

1 **Concentrations of trace elements and PCDD/Fs around a municipal solid waste**
2 **incinerator in Girona (Catalonia, Spain). Human health risks for the population living**
3 **in the neighborhood.**

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5 Joaquim Rovira ^{1,2}, Martí Nadal ¹, Marta Schuhmacher ^{1,2}, José L. Domingo ¹

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9 *1. Laboratory of Toxicology and Environmental Health, School of Medicine, IISPV,*
10 *Universitat Rovira i Virgili, Sant Llorenç 21, 43201 Reus, Catalonia, Spain*

11 *2. Environmental Engineering Laboratory, Departament d'Enginyeria Química, Universitat*
12 *Rovira i Virgili, Av. Països Catalans 26, 43007 Tarragona, Catalonia, Spain*

13

14 Corresponding author: J. Rovira, joaquim.rovira@urv.cat; *Phone:* +34 977 55 85 53; *Fax:*
15 *+34 977 55 96 21*

16

17 **Abstract**

18 Previously to the modernization of the municipal solid waste incinerator (MSWI) of
19 Campdorà (Girona, Catalonia, Spain) two sampling campaigns (2015 and 2016) were
20 conducted. In each campaign, 8 soil and 4 air samples (PM₁₀ and total particle phase and
21 gas phase) were collected. The levels of As, Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, Sb, Sn, Tl and
22 V, and PCDD/Fs were analysed at different distances and wind directions around the
23 MSWI. Environmental levels of trace elements and PCDD/Fs were used to assess exposure
24 and health risks (carcinogenic and non-carcinogenic) for the population living around the
25 facility. In soils, no significant differences were observed for trace elements and PCDD/Fs
26 between both campaigns. In air, significant higher levels of As, Cd, Co, Mn, Ni, Pb, Tl and
27 V were detected in 2016. Regarding soil levels, only Cd (distances) and As, Cu, Mn, and Ni
28 (wind directions) showed significant differences. No differences were noted in the
29 concentrations of trace elements and PCDD/Fs in air levels with respect to distances and
30 directions to the MSWI. No differences were registered in air levels (elements and
31 PCDD/Fs) between points influenced by MSWI emissions and background point. However
32 some differences in congener profile were noted regarding from where back-trajectories
33 come from (HYSPLIT model results), pointing some influence of Barcelona metropolitan
34 area. The concentrations of trace elements and PCDD/Fs were similar -or even lower- than
35 those reported around other MSWIs in Catalonia and various countries. Non-carcinogenic
36 risks were below the safety limit (HQ < 1). In turn, carcinogenic risks due to exposure to
37 trace elements and PCDD/Fs were in acceptable ranges, according to national and
38 international standard regulations.

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40 Keywords: Trace elements, dioxins and furans, soil, air, MSWI, back-trajectories, risk
41 assessment

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44 **1. INTRODUCTION**

45 In 2015, each habitant in Catalonia generated a mean of 474 kg of waste, being 301 kg
46 (64%) treated as residual waste fraction (ARC, 2016). Educational and advertising
47 campaigns focused on enhancing the recycle among general public have been conducted.
48 Since 2000, residual waste fraction decreased from 2.99 M tones to 2.26 M tones in 2015
49 (ARC, 2016). This means a reduction from 86% to 61% regarding total waste generated
50 (ARC, 2016). Despite the increase of reuse and recycle practice, the residual waste fraction
51 should be treated. The Directive 2008/98/EC (European Union Parliament and Council,
52 2008a) establishes a waste hierarchy with prevention, re-use and recycling being the three
53 top of the rank. The waste hierarchy also indicates that “other recovery”, such energy
54 recovery, is a better option than disposal, which is the last option in waste treatment
55 hierarchy.

56 Modern municipal solid waste incinerators (MSWIs) are adapted to recovery energy (heat
57 or electricity) through the incineration process. In addition to energy recovery, MSWIs
58 minimize the volume of residues. Moreover, modern MSWIs are operating with the best
59 available technologies (BATs), which ensure a high energetic efficiency in the incineration
60 process, as well as a rather low pollutant emission to the environment. However, in spite of
61 this, all combustion processes can generate organic compounds that could be released to the
62 environment with other pollutants such particle matter containing metals and metalloids
63 (Lin et al., 2016; Margallo et al., 2015; Yang et al., 2016). Among other substances,
64 MSWIs release to the environment polychlorinated dibenzo-p-dioxins and dibenzofurans
65 (PCDD/Fs) (Wang et al., 2010; Wang et al., 2013). In relation to this, a considerable
66 number of studies over the world have been carried out to monitor, control, and assess the
67 impact of MSWI emissions (Caserini et al., 2004; Jin et al., 2012; Liu et al., 2013; Nzihou
68 et al., 2012; Venturini et al., 2013). Our research group has been one of the most active in
69 this topic (Nadal et al., 2002; Schuhmacher et al., 1997a,b, 1998, 1999; Vilavert et al.,
70 2011, 2013, 2015a,b).

