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# Health risks for the population living in the vicinity of an Integrated Waste Management Facility: Screening environmental pollutants

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## HIGHLIGHTS

- Health risks of an Integrated Waste Management Facility in Catalonia are assessed.
- PCDD/F exposure near this facility is up to 10-times higher than that near others.
- Environmental monitoring of incineration plants should be performed case-by-case.
- Since results are very preliminary, confirmatory studies should be conducted.

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## ABSTRACT

We performed a screening investigation to assess the human health risks of the Integrated Waste Management Facility (IWMF: mechanical–biological treatment (MBT) plant plus municipal solid waste incinerator (MSWI); Ecoparc-3) of Barcelona (Spain). Air concentrations of pollutants potentially released by the MBT plant (VOCs and bioaerosols) and the MSWI (trace elements, PCDD/Fs and PCBs) were determined. Trace elements, PCDD/Fs and PCBs were also analyzed in soil samples. The concentrations of trace elements and bioaerosols were similar to those previously reported in other areas of similar characteristics, while formaldehyde was the predominant VOC. Interestingly, PCDD/F concentrations in soil and air were the highest ever reported near a MSWI in Catalonia, being maximum concentrations 10.8 ng WHO-TEQ/kg and 41.3 fg WHO-TEQ/$m^3$, respectively. In addition, there has not been any reduction in soils, even after the closure of a power plant located adjacently. Human health risks of PCDD/F exposure in the closest urban nucleus located downwind the MSWI are up to 10-times higher than those nearby other MSWIs in Catalonia. Although results must be considered as very preliminary, they are a serious warning for local authorities. We strongly recommend to conduct additional studies to confirm these findings and, if necessary, to implement measures to urgently mitigate the impact of the MSWI on the surrounding environment. We must also state the tremendous importance of an individual evaluation of MSWIs, rather than generalizing their environmental and health risks.



## 1. Introduction

In 2012, the annual amount of municipal solid waste (MSW) per capita generated in the EU-27 was 492 kg, following a slight decreasing trend (Eurostat, 2012). The disposal of such quantity of waste has become one of the most crucial environmental problems. A few years ago, landfills were predominantly used for MSW management, mainly because of the notably reduced operation costs. However, while materials and energy are not valorized in this practice, it is also associated with potential health risks (Rovira et al., 2012). Therefore, alternative methods have been being implemented. The European waste hierarchy developed in the EU 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, recovery, and disposal (Mari et al., 2009).

Waste incineration, also known as waste-to-energy, has become a serious option in developed and emerging countries (Cheng and Hu, 2010; Reddy, 2014). It presents numerous advantages such as energy recovery and volume minimization (Antonopoulos et al., 2014; Ferré-Huguet et al., 2006; Nadal et al., 2013; Pirotta et al., 2013; Schuhmacher et al., 1997). In addition, their contribution as releasers of a wide variety of chemical pollutants, including polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and heavy metals has notably decreased after the implementation of new legislative and technical regulation measures (Mari et al., 2010). However, public

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controversy is also present around municipal solid waste incinerators (MSWIs), which are commonly affected by the "Not In My Back Yard" (NIMBY) syndrome. Because of social and legislative pressures, complementary methods for managing the MSW have been introduced, being mechanical–biological treatment (MBT) one of the most extended (Garg, 2014). In Catalonia (Spain), MBT plants are called Ecoparcs. At the moment, there are 4 Ecoparcs within the metropolitan area of Barcelona (Catalonia, Spain), as well as other MBT plants outside (Mataró, Lloret de Mar, and Botarell). In 2006, Ecoparc-3 started to operate regularly. This MBT plant, located in Sant Adrià de Besòs, was built next to a MSWI operating since 1975. Overall, the MSWI and the MBT plant constitute the Integrated Waste Management Facility (IWMF) of Sant Adrià de Besòs, whose capacity is 192,000 tons/year for mechanical–biological treatment, and 350,000 tons/year for energy recovery. In the period 1998–2001, we performed a monitoring study aimed at determining the environmental impact of the MSWI after the assembly of a new gas cleaning system. We analyzed the levels of PCDD/Fs in samples of soil and vegetation collected nearby (Domingo et al., 2000, 2002a,b; Schuhmacher et al., 2000). In 2005, the facility was adapted to the environmental requirements of the European Directive 2000/76/EC, and the occurrence of PCDD/Fs, polychlorinated biphenyls (PCBs) and other persistent organic pollutants (POPs) in air around the facility was also determined by means of active and passive air sampling devices (Mari et al., 2008a,b). However, environmental data regarding the same chemicals, as well as other potentially generated by the MBT plants, both chemical and microbiological, are currently inexistent, even though Ecoparc-3 started to operate in 2006.

The purpose of the present preliminary study was to screen the concentrations of PCDD/Fs, dioxin-like PCBs (dl-PCBs), non-dioxin-like PCBs (ndl-PCBs), and elements in samples of air and soil collected in the vicinity of the IWMF of Sant Adrià de Besòs. Airborne levels of volatile organic compounds (VOCs) and bioaerosols (total bacteria and fungi) were also determined. Analytical data were used to assess human exposure to each one of the chemical pollutants, as well as to characterize the associated health risks for the adult population living in the neighborhood.

