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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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Abstract 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-mentioned pollutants have also been 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. Decreases 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 slight temporal decrease of the PCDD/F levels was also found with the remaining concentrations staying 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 Tarragona MSWI is not significant, in terms of PCDD/Fs and heavy metals, after >20 years of continuous operation.

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According to data of the European Commission (EC), people use 16 tons of material/person/y, 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 reuse, recycling, other recoveries, and disposal. Incineration is a well-known option for MSW treatment. In the Europe-27, approximately 20 % (in weight) of MSW is 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 decrease and energy recovery (Nadal et al. 2009). The EU sets a series of measures to prevent or decrease air, water, and soil pollution caused by incineration and coincineration of waste as well as the human health risks derived from such operations. These measures include the obligation to obtain permission for incineration or coincineration 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 shown 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 showed that at the 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 decrease 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 Tarragona MSWI (Catalonia, Spain) has been operating since 1991. Based on a wide surveillance program initiated approximately 20 years ago, concentrations of PCDD/Fs and heavy metals are 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- 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 compared with other potential sources of metals and PCDD/Fs in the same area. Taking into account the notable decrease 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 decrease 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 to determine 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 [Ni]), we considered reestablishing 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 discernment 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 by analyzing heavy metals in soil and vegetation samples. Table 1 lists 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, i.e., by either active or passive samplers, is also specified. The objectives of the survey were the following: (1) 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; (2) to determine the temporal trends of metals and PCDD/Fs by comparing the data of the current study with those from previous surveys; and (3) to assess the human exposure to these pollutants, as well as characterize the associated noncarcinogenic and carcinogenic risks (CRs), for the population living in the neighborhood of the facility.

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

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

Materials and Methods

Sample Collection

In May and June 2013, 24 vegetation samples and 8 air samples were collected for metal analyses in the vicinity of

the Tarragona MSWI, whereas 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. Mean sampling volumes were 522 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 (northeast, northwest, southeast, and southwest) and at various distances (250, 500, 750, 1000, 1250, and 1500 m) from the facility (Fig. 1). In June 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 mesh screen. In turn, polyurethane foam (PUF) passive air samplers (Pac-Will Environmental, Stoney Creek, Ontario, 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 to be 182 m^3. Average temperatures during the sampling periods were 17.6 °C and 21.1 °C in May/June 2013 and June 2014, respectively, whereas 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) (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 increasing 165 °C 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 −20 °C until 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) and 3 mL of hydrofluoric acid (37 % [Panreac SA,