71 The current study, immision levels of trace elements (metals and metalloids) and PCDD/Fs
72 were determined in air and soils around the Campdorà MSWI, in two sampling campaigns
73 (2015 and 2016), previously to a modernization of the facility. By 2020, the waste

74 treatment capacity will be increased up to 45,000 tons annually, whilst several technical
75 improvements, including new gas control systems, will be also performed. Nowadays, this
76 facility, located in Girona (Catalonia, NE Spain), have a capacity to treat 29,000 tons of
77 waste annually. The waste treated (residual waste fraction) comes from three municipalities
78 (Girona, Salt, and Sarrià de Ter) with a total population of 115,000 inhabitants. The facility
79 is placed a hundred kilometers north from Barcelona metropolitan area and could be
80 influenced by mid-range transport of pollutants. Air and soil levels were used to assess
81 human exposure and health risks for the population living around the facility. The influence
82 of mid and long-range transport of pollutants was studied. The design of the present study
83 was similar to those previously conducted around some MSWIs in Catalonia (NE Spain),
84 during the last 20 years long (Domingo et al., 2015; Mari et al., 2007; Rovira et al., 2010,
85 2015; Vilavert et al., 2012b, 2014, 2015a).

86

87 **2. MATERIALS AND METHODS**

88 **2.1. Sampling**

89 In February 2015 and again in February 2016, 8 soil and 4 air samples were collected in the
90 surroundings of the Campdorà's MSWI. Sampling points are shown in Fig. 1. They are
91 located at different distances (<1km (sampling points #2, 6, and 7) and >1km (sampling
92 points #1, 3, 4, 5, and 8)) and wind directions (NE, sampling points #3, 4, 6, and 8; SW,
93 sampling points #1 and 7, and W, sampling points #2 and 5) (Supplementary information
94 Table S1). Selection of sampling point where done according predominant winds in the
95 area, blowing from south, south-west, and east following the valley where the MSWI is
96 placed, and the presence of inhabited nuclei. According to these prevalent winds, point 1
97 was considered for their location (SW) and distance (4.2 km) regarding MSWI as a
98 background point. Mean temperatures were 7°C (range: -1 to 17°C) and 8°C (range: -4 to
99 22°C) during the 2015 and 2016 sampling campaigns, respectively. No precipitation was
100 detected during the samplings.

101 Soil sampling was described previously (Vilavert et al., 2015b). Briefly, around 500 g of
102 superficial soil (0-5 cm) constituted by four subsamples in an area of 10 m² were collected

103 in a polyethylene bag. Once in the laboratory, soil samples were dried at room temperature,
104 and sieved through a 2 mm mesh screen. Air sampling is described elsewhere (Nadal et al.,
105 2015). Two kind of air samples were collected. 1) PM₁₀ in quartz fiber filter (QFF) were
106 sampled in order to analyze metals, and 2) total suspended particles (particle phase) in
107 quartz fiber filter (QFF) and gas phase were collected in polyurethane foam (PUF) to
108 determine PCDD/Fs. PM₁₀ and Particle (QFF) plus gas phase (PUF) were collected using
109 high volume samplers (TE-1000-PUF and TE-6070-DV, respectively). Sampling volumes
110 were within the range 480-620 m³ in 48 hr for QFF+PUF, and 1734-1945 m³ in 24 hr for
111 particulate matter (PM₁₀).

112 **2.2. Analytical determinations**

113 For determining trace elements in PM₁₀, a part of QFF sheet was digested with HNO₃ and
114 HF in hermetic Teflon bombs for 8 hr at room temperature, and for 8 hr at 80°C (Mari et
115 al., 2009). For soil samples, acid digestion with HNO₃ was done in a Microwave digestion
116 system (Milestone Start D) for 10 min, until reaching 165°C, and maintaining this
117 temperature for 20 min (Vilavert et al., 2015a). Finally, digested extracts were fill up with
118 distilled water and kept frozen until the analyses. The concentrations of As, Cd, Co, Cr, Cu,
119 Hg, Mn, Ni, Pb, Sb, Sn, Tl and V were measured by inductively coupled plasma-mass
120 spectrometry (ICP-MS). Recovery percentages for air and soil samples were from 93 to
121 109% and from 82 to 108%, respectively.

