.24	-47**	-15
Pb	2.30 ± 2.53	4.40 ± 1.27	5.38 ± 1.97	134***	22
Sb	ND	1.59 ± 0.33	1.33 ± 0.63	-	-16
Tl	0.02 ± 0.01	0.03 ± 0.03	ND	-	-
V	12.2 ± 4.19	8.14 ± 3.12	3.65 ± 2.99	-70***	-55*

[a]Units in soil and vegetation: μg/g
[b]Units in air: ng/m^3
Asterisks indicate significant differences at *p <0.05, **p <0.01, and ***p <0.001.
ND: Not detected
NA: Not analysed

Table 3 Levels of PCDD/Fs in samples of soil, vegetation and air collected in the vicinity of the MSWI of Tarragona (Catalonia, Spain). Temporal variations

				%Variation	
Soil	**1999**	**2010**	**2014**	**1999-2014**	**2010-2014**
ng I-TEQ/kg	1.20	0.58	0.63	-47	9
Vegetation	**1999**	**2009**	**2013**	**1999-2013**	**2009-2013**
ng I-TEQ/kg	0.11	0.06	0.07	-35	12
Air	**2010**	**2013**	**2014**	**2010-2014**	**2013-2014**
fg WHO-TEQ/m^3	10.5	3.56	10.1	-3	184

Table 4 Summary of PCDD/F concentrations in soil samples from recent studies. Data obtained from the scientific literature

Location	Assessed area	Mean (Min-Max)	Units	References
Tarragona (previous study)	MSWI	0.58	ng I-TEQ/kg	Vilavert et al. 2012
Taiwan	MSWI	2.00±0.97 (0.85-4.50)	pg-TEQ/g	Chen et al. 2011
The Republic of Maldives	Developed and undeveloped area	(0.01-49.3)	pg WHO_{2005}-TEQ/g	Colombo et al. 2014
Shanghai, China	MSWIs	(0.64-61.15)	pg I-TEQ/g	Deng et al. 2011
Zhejiang Province, China	HWI (2007)	1.09	pg I-TEQ/g	Li et al. 2012
	HWI (2010)	2.54	pg I-TEQ/g	
Tianjin, China	MSWI	1.08 (0.47-2.07)	pg I-TEQ/g	Liu et al. 2013
Tarragona County, Spain	Chemical	0.71±0.83	ng WHO-TEQ/kg	Nadal et al. 2009
	Petrochemical	0.76±0.47	ng WHO-TEQ/kg	
	Residential	1.10±0.78	ng WHO-TEQ/kg	
	Unpolluted	0.23±0.03	ng WHO-TEQ/kg	
Mataró, Catalonia, Spain	MSWI	0.14-0.46	ng WHO-TEQ/kg	Rovira et al. 2010
Moscow, Russia	Urban	8.2 (0.27-16.1)	ng WHO-TEQ/kg	Shelepchikov et al. 2011
Japan	MSWI	390-8800	pg TEQ/g	Takeda and Takaoka 2013
Taurus Mountains, Turkey	forest	0.19-1.05	pg WHO-TEQ/g	Turgut et al. 2012
Italy	suburban area, MSWI	58-164	ng TEQ/kg	Vassura et al. 2011
Beijing, China	mountain soil	0.29 (0.086-0.59)	ng I-TEQ/kg	Li et al. 2014
	park soil	0.68 (0.39-0.88)	ng I-TEQ/kg	
Sant Vicens dels Horts and Pallejà, Spain	Cement plant, July 2012 (traditional fuel)	0.37±0.33	ng WHO-TEQ/kg	Rovira et al. 2014b
	Cement plant, June 2013 (alternative fuel)	0.41±0.29	ng WHO-TEQ/kg	
Sta. Margarida i els Monjos, Spain	Cement plant	0.2	ng WHO-TEQ/kg	Rovira et al. 2014a

Table 5 Summary of PCDD/F concentrations in air samples from recent studies. Data obtained from the scientific literature