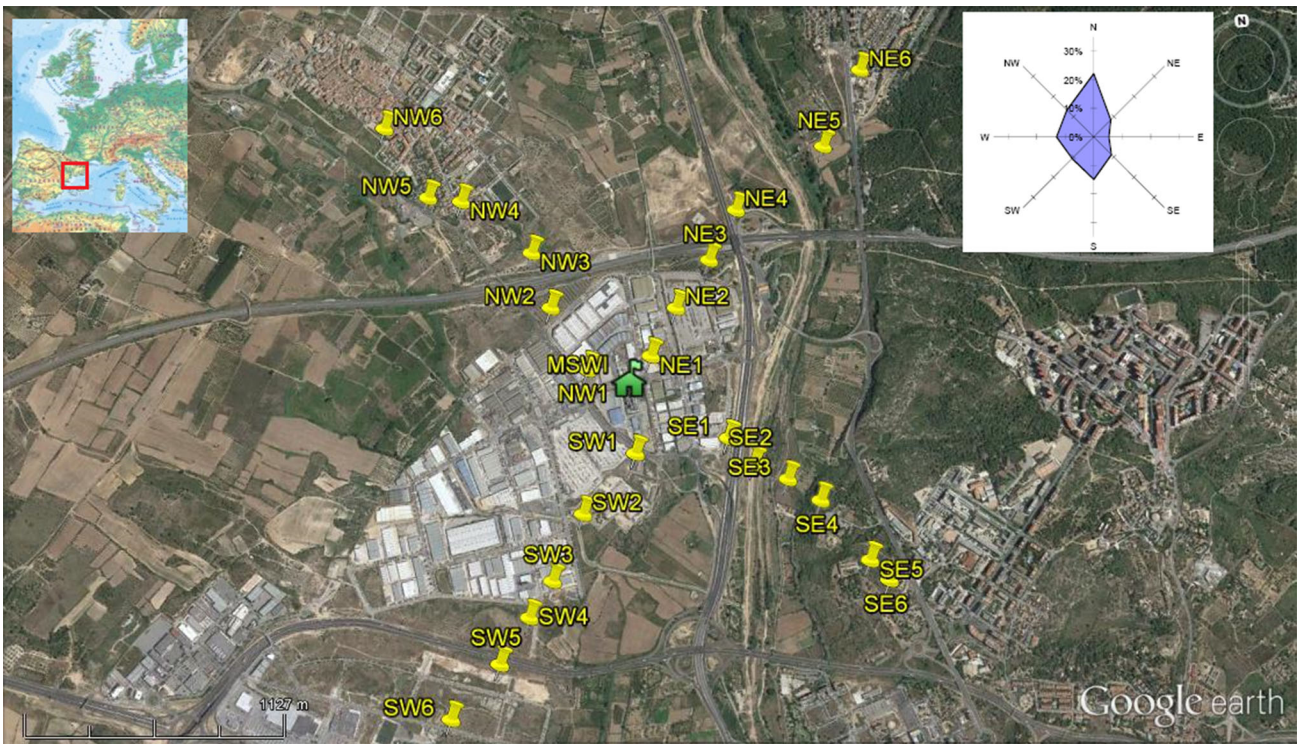

Fig. 1 Sampling sites. The wind rose indicates where the wind blows from

Castellar del Vallès, Spain]) in Teflon bombs for 8 h at room temperature followed by an additional 8 h at 80 °C. The accuracy of the instrumental methods was performed 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 elements 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-resolution gas chromatography coupled to high-resolution mass spectrometry (HRGC/HRMS) according to United States Environmental Protection Agency (USEPA) method 1613 (USEPA 1994). In turn, PCDD/F levels in air were also determined by HRGC/HRMS according to the German VDI method 3499 (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 USEPA Risk Assessment Guidance for Superfund Methodology (USEPA 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 (Risk Assessment Information System [available at http://rais.ornl.gov/]); Vilavert et al. 2012, 2014). The characterization of noncarcinogenic risks involves calculation of the hazard quotient (HQ), which is defined as the relation between the predicted exposure and the reference dose (RfD). In contrast, CRs 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 RfD by the gastrointestinal absorption factor, whereas dermal SFs were obtained by dividing the respective oral factor by the same gastrointestinal absorption factor (USEPA 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 performed by means of the statistical software package SPSS 19.0 (SPSS, Chicago, IL, USA). Significant differences ($p < 0.05$) were analysed by applying Levene test followed by analysis of variance or Mann–Whitney's U test depending on whether data followed (or did not follow) a parametric distribution.

Results and Discussion

Environmental Concentrations

Metal concentrations in soil, vegetation, and air samples collected in the vicinity of the Tarragona MSWI are listed in Table 2. Regarding the soil samples, greater levels were observed with respect to the background (1999) (Llobet et al. 2002) data and data from 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). Compared with that study (1999), only a decrease in As levels was noted. For other metals, there were increases of different degrees, being significant for Be ($p < 0.05$), Cr ($p < 0.001$), Tl ($p < 0.01$), and V ($p < 0.001$). In contrast, 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 higher levels observed for Co, Cr, Cu, Mn, Pb, Sb, and V. In contrast, As, Be, and Cd presented nonsignificant 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 was 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 with none of them being statistically significant ($p > 0.05$). When comparing the current results with those obtained 1 year before (2013) (data also presented here), decreases in 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 greater concentrations with mean values of 236 and 40.0 μg/g, respectively. In contrast, Hg was the element showing the lowest levels, although in the present study (2014), it was found greater than its LOD in both matrices. In the previous study (2013), the different elements showed similar levels in air samples to those found in previous monitoring campaigns, although some variations could be observed. Compared 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. Compared with the 2007 study (Vilavert et al. 2009), there has been a decrease in the concentration of most metals, being

