Regulatory compliance of PCDD/F emissions by a municipal solid waste incinerator. A case study in Sant Adrià de Besòs, Catalonia, Spain

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ABSTRACT

Despite incineration is an important emission source of toxic pollutants, such as metals and PCDD/Fs, it is still one of the most widely used methods for the management of municipal solid waste. This paper summarizes the results of a follow-up study of PCDD/Fs emissions by the municipal solid waste incinerator (MSWI) of Sant Adrià de Besòs (Catalonia, Spain), study conducted in our laboratory for 20 years. Air, soil and herbage samples were periodically collected near the facility and the content of PCDD/Fs was analyzed. In our last (2017) survey, mean levels in soil were 3.60 ng WHO-TEQ/kg (range: 0.40-10.6), being considerably higher than the mean concentrations of PCDD/Fs in soil samples collected near other MSWIs in Catalonia. Moreover, air PCDD/F concentrations were even higher than those found in a previous (2014) survey, as they increased from 0.026 to 0.044 pg WHO-TEQ/m^3. Ultimately, PCDD/F exposure would be associated to a cancer risk (2.5 x 10^{-6}) for the population living in the surrounding area. Globally, this information indicates that the MSWI of Sant Adrià de Besòs could have had a negative impact on the environment and potentially on the public health, being an example of a possible inappropriate management for years.

KEYWORDS: Municipal solid waste incinerator; PCDD/Fs; environmental impact; health risks; regulatory compliance.

1. Introduction: municipal solid waste incineration

One of the methods used to manage municipal solid waste (MSW) is incineration. This process began to gain popularity in the mid-20th century, especially in the last decades of that century. In the municipal solid waste incinerators (MSWIs), the waste is burned at high temperatures -using grate-firing, fluidized bed and rotary furnace as combustion devices- in controlled environments, reducing the volume of waste, while simultaneously energy is generated.[1-4] Unfortunately, MSW incineration also has some inconveniences. Thus, various environmental and health risks are directly related with MSWI activities.[5-8] One of the most relevant is the air pollution due to the emissions of trace amounts of various metals with well-known toxic effects[9-12] and a variety of organic compounds, including polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs).[9,13-15] The health impact derived from these emissions is obviously an issue of considerable concern.[16-18] Greenhouse gas emissions is another drawback of these facilities,[19,20] while the regulatory compliance is always a key issue. Given the potentially remarkable environmental and health risks of the MSWIs, ensuring the proper operations and maintenance of these facilities requires stringent regulations. As result of an inadequate monitoring or enforcement, the allowable emissions can be easily exceeded, consequently exacerbating the environmental and human health risks.

Based on the above concerns on MSWIs, we here summarize a series of data corresponding to an example of a potentially poor management of a MSWI, located in Sant Adrià de Besòs, Catalonia, Spain. A revision of studies currently available in the scientific literature about pollutant emissions near that facility, is here presented. This revision has been focused only on PCDD/Fs, while the presence of other co-emitted substances is not considered, despite in the past the facility has been linked to problems with the emissions of other pollutants (e.g., mercury).[21]

2. Case-study: the MSWI of Sant Adrià de Besòs (Catalonia, Spain). A description

Since 1975, a MSWI has been operating in Sant Adrià de Besòs, a town with approximately 38,000 inhabitants, which is situated at the northeast of the city of Barcelona (Catalonia, Spain). The facility is placed in an urban area close to an important industrial port. In that zone, and during various decades, a considerable number of different industrial activities had been carried out. High concentrations of arsenic, heavy metals and persistent organic pollutants (POPs), including PCDD/Fs, were found in underground samples collected in a beach nearby, where waste had been buried.[22,23] In addition to those industries, either active or not, a motorway with a heavy vehicle traffic is close to the MSWI. The facility has a capacity of incineration of approximately 360,000 tons of MSW per year, which is about a quarter part of the amount generated in the metropolitan area of Barcelona. It is a clear indicator of the relevance of that MSWI in the context of the management of the MSW in the populous area of Barcelona and nearby cities. Until 1998, an electrostatic precipitator was used as control device to reduce the emissions of pollutants, mainly focused on heavy metals and PCDD/Fs. In March 1999, due to the newly implemented EU legislation on pollutant emissions, an adaptation of the stack was carried out. In order to replace the electrostatic precipitator, an acid gas (HCl–SO2) and metal emission limit equipment were then installed, together with an active-carbon adsorption filter.

