High cancer risks by exposure to PCDD/Fs in the neighborhood of an Integrated Waste Management Facility

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ABSTRACT

In 2014, we conducted a study aimed at screening the concentrations of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs) and various trace elements in air and soil samples collected in an urban area of Sant Adrià de Besòs (Barcelona, Spain) in the vicinity of an Integrated Waste Management Facility (IWMF). It consists of a mechanical-biological treatment plant (MBT) and an old municipal solid waste incinerator (MSWI). Human health risks for the population living in the area were also assessed. The most worrying result was the high cancer risks estimated for the area (2.5×10^{-6}). In March 2017, we have carried out a new survey to check if the authorities had taken the necessary and urgent measures to reduce the environmental concentrations of PCDD/Fs -and the human health risks- until acceptable levels. Although the concentrations of PCDD/Fs in soils are currently lower (mean value: 1.66 vs. 3.6 ng WHO-TEQ/kg in 2014), they are still are notably higher than those found near other MSWIs of Catalonia. In turn, the levels of PCDD/Fs in air are even higher than in 2014 (mean value: 0.044 vs. 0.026 pg WHO-TEQ/m^3 in 2014), being also the highest detected in similar zones of Catalonia. The current cancer risk due to PCDD/F exposure for the residents in the neighborhood of the IWMF is 2.3×10^{-6}, a worrying fact as the 10^{-6} threshold continues to be exceeded.

Keywords:

Integrated Waste Management Facility

Municipal Solid Waste Incinerator

Urban area

PCDD/Fs

Cancer risks

1. Introduction

The Integrated Waste Management Facility (IWMF) of Sant Adrià de Besòs (Barcelona, Spain) is constituted by a municipal solid waste incinerator (MSWI), which is operating since 1975, and a mechanical-biological treatment (MBT) plant, which started to operate regularly in 2006. The main purpose of the IWMF is to manage part of the municipal solid waste (MSW) generated in the Metropolitan Area of Barcelona. The capacities of treatment of wastes are 192,000 and 350,000 tons per year, for the MBT and the MSWI, respectively. In spite of the advantages that, doubtless, a facility like this one means, both the MBT and the MSWI also own some important inconveniences. A varied typology of microbiological and chemical agents may be generated and released by MBTs. During the waste sorting, composting, and compost refining, MBTs can release into the atmosphere a significant amount of hazardous compounds, including volatile organic compounds (VOCs), a kind of pollutants characterized by their malodorous and dangerous properties (Domingo and Nadal, 2009; Vilavert et al., 2012, 2014). Moreover, biological particulates suspended in air, known as bioaerosols, or "organic dust", may lead to different pathologies (Domingo and Nadal, 2009; Nadal et al., 2009; Vilavert et al., 2012a).

On the other hand, MSW incineration has become a serious option of waste management in developed countries. While MSWIs means energy recovery and volume minimization, public controversy is also frequent where this kind of facilities are already operating or being planned. MSWIs have been historically associated to emissions of toxic chemicals, mainly polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and heavy metals, among other pollutants, which can mean carcinogenic and non-carcinogenic risks for the population living in the neighborhood (Chen et al., 2006; Giusti, 2009; Schuhmacher et al., 2001; Vinceti et al., 2008; Vilavert et al., 2012b). During 1998-2001, we conducted a survey whose main goal was to determine the environmental impact of the MSWI of Sant Adrià de Besòs after installation of a new gas cleaning system. The concentrations of a number of metals and PCDD/Fs were determined in soil and herbage samples collected in various points in the vicinity of the facility (Domingo et al., 2000, 2002a,b; Schuhmacher et al., 2000). In subsequent years, other studies, including the measurement of PCDD/F concentrations in air samples

collected in the area under potential influence of the MSWI, were also carried out (Mari et al., 2008a,b).

In 2014, we conducted a screening-survey to assess the human health risks of the IWMF. Among other interesting results, we observed that PCDD/F concentrations in soil and air were the highest ever reported near a MSWI in Catalonia (Domingo et al., 2015). We also noted that human health risks of PCDD/F exposure in the closest urban nucleus -located downwind the MSWI- were up to 10-times higher than those nearby other MSWIs in Catalonia. Given the transcendence on the public health of these results, we immediately warned the technical and political managers of the IWMF about the importance of reducing the levels of PCDD/Fs in the area. Three years after, we have conducted a new study -with similar characteristics to that of 2014- in order to assess if the current environmental levels of PCDD/Fs, as well as the human health risks were already significantly reduced. The results are here presented.