122 For PCDD/Fs analysis, the contents in particle (QFF) and gas (PUF) phase were analysed
123 together. Samples were extracted with Soxhlet using toluene. A multi-step clean-up with
124 multilayer silica columns was performed to remove the matrix and potential interfering
125 components. The PCDD/F fraction was collected and concentrated near dryness.
126 Determinations were done by high resolution gas chromatography coupled to high
127 resolution mass spectrometry (HRGC/HRMS) (Mari et al., 2009). Soil and air analyses
128 were based on the US EPA method 1613 and the German VDI 3499 method, respectively.
129 Appropriate labeled extraction standards (¹³C₁₂-PCDD/Fs substituted congeners) were
130 added in order to control the whole sample preparation process and to evaluate potential
131 losses. Recovery percentages for air and soil samples were ranged from 52 to 107% and
132 from 42 to 120%, respectively.

133 Blanks and standards, as well as duplicate of samples were used to assure the quality of the
134 analytical results.

135 **2.3 Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model**

136 HYSPLIT model, one of the most widely used, is “a complete system for computing simple
137 air parcel trajectories as well as complex transport, dispersion, chemical transformation,
138 and deposition simulations” developed by NOAA Air Resources Laboratory's (ARL)
139 (Rolph et al. 2017; Steinet al. 2016). HYSPLIT model was applied to calculate 24-hours
140 back-trajectories during air sampling campaign at 2-hour time resolution and a new
141 trajectory was calculated every 2 hours during the 48 hours of sampling campaign. In the
142 present work the calculation of back-trajectories has two main objectives: 1) to establish
143 possible long-mid range transport of pollutants, and 2) to determine the local sources
144 influence during air sampling collection.

145 **2.4. Human health risks**

146 The concentrations of trace elements and PCDD/Fs in soil and air were used to evaluate the
147 exposure of residents nearby the MSWI. Exposure pathways taken into account were soil
148 ingestion, dermal contact with soil and dust, and air inhalation. Expressions to calculate the
149 soil ingestion and dermal contact exposure were taken from Spanish Royal Decree 9/2005
150 (Ministerio de Medio Ambiente, 2007), which in turn is based on US EPA RAGS
151 methodology (US EPA, 1989). Expressions to calculate inhalation exposure were taken
152 from inhalation dosimetry methodology of US EPA RAGS (US EPA, 2009a).
153 Toxicological values were obtained from the Risk Assessment Information System
154 database (RAIS, 2013), with the exception of the oral reference dose for Pb, which was
155 taken from the WHO (Seiler et al., 1988). To assess carcinogenic and non-carcinogenic
156 risks from dermal exposure, the oral reference dose (RfDo) and the oral slope factor (SFo)
157 were multiplied and divided, respectively, by the gastrointestinal absorption factor (GI_{ABS})
158 (US EPA, 1989).

159 **2.5. Data analysis**

160 For non-detected (ND) congeners of PCDD/Fs and trace elements, levels below the
161 respective limit of detection (LD) were assumed to be equal to one-half of that limit (ND =
162 $\frac{1}{2}$ LD). Data analysis was carried out with the software package XLSTAT (Version
163 2015.2.02.18681). The level of significance was set at a probability lower than 0.05
164 ($p < 0.05$). To evaluate significant differences between groups, the Levene test was applied
165 to verify the equality of variances. ANOVA or Kruskal Wallis tests were subsequently
166 applied, depending on whether the data followed a normal distribution or not, respectively.
167 Principal component analysis (PCA) was applied to reduce the number of variables
168 extracting as much information as possible. It has been successfully applied to study the
169 influence of pollutant emission sources as well as to detect particular “hot spots” of
170 contamination (Mari et al. 2008; Vilavert et al. 2010).

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172 **3. RESULTS AND DISCUSSION**

173 **3.1. Environmental levels**

174 Table 1 and Table S2 summarize the concentrations of trace elements and PCDD/Fs levels
175 in soil samples collected around Campdorà’s MSWI in 2015 and 2016. Mercury was
176 detected only in one point in 2015 (0.04 mg kg^{-1} in point 7) and in 3 points in 2016 (0.03 ,
177 0.02 , and 0.02 mg kg^{-1} in points 3, 4, and 7, respectively), while Sb was not detected in
178 2015 ($< 0.05 \text{ mg kg}^{-1}$) at any sampling point. In contrast, Mn (316 and 344 mg kg^{-1}), Cu
179 (20.6 and 25.3 mg kg^{-1}), Pb (16.1 and 18.2 mg kg^{-1}) and V (15.4 and 18.7 mg kg^{-1}) showed
180 the highest mean values in 2015 and 2016. No significant differences were noted between
181 2015 and 2016 mean levels. Cadmium was the only element showing significant
182 differences ($p < 0.05$) in the sampling points located at less than 1 km from the MSWI (0.32
183 mg kg^{-1}) (points 2, 6, and 7)), in relation to those located further (0.09 mg kg^{-1}) (points 1, 3,
184 4, 5, and 8) (Fig. 2a). Taking into account directions from the MSWI, West direction
185 (impacted by east winds, prevalent in the area) presented the lowest levels ($p < 0.05$) of As
186 and Ni, and higher levels ($p < 0.05$) of Mn (Fig. 2b). On the other hand, South direction, not
187 impacted by prevalent winds, presented significant higher ($p < 0.05$) levels of Cu (Fig.2b).