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

Soil[a]	Years				
	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*[c]	−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]	Years				
	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]	Years				
	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*

ND not detected, *NA* not analysed

[a] Units in soil and vegetation are μg/g

[b] Units in air are ng/m^3

[c] Significant differences at * $p < 0.05$, ** $p < 0.01$, and *** $p < 0.001$

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

Soil (ng I-TEQ/kg)	Years			Variation (%)	
	1999	2010	2014	1999–2014	2010–2014
	1.20	0.58	0.63	−47	9
Vegetation (ng I-TEQ/kg)	Years				
	1999	2009	2013	1999–2013	2009–2013
	0.11	0.06	0.07	−35	12
Air (fg WHO-TEQ/m^3)	Years				
	2010[a]	2013[b]	2014[a]	2010–2014	2013–2014
	10.5	3.56	10.1	−3	184

By means of [a]passive or [b]active sampling means

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, whereas Hg presented the lowest levels, and Tl was found to be lower than its LOD 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 listed in Table 3. The mean levels determined in our initial/baseline study (1999) and in our previous studies are also shown. The concentrations of PCDD/Fs measured in eight 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 nonsignificant 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 because the two points closest to the facility presented the highest levels. Nonetheless, both are close to sampling points that are strongly affected by the heavy traffic in the area. Furthermore, a similar increase was not observed in the other two monitors, i.e., 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 decrease 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 distance to the MSWI.

Finally, the levels of PCDD/Fs in air, obtained by means passive samplers, in 2014 were 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 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 with 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 with previous studies, although some specific congeners have increased with respect to the survey performed 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 are the concentrations of PCDD/Fs very similar but also that no changes occurred in terms of congener profiles.