2. Materials and methods

2.1. Sampling

In March 2017, three air samples were collected at the School/High School of Sant Adrià de Besòs, the sports area of "La Mina", and the IES "Manuel Vázquez Montalbán", all 3 belonging to the city of Sant Adrià de Besòs (Barcelona, Spain). Soil samples (3) were also collected at the same three locations (Fig. 1).

Air sampling was performed by following the US EPA Method TO-9 (Rovira et al., 2015), and using TE-1000 high-volume active samplers (Tisch Environmental, Cleves, OH, USA) The particle phase of PCDD/Fs was collected on quartz microfiber filters (QFFs), while the gas phase was collected by means of polyurethane foams (PUFs). In order to remove any potential contamination, PUFs were pre-cleaned in a Soxhlet with dichloromethane (Scharlau, Sentmenat, Spain) for 24 h. Air sampling lasted for approximately 48 h, during which a total air volume of 600 m^3 was collected. To prevent any possible photo-degradation of the compounds, QFFs and PUFs were stored together in amber glass bottles, with Teflon stoppers. Samples were stored at a temperature of -20°C until analysis.

Surface soil samples were taken from the upper 5 cm of ground and stored in polyethylene bags. Each sample (500 g approximately) consisted of 4 subsamples collected in an area of approximately 25 m^2. The bulked samples were dried at room temperature and sieved with a 2 mm mesh screen until analysis.

2.2. Analytical procedure

The analysis of the concentrations of PCDD/Fs is air and soil samples were done by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS), in combination with the isotope dilution technique, according to the US EPA method 1613 (Rovira et al., 2015). To control the whole process and to evaluate potential losses, labeled standards ($^{13}C_{12}$ PCDD/Fs) were added. An Accelerated Solvent Extraction (ASE) was carried out by using toluene. The extract was subsequently subjected to an acid/base clean-up procedure, followed on micro columns of silica gel and alumina. The final extract was again spiked with isotopic labeled internal standards, being then analyzed for PCDD/F concentrations. Blank samples were also analyzed.

The limits of detection (LODs) of the PCDD/F congeners in soils were: 0.04 ng/kg for 2,3,7,8-TCDD and 2,3,7,8-TCDF, 0.08 ng/kg for penta- and hexadioxins and for penta-, hexa- and heptafurans, and 0.10 ng/kg for heptadioxins, OCDD and OCDF. Recovery percentages of PCDD/Fs ranged between 51 and 96%. In air, the LODs of the PCDD/Fs were the following: 0.001 pg/m^3 for 2,3,7,8-TCDD, 2,3,7,8-TCDF and the penta- and hexadioxins and furans, 0.008 pg/m^3 for heptadioxins and furans, and 0.032 pg/m^3 for OCDD and OCDF. Recovery percentages of PCDD/Fs ranged between 56 and 97%. Total concentrations of PCDD/Fs were calculated by using the toxic equivalency factors (TEFs) of the World Health Organization (WHO-TEFs) (van den Berg et al., 2006).

2.3. Human health risks

Human exposure through ingestion, dermal absorption and inhalation, as well as the associated health risks, were estimated as previously described (Rovira et al., 2010, 2015). The numerical expressions were taken from the Technical Guide of the "Spanish

Royal Decree 9/2005", which establishes the list of activities potentially contaminants of soils, as well as the criteria and standards for the declaration of contaminated soils (Ministerio de Medio Ambiente, 2007). It is based on the US EPA methodology. In addition, inhalation risks were calculated based on the most updated US EPA methodology for this route of exposure (US EPA, 2009). To assess the carcinogenic and non-carcinogenic risks from oral exposure, the oral reference dose (RfD_o) and the oral slope factor (SF_o) were multiplied and divided, respectively, by the gastrointestinal absorption factor (GI_{ABS}) (US EPA, 1989). Specific equations of human exposure and risk characterization were those previously used by Rovira et al. (2010).