Location	Assessed area	Mean (Min-Max)	Units	References
Tarragona (previous study)	MSWI	10.5	fg WHO-TEQ/m^3	Vilavert et al. 2012
Taiwan	MSWI	0.0500±0.0163 (0.039-0.088)	pg-TEQ/m^3	Chen et al. 2011
Mataró, Spain	MSWI	(0.008-0.015)	pg WHO-TEQ/m^3	Rovira et al. 2010
Southern Taiwan	MSWI	59.6 (33.5-105)	fg WHO-TEQ/m^3	Wang et al. 2010
Sant Vicens dels Horts and Pallejà, Spain	Cement plant, July 2011 (traditional fuel)	0.009±0.001	pg WHO-TEQ/m^3	Rovira et al. 2014b
	Cement plant, June 2013 (alternative fuel)	0.018±0.011	pg WHO-TEQ/m^3	
Zurich, Switzerland	Urban	62* (32-190)	fg WHO_{98}-TEQ/m^3	Bogdal et al. 2014
Sta. Margarida i els Monjos, Spain	Cement plant	7	fg WHO-TEQ/m^3	Rovira et al. 2014a
Pacific Ocean, near southern Taiwan and the northern Philippines	Oceanic atmosphere	0.00438	pg WHO_{2005}-TEQ/m^3	Chao et al. 2014
	Ambient air over the land	0.0113	pg WHO_{2005}-TEQ/m^4	
China	Cement plant	$9.3\cdot10^{-3}$-$90.8\cdot10^{-3}$	ng I-TEQ/m^3	Chen et al. 2014a
Italy	Industrialized city	0.01-0.19	pg WHO_{2005}-TEQ/m^3	Colombo et al. 2013
Istanbul	Urban zone	123 (52-229)	fg I-TEQ/m^3	Gunes et al. 2014
China	Aluminum metallurgical facilities	0.61 (0.31-0.84)	pg TEQ/m^3	Hu et al. 2014
	Copper metallurgical facilities	5.26 (0.32-13.6)	pg TEQ/m^3	
Korea	Industrialized city	0.052	pg TEQ/m^3	Kim and Yoon 2014
United States	Remote and rural area	10.4±33.2	fg WHO_{2005}-TEQ/m^3	Lorber et al. 2013
Tropical and subtropical oceans	Remote area	(1-10)	fg I-TEQ/m^3	Morales et al. 2014
Trieste, Italy	Industrial, urban and unpolluted area	(5-38)	fg TEQ/m^3	Mosca et al. 2012
India	MSWI	(0.007-26814)	ng TEQ/Nm^3	Thacker et al. 2013
China	MSWI (running periods)	(0.156-1.44)	pg I-TEQ/m^3	Zhang et al. 2014
	MSWI (non-running periods)	(0.158-0.648)	pg I-TEQ/m^3	

*Median value

Table 6 Environmental exposure to metals and PCDD/Fs (mg/kg/day) for the adult population living near the MSWI of Tarragona (Catalonia, Spain)