At that time, our research group was contacted by the plant managers and the politicians and technicians of Barcelona City Council and the Metropolitan Environmental Entity of Barcelona, ultimately responsible of the facility. They commissioned us to carry out a follow-up study to evaluate the environmental impact of the emissions of metals and PCDD/Fs near the facility. Between 1998 and 2006, our laboratory conducted five studies aimed at determining the concentrations of PCDD/Fs and several trace elements in samples of air, soils, and herbage collected in the vicinity of the MSWI. The health risks for the population living in the area under potential influence of the emissions of the facility were also assessed. In 2006, a MSW biological-mechanical treatment plant was built adjacent to the incinerator. Therefore, the facility was transformed into a comprehensive MSW recovery plant.

3. PCDD/Fs is samples of air, soils and herbage in the vicinity of the MSWI of Sant Adrià de Besòs

We next summarize the main results of the studies conducted in our laboratory on the environmental/health impact of the MSWI of Sant Adrià de Besòs. Although PCDD/Fs and heavy metals are not the only pollutants emitted by MSWIs, they are those raising more concern. Moreover, they are also the most investigated in studies on emissions of pollutants by MSWIs carried out over the world. It must be noted that emissions of PCDD/Fs by MSWIs were not detected until the decade of the 1970's, when Olie et al.[24] reported the presence of these highly toxic substances as trace components of flue gas in municipal incinerators of the Netherlands. Since then, the emission of PCDD/Fs by

MSWIs has been/is an issue of great concern for the environment and public health.[9,13-18]

In March 1998, 24 soil and 24 herbage samples were collected in sampling sites established according to considerations on the prediction of the time averaged emission plume, which were obtained from a Gaussian model (ISC-LT). Duplicate samples were collected at 250 m (six samples), 500 m (five samples), 750 m (four samples), 1000 m (three samples), 1500 m (three samples) and 3000 m (three samples) from the stack of the MSWI. PCDD/F concentrations in all the samples were calculated as 2,3,7,8-TCDD toxic equivalents (I-TEQ) by means of the NATO/CCMS factors (ref. of these factors missing). The predominant congeners in soils were hepta- and octa-CDDs, while in herbage TCDF and TCDD (the lowest substituted congeners) were the most abundant. In soils, the levels of PCDD/Fs varied between 1.22 and 34.28 ng I-TEQ/kg (dry matter, dm), with 9.06 and 12.24 ng I-TEQ/kg (dm) as median and mean values, respectively. In turn, the median and mean concentrations of PCDD/Fs in herbage samples were 0.58 and 0.70 ng I-TEQ/kg (dm), respectively, ranging between 0.33 and 1.98 ng I-TEQ/kg (dm).[25] A careful analysis of the individual data showed that PCDD/F levels between 10 and 20 ng I-TEQ/kg (dm) were found in seven soil samples, while levels of PCDD/Fs higher that 20 ng I-TEQ/kg (dm) were detected in six samples. It indicated that the stack emissions of PCDD/Fs had to be reduced to diminish the health risks for the population living near the MSWI.

In March 1999, one year after the first survey, samples of soils and herbage were again collected at the same 24 sampling points.[26] The main goal of the new study was to establish the temporal variation in the levels of PCDD/Fs in both environmental matrices. In the study conducted in 1999, PCDD/Fs concentrations in soils ranged from 1.33 to 54.23 ng I-TEQ/kg (dm), with 11.85 and 14.41 ng I-TEQ/kg (dm) as median and mean values, respectively. A comparison of the results in soils showed an average increase of 31% in the median (I-TEQ) levels of PCDD/Fs, with increases detected in 16 of the 24 analyzed samples. Regarding herbage samples, the levels of PCDD/Fs observed in 1999 ranged between 0.32 and 2.52 ng I-TEQ/kg (dm), being the median and mean values, 0.82 and 0.97 ng I-TEQ/kg (dm), respectively. The comparison of the median (I-TEQ) values with those found in the previous survey[25] showed an average increase of 41%, with increases detected in 17 of the 24 herbage samples. Considering values <5 ng/kg (dm) as a concentration of reference PCDD/Fs in soils,[27] the levels of PCDD/Fs exceeded the threshold in 20 of the 24 analyzed soil samples. Twelve of these concentrations were higher than 10 ng/kg, while 6 even exceeded the 20 ng/kg. Furthermore, it was concluded that human health the risks might not be underrated and, consequently, they had to be reduced.