3. Results and discussion

3.1. PCDD/F levels in soil samples

Table 1 summarizes the total levels (in WHO-TEQ) of PCDD/Fs and the concentrations of each individual congener in soil samples collected in March 2017 in the vicinity of the IWMF of Sant Adrià de Besòs. Total levels of PCDD/Fs in the samples collected in the current study (2017), and those obtained in a previous survey (2014), are also depicted in Fig. 2. The highest concentrations corresponded to the most chlorinated dioxin congener, OCDD, followed by 1,2,3,4,6,7,8-HpCDD, OCDF and 1,2,3,4,6,7,8-HpCDF. In turn, 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD and 1,2,3,7,8,9-HxCDF could not be detected at the sampling point located in the vicinity of the School/High School of Sant Adrià de Besòs. The most toxic congener, 2,3,7,8-TCDD, presented a range between <0.04 and 0.26 ng/kg. Total PCDD/Fs reached a mean level of 1.66 ng WHO-TEQ/kg (range: 0.36-3.23 ng WHO-TEQ/kg). The comparatively high total level found in the point closest to the IES "Manuel Vázquez Montalbán" (3.23 ng WHO-TEQ/kg) was already observed in the 2014-sampling, in which a considerable concentration of PCDD/Fs was found: 10.1 ng WHO-TEQ /kg (Domingo et al., 2015). In previous surveys, this point already showed similar -or even higher levels- of PCDD/Fs: 19.1 ng I-TEQ/kg in 1998 and 7.9 ng I-TEQ/kg in 1999 (Domingo et al., 2002b).

Table 2 and Fig. 3 show the levels of PCDD/Fs in soils obtained in similar surveys conducted around various MSWIs (and a HWI) in Catalonia. It can be seen that the

previous (2014 sampling) and the present (2017) concentrations of PCDD/Fs near the IWMF of Sant Adrià de Besòs, which includes an old MSWI, are the highest we have ever detected. In fact, the current levels of PCDD/Fs are 3-5 times higher than those found near other incinerators of Catalonia.

3.2. PCDD/Fs in air samples

The total concentrations of PCDD/Fs (WHO-TEQ), as well as the levels of each individual congener in air samples collected in March 2017 in the vicinity of the IWMF of Sant Adrià de Besòs, are summarized in Table 3. As for soils, the highest concentrations corresponded to OCDD, followed by 1,2,3,4,6,7,8-HpCDD and 1,2,3,4, 6,7,8-HpCDF. On the other hand, OCDF was not detected in any of the air samples analyzed. The most toxic congener, 2,3,7,8-TCDD, showed a concentration of 0.001 pg/m^3 in the three samples. Regarding the total level of PCDD/Fs (WHO-TEQ), similar values were found in the 3 sampling points, with a mean concentration of 0.044 pg WHO-TEQ/m^3 (range: 0.042-0.048 pg WHO-TEQ/m^3). Total levels of PCDD/Fs in air samples of the present survey, as well as those obtained in the 2014 study, are also depicted in Fig. 4. It can be observed that for the three sampling points, the current PCDD/F levels are higher than those found in the 2014 survey.

As for soils, we have also summarized the results of a number of studies on the concentrations of PCDD/Fs in air samples, which we have conducted in the vicinity of various MSWIs of Catalonia (Table 4 and Fig. 5). It can be seen that the current concentrations of PCDD/Fs in air samples collected in the neighborhood of the IWMF – which includes an old MSWI- of Sant Adrià de Besòs, are the highest ever detected in studies performed in Catalonia. In contrast to soils, where we found a decrease of the current mean concentration with respect to that observed in the 2014 survey, the mean PCDD/F concentrations in air have shown a considerable increase (2-3 times) in relation to the levels found in 2014, which is certainly a worrying question.

3.3. Human health risks

The exposure to PCDD/Fs, depending on the route (soil ingestion, air inhalation and dermal contact with soil and dust), for the individuals living in the area under influence of the IWMF of Sant Adrià de Besòs, is summarized in Table 5. It can be noted that the main route of human exposure to PCDD/Fs in the zone under evaluation is air inhalation, with contributions of 38% and 68%, in 2014 and 2017, respectively. The hazardous quotient (HQ) is used to evaluate the non-carcinogenic effects of exposure to a certain pollutant. It represents the relationship between exposure to the pollutant and the reference dose (RfD), which is defined as the exposure for which there are no adverse effects. HQ values below the unity are considered as safe. The current HQ was 0.01, versus 0.02 in 2014. Both values are < 1, which indicate that there are no significant non-carcinogenic risks due to human exposure to PCDD/Fs in the neighborhood of the IWMF.