		Scenario A		Scenario B		Scenario C	
	Pathways	**Exposure**	**Contribution (%)**	**Exposure**	**Contribution (%)**	**Exposure**	**Contribution (%)**
As	Soil ingestion	8.03E-06	48.1	8.76E-06	46.5	7.31E-06	46.5
	Dermal contact	8.56E-06	51.2	9.96E-06	52.8	8.31E-06	52.9
	Air inhalation	1.14E-07	0.7	1.34E-07	0.7	1.02E-07	0.6
Be	Soil ingestion	6.55E-07	95.6	7.25E-07	95.5	5.85E-07	95.6
	Dermal contact	2.33E-08	3.4	2.57E-08	3.4	2.08E-08	3.4
	Air inhalation	6.60E-09	1.0	8.61E-09	1.1	5.84E-09	1.0
Cd	Soil ingestion	2.75E-07	87.5	2.94E-07	86.5	2.56E-07	88.0
	Dermal contact	9.76E-09	3.1	1.04E-08	3.1	9.09E-09	3.1
	Air inhalation	2.94E-08	9.4	3.54E-08	10.4	2.58E-08	8.9
Co	Soil ingestion	6.41E-06	95.7	6.84E-06	95.5	5.97E-06	95.8
	Dermal contact	2.28E-07	3.4	2.43E-07	3.4	2.12E-07	3.4
	Air Inhalation	5.97E-08	0.9	7.57E-08	1.1	5.00E-08	0.8
Cr	Soil ingestion	2.78E-05	94.0	2.94E-05	93.3	2.63E-05	93.8
	Dermal contact	9.89E-07	3.4	1.04E-06	3.5	9.35E-07	3.4
	Air inhalation	7.75E-07	2.6	9.37E-07	3.2	7.97E-07	2.8
Cu	Soil ingestion	4.41E-05	72.1	6.17E-05	73.1	2.65E-05	66.3
	Dermal contact	1.57E-06	2.5	2.19E-06	2.6	9.41E-07	2.3
	Air inhalation	1.55E-05	25.4	2.05E-05	24.3	1.25E-05	31.4
Hg	Soil ingestion	1.18E-06	96.0	2.27E-06	96.3	7.81E-08	88.1
	Dermal contact	4.18E-08	3.4	8.07E-08	3.4	2.77E-09	3.1
	Air Inhalation	7.56E-09	0.6	6.85E-09	0.3	7.79E-09	8.8
Mn	Soil ingestion	3.38E-04	95.9	5.78E-04	96.1	3.15E-04	95.9
	Dermal contact	1.20E-05	3.5	2.05E-05	3.4	1.12E-05	3.4
	Air Inhalation	2.47E-06	0.6	3.03E-06	0.5	2.13E-06	0.7
Ni	Soil ingestion	1.43E-05	90.7	1.60E-05	89.0	1.26E-05	91.7
	Dermal contact	5.08E-07	3.2	5.69E-07	3.2	4.47E-07	3.3
	Air Inhalation	9.62E-07	6.1	1.41E-06	7.8	6.94E-07	5.0
Pb	Soil ingestion	6.17E-05	94.4	7.23E-05	94.1	5.11E-05	94.5
	Dermal contact	2.19E-06	3.5	2.57E-06	3.3	1.82E-06	3.4
	Air inhalation	1.47E-06	2.2	1.99E-06	2.6	1.16E-06	2.1
Sb	Soil ingestion	2.57E-07	40.7	3.34E-07	38.8	1.79E-07	38.9
	Dermal contact	9.11E-09	1.4	1.19E-08	1.4	6.38E-09	1.3
	Air Inhalation	3.65E-07	57.9	5.14E-07	59.8	2.76E-07	59.8
Tl	Soil ingestion	6.79E-07	96.4	6.65E-07	96.4	6.93E-07	96.4
	Dermal contact	2.41E-08	3.4	2.36E-08	3.4	2.46E-08	3.4
	Air inhalation	9.37E-10	0.1	9.37E-10	0.2	9.37E-10	0.2
V	Soil ingestion	3.86E-05	94.2	3.91E-05	93.2	3.80E-05	94.8
	Dermal contact	1.37E-06	3.3	1.39E-06	3.4	1.35E-06	3.4
	Air Inhalation	9.99E-07	2.5	1.45E-06	3.4	7.30E-07	1.8
PCDD/Fs	Soil ingestion	9.23E-13	19.8	1.68E-12	26.0	4.70E-13	13.0
	Dermal contact	9.84E-13	21.0	1.79E-12	27.7	5.01E-13	13.8
	Air inhalation	2.77E-12	59.2	2.98E-12	46.3	2.65E-12	73.2

A: all sampling points, B: sampling points closer to the facility (from 250 to 750 m), and C: sampling points located at 1000-1500 m from the MSWI.