## 2. Materials and methods

### 2.1. Sampling

In May and June of 2014, samples of ambient air were collected in the vicinity of the IWMF of Sant Adrià de Besòs (Barcelona, Catalonia, Spain). The concentrations of a number of environmental pollutants potentially released by the MSWI and the MBT plant were determined. Two parallel samplings were conducted, depending on the analyzed contaminants. Four sampling points around the MSWI were selected to measure the levels of PCDD/Fs, PCBs and elements (Fig. 1). Because data were also used for health risk assessment, air sampling equipment was deployed in schools and sport centers, therefore highlighting children exposure to chemical pollutants. Prior to sampling, the AERMOD dispersion modeling software was executed to determine the main direction and spatial range of the air pollutants emitted by both facilities. Predominant wind directions were especially taken into account. Since

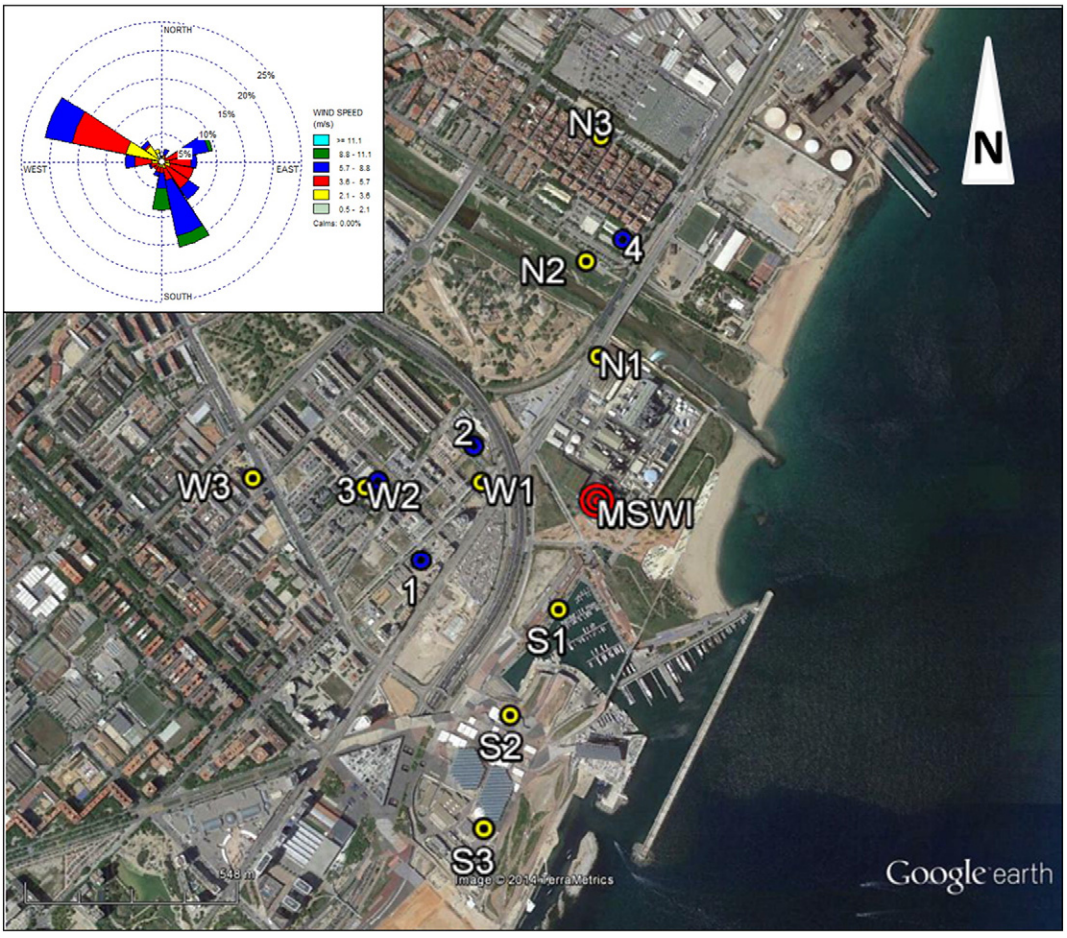


**Fig. 1.** Area of study and wind rose. Sampling points of PCDD/Fs, PCBs and metals (1, 2, 3, and 4) are marked in blue, while those of VOCs (N1, N2, S1, S2, W1, and W2) and bioaerosols (N1, N2, N3, S1, S2, S3, W1, W2, and W3) are marked in yellow. In red, MSWI.

wind mostly blows from W and S (Fig. 1), a specific air sample (#4) was also collected at the N of the MSWI. In contrast, no sampling was conducted in the E transect because the IWMF is situated on the shore. The methodology of sampling and analysis has been widely described in previous studies (Rovira et al., 2011, 2014), as well as in Supporting Information.

A different sampling was performed for VOCs and bioaerosols, following standard procedures (Vilavert et al., 2009, 2012a). A detailed description is given in Supporting Information. These agents are potentially released by MBT and composting plants (Domingo and Nadal, 2009). Points were located at different distances (300, 600 and 900 m) and directions (N, W and S) from the IWMF. Nine air samples were obtained for the analysis of microbiological pollutants, while VOCs were determined in the 6 points closest to the plant (N1, N2, W1, W2, S1 and S2). With respect to bioaerosols, five replicates of air samples were collected in each point. Airborne levels of total bacteria and total fungi, grown at two temperatures (25 °C and 37 °C), were subsequently determined. *Aspergillus fumigatus*, a fungus which may present a potentially remarkable risk for the population, and one of the most frequent species in the air of compost plants (Fischer et al., 1999), was also closely monitored.

### 2.2. Analytical procedure

Metals were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS), while the analysis of PCDD/Fs and PCBs in soil and air was done by high resolution gas chromatography coupled to high resolution mass spectrometry (HRGC/HRMS). The analytical determination of most VOCs was carried out by using a gas chromatograph coupled to a mass spectrometer (GC–MS). In turn, the analysis of formaldehyde was performed by high pressure liquid chromatography with UV detection. Further information on the analytical methods for chemicals is provided in Supporting Information. With respect to bioaerosol determination, once in the lab, Petri dishes were incubated for 48 h at 37 °C for bacterial analyses, as well as at 25 °C and 37 °C for fungi determination. After the incubation period, the number of colony-forming units (cfu) per air volume was counted by using microscopy. The presence of *A. fumigatus* in two of the five fungi dishes was also studied. Quality control/quality assurance (QC/QA) was checked by the analysis of blanks, internal reference samples and standard materials in each batch. Detection limits are detailed in Supporting Information.