HQs are only applicable to non-carcinogenic risks. Carcinogenic risks are expressed in terms of probability of developing cancer, due to exposure over a lifetime (estimated in 70 years). The US EPA considers as negligible a carcinogenic risk of less than 10^{-6}. The carcinogenic risks of the 2014 and 2017 surveys, as well as those corresponding to areas under the potential influence of emissions by other MSWIs in Catalonia, are depicted in Fig. 6. The carcinogenic risks due to PCDD/F exposure for the residents in the neighborhood of the IWMF of Sant Adrià de Besòs were 2.5 x 10^{-6} and 2.3 x 10^{-6}, in 2014 and 2017, respectively. Therefore, both values exceed the 10^{-6} threshold, which according to the US EPA, cannot be considered as a negligible risk of cancer. We have compared these risks with those estimated in other areas of Catalonia, also under the potential influence of PCDD/Fs emitted by MSWIs. The residents in the neighborhood of the IWMF of Sant Adrià de Besòs are 3-4 times more likely of developing cancer over a lifetime (due to exposure to PCDD/Fs) than residents of cities such as Girona, Mataró and Tarragona, where there are also MSWIs operating since various years ago.

4. Conclusions

Based on the results of the present study, as well as those obtained in a recent survey (Domingo et al., 1015), the carcinogenic risks due to environmental exposure to PCDD/Fs for the population living in the vicinity of the IWMP here evaluated, are considerable. It must mean a serious warning for those authorities with responsibilities

on the environment and the protection of the public health. Unfortunately, the situation in the area has not improved during the last 3 years, being the cancer risks due to exposure to PCDD/Fs very similar between 2014 and 2017. While there has been a notable decrease in the concentrations of PCDD/Fs in soils, unfortunately it has been counteracted by the important increase observed in the mean concentrations of these pollutants in air.

The most important limitation of the current survey is the small number of air and soil samples. Consequently, the results should be taken with caution. However, the increases in the concentrations of PCDD/Fs in air are evident for the 3 sampling points. Anyway, the current results corroborate data previously obtained in another rather limited study conducted in 2014 (Domingo et al., 2015).

With the present information, we cannot assign the levels of PCDD/Fs in air only to the MSWI included in the IWMF. Very probably, traffic is also an important emission source of these pollutants in the area, while other masked sources cannot be discarded (Dopico and Gómez, 2015; Rahman et al., 2014; Rey et al., 2014; Zhang et al., 2014). An exhaustive study of all emission sources of PCDD/Fs in this zone is a previous and essential step to take decisive actions, which should allow significant reductions in the concentrations of these pollutants in the area, and consequently, a significant reduction of the carcinogenic risks for the population until negligible/acceptable levels.

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Table 1

Concentrations (ng/kg) of PCDD/Fs in soil samples collected in 2017 around the IWMF of Sant Adrià de Besòs

	School/High School Sant Adrià	**Sports Zone "La Mina"**	**IES Vázquez Montalbán**
2,3,7,8-TCDD	<0.04	0.09	0.26
1,2,3,7,8-PeCDD	<0.08	0.30	0.75
1,2,3,4,7,8-HxCDD	<0.08	0.39	1.30
1,2,3,6,7,8-HxCDD	0.20	0.88	6.00
1,2,3,7,8,9-HxCDD	0.13	0.57	3.10
1,2,3,4,6,7,8-HpCDD	3.70	29.0	42.0
OCDD	31.0	200	140
2,3,7,8-TCDF	0.38	0.78	0.42
1,2,3,7,8-PeCDF	0.23	0.58	0.43
2,3,4,7,8-PeCDF	0.25	0.51	0.85
1,2,3,4,7,8-HxCDF	0.30	0.66	2.00
1,2,3,6,7,8-HxCDF	0.23	0.45	0.72
1,2,3,7,8,9-HxCDF	<0.08	0.18	0.16
2,3,4,6,7,8-HxCDF	0.21	0.56	0.51
1,2,3,4,6,7,8-HpCDF	1.30	2.90	5.70
1,2,3,4,7,8,9-HpCDF	0.18	0.46	1.10
OCDF	1.40	6.50	12.0
Total WHO-TEQ	**0.36**	**1.39**	**3.23**

Table 2
Concentrations of PCDD/Fs in soil samples collected near various incinerators in Catalonia (Spain)