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

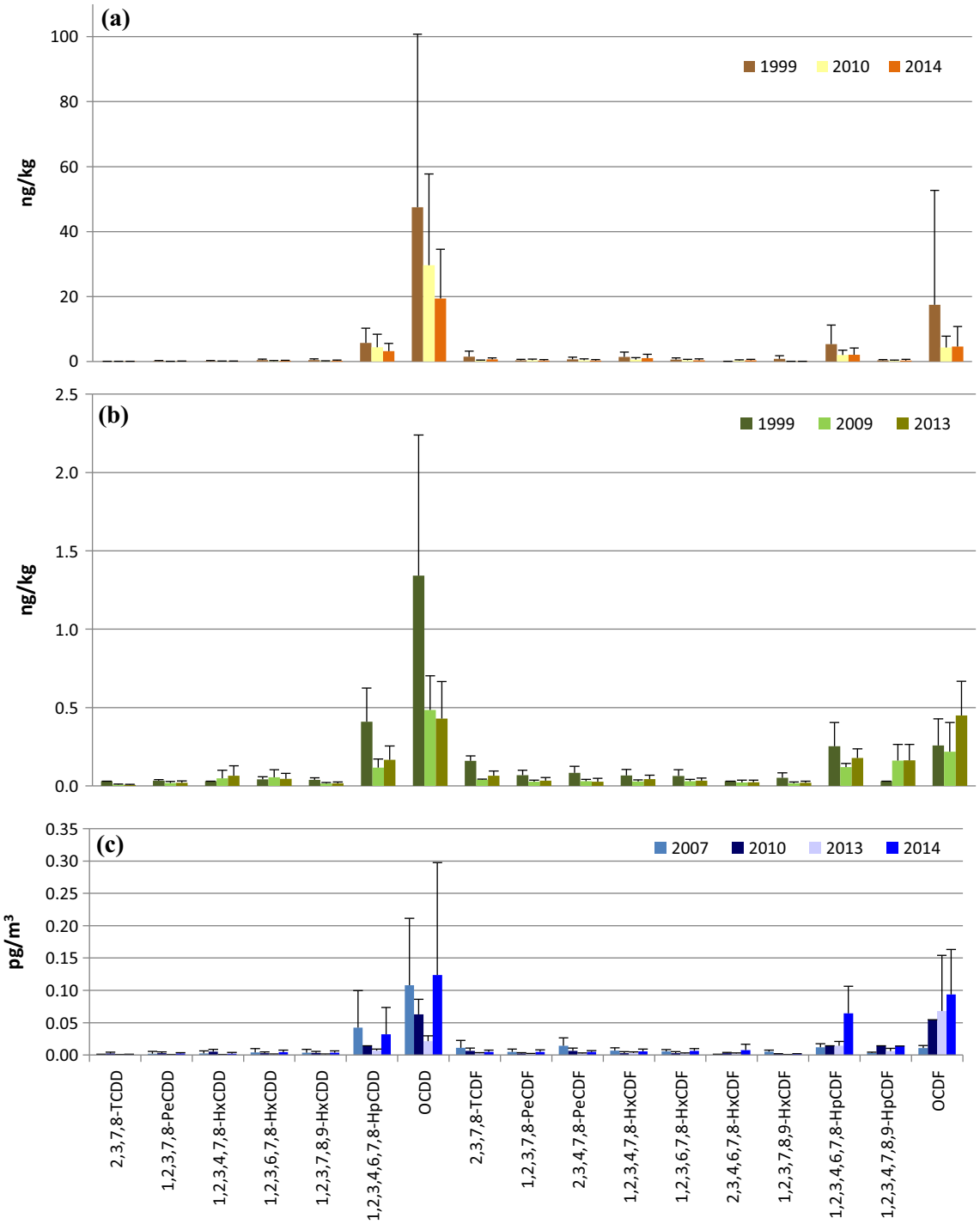

Fig. 2 PCDD/F congener profiles in **a** soil, **b** vegetation, and **c** air samples collected in the vicinity of the Tarragona MSWI (Catalonia, Spain)

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, whereas 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 investigators found

Table 4 Summary of PCDD/F concentrations in soil samples from recent studies

Location	Assessed area	Mean (minimum–maximum)	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 areas	(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	Hospital waste incinerator (2007)	1.09	pg I-TEQ/g	Li et al. (2012)
	Hospital waste incinerator (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 & 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)

Data obtained from the scientific literature

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 very similar to those obtained in the current study. In another survey performed 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) performed 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 greater 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 United States 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 to or lower than those reported in other studies performed in Catalonia and other countries where the same species were analysed. Investigations performed 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). In contrast, 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 sawmill of northern Sweden ranging in concentration from

Table 5 Summary of PCDD/F concentrations in air samples from recent studies

Location	Assessed area	Mean (minimum–maximum)	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)[a]	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	9.3×10^{-3}–90.8×10^{-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 & 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 (nonrunning periods)	(0.158–0.648)	pg I-TEQ/m^3	

Data obtained from the scientific literature

[a] Median value

0.26 to 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 and obtained a mean value of 8.8 ng I-TEQ/g, whereas 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 listed in Table 6. We considered three different scenarios: (1) all sampling points; (2) sampling points closer to the facility from 250 to 750 m; and (3) sampling points located 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 scenario B. For most metals, the exposure pathway with the highest percentage of contribution was soil ingestion followed by dermal contact. In contrast, air inhalation was the minority route for metals, with the only exception of Sb. Arsenic was the only element for which dermal contact was the main route of exposure, which was followed closely by soil ingestion. The PCDD/F profile was found to be quite different with respect to those for metals with inhalation being the predominant route, i.e., 60 % of total exposure. The

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

	Pathways	Scenario A		Scenario B		Scenario C	
		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 (250–750 m), *C* sampling points located at 1000–1500 m from the MSWI