### 2.3. Data analysis

Human exposure to chemical pollutants was evaluated by applying the US EPA Risk Assessment Guidance for Superfund (RAGS) methodology. For metals, PCDD/Fs and PCBs, exposure through 3 different pathways (soil ingestion, dermal absorption, and air inhalation) was assessed. In contrast, exposure to VOCs was only assessed by air inhalation, since this has been identified as the key route (Tan et al., 2013). Furthermore, a health risk assessment of exposure to chemicals was performed by differentiating non-cancer and cancer risks. The characterization of non-carcinogenic risks consisted of calculating the Hazard Quotient (HQ), which is defined as the relation between the predicted exposure and the reference dose (RfD). Cancer risks (CR) through ingestion and dermal exposure were estimated by multiplying the predicted exposure by the slope factor. The numerical expressions and parameters used for human exposure assessment and risk characterization were collected from US EPA (RAIS, 2013; Rovira et al., 2011). Because of the lack of threshold values to assess human health risks derived from microbiological exposure, these were only evaluated for chemical pollutants.

### 2.4. Statistics

Undetected pollutants were assumed to have a concentration equal to one-half of the limit of detection (ND = ½ LOD). Data analysis was carried out by means of the statistical software package SPSS 20.0. Because of the rather low number of samples, the significance of the differences in chemical levels between data sets was not determined. Regarding bioaerosols, significant differences ($p < 0.05$) were analyzed by applying the Levene test, and subsequently, an ANOVA or Mann–Whitney's U-test.

## 3. Results and discussion

### 3.1. Environmental concentrations

The metal levels of air and soil samples collected in the vicinity of the IWMF are summarized in Table 1. In air, Zn and Cu showed the highest mean concentrations (51.1 and 33.2 ng/m$^3$, respectively). By contrast, Be, Cr, Mo, and Tl were below their respective detection limits in all samples, while As, Cd and Hg were detected in only one sample. In soils, Mn and Zn showed the most elevated values (332 and 82.7 mg/kg, respectively), while Hg was the only element with undetected values (<0.10 mg/kg) in all samples. Metal concentrations in air and soils were within the same order of magnitude as those recently reported near other MSWIs in Catalonia, as well as worldwide (Bretzel and Calderisi, 2011; Vilavert et al., 2012b).

The total concentrations of PCDD/Fs and dioxin-like PCBs in air and soil samples from the surroundings of the IWMF are depicted in Fig. 2. The mean airborne TEQ level was 26.4 fg WHO-TEQ/m$^3$ (median: 22.9 fg WHO-TEQ/m$^3$), with minimum and maximum values of 18.5 and 41.3 fg WHO-TEQ/m$^3$, respectively. The contribution percentage of PCDD/Fs to the total concentration was very similar regardless of the sampling point, ranging between 82 and 85%. For PCDD/Fs, the highest level in air (35.1 fg WHO-TEQ/m$^3$) was observed in sample #4, located N and downwind the IWMF. Interestingly, this concentration is the highest ever reported near a MSWI in Catalonia. Since 1996, we have measured the environmental concentrations of PCDD/Fs and other chemical pollutants in the surroundings of the MSWI of Tarragona County (Spain) (Vilavert et al., 2012b). In 2007 and 2009, the mean

**Table 1**
Metal concentrations in air and soil samples collected around the IWMF of Sant Adrià de Besòs.

| | AIR (n = 4) | | | | | SOIL (n = 4) | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Mean | SD | Min | Max | Limit value[a] | Mean | SD | Min | Max | Limit value[b] |
| As | 0.20 | 0.15 | <0.26 | 0.42 | 6 | 5.82 | 5.91 | 1.36 | 14.4 | 30 |
| Be | <0.63 | – | – | – | – | 0.61 | 0.33 | 0.31 | 1.02 | 10 |
| Cd | 0.08 | 0.10 | <0.06 | 0.23 | 5 | 0.34 | 0.26 | 0.12 | 0.71 | 2.5 |
| Co | 0.26 | 0.10 | 0.15 | 0.39 | – | 4.59 | 1.93 | 2.87 | 6.45 | 25 |
| Cr | <0.63 | – | – | – | – | 11.7 | 7.23 | 6.32 | 22.3 | 400 |
| Cu | 33.2 | 13.5 | 20.8 | 52.5 | – | 19.5 | 10.9 | 11.1 | 35.4 | 90 |
| Hg | 0.21 | 0.17 | <0.25 | 0.47 | – | <0.10 | | | | 2 |
| Mn | 9.83 | 4.37 | 6.49 | 16.2 | – | 332 | 149 | 212 | 531 | – |
| Mo | <0.13 | – | – | – | – | 0.72 | 0.43 | 0.27 | 1.29 | 3.5 |
| Ni | 5.14 | 2.59 | 3.04 | 8.65 | 20 | 6.81 | 5.47 | 2.64 | 14.9 | 45 |
| Pb | 4.48 | 0.84 | 3.51 | 5.31 | 500 | 24.4 | 21.5 | 8.68 | 56.0 | 60 |
| Sb | 2.47 | 0.68 | 1.66 | 3.24 | – | 0.12 | 0.15 | 0.05 | 0.34 | 6 |
| Se | 0.59 | 0.32 | 0.26 | 1.01 | – | 0.72 | 0.18 | 0.52 | 0.96 | 0.7 |
| Sn | 3.75 | 0.65 | 2.85 | 4.39 | – | 1.15 | 0.84 | 0.54 | 2.37 | 50 |
| Sr | 3.31 | 0.67 | 2.75 | 4.28 | – | 66.7 | 61.4 | 20.5 | 155 | – |
| Tl | <0.06 | – | – | – | – | 0.12 | 0.03 | 0.09 | 0.17 | 1.5 |
| V | 9.40 | 3.53 | 6.88 | 14.39 | – | 35.1 | 8.39 | 27.2 | 45.6 | 135 |
| Zn | 51.1 | 6.37 | 43.8 | 58.7 | – | 82.7 | 34.6 | 54.1 | 129 | 170 |
| $PM_{10}$ | 33.3 | 7.30 | 26.5 | 42.0 | 40 | – | – | – | – | |

Units: Air: ng/m$^3$; Soil: mg/kg; $PM_{10}$: μg/m$^3$.
[a] According to Directives 2007/107/EC and 1999/30/CE.
[b] Most restrictive scenario according to the Reference Threshold Levels of the regional government (Agency of Waste of Catalonia).