Location	Year	Mean (ng TEQ/kg)	Range (ng TEQ/kg)	Reference
Tarragona (MSWI)	1999	1.20	0.15 - 4.89	Vilavert et al. (2009)
	2005	6.01	0.33 - 46.4	
	2008	0.64	0.13 - 2.41	
	2010	0.58	0.11 - 1.35	Vilavert et al. (2012b)
Constantí (HWI)	2009	0.75	0.09 - 2.99	Vilavert et al. (2010)
Campdorà, Girona (MSWI)	2015	0.39	0.13 - 0.76	Not published data
	2016	0.36	0.12 – 0.97	
Mataró (MSWI)	2008	0.34	0.14 - 0.46	Rovira et al. (2010)
	2011	0.23	0.13 - 0.56	Rovira et al. (2015)
	2013	0.34	0.12 - 0.61	Rovira et al. (2015)
Sant Adrià de Besòs (IWMF/MSWI)	1998	9.06	1.22 - 34.28	Domingo et al (2002a)
	1999	11.9	1.33 - 54.23	Domingo et al (2002a)
	2000	7.09	0.41 - 121	Domingo et al (2002b)
	2014	3.60	0.40 - 10.6	Domingo et al. (2015)
	2017	1.66	0.36 – 3.23	This study

MSWI: Municipal solid waste incinerator
HWI: Hazardous waste incinerator
IWMF: Integral waste management facility

Table 3
Concentrations (pg/m^3) of PCDD/Fs in air samples collected in 2017 around the IWMF of Sant Adrià de Besòs

	School/High School Sant Adrià	**Sports Zone "La Mina"**	**IES Vázquez Montalbán**
2,3,7,8-TCDD	0.001	0.001	0.001
1,2,3,7,8-PeCDD	0.002	0.005	0.002
1,2,3,4,7,8-HxCDD	0.002	0.004	0.002
1,2,3,6,7,8-HxCDD	0.005	0.007	0.004
1,2,3,7,8,9-HxCDD	0.003	0.008	0.004
1,2,3,4,6,7,8-HpCDD	0.041	0.059	0.043
OCDD	0.147	0.165	0.181
2,3,7,8-TCDF	0.041	0.040	0.043
1,2,3,7,8-PeCDF	0.025	0.023	0.021
2,3,4,7,8-PeCDF	0.073	0.076	0.071
1,2,3,4,7,8-HxCDF	0.033	0.038	0.035
1,2,3,6,7,8-HxCDF	0.026	0.031	0.026
1,2,3,7,8,9-HxCDF	0.009	0.008	0.003
2,3,4,6,7,8-HxCDF	0.036	0.036	0.030
1,2,3,4,6,7,8-HpCDF	0.049	0.049	0.040
1,2,3,4,7,8,9-HpCDF	<0.008	0.011	<0.008
OCDF	<0.032	<0.032	<0.032
Total WHO-TEQ	**0.042**	**0.048**	**0.041**

Table 4
Concentrations of PCDD/Fs in air samples collected near various MSWIs in Catalonia (Spain)

Location	Year	Mean (pg TEQ/m^3)	Range (pg TEQ/m^3)	Reference
Tarragona	2007	0.012	0.004 - 0.033	Vilavert et al. (2009)
	2008	0.015	0.007 - 0.031	
	2009	0.009	0.007 - 0.022	Vilavert et al. (2012b)
	2010	0.010	0.004 - 0.022	
	2013	0.004	0.002 - 0.005	Vilavert et al. (2015)
	2014	0.010	0.006 - 0.017	
Mataró	2008	0.011	0.008 - 0.015	Rovira et al. (2010)
	2011	0.010	0.006 - 0.013	Rovira et al. (2015)
	2013	0.014	0.010 - 0.018	Rovira et al. (2015)
Campdorà, Girona	2015	0.015	0.009 - 0.028	Unpublished data
	2016	0.011	0.009 – 0.014	
Sant Adrià de Besòs	2005-2006			Mari et al. (2008a)
Near the MSWI		0.018	0.010 - 0.024	
Control area		0.012	0.008 - 0.019	
Sant Adrià de Besòs	2014	0.026	0.018 - 0.041	Domingo et al. (2015)
Sant Adrià de Besòs	2017	0.044	0.042 - 0.048	This study

Table 5
Environmental levels of PCDD/Fs and human exposure for the population living in the neighborhood of the IWMF of Sant Adrià de Besòs. Data for the 2014 and 2017 surveys

	PCDD/Fs*		Exposure (ng WHO-TEQ/(kg·day))		
	Soils	**Air**	**Soil ingestion**	**Dermal contact**	**Air inhalation**
2014	3.60	0.026	5.6E-06	6.0E-06	7.2E-06
2017	1.66	0.044	2.6E-06	2.8E-06	1.2E-05

*Units: PCDD/Fs in soils: ng WHO-TEQ/kg; PCDD/Fs in air: pg WHO-TEQ/m^3

Fig. 1. Location of sampling points for air (blue) and soils (orange). The facility is marked in red.