188 Only for Pb levels (67.6 mg kg^{-1} in sampling point #7 in 2016), the threshold levels (60 mg
189 kg^{-1}) for protection of human health applicable to Catalonia were exceeded (ARC, 2009).

190 In relation to the concentrations of PCDD/Fs in soils, 2,3,7,8-TCDD and 1,2,3,7,8,9-
191 HxCDF were not detected in any sampling point in 2016. OCDD, OCDF, 1,2,3,4,6,7,8-
192 HpCDD, and 1,2,3,4,6,7,8-HpCDF showed the highest levels. Only OCDF showed
193 significant differences ($p < 0.05$) between the 2015 (5.23 ng kg^{-1}) and 2016 (0.91 ng kg^{-1})
194 sampling campaigns. Total PCDD/Fs levels in soil were 0.39 and $0.36 \text{ ng WHO-TEQ kg}^{-1}$
195 in 2015 and 2016, respectively. No significant differences ($p > 0.05$) were noted for the total
196 PCDD/Fs levels with respect to the distance to the MSWI. Only OCDD showed significant
197 higher levels at North-east direction (19.1 ng kg^{-1}), compared with South-west (8.90 ng kg^{-1})
198 and West (2.35 ng kg^{-1}) directions. No significant differences were noted in total
199 PCDD/Fs levels with respect to wind directions (0.37 , 0.46 , and $0.19 \text{ ng WHO-TEQ kg}^{-1}$ at
200 South-west, North-east, and West directions, respectively) (Fig. 2b). In the current survey,
201 total PCDD/F levels were low compared with soil screening levels reported in the scientific
202 literature and belonging to countries such as Finland, Sweden, Austria ($10 \text{ ng I-TEQ kg}^{-1}$),
203 or Canada ($4 \text{ ng I-TEQ kg}^{-1}$) (US EPA, 2009b). Soil PCDD/F levels around the
204 Campdorà's MSWI were also in the low range of other concentrations previously reported
205 for Catalonia and other countries (Table 2). Principal components analysis (PCA), that
206 explain 87% of the variance in a three dimension plot, (Fig. 3) showed that soil sampling
207 points are clustered in 3 groups. One of this is made with sampling points #1, 2, and 5, in
208 both campaigns and point #6 in 2016, while the second one contains points #3, 4, and 8 in
209 2015 and 2016, and point #6 in 2015. Finally, the third cluster is formed by point #7 in both
210 campaigns. These clusters include points belonging at different distances ($< 1 \text{ km}$ and $> 1 \text{ km}$)
211 and wind directions (South-west and North-east), which indicates a low influence of the
212 MSWI on the concentrations of trace elements and PCDD/Fs, at those wind directions and
213 distances to the facility.

214 Table 3 and Table 3S show the concentrations of trace elements and PCDD/Fs in air
215 samples collected around the Campdorà's MSWI in 2015 and 2016. Mercury levels were
216 below the detection limit (0.01 ng m^{-3}) in both sampling periods, while Cr and Tl were
217 below the detection limit (0.45 and 0.01 ng m^{-3} , for Cr and Tl, respectively) in the 2015

218 sampling campaign. The highest air metal and metalloid levels corresponded to Cu (39.0
219 and 170 ng m⁻³, in 2015 and 2016, respectively). In contrast to soils, which did not register
220 any difference between campaigns, air presented higher levels in 2016, being the
221 differences statistically significant for As, Cd, Co, Mn, Ni, Pb, Tl and V. This can be
222 explained by the fact that soils are long-term environmental monitors, while air tends to
223 reflect the current status of pollution with a higher temporal variability (Schuhmacher and
224 Domingo, 2006). However, no significant differences regarding the distance (<1km and
225 >1km) and wind directions (NE, SW, and W) were noted (Fig. 4). A maximum annual
226 average air concentration was established by EU directives (500, 20, 5, and 6 ng m⁻³, for
227 Pb, Ni, Cd and As) (European Union Parliament and Council, 2008b, 2004). The levels of
228 the present study, although correspond to daily values and not annual, were below the
229 annual maximum levels established by legislation. Regarding PCDD/Fs, 1,2,3,4,7,8,9-
230 HpCDF was the only congener below the detection limit (0.005 pg m⁻³) in all samples and
231 for both periods (2015 and 2016). The highest levels corresponded to OCDD, 1,2,3,4,6,7,8-
232 HpCDD, 1,2,3,4,6,7,8-HpCDF, and OCDF. Only 1,2,3,7,8,9-HxCDF showed significant
233 differences (p<0.05) between 2015 (0.003 pg m⁻³) and 2016 (0.001 pg m⁻³). As in soil, no
234 significant differences were noted with respect to distances and wind directions for
235 PCDD/Fs levels (Fig. 4). The concentrations found in the present study are similar than
236 those reported for other incinerators in Catalonia, being also in the low range of a number
237 of worldwide studies (Table 4). These results do not show any tendency in relation to the
238 distances or wind direction to MSWI. Hybrid Single Particle Lagrangian Integrated
239 Trajectory (HYSPLIT) model (Draxler and Rolph, 2017; Rolph et al., 2017) was applied in
240 order to calculate back trajectories during air sampling periods in order to establish local
241 emission sources and long-mid range transport influence during samples collection. Fig. 5
242 summarizes these back-trajectories for each sampling points and period. In general, three
243 long-mid range 24-hours back-trajectories were noted: 1) trajectories coming from west
244 following Pyrenees Mountains starting over Atlantic Ocean (points 1 and 2 in both
245 campaigns and point 3 in 2015); 2) trajectories coming from north crossing France (point 4
246 in 2015); 3) trajectories cross Barcelona metropolitan area (Point 3 and 4 in 2016). These
247 back-trajectories classification are in accordance with air PCDD/Fs congener PCA results
248 (Fig. 6) that show a cluster with points 1 and 2, both periods and point 3 in 2015. Point 3 in