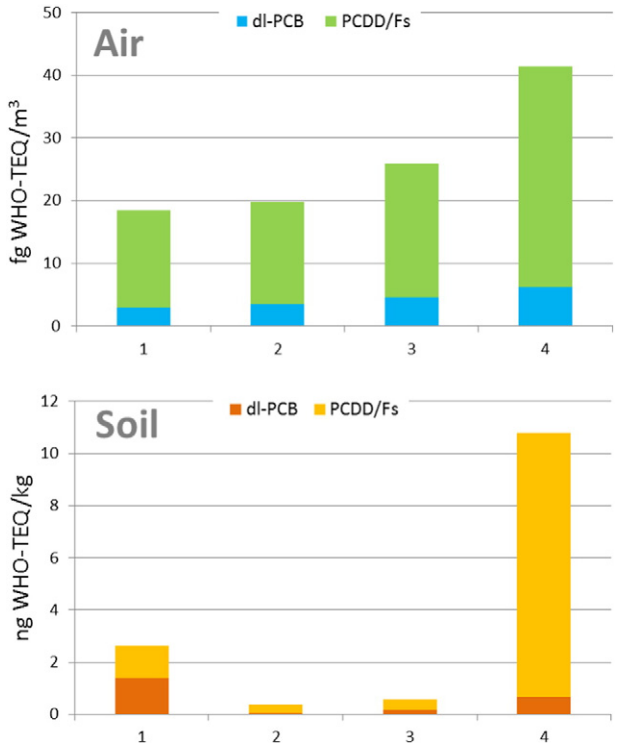


**Fig. 2.** Concentrations (WHO-TEQ) of PCDD/Fs and dioxin-like (dl) PCBs in 4 soil and 4 air samples collected near the IWMF of Sant Adrià de Besòs (Spain).

PCDD/F levels in 8 air samples collected by means of active air sampling devices were 12.0 and 8.6 fg WHO-TEQ/m$^3$, respectively, being 3- and 4-fold lower than those currently observed near the IWMF of Sant Adrià de Besòs. In addition, a periodical assessment of airborne burdens of PCDD/Fs, PCBs and polychlorinated naphthalenes near the MSWI of Tarragona County was also performed by means of passive air sampling devices in the period 2011–2012, being PCDD/F mean levels 16.3 fg WHO-TEQ/m$^3$ (range: 11.7–26.5 fg WHO-TEQ/m$^3$). On the other hand, near the MSWI of Mataró (Catalonia, Spain), the mean concentration of PCDD/Fs in air was between 8 and 15 fg WHO-TEQ/m$^3$ (Rovira et al., 2010), sensitively lower levels than the maximum concentration found near the plant here evaluated. The MSWI of Sant Adrià de Besòs, which started to operate in 1975, has a treatment capacity of 350.000 tons/year. Contrastingly, the MSWIs of Tarragona and Mataró began their regular operations more recently, in 1991 and 1994, respectively, while their contractual capacity for treating MSW is 144.000 and 160.000 tons/year, respectively. In the area of Po Valley (Italy), air PCDD/F concentrations ranging between 22 and 39 fg I-TEQ/m$^3$ were found in 2000, three years after a MSWI started to operate (Caserini et al., 2004). In China, Gao et al. (2014) recently studied the spatial and seasonal distributions of PCDD/Fs around a MSWI by means of passive air samplers. Mean concentrations in air were 54.3 and 52.1 fg WHO-TEQ/m$^3$. However, data comparison between our study and that performed in China is rather difficult because sampling was conducted by different methods.

Some years ago, environmental levels of PCDD/Fs in the vicinity of the MSWI of Sant Adrià de Besòs were also monitored, and compared with data from background areas in the city of Barcelona. The first air monitoring study included the collection of samples in site #4. Air levels of PCDD/Fs in that point were 22, 20, 11 and 14 fg WHO-TEQ/m$^3$ in spring 2005, autumn 2005, spring 2006 and autumn 2006, respectively (Mari et al., 2008a). Consequently, a clear increase has been observed between 2006 and 2014 downwind the MSWI, even after the closure of a combined cycle power plant located nearby.

In soils, the mean concentration of PCDD/Fs and dioxin-like (dl) PCBs in 4 points around the IWMF of Sant Adrià de Besòs was 3.6 ng WHO-TEQ/kg (median: 1.6 ng WHO-TEQ/kg), with a range between 0.4 and 10.8 ng WHO-TEQ/kg. Similar to air, the highest levels of PCDD/Fs and dl-PCBs were found in sample #4, downwind the incineration plant. This clearly evidences that wind from S plays a key role in the dispersion of PCDD/Fs potentially emitted by the MSWI. The contribution percentage of dioxins and furans in soils with respect to the total TEQ was more variable than in the air matrix, ranging from 48 to 94%. The soil concentration of PCDD/Fs in sample #4 (10.1 ng WHO-TEQ/kg) is also notably higher than the levels previously found in the vicinity of other incineration plants across Catalonia. In 2007 and 2009, mean PCDD/F concentrations in 8 soil samples collected near the MSWI of Tarragona County were 0.64 and 0.58 ng I TEQ/kg, respectively (Vilavert et al., 2012b). These levels were very similar to those also found in 30 sampling points around the hazardous waste incinerator (HWI) of Constantí (Catalonia), where the mean PCDD/F concentrations were 0.75 and 0.67 ng I-TEQ/kg in 2009 and 2011, respectively (Mari et al., 2013). In 1998, a wide program was initiated to determine the levels of PCDD/Fs in soil and herbage samples collected in the neighborhood of the MSWI of Sant Adrià de Besòs. Three campaigns were performed between 1998 and 2000 (Domingo et al., 2000, 2002a,b; Schuhmacher et al., 2000). High concentrations of PCDD/Fs were noted in soils, with mean values ranging from 7.09 to 11.85 ng I-TEQ/kg (2000 and 1999, respectively). The current concentration of PCDD/Fs in sample #4 confirms that there has not been a reduction of pollutant levels downwind. This contrasts with the significant decrease of PCDD/F soil burdens observed in the remaining 3 sampling points.