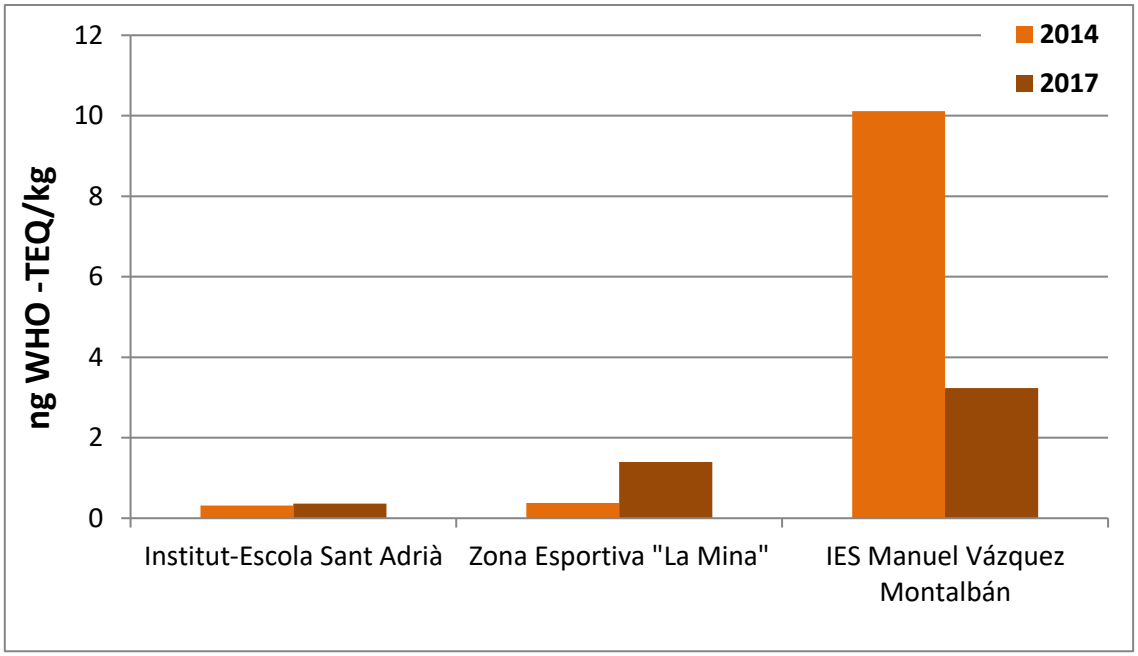

Fig. 2. Total levels of PCDD/Fs (ng WHO-TEQ/kg) in soil samples collected around the IWMF of Sant Adrià de Besòs in the previous (2014) and current (2017) surveys.