249 2016 and point 4 in 2015 and 2016 were plotted separately from the cluster and between
250 them. These highlighted the influence of long and also medium (from Barcelona
251 metropolitan area located around 100 km away) range transport. Regarding local transport,
252 some of the 2-hours resolution back-trajectories cross the location of MSWI before
253 reaching sampling points 2, 3 and 4 in both campaigns (2015 and 2016) but not for
254 sampling point 1 in both campaigns. This confirms the adequacy of the selection of air
255 sampling points 2, 3 and 4 to control inmission levels around the MSWI and the selection
256 of point 1 as a control or background point. No significant differences ($p>0.05$) were noted
257 in air elements and PCDD/Fs (congeners and total) levels between point 1 (background)
258 and points 2, 3, and 4 (under MSWI influence).

259 **3.2. Human exposure and risk assessment**

260 Human exposure levels to trace elements and PCDD/Fs during 2015 and 2016 through soil
261 ingestion, dermal contact with soils, and air inhalation are shown in Table 5. Manganese
262 presented the highest exposure: $6.7 \cdot 10^{-4}$ and $7.3 \cdot 10^{-4}$ $\text{mg kg}^{-1} \text{day}^{-1}$, during 2015 and 2016,
263 respectively, followed by Cu ($5.4 \cdot 10^{-5}$ and $1.0 \cdot 10^{-4}$ $\text{mg kg}^{-1} \text{day}^{-1}$), during 2015 and 2016,
264 respectively. In general terms, soil ingestion was the main exposure pathway for trace
265 elements, followed by dermal contact. The exceptions were As for which the main pathway
266 was dermal contact followed by soil ingestion, and Sb, which was not detected, or it was
267 present only at low levels in soil. On the other hand, PCDD/Fs principal exposure pathway
268 was air inhalation, with a contribution between 73 and 77%. Similar values regarding the
269 contribution of air inhalation with respect to the total intake of PCDD/Fs, were found in
270 previous studies around other waste incinerators (Vilavert et al., 2015a; Rovira et al., 2015).

271 In relation to non-cancer risks (Fig. 7), Mn showed the highest HQ values (0.2 and 0.3, in
272 2015 and 2016, respectively), while other elements presented HQ below 0.1. For PCDD/Fs,
273 HQ had a value of 0.002 in both sampling campaigns. It indicates that for all elements and
274 PCDD/Fs the threshold value defined as 1, was not reached, meaning a safe situation
275 regarding non-carcinogenic risks. Cancer risks due the exposure to elements and PCDD/Fs
276 are depicted in Fig. 7. Cancer risks for any pathway were below 10^{-5} . For the sum of the
277 three pathways, total cancer risk for As almost reached 10^{-5} ($9.9 \cdot 10^{-6}$ in both campaigns)
278 and for Cr(VI) slightly exceeded 10^{-5} ($7.0 \cdot 10^{-6}$ and $1.2 \cdot 10^{-5}$ in 2015 and 2016, respectively).

279 However, cancer risks are still in a range considered as acceptable (between 10^{-6} and 10^{-4}).
280 It should be noted that total (different chemical species) As and Cr were analyzed, while
281 toxicological parameters for hexavalent Cr and inorganic As were used. For Cr, 1/6 of total
282 Cr was assumed to be in the hexavalent (Cr(VI)) form (Brown et al., 2014), while for As,
283 total environmental As was assumed to be in inorganic form (Huang et al., 2014).