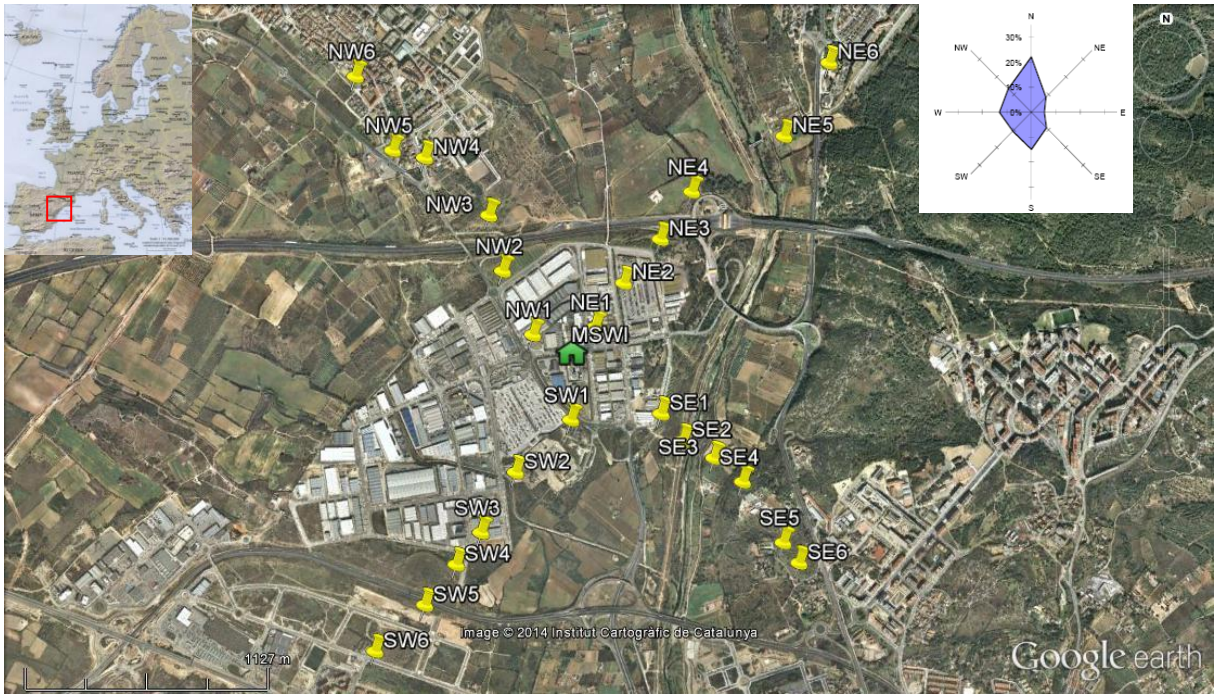

Fig. 1 Sampling sites. The wind rose indicates where the wind blows from

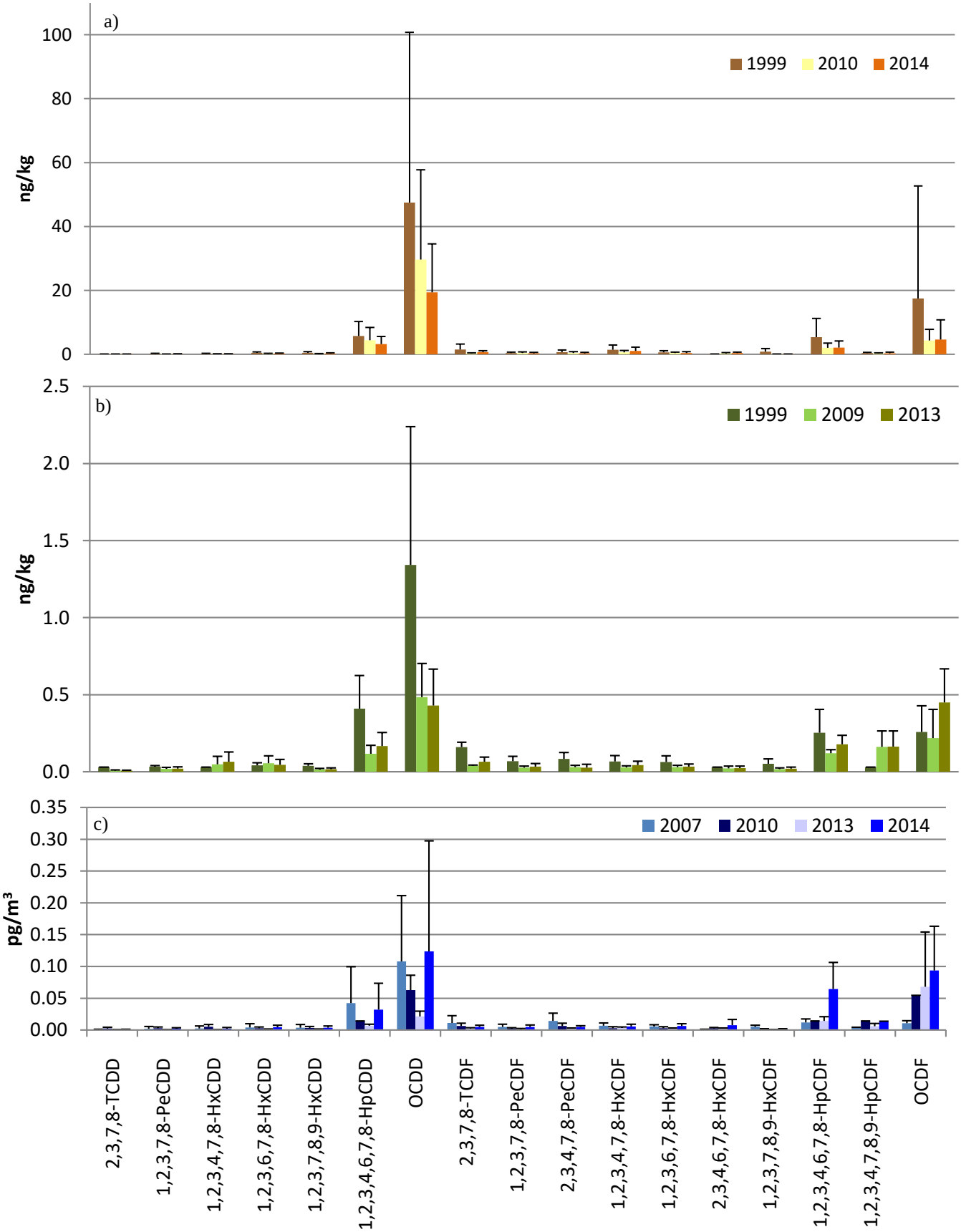

Fig. 2 PCDD/F congener profiles in (a) soil, (b) vegetation and (c) air samples collected in the vicinity of the MSWI of Tarragona (Catalonia, Spain)