As above indicated, in March 1999, an acid gas (HCl-SO_2) and metal emission limit equipment were installed in the MSWI, while an active-carbon adsorption filter was also added to the fabric filter. As result of this modernization, PCDD/F stack emissions decreased -on average- from 1.4 to 0.06 ng I-TEQ/Nm^3, with the legal limit of 0.1 ng I-TEQ/Nm^3 clearly fulfilled. In March 2000, a third survey was carried out. The main purpose was to examine if the decreases detected in the emissions of PCDD/Fs from the MSWI were also resulting in similar reductions in the PCDD/Fs levels in soil and herbage samples collected again near the MSWI.[28] At that time, 23 soil and 23 herbage samples were collected in 23 of the previous 24 sampling points, located between 250 and 3000 m from the stack. In the new survey, the concentrations of PCDD/Fs were found in the range 0.41-121.46 ng I-TEQ/kg (dm), being 7.09 and a 14.95 ng I-TEQ/kg (dm) the median and mean values, respectively. It meant a 40% reduction of median value in the period 1999-2000. However, this reduction in soils was not correlated with the very considerable decreases found in the emissions of PCDD/Fs from the stack. The concentrations of PCDD/Fs in herbage samples followed a similar trend than in the soil matrix, with a 30% decrease in the period 1999-2000.[28] Despite this notable reduction, the mean concentrations of PCDD/F levels in herbage were found to be still relatively high after the introduction of the technical improvements in the MSWI. To assess if that result was sporadic or it had continuity over time, in March 2001 herbage samples were again collected, and the concentrations of PCDD/Fs determined. In that survey,[29] 20 duplicate herbage samples were collected at the same sampling points than those of the previous surveys. Four of the 24 usual sampling points were not available that time. The median and mean PCDD/F concentrations were 0.58 and 0.66 ng I-TEQ/kg (dm), respectively, with a range from 0.23 to 1.43 ng I-TEQ/kg (dm). For the period 2000-2001, this meant a very low percentage of I-TEQ variation (increasing only 1%), in contrast to the reduction of 30% observed in the period 1999-2000.[28] The individual analysis of the results showed decreases in the levels of PCDD/Fs at 8 sampling points and increases at 11 sampling points, while the concentration did not change at the sample collected at 500 m from the MSWI. The results of the studies carried out in 2000 and 2001 were globally analyzed,[28,29] together with an exhaustive evaluation of these results by means of principal component analysis (PCA). The outcomes suggested that in addition to the

MSWI, there were also other emissions sources of PCDD/Fs affecting the area under direct influence of the MSWI here examined.

In 2005, we carried out a new survey, which was focused on the analyses of air concentrations of PCDD/Fs using active and passive air samplers.[30] It is well established that soils reflect the cumulative deposition of environmental pollutants in general, including PCDD/Fs, during long periods of time. In contrast, herbage is a more suitable monitor to be correlated with air emissions of PCDD/Fs at short term.[31,32] In turn, air concentrations of PCDD/Fs are a direct indicator of the current atmospheric emissions of these compounds from any emission source. In March-April 2005, using high-volume active samplers, air samples were collected at 4 sampling points: 3 of them in urban/industrial zones of Barcelona and one in a background/control site. Four PUF passive samplers were also deployed during 3 months at the same sampling points. Concentrations as WHO-toxic equivalents (WHO-TEQ) were calculated by using WHO toxicity equivalency factors (WHO-TEF). Air levels of PCDD/Fs were 0.027 and 0.011 pg WHO-TEQ/m^3 at the 3 urban/industrial and at the background site, respectively. Based on those results, the following survey of this long-term monitoring study consisted of measuring the levels of PCDD/Fs in air samples collected near the MSWI of Sant Adrià de Besòs.[32] In 2006, three air samples were collected (using high-volume active samplers) at 500 m (2 samples) and 1000 m (one sample) from the MSWI, while a fourth air sample (background/control)) was obtained in a close zone (green space), without any known source of PCDD/Fs nearby. The mean levels of PCDD/Fs in the industrial and the control areas were 0.018 and 0.012 pg WHO-TEQ/m^3, respectively, with the highest level found in a sample collected in the industrial area (0.024 pg WHO-TEQ/m^3). As expected, the lowest PCDD/F concentration (0.008 pg WHO-TEQ/m^3) corresponded to the sample collected in the control area. In comparison with our previous air data,[33] the temporal variation was rather irrelevant. A PCA was performed to obtain information on the relationship among samples, pollutants, and emission sources. No significant seasonal, temporal, and spatial variations were found. However, differences between the profiles of the PCDD/F congeners in air samples, as well as those in emission gases, were observed. These findings suggested the presence of other potential emission sources of PCDD/Fs in the same area.[33]