284 **3.3. Risk assessment around Catalan incinerator plants**

285 Since 1996, we have conducted a number of studies around incineration plants (MSWI and
286 HWI) placed in Catalonia, with similar objectives than those of the present survey; that is to
287 say, to monitor environmental levels of trace elements and PCDD/Fs, and to assess human
288 health risks for the population living in the neighborhood (Domingo et al., 2000; Rovira et
289 al., 2010, 2015; Schuhmacher et al., 2006; Schuhmacher and Domingo, 2006; Vilavert et
290 al., 2009, 2010, 2012ab, 2015a). In fact, we have evaluated all the facilities that are
291 currently operating, or have been operating in Catalonia in recent years. Tables 2 and 4
292 summarize air and soil PCDD/F levels. In order to update and harmonize results, levels of
293 trace elements and PCDD/Fs in air and soil were used to assess human health risks (cancer
294 and non-cancer), the same methodology and same toxicological parameters than those in
295 previous studies have been used here. Risks for population living around MSWIs in
296 Tarragona, Sant Adrià del Besòs, Mataró, Montcada i Reixac (only for levels of
297 metals/metalloids in soil), and the hazardous waste incinerator (HWI) of Constantí (only for
298 levels of metals/metalloids in soil) were assessed (Domingo et al., 2000; Rovira et al.,
299 2010, 2015; Schuhmacher and Domingo, 2006; Schuhmacher et al., 2006; Vilavert et al.,
300 2009, 2010, 2012ab, 2015a). HQ levels for metals, metalloids, and PCDD/Fs for all
301 facilities were below the threshold limit ($HQ < 1$). Results of cancer risks around each
302 facility are shown in Fig. 8. With respect to the PCDD/Fs carcinogenic risks of the MSWI
303 of Sant Adrià del Besòs and Montcada i Reixac, these exceeded 10^{-6} , but not reached 10^{-5} ,
304 while other MSWIs in Catalonia showed risks below the 10^{-6} threshold. A clear decrease
305 temporal tendency was noted in soil PCDD/Fs levels, especially in the facility of Tarragona
306 during the last 15 years (Table 2). For the risks of Cd, Co and Ni inhalation in the
307 neighborhood of all facilities, all values were around, or lower, than 10^{-7} . Arsenic and
308 Cr(VI) carcinogenic levels presented similar values, approximately 10^{-5} , with the same

309 assumptions (Inorganic As=Total As and Cr (VI)=1/6 Total Cr) than those made for the
310 Campdorà's MSWI. Generally, Campdorà's MSWI showed lower carcinogenic risk levels
311 than other incinerators of Catalonia. The differences clearly reveal that individual
312 environmental monitoring campaigns should be conducted for each specific facility. A
313 generalization is very inappropriate. Not only technical and operational parameters should
314 be taken into account, but also external factors to the facilities, such the presence of other
315 emission sources in the specific zone, and the meteorological conditions and orographic
316 characteristics of the area, among others, are also essential for this kind of studies.
317 Individual monitoring of each plant seems the proper strategy to ensure the protection of
318 human health and the environment.

319 **4. CONCLUSIONS**

320 In general terms, no significant differences were observed in the concentrations of trace
321 elements and PCDD/Fs congeners in soils between the 2015 and 2016 campaigns. In 2016,
322 significant higher air levels of As, Cd, Co, Mn, Ni, Pb, Tl, and V were noted, but not
323 ($p>0.05$) for PCDD/Fs. In soils, only Cd regarding distances to MSWI and As, Cu, Mn and
324 Ni regarding direction to MSWI presented differences. No differences were noted in the
325 levels of trace elements and PCDD/Fs in air with respect to the distances to the facility and
326 wind directions. No differences were registered in air levels (elements and PCDD/Fs)
327 between points influenced by MSWI emissions and background point. However some
328 differences in congener profile were noted regarding from where back-trajectories come
329 from (HYSPLIT model results), pointing at some influence of Barcelona metropolitan area.
330 The concentrations here found were similar, or even lower, to those reported for other
331 MSWI, in Catalonia, as well as in studies performed in various countries. Non-carcinogenic
332 risks were below the safety limit ($HQ<1$). In turn, carcinogenic risks due to exposure to
333 trace elements and PCDD/Fs were in an acceptable range according to national and
334 international standards regulations. The impact on the environment and human health of
335 MSWIs should be individually assessed. A generalization is not possible due technical and
336 operational differences, as well as external differences between facilities.