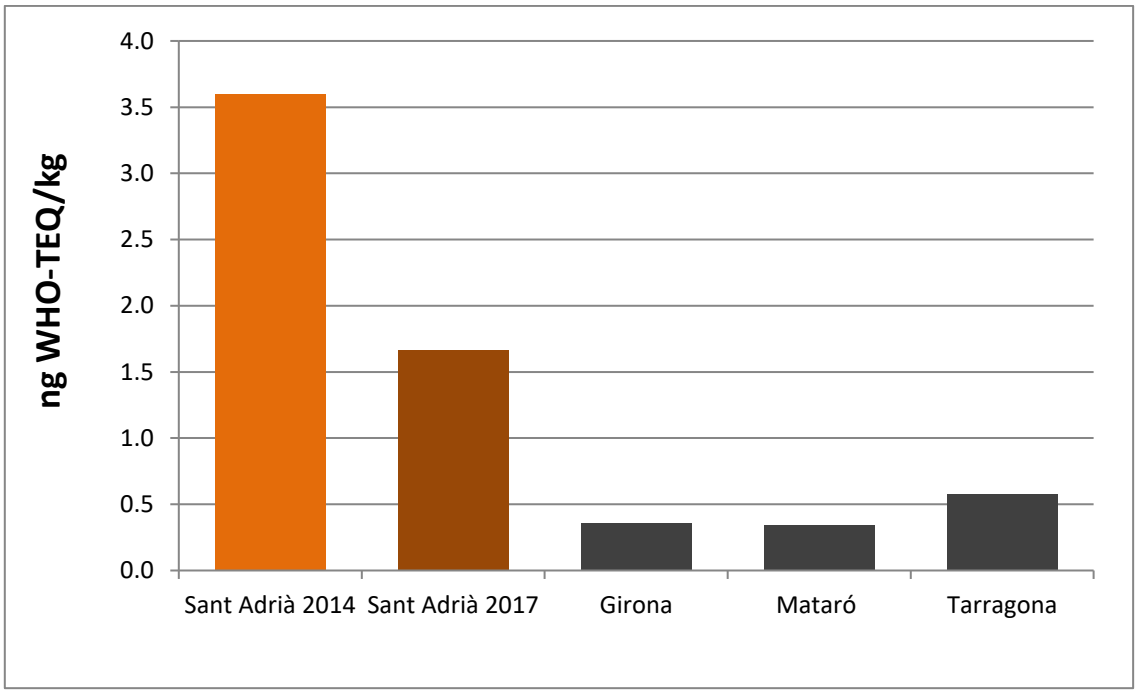

Fig. 3. Levels of PCDD/Fs in soils collected around the IWMF of Sant Adrià de Besòs in 2014 and 2017, as well as in the vicinity of other MSWIs located in Catalonia.

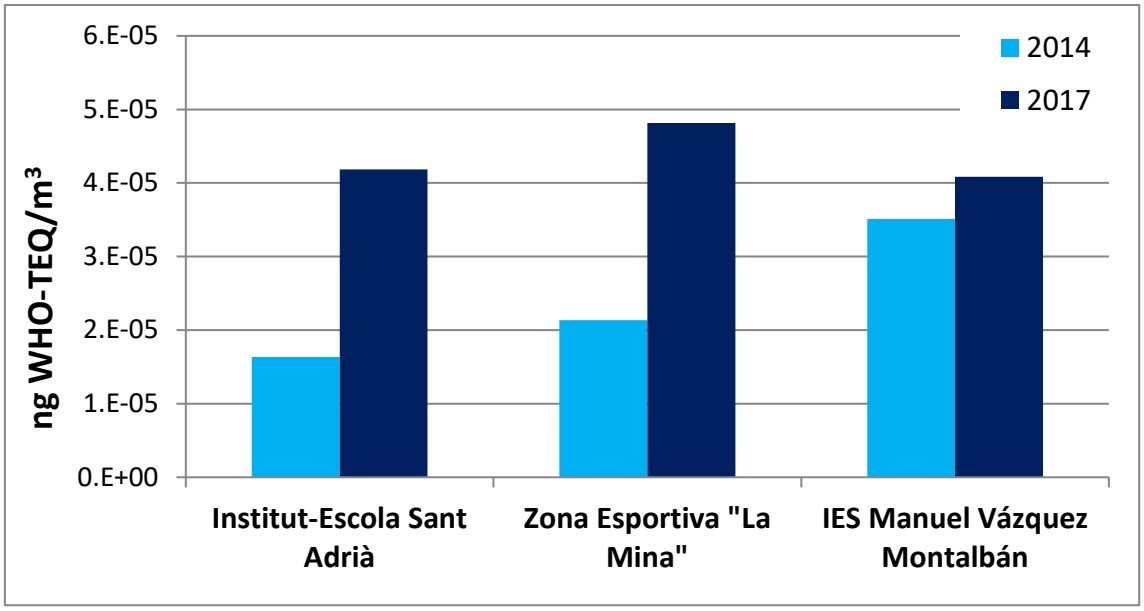

Fig. 4. Total levels of PCDD/Fs (ng WHO-TEQ/m^3) in air samples collected around the IWMF of Sant Adrià de Besòs in the previous (2014) and current (2017) surveys.