After the studies by Mari et al.[32,33], and considering the already important age of the plant, as well as the rather high concentrations of PCDD/Fs found in the studies carried out between 1998 and 2006, in May-June 2014, our laboratory decided to perform a screening investigation aimed at determining the concentrations of PCDD/Fs, dioxin-like PCBs (dl-PCBs) and non-dioxin-like PCBs (ndl-PCBs), in samples of air and soils collected in the vicinity of the MSWI.[34] The results were used to assess the health risks for the adult population living in the neighborhood. The levels of a number of trace elements and volatile organic compounds (VOCs) were also analyzed in that survey. The most striking result concerned the levels of PCDD/Fs found in soil and air samples. The concentrations were the highest amongst those previously reported near MSWIs in Catalonia, being the maximum levels 10.8 ng WHO-TEQ/kg (for soils) and 41.3 fg WHO-TEQ/m^3 (for air). This could mean a sign of the possible poor operations in the MSWI, taking also into account that there had not any reduction in the levels of PCDD/Fs in soils. It even after the closure of a power plant located adjacently to the MSWI. Human health risks of PCDD/Fs exposure in the closest urban nucleus, located downwind the MSWI, were up to 10-times higher than those estimated nearby other MSWIs of Catalonia. It must be highlighted that these findings were only the result of a screening conducted with a few number of samples, which was an obvious limitation of the study. To clear our doubts about the functioning of the MSWI, a new survey was carried out in 2017.[35] It was focused on assessing whether the environmental concentrations of PCDD/Fs, and the associated health risks, were being reduced. As in previous surveys, the concentrations of various metals were determined. The mean PCDD/F concentration in soils was 1.66 (range 0.36-3.23) ng WHO-TEQ/kg. Although lower than in 2014 (3.60 ng WHO-TEQ/kg, range: 0.40-10.6), it was still considerably higher than the mean values found in soil samples collected near other MSWIs in Catalonia.[36,37] Interestingly, the concentrations of PCDD/Fs in air samples were even higher than those observed three years before, with a mean value of 0.044 pg WHO-TEQ/m^3 (0.026 pg WHO-TEQ/m^3 in 2014). They were also in the highest part of the range among typical values previously found in other industrial zones of Catalonia (Fig. 1).[34] However, the most notable result for the population living in the area was the cancer risk (2.5 x 10^{-6}) due to PCDD/F exposure, which exceeded the 10^{-6} threshold. In fact, cancer risks were not reduced between 2014 and 2017, which was certainly a very negative result (Table 1 and Fig. 2).[35] With respect to the cancer risks, Garcia-Pérez et al.[38] published an interesting review on cancer mortality in towns located near Spanish-based incinerators, including the MSWI of Sant Adrià de Besòs. The authors reported higher relative risks of dying from cancers (e.g., bone cancer, non-Hodgkin's lymphoma, and thyroid cancer) for the population living at distances of less than 5 km of that facility.