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543

544 Table 1. Levels of trace elements (mg kg⁻¹) and PCDD/Fs (ng kg⁻¹) in soils around the
 545 Campdorà's MSWI.

	2015		2016		<i>p</i>
	Mean	SD	Mean	SD	
As	4.77	2.44	4.62	2.35	0.898
Cd	0.16	0.12	0.20	0.19	0.665
Co	4.87	1.28	4.83	1.81	0.963
Cr	7.53	3.21	9.31	5.75	0.455
Cu	20.6	37.7	25.3	46.3	0.829
Hg	0.01	0.01	0.01	0.01	0.532
Mn	316	135	344	156	0.703
Ni	10.8	6.04	11.4	6.53	0.857
Pb	16.1	12.5	18.2	20.5	0.809
Sb	<0.05	-	0.24	0.34	0.104
Sn	0.98	0.21	1.61	1.47	0.251
Tl	0.08	0.05	0.09	0.05	0.563
V	15.4	8.66	18.7	9.04	0.479
2,3,7,8-TCDD	0.04	0.04	<0.04	-	0.223
1,2,3,7,8-PeCDD	0.11	0.07	0.07	0.05	0.213
1,2,3,4,7,8-HxCDD	0.15	0.12	0.10	0.07	0.272
1,2,3,6,7,8-HxCDD	0.22	0.17	0.63	1.37	0.416
1,2,3,7,8,9-HxCDD	0.21	0.16	0.34	0.60	0.573
1,2,3,4,6,7,8-HpCDD	2.32	1.81	3.45	4.57	0.526
OCDD	13.1	11.8	11.7	8.97	0.802
2,3,7,8-TCDF	0.12	0.00	0.15	0.07	0.297
1,2,3,7,8-PeCDF	0.19	0.14	0.18	0.20	0.850
2,3,4,7,8-PeCDF	0.17	0.09	0.12	0.11	0.433
1,2,3,4,7,8-HxCDF	0.21	0.11	0.19	0.17	0.774
1,2,3,6,7,8-HxCDF	0.20	0.10	0.15	0.13	0.395
1,2,3,7,8,9-HxCDF	0.05	0.02	<0.08	-	0.441
2,3,4,6,7,8-HxCDF	0.25	0.16	0.20	0.20	0.614
1,2,3,4,6,7,8-HpCDF	1.38	0.75	1.02	0.86	0.392
1,2,3,4,7,8,9-HpCDF	0.16	0.10	0.17	0.11	0.861
OCDF	5.23	2.40	0.91	0.66	<0.001
Total WHO-TEQ	0.39	0.21	0.36	0.30	0.855

In bold significant differences at p<0.05

546

547

548 Table 2. A summary of concentrations of PCDD/Fs in soils around a number of MSWIs
 549 and hazardous waste incinerators (HWI). Data from the scientific literature.

Place	Year	Mean (ng TEQ kg ⁻¹)	Range (ng TEQ kg ⁻¹)	Reference	
Taiwan	2006	2.00	0.85 - 4.50	(Chen et al., 2012)	
China	2014	1.49	0.52 - 3.40	(Zhou et al., 2015)	
China	-	1.33	0.54 - 1.96	(Liu et al., 2012)	
Shanghai (China)	2007	4.73	0.64 - 61.2	(Deng et al., 2011)	
Hangzhou (China)	2007	1.50	0.60 - 6.38	(Xu et al., 2009)	
Trondheim (Norway)	2004	-	0.16 - 14.0	(Andersson and Ottesen, 2008)	
Besançon (France)	-	2.70	0.25 - 28.1	(Floret et al., 2007)	
Korea	-	19.1	1.25 - 75.0	(Oh et al., 2006)	
Po valley (Italy)	1997	-	0.85 - 1.5	(Caserini et al., 2004)	
Veneto Region (Italy)	2000	-	1.1 - 1.4	(Caserini et al., 2004)	
Adige valley (Italy)	2001	-	0.08 - 1.2	(Caserini et al., 2004)	
Reggio Emilia (Italy)	-	-	1.8 - 7.1	(Capuano et al., 2005)	
Tarragona (Spain)	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)
	2014	0.63	-		(Vilavert et al., 2015a)
Constantí (Spain)*	1998	1.59	0.12 - 17.2	(Vilavert et al., 2010)	
	2003	0.77	0.10 - 3.66		
	2008	2.89	0.07 - 50.6		
	2009	0.75	0.09 - 2.99		
Sant Adrià del Besòs (Spain)	1998	9.06	1.22 - 34.3	(Domingo et al., 2000)	
	1999	11.8	1.33 - 54.2		
	2014	3.6	0.4 - 10.6		(Domingo et al., 2015)
Mataró (Spain)	2009	0.34	0.14 - 0.46	(Rovira et al., 2010)	
	2011	0.23	0.13 - 0.56		
	2013	0.34	0.12 - 0.61		(Rovira et al., 2015)
Montcada i Reixac	1996	3.5	0.3 - 44.3	(Schuhmacher and Domingo, 2006)	
	1997	2.2	0.2 - 29.3		
	1998	3.4	0.1 - 127		
	2000	2.3	0.2 - 49.4		
	2002	2.4	0.4 - 16.9		
Campdorà, Girona (Spain)	2015	0.39	0.13 - 0.76	Present study	
	2016	0.36	0.12 - 0.97		