From an international perspective, the value associated with PCDD/Fs in that location is relatively high. In Italy, dioxin concentrations within a range of 0.7–1.5 ng I-TEQ/kg[1] were found in soil samples collected near a MSWI in Po Valley (Caserini et al., 2004). Similarly, the mean PCDD/F concentration in 21 soil samples collected in the Tianjin municipality (China) was 1.08 ng I-TEQ/kg, with minimum and maximum concentrations of 0.47 and 2.07 ng I-TEQ/kg (Liu et al., 2013). Also in China, PCDD/F levels in agricultural soils at 41 sites within a radius of 3 km from two municipal solid waste incinerators in Shanghai were ranged from 0.64 to 61.15 ng I-TEQ/kg (Deng et al., 2011). This wide range of concentrations has been found not only in soils potentially impacted by MSWIs but also in industrial, urban and agricultural soils of different characteristics. For instance, in their extensive US review, Urban et al. (2014) identified an important variability of background levels of dioxin-like compounds in urban/suburban soils (0.1–186 ng/kg TEQ), as well as in rural soils (0.1–22.9 ng/kg TEQ). In Germany, reference values and recommended action for agricultural and horticultural land uses were issued in 1992. For preventive reasons and as a long-term objective, PCDD/F concentrations in soils used for agricultural purposes should be reduced below 5 pg TEQ/g. Moreover, the Canadian Council of Ministers of the Environment set a soil quality guideline of 4 ng TEQ/kg, irrespective of the land use.

The PCDD/F and PCB congener profiles in samples of air and soil collected in the vicinity of the IWMF of Sant Adrià de Besòs are depicted in Fig. S1 (Supporting Information). High-chlorinated PCDD/F congeners (OCDD, 1,2,3,4,6,7,8-HpCDD and OCDF) showed the highest levels in soils, while a certain increase of low-substituted furans was also noted. All 7 environmental marker PCBs were detected in samples of both monitors. However, a different PCB congener profile was found in air and soil. In agreement with data obtained from the scientific literature (Schuhmacher et al., 2004; Vilavert et al., 2014b), airborne concentrations of PCBs decreased with the chlorination degree, being the low-substituted congeners (#28 and #52) the predominant. In contrast, the contribution of heavy PCBs was more important in soil. Finally, all dioxin-like PCBs were detected in soil samples, while only PCBs #77, #126, #123, #105, and #156 showed levels above their respective detection limits in air.

The individual concentrations of 19 VOCs in 6 air samples collected in the surroundings of the IWMF are summarized in Table 2. Mean total VOC levels were 11.4 μg/m$^3$, ranging from 6.00 to 17.6 μg/m$^3$.

**Table 2**
Levels of 19 VOCs in air in 6 sampling points around the IWMF of Sant Adrià de Besòs (Spain).

| | N1 | N2 | S1 | S2 | W1 | W2 |
|---|---|---|---|---|---|---|
| Benzene | 0.12 | <0.06 | <0.06 | 0.31 | 0.61 | 0.33 |
| Toluene | 4.72 | 0.19 | 0.56 | 2.03 | 5.00 | 3.33 |
| Ethylbenzene | 0.22 | <0.06 | <0.06 | 0.44 | 0.56 | 0.61 |
| *m*,*p*-Xylene | 0.39 | <0.11 | 0.24 | 0.61 | 1.14 | 1.03 |
| *o*-Xylene | 0.26 | 0.06 | 0.19 | 0.39 | 0.86 | 0.61 |
| Styrene | <0.06 | <0.06 | <0.06 | <0.06 | <0.06 | <0.06 |
| Naphthalene | <0.06 | <0.06 | <0.06 | <0.06 | <0.06 | <0.06 |
| Methylene chloride | <0.06 | <0.06 | <0.06 | <0.06 | <0.06 | <0.06 |
| 1,2-Dichloroethane | <0.06 | <0.06 | <0.06 | 0.08 | 0.09 | 0.09 |
| Chloroform | 0.08 | <0.06 | <0.06 | 0.07 | 0.31 | 0.47 |
| Trichloroethylene | 0.09 | <0.06 | <0.06 | 0.23 | 0.18 | 0.31 |
| Tetrachloroethylene | 0.47 | <0.06 | <0.06 | 0.16 | 0.44 | 0.83 |
| 1,3-Butadiene | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 |
| 1,3,5-Trimethylbenzene | <0.06 | <0.06 | <0.06 | 0.08 | 0.16 | 0.10 |
| 1,2,4-Trimethylbenzene | 0.28 | <0.06 | 0.16 | 0.31 | 0.47 | 0.39 |
| *p*-Isopropyltoluene | 2.00 | <0.06 | <0.06 | 0.06 | 0.08 | 0.09 |
| *n*-Propylbenzene | <0.06 | <0.06 | <0.06 | <0.06 | <0.06 | 0.08 |
| Isopropylbenzene | <0.06 | <0.06 | <0.06 | <0.06 | <0.06 | ND |
| Formaldehyde | 5.56 | 3.89 | 3.50 | 6.67 | 6.11 | <0.06 |
| BTEX | 5.71 | 0.37 | 1.04 | 3.78 | 8.17 | 5.92 |
| VOCs | 15.8 | 6.00 | 6.42 | 13.0 | 17.6 | 9.75 |

Units: μg/m$^3$; BTEX: benzene, ethylbenzene, toluene and *o*-, *m*-, *p*-xylenes. Limit value for benzene in air is 5 μg/m$^3$ according to the Directive 2008/50/EC.