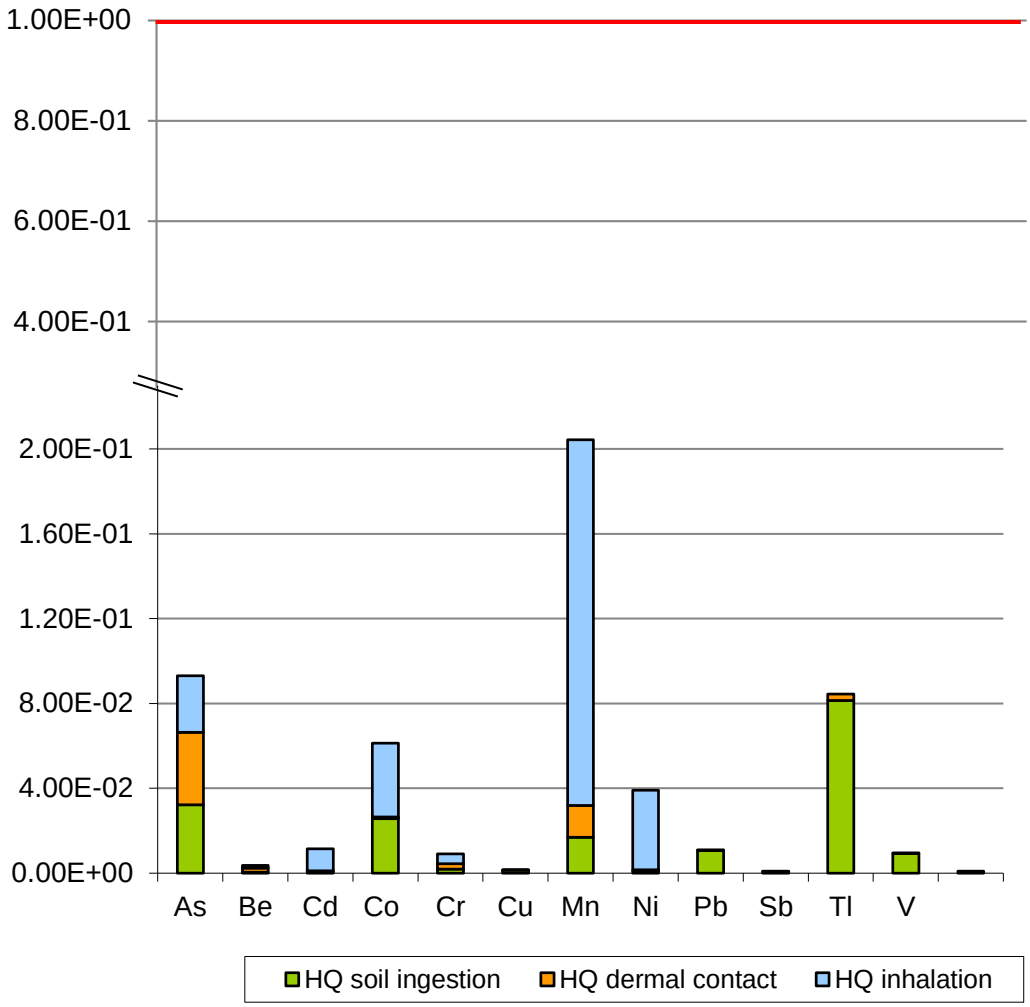

Fig. 3 Non-carcinogenic risks (Hazard Quotient) of environmental exposure to metals and PCDD/Fs in the surroundings of the MSWI of Tarragona (Catalonia, Spain).