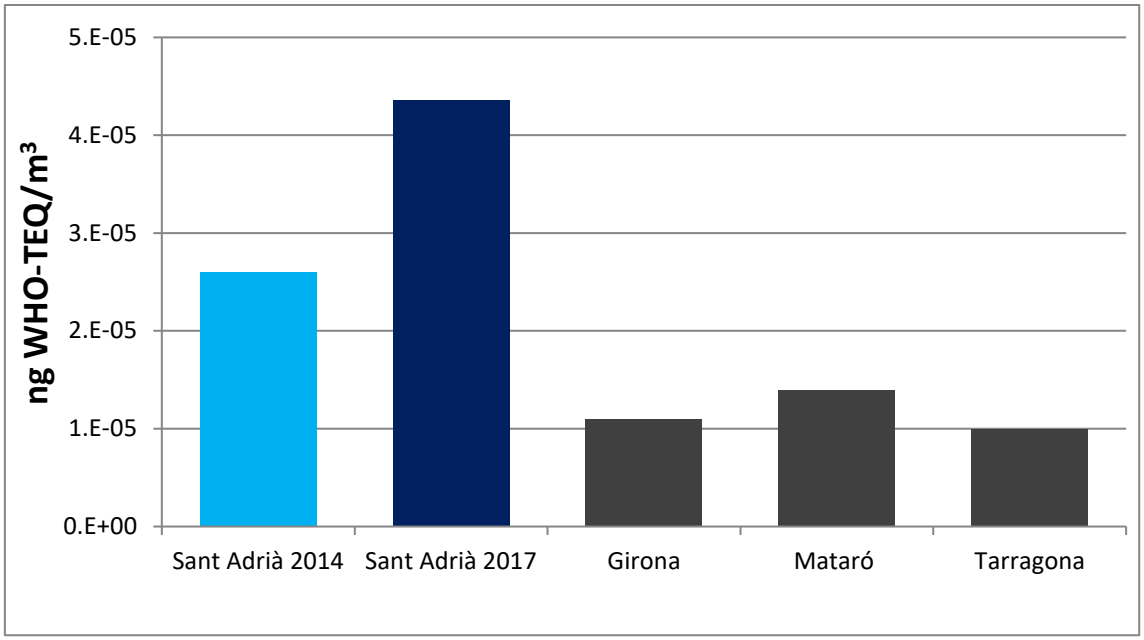

Fig. 5. Levels of PCDD/Fs in air samples collected around the IWMF of Sant Adrià de Besòs in 2014 and 2017, as well as in the vicinity of other MSWIs located in Catalonia.

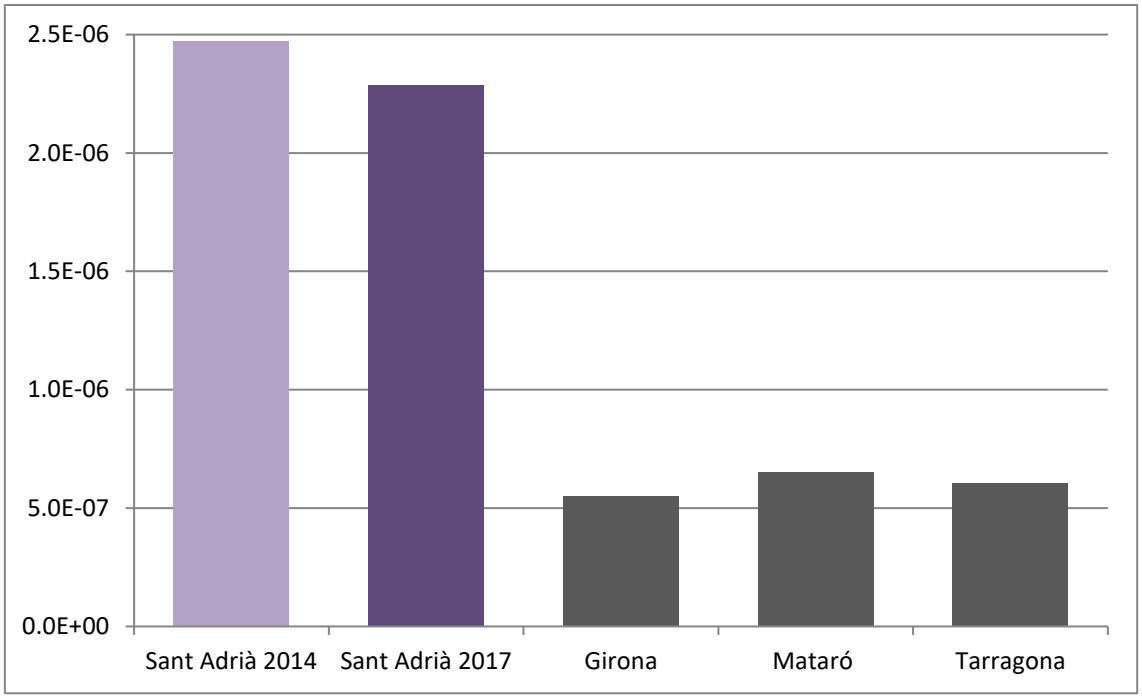

Fig. 6. Cancer risks due to exposure to PCDD/Fs for the population living near the IWMF of Sant Adrià de Besòs (2014 and 2017) and for residents in the neighborhood of other MSWIs located in Catalonia.