Formaldehyde was the predominant compound, with a mean concentration of 4.29 μg/m$^3$ and a range of <0.06–6.67 μg/m$^3$, and being detected in 5 of the 6 sampling points. BTEX (benzene, toluene, ethylbenzene, *m*,*p*-xylene, and *o*-xylene) were detected in the whole set of samples, excepting N2 and S1, in which benzene and ethylbenzene showed levels below their respective detection limits (<0.06 μg/m$^3$). The levels of BTEX ranged from 0.37 to 8.17 μg/m$^3$, with a mean contribution to $\sum$VOCs of 36%. However, the important range in the contribution percentage (6–61%) indicated the presence of a number of potential emission sources. Benzene is the only one of the measured VOCs whose levels are currently legislatively restricted in urban air. The maximum concentration of benzene (0.61 μg/m$^3$) was well below the threshold value (5 μg/m$^3$) set in the EC Air Quality Framework Directive (2000/69/EC). A detailed study of the concentrations of VOCs with respect to the distance to the MBT plant was also performed, comparing airborne levels at 300 and 600 m from the facility. A certain increase of VOC concentrations was found at the N (downwind) and W directions, being levels around 3- and 2-times higher, respectively, in the closest site. Despite the rather low number of samples, our findings suggest a potential impact of the MBT plant on the surrounding area. Anyhow, this pattern should be confirmed by a more extensive study, in which the number of samples is increased.

The environmental concentrations of VOCs were also low compared with those previously reported in the surroundings of similar waste management plants. In general terms, VOC levels are in the lowest part of the range, when compared to those observed near MBT plants and MSWIs. Recently, Vilavert et al. (2014a) found that mean concentrations of the same 19 VOCs here analyzed near Ecoparc-2, another MBT plant located in Montcada i Reixac (Barcelona, Spain), ranged between 9.12 and 45.7 μg/m$^3$. A clear seasonal trend was also noted, with higher VOC concentrations in winter campaigns. Since the sampling of the IWMF of Sant Adrià de Besòs was conducted in summer, an increase of VOCs should be presumably expected in winter times. The occurrence of these chemicals was also studied in the vicinity of the MSWI of Tarragona, prior to the construction of an MBT plant (Vilavert et al., 2009, 2011). Although this facility was not finally built, data were used to determine the impact of a waste incinerator on the surrounding environment, in terms of chemical and microbiological pollutants other than PCDD/Fs and metals. Mean VOC values ranged from 7.6 to 18.2 μg/m$^3$, being of the same order of magnitude as those currently observed near Ecoparc-3. Special attention was paid to formaldehyde, a chemical of concern in the vicinity of MBT plants of solid waste. Near the Ecoparc-2, mean air levels of formaldehyde were up to 5.48 μg/m$^3$, while this chemical was undetected in the surroundings of the MSWI of Tarragona. Airborne concentrations of formaldehyde near the Ecoparc-3 are very similar to those previously observed near other MBT plants. However, they are sensitively higher than typical values from urban areas across the European Union (Bruinen De Bruin et al., 2008).

The airborne concentrations of total bacteria and fungi were also determined in a number of sampling sites around the IWMF of Sant Adrià de Besòs. Mean levels of total bacteria were 521 cfu/m$^3$, while those of total fungi, grown at 25 °C and 37 °C, were 616 and 70 cfu/m$^3$, respectively. An important variability was noted for all bioaerosols, whose concentration ranges were 80–3190, 20–1970, and ND-260 cfu/m$^3$ for total bacteria, fungi at 25 °C, and fungi at 37 °C, respectively. The airborne levels of microbiological pollutants according to the distance and direction from the MBT plant are shown in Fig. S2 (Supporting Information). Although a certain reduction of total bacteria with respect to the distance was noted, the difference between sampling points did not reach the level of statistical significance. However, a significant increase of fungi (at both temperatures) was observed at 600 m ($p < 0.05$). When evaluating the potential release of bioaerosols from the MBT plant according to the sampling direction, a higher impact was not observed in those sites located downwind. S points presented a significantly greater concentration of total bacteria, while W direction showed significantly higher levels of fungi ($p < 0.05$). These findings are in disagreement with those previously found near the Ecoparc-2, where a significant correlation of fungi at 37 °C and the distance from the MBT plant was observed (Vilavert et al., 2014a). Furthermore, the levels of *A. fumigatus* were up to 12-fold higher near (300 m) the plant than in the farthest (900) points. In the present study, *A. fumigatus* was not detected in any of the 9 samples. The current concentrations of total bacteria are similar to those found near another MBT plant (Ecoparc-2), where the mean airborne level of 4 sampling campaigns was 628 cfu/m$^3$. In turn, they are slightly higher than those found near the MSWI of Tarragona (521 vs. 279 cfu/m$^3$). In contrast to the current data, fungi levels were substantially lower than those previously reported elsewhere (Vilavert et al., 2011, 2014a).

### 3.2. Human health risks

The concentrations of elements, PCDD/Fs, PCBs, and VOCs were used to estimate the human exposure to these chemical pollutants. Three different exposure pathways (soil ingestion, dermal absorption, and air inhalation) were considered for metals and chlorinated compounds, while VOC exposure was assumed to occur exclusively via inhalation. The human exposure to each one of the chemicals here evaluated is summarized in Table 3. Metal exposure ranged between $5.81 \cdot 10^{-8}$ and $5.39 \cdot 10^{-4}$ mg/kg/day (Hg and Mn, respectively). Ingestion was the predominant exposure pathway for metals, except As, for which dermal absorption contributed 52%, as well as for Hg and Sb, whose exposure mainly occurred through air inhalation. In terms of TEQ, exposure to PCDD/Fs and dl-PCBs was $1.58 \cdot 10^{-5}$ and $6.50 \cdot 10^{-6}$ ng WHO-TEQ/kg/day, respectively, while environmental exposure to PCBs in terms of total concentration was 0.008 and 0.061 ng/kg/day for dl-PCBs and ndl-PCBs, respectively. Dermal absorption was the main pathway for PCBs, while similar contributions of PCDD/F exposure were estimated for each route (30%, 32% and 38% for soil ingestion, dermal absorption and air inhalation, respectively).