To the best of our knowledge, since the publication of the results of our last survey,[35] only a study regarding the MSWI here examined is available in the scientific literature. Van Drooge et al.[39] collected air and soil samples in four sampling stations in the zone under direct influence of the MSWI of Sant Adrià de Besòs, in 2018-2019. The concentrations of PCDD/Fs in soils were between 9.0 and 22 pg WHO-TEQ/g in the two sampling points nearest to the facility. These levels were higher than those found in other urban areas, being above the value of 5 pg WHO-TEQ/g, which is a reference value established in various countries of the European Union. In the other two sampling points, including the traffic site, PCDD/F concentrations were 0.8 and 1.9 pg WHO-TEQ/g. Regarding air samples, the median and the mean were 7.5 and 11 fg WHO-TEQ/m^3 (SD: 8.3 fg WHO-TEQ/m^3), respectively. Human health risks were not assessed in that study.[39]

4. Discussion and conclusions

Concern among the population living in the neighborhood of MSWIs has been increasing all over the world. A good example is the MSWI of Sant Adrià de Besòs, here examined. It concerns not only the health risks of living in the neighborhood of the MSWI, as the zone is also subjected to the impact of other potentially polluting infrastructures, such as thermal power plants, a large wastewater treatment plant, or motorways with a heavy traffic, among others. As result of that concern, residents formed a platform called 'Airenet' (www.airenet.eu), which was originally created to control and report the environmental irregularities of the MSWI to local and regional authorities. Since 2017, Airenet has detected various irregularities, affecting emissions of PCDD/Fs and heavy metals such as mercury, but also internal deficiencies, which were reported to the Environmental Prosecutor's Office of Catalonia.[21] Right now, all these issues related with the internal/external issues and irregularities of the facility are awaiting trial. With the activity of the MSWI (which is publicly owned) under scrutiny by the Environmental Prosecutor's Office since 2018, as well as by a court since 2022, the Council of San Adrià de Besòs is no longer resigned to keep the MSWI in its territory. In a demand, unprecedented in half a century, the city Council has just proposed that the MSWI be closed and abandon the mouth of the Besòs River.[40] Without yet knowing what politics or justice will end up deciding about this MSWI, it seems even probable that a deficient/poor management -for years- of the plant will end up leading to a possible closure. It would have important consequences for an efficient global management of the MSW in the Metropolitan Area of Barcelona.

The above is a clear example on the great importance of conducting exhaustive processes of risk characterization, risk assessment, and very especially risk communication on sensitive facilities, such as MSWIs, that involve emissions of toxic substances to the environment. Addressing the gap between experts and the general population on the knowledge of technical topics is an essential issue.

In conclusion, the incineration of MSW -in itself- is not a good or bad process of waste management, which is neither safe nor unsafe. Numerous MSWIs are likely to operate adequately and with a high level of environmental efficiency. However, others, such as that here analyzed, have been having too many problems, probably because of the deficient or inappropriate management of the plant. Based on the studies above discussed, the MSWI of Sant Adrià de Besòs might have had a negative impact on the environment and potentially on the public health.

Author contributions

José L. Domingo: Conceptualization, Methodology, Investigation, Writing-original draft, Visualization. Martí Nadal: Writing-review & editing. Joaquim Rovira: Investigation, Writing-original draft.

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Data availability statement

The data that support the findings of our studies on the topic are available from the corresponding author at reasonable request.

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TABLE and FIGURES

Table 1. Environmental levels of PCDD/Fs in soils and air samples, and human exposure for the population living in the neighborhood of the MSWI of Sant Adrià de Besòs. Data for the 2014 and 2017 surveys (Source: Domingo et al.[34])

	PCDD/Fs [a]		**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

[a]Units: Soils: ng WHO-TEQ/kg; Air: pg WHO-TEQ/m^3.