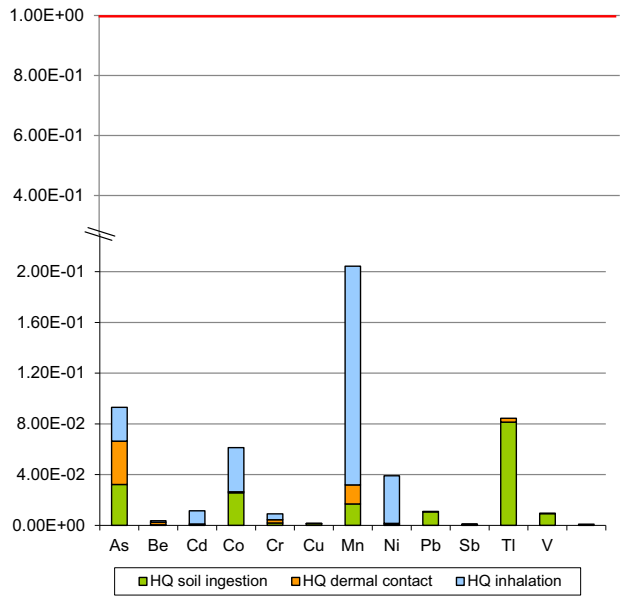

Fig. 3 Noncarcinogenic risks (HQs) of environmental exposure to metals and PCDD/Fs in the surroundings of the Tarragona MSWI (Catalonia, Spain)

other two routes, soil ingestion and dermal contact, contributed almost equally with approximately 20 % for each one.

Health risks associated with environmental exposure to metals and PCDD/Fs were also characterized (Figs. 3, 4). Noncarcinogenic risks were assessed by comparing metal and PCDD/F exposure with the RfD. The HQ for all metals and PCDD/Fs did not exceed (in any case) unity, which is considered the safety threshold. Mn, followed by As, was the element showing the maximum HQ, whereas PCDD/Fs had a HQ value of 2.15×10^{-3}. With respect to cancer risks, Cr was the metal showing the highest value (risk index $= 1.73 \times 10^{-5}$) followed by As (risk index $= 1.14 \times 10^{-5}$). 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 one sixth of the total Cr was Cr-VI, which is the carcinogenic form (USEPA 1998). Thus, we could have overestimated the exposure and the associated health risks. Similarly, we only analysed the levels of total As, assuming that it was completely 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 (approximately 20 years) environmental monitoring program involving analyses of PCDD/Fs and heavy metals in environmental samples collected near the Tarragona MSWI. The inclusion of air samples taken since 2007 allows an integrated diagnosis of the impact of the facility by 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- and immediate-term environmental monitors, respectively) reflects the low impact of the facility on the neighborhood. Human exposure to PCDD/Fs and metals and the associated noncarcinogenic and carcinogenic risks have shown that the emissions of these pollutants by the MSWI do not

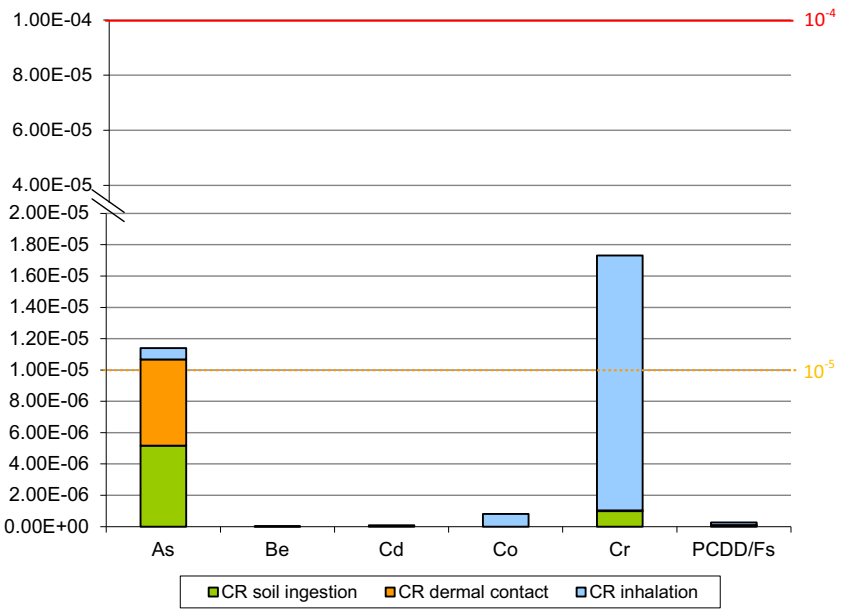

Fig. 4 CRs of environmental exposure to metals and PCDD/Fs near the Tarragona MSWI (Catalonia, Spain)

generate additional health risks for the populations living nearby.

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

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