With respect to VOCs, formaldehyde showed the highest exposure, with a mean level of $1.40 \cdot 10^{-3}$ mg/kg/day, a close value to that calculated for the adult population living near the Ecoparc-2. In fact, the overall exposure to chemicals was very similar to those obtained in previous investigations performed in Catalonia, with the only exception of PCDD/Fs. According to our estimations, the population living in the surroundings of the MSWI of Sant Adrià de Besòs is, at least, 3-times more

**Table 3**
Environmental exposure (in mg/kg/day) to chemical pollutants for the adult population living near the IWMF of Sant Adrià de Besòs (Spain). Contribution percentage of three exposure routes.

| | Total exposure | Contribution (%) | | |
|---|---|---|---|---|
| | | Ingestion | Dermal | Inhalation |
| As | 1.88E−05 | 48 | 52 | 0 |
| Be | 9.83E−07 | 97 | 3 | NC |
| Cd | 5.64E−07 | 93 | 3 | 4 |
| Co | 7.49E−06 | 96 | 3 | 1 |
| Cr | 1.89E−05 | 97 | 3 | NC |
| Cu | 4.06E−05 | 75 | 3 | 22 |
| Hg | 5.81E−08 | NC | NC | 100 |
| Mn | 5.39E−04 | 96 | 3 | 1 |
| Mo | 1.17E−06 | 97 | 3 | NC |
| Ni | 1.24E−05 | 86 | 3 | 11 |
| Pb | 4.08E−05 | 94 | 3 | 3 |
| Sb | 8.77E−07 | 22 | 1 | 77 |
| Se | 1.32E−06 | 85 | 3 | 12 |
| Sn | 2.88E−06 | 62 | 2 | 36 |
| Sr | 1.09E−04 | 96 | 3 | 1 |
| Tl | 1.91E−07 | 97 | 3 | NC |
| V | 5.94E−05 | 93 | 3 | 4 |
| Zn | 1.48E−04 | 87 | 3 | 10 |
| PCDD/Fs | 1.58E−11 | 30 | 32 | 38 |
| dl-PCBs | 8.52E−09 | 13 | 67 | 20 |
| ndl-PCB | 6.16E−08 | 11 | 54 | 35 |
| Benzene | 9.36E−05 | NA | NA | 100 |
| Toluene | 7.23E−04 | NA | NA | 100 |
| Ethylbenzene | 1.26E−04 | NA | NA | 100 |
| *m*-, *p*-Xylene | 1.87E−04 | NA | NA | 100 |
| *o*-Xylene | 1.08E−04 | NA | NA | 100 |
| Styrene | NC | NA | NA | NC |
| Naphthalene | NC | NA | NA | NC |
| Methylene chloride | NC | NA | NA | NC |
| 1,2-Dichloroethane | 2.41E−05 | NA | NA | 100 |
| Chloroform | 6.35E−05 | NA | NA | 100 |
| Trichloroethylene | 5.52E−05 | NA | NA | 100 |
| Tetrachloroethylene | 1.31E−04 | NA | NA | 100 |
| 1,3-Butadiene | NC | NA | NA | NC |
| 1,3,5-Trimethylbenzene | 3.07E−05 | NA | NA | 100 |
| 1,2,4-Trimethylbenzene | 8.77E−05 | NA | NA | 100 |
| *p*-Isopropyltoluene | 1.52E−04 | NA | NA | 100 |
| *n*-Propylbenzene | 2.13E−05 | NA | NA | 100 |
| Isopropylbenzene | NC | NA | NA | NC |
| Formaldehyde | 1.40E−03 | NA | NA | 100 |

NC: Not calculated because environmental levels were below detection limit; NA: Not assessed.

exposed to PCDD/Fs than people living in the vicinity of other waste incinerators across Catalonia. Moreover, PCDD/F exposure is one order of magnitude (10-times) higher in the downwind area, where the urban nucleus of Sant Adrià de Besòs is specifically located. The environmental exposure to PCDD/Fs for the population living in the vicinity of the MSWIs of Tarragona and Mataró has been estimated in $4.48 \cdot 10^{-6}$ and $4.47 \cdot 10^{-6}$ ng WHO-TEQ/kg/day, respectively (Rovira et al., 2010; Vilavert et al., 2012b). Similarly, the human exposure of PCDD/Fs for residents nearby the hazardous waste incinerator of Constantí (Tarragona, Spain) has been estimated in $5.04 \cdot 10^{-6}$ ng WHO-TEQ/kg/day (Mari et al., 2013). Despite the low number of current sampling sites, this is a clear evidence of the high impact of the MSWI of Sant Adrià de Besòs on the population living in the neighborhood. It is well established that diet is the main exposure pathway of PCDD/Fs, with percentages of >95% (Malisch and Kotz, 2014; Linares et al., 2010). In Catalonia, the dietary intake of PCDD/Fs has been estimated in $1.12 \cdot 10^{-3}$ ng WHO-TEQ/kg body weight per day (Llobet et al., 2008). When comparing the environmental (direct) and dietary (indirect) exposure pathways, the intake of PCDD/Fs through food consumption continues to be the predominant route. For the citizens of Sant Adrià de Besòs, the environmental exposure to PCDD/Fs, occurring through air inhalation, soil ingestion and dermal absorption, was calculated in 3.6%.

Health risks associated with environmental exposure to chemical pollutants were also characterized. With respect to non-carcinogenic risks (HQ < 1), none of the evaluated chemicals exceeded the threshold value (Table 4). However, the Hazard Index (HI), calculated as the sum of the HQ corresponding to each chemical, exceeded the safe value (HI = 1.6). As it is well known, people are not environmentally exposed to individual pollutants, but to chemical mixtures. Consequently, non-cancer risks associated with this exposure must be evaluated from an integrated perspective. Although potential interactions occur in the environment, one of the most usual approaches is considering an additive exposure (Kumar et al., 2013).