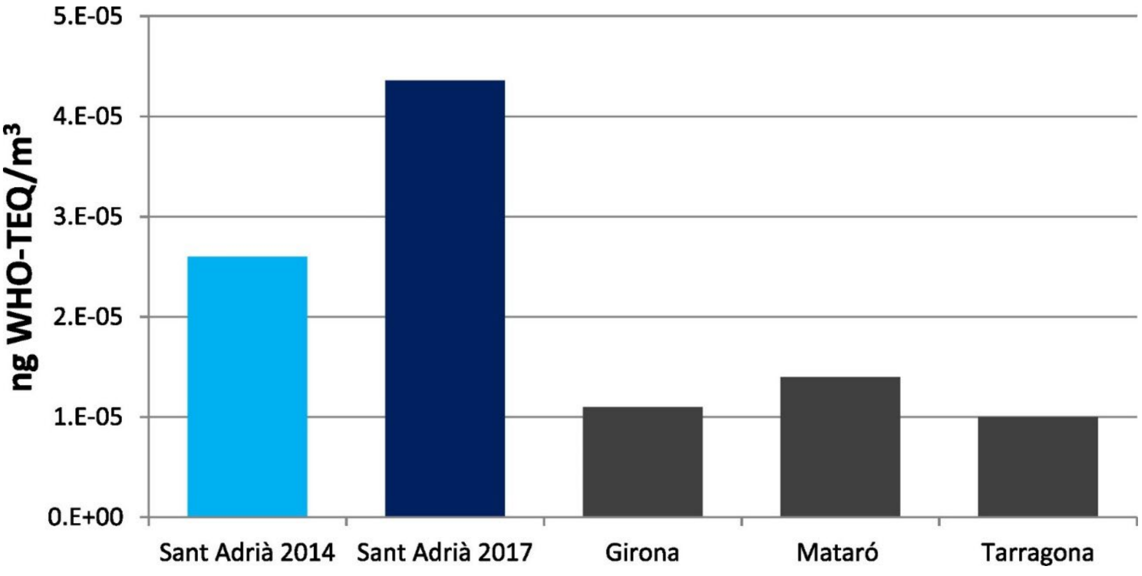

Figure 1. Levels of PCDD/Fs in air samples collected around the MSWI of Sant Adrià de Besòs in 2014 and 2017, as well as in the vicinity of other incinerators located in Catalonia (Source: Domingo et al.[34])

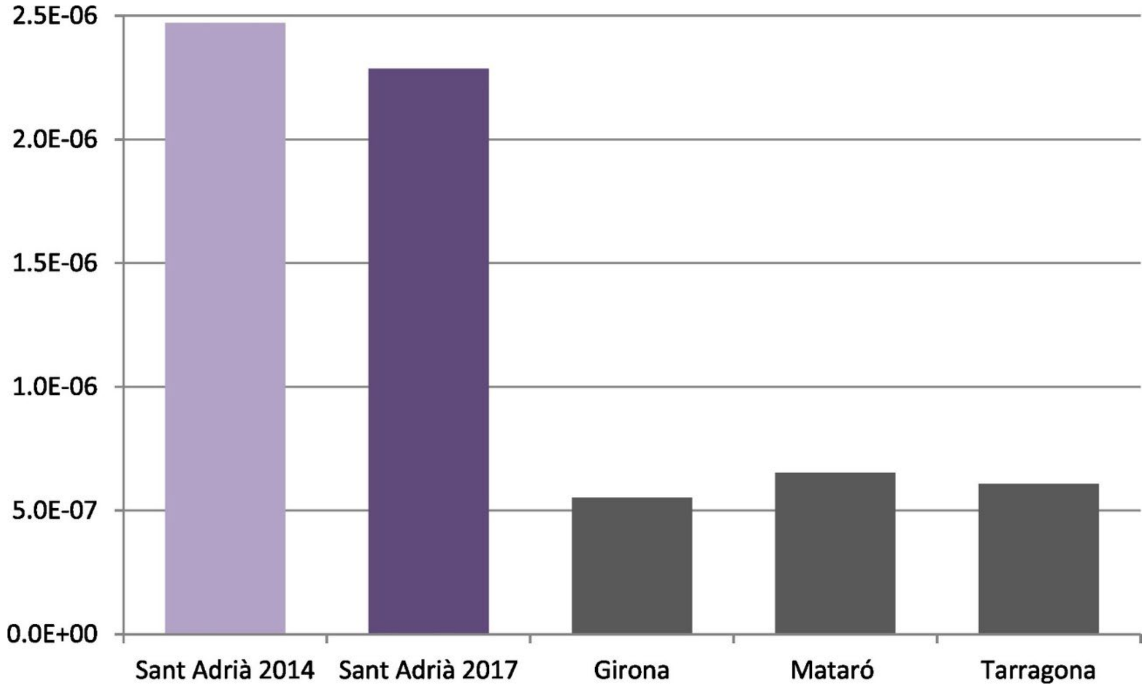

Figure 2. Cancer risks due to exposure to PCDD/Fs for the population living near the MSWI of Sant Adrià de Besòs (2014 and 2017), as well as for residents in the vicinity of other facilities located in Catalonia (Source: Domingo et al.[34])