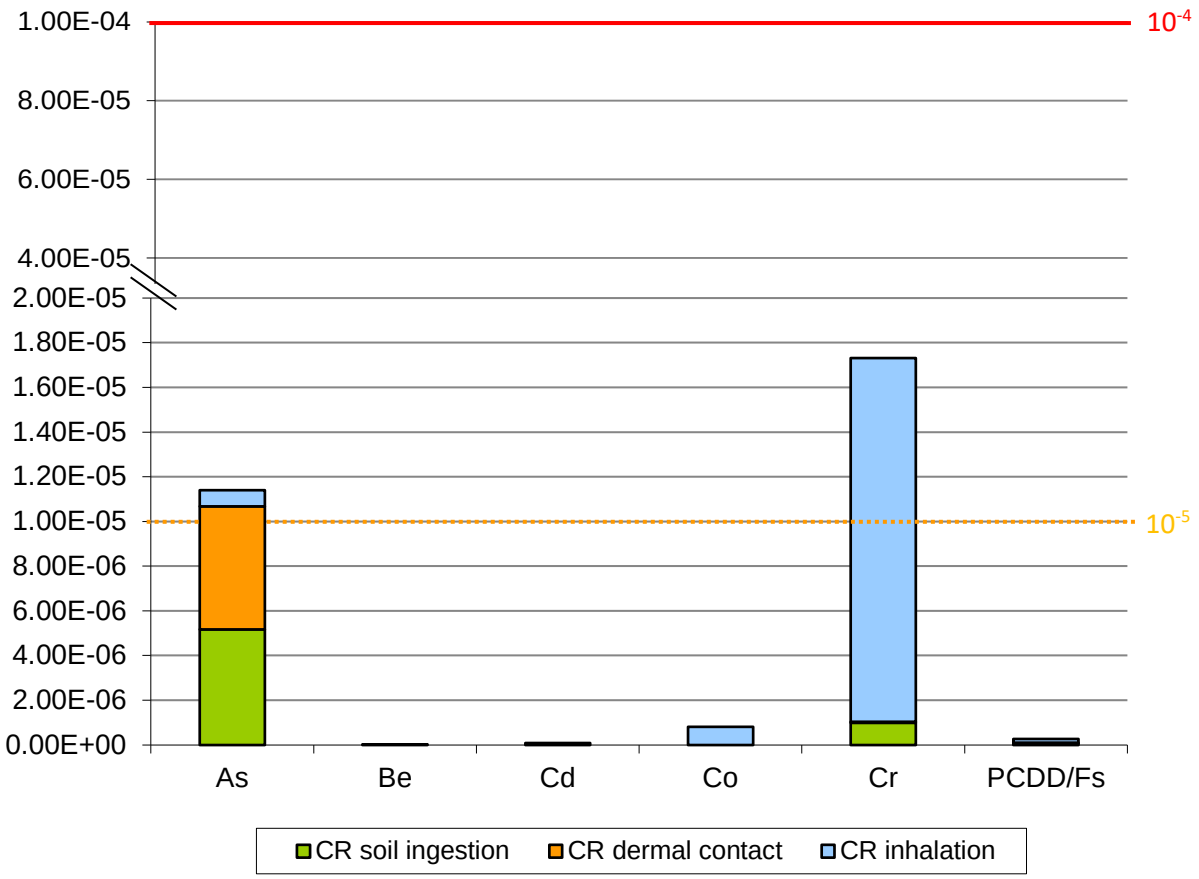

Fig. 4 Cancer risks (CRs) of environmental exposure to metals and PCDD/Fs near the MSWI of Tarragona (Catalonia, Spain).