We also assessed cancer risks associated with exposure to carcinogenic substances (Fig. 3). Formaldehyde was the chemical presenting the highest risk, with a risk index of $2.7 \cdot 10^{-5}$. Arsenic and Cr also exceeded the threshold value set by the Spanish legislation at $10^{-5}$. However, this would be due to a risk overestimation related to elemental speciation. The carcinogenicity of elements depends basically on the species and the exposure pathway. Thus, inorganic As is the carcinogenic species of this element, although it does not mean 100% of the total As in the environment. Similarly, only Cr-VI is carcinogenic, while most of the Cr present in the environment is in the form of Cr-III. Since the concentrations of elemental species were not determined, a worst-case scenario was taken into account for health risk assessment calculations: a) 100% of total As was here assumed to be present as inorganic As, and b) the concentration of Cr-VI was estimated to be 1/6 of total Cr. These unlikely scenarios gave place to an overestimation of the exposure and the associated health risks. In any case, the current cancer risks due to metal exposure are similar to those previously found in other industrial and urban areas worldwide.

To the best of our knowledge, this is the first human health risk assessment study performed in the vicinity of the IWMF of Sant Adrià de Besòs, after the construction of the MBT plant. A number of contaminants potentially generated during thermal (incineration) and mechanical–biological treatment processes of MSW have been considered. Although none of chemicals exceeded the maximum recommended values according to national and EU guidelines, non-cancer risks associated with the exposure to all of them were above threshold, reaching the HI a level of 1.6. Humans are exposed to chemical mixtures instead of single substances. It means that the exposure assessment of environmental pollutants must be conducted by considering all the chemicals as a whole. Additional efforts should be made to assure that the human exposure to all environmental contaminants is minimized, beyond the simple law accomplishment of the industrial sector regarding air emissions. It means the involvement of a number of stakeholders,

**Table 4**
Non-carcinogenic risks (Hazard Quotient) of environmental exposure to chemical pollutants in the surroundings of the IWMF of Sant Adrià de Besòs (Spain).

| | | | |
|---|---|---|---|
| dl-PCBs | 1.1E−03 | VOCs | |
| PCDD/Fs | 1.4E−02 | Benzene | 1.1E−02 |
| Metals | | Toluene | 5.1E−04 |
| As | 7.5E−02 | Ethylbenzene | 4.4E−04 |
| Be | 2.9E−03 | *m*-, *p*-Xylene | 6.5E−03 |
| Cd | 8.9E−03 | *o*-Xylene | 3.8E−03 |
| Co | 6.7E−02 | Styrene | NC |
| Cr | 1.5E−02 | Naphthalene | NC |
| Cu | 3.5E−01 | Methylene chloride | NC |
| Mn | 2.3E−01 | 1,2-Dichloroethane | 1.2E−02 |
| Mo | 2.3E−04 | Chloroform | 2.3E−03 |
| Ni | 1.0E−03 | Trichloroethylene | 9.7E−02 |
| Pb | 5.6E−03 | Tetrachloroethylene | 1.1E−02 |
| Sb | 6.0E−04 | 1,3-Butadiene | NC |
| Se | 2.6E−04 | 1,3,5-Trimethylbenzene | 1.8E−02 |
| Sn | 3.1E−06 | 1,2,4-Trimethylbenzene | 4.4E−02 |
| Sr | 1.8E−04 | *p*-Isopropyltoluene | NC |
| Tl | 1.9E−02 | *n*-Propylbenzene | 7.5E−05 |
| V | 1.2E−01 | Isopropylbenzene | NC |
| Zn | 4.5E−04 | Formaldehyde | 5.0E−01 |

NC: Not calculated because environmental levels were below detection limit.

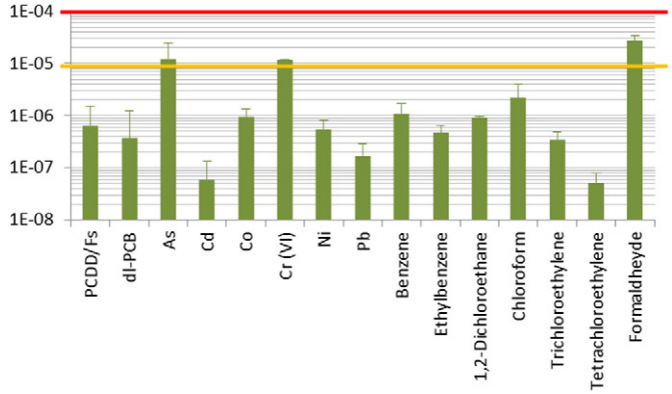


**Fig. 3.** Cancer risks of environmental exposure to chemical pollutants near the IWMF of Sant Adrià de Besòs (Spain).

including waste management companies, national, regional and local administrations, civil society, healthcare facilities, as well as universities and research institutions, among others.

## 4. Conclusions

Our results clearly show the tremendous importance of performing monitoring studies in a case-by-case basis, when evaluating the human health risks associated with exposure to chemical and microbiological contaminants released by MSWIs and MBT plants, among other industrial activities (i.e., cement plants, oil refineries, power plants, iron and steel industries, landfills, etc.). Even working under similar operational conditions, the characteristics of each waste management plant (MSWI/MBT) may substantially differ. Moreover, the environmental status of each particular area, and the presence of other potential emission sources, such as traffic, plays a key role in the occurrence of environmental pollutants in the vicinity of those facilities. If information regarding the environmental impact of new waste management facilities is important to assure the harmlessness for the surrounding population, these data become fundamental when evaluating the potential consequences for the environment and the human health of old, big incineration plants, such as the MSWI here assessed. Unfortunately, human health risk assessment studies around the IWMF of Sant Adrià de Besòs are scarce (if any), either before or after the construction of the MBT plant.

Despite the current findings are rather preliminary, given the reduced number of samples, our results are a serious warning for policymakers. Subsequent studies should be immediately performed to assure that current concentrations of PCDD/Fs are not higher in the downwind area by enlarging the number of analyzed samples in both soil and air. Furthermore, a biological monitoring of PCDD/Fs in the population of Sant Adrià de Besòs would be desirable to determine the human exposure to those chemical pollutants. In order to protect the health of the inhabitants of the area under potential influence of the MSWI, measures to reduce the levels of chemical pollutants in those zones and to mitigate the impact of that facility on the surrounding environment, must be urgently carried out.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2015.03.010.

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