*Hazardous waste incinerator

550

551

552 Table 3. Levels of trace elements (ng m⁻³) and PCDD/Fs (pg m⁻³) in air samples around the
 553 Campdorà's MSWI

	2015		2016		<i>p</i>
	Mean	SD	Mean	SD	
As	0.04	0.02	0.36	0.18	0.013
Cd	0.02	0.01	0.13	0.06	0.013
Co	0.04	0.02	0.16	0.07	0.013
Cr	<0.45	-	1.50	1.12	0.062
Cu	39.0	45.8	170	101	0.056
Hg	<0.01	-	<0.01	-	-
Mn	1.58	0.35	10.7	6.07	0.029
Ni	0.38	0.21	0.45	0.35	0.012
Pb	0.72	0.07	5.15	2.51	0.029
Sb	0.20	0.13	1.12	0.82	0.200
Sn	0.35	0.26	2.21	1.67	0.057
Tl	<0.01	-	0.02	0.01	0.013
V	0.47	0.45	2.22	0.11	0.013
2,3,7,8-TCDD	0.001	0.001	0.001	0.001	0.243
1,2,3,7,8-PeCDD	0.005	0.005	0.002	0.000	0.406
1,2,3,4,7,8-HxCDD	0.004	0.001	0.002	0.001	0.159
1,2,3,6,7,8-HxCDD	0.006	0.002	0.004	0.000	0.310
1,2,3,7,8,9-HxCDD	0.004	0.002	0.004	0.001	0.821
1,2,3,4,6,7,8-HpCDD	0.054	0.016	0.045	0.008	0.336
OCDD	0.118	0.024	0.097	0.021	0.231
2,3,7,8-TCDF	0.007	0.001	0.006	0.001	0.158
1,2,3,7,8-PeCDF	0.007	0.003	0.005	0.002	0.355
2,3,4,7,8-PeCDF	0.009	0.003	0.009	0.003	0.737
1,2,3,4,7,8-HxCDF	0.007	0.003	0.013	0.005	0.119
1,2,3,6,7,8-HxCDF	0.006	0.003	0.007	0.002	0.773
1,2,3,7,8,9-HxCDF	0.003	0.001	0.001	0.001	0.034
2,3,4,6,7,8-HxCDF	0.011	0.004	0.010	0.005	0.809
1,2,3,4,6,7,8-HpCDF	0.030	0.008	0.030	0.011	0.974
1,2,3,4,7,8,9-HpCDF	<0.005	-	<0.005	-	-
OCDF	0.027	0.014	0.024	0.013	0.738
Total WHO-TEQ	0.015	0.009	0.012	0.002	0.512

In bold significant differences at p<0.05

554

555

556 Table 4. A summary of concentrations of PCDD/Fs around a number of MSWIs. Data from
 557 the scientific literature.

Place	Year	Mean (pg TEQ m ⁻³)	Range (pg TEQ m ⁻³)	Reference
Taiwan	2006	0.050	0.039 - 0.088	(Chen et al., 2012)
Taiwan	-	0.060	0.033 - 0.105	(Wang et al., 2010)
Taiwan	-	0.078	0.022 - 0.155	(Shih et al., 2006)
Hangzhou (China)	2007/08	0.495	0.059 - 3.03	(Xu et al., 2009)
China	2014	0.237	0.030 - 1.03	(Zhou et al., 2015)
Korea	-	0.66	0.22 - 1.16	(Oh et al., 2006)
Po valley (Italy)	2000	-	0.022 - 0.039	(Caserini et al., 2004)
Veneto Region (Italy)	2000	-	0.144 - 0.337	(Caserini et al., 2004)
Adige valley (Italy)	2000	-	0.010 - 0.060	(Caserini et al., 2004)
Portugal				
Lisbon	1998/2004	0.034	0.002 - 0.153	(Coutinho et al., 2007)
Porto		0.149	0.009 - 0.817	
Madeira		0.015	0.002 - 0.060	
	2007	0.012	0.004 - 0.033	(Vilavert et al., 2009)
	2008	0.015	0.007 - 0.031	
Tarragona	2009	0.009	0.007 - 0.022	(Vilavert et al., 2012b)
	2010	0.010	0.004 - 0.022	
	2013	0.004	-	(Vilavert et al., 2015a)
Sant Adrià del Besòs (Spain)				
MSWI influenced	2005/06	0.018	0.010 - 0.024	(Mari et al., 2008)
Control		0.012	0.008 - 0.019	
Sant Adrià del Besòs (Spain)	2014	0.026	0.018 - 0.041	(Domingo et al., 2015)
	2009	0.011	0.008 - 0.015	(Rovira et al., 2010)
Mataró (Spain)	2011	0.010	0.006 - 0.013	(Rovira et al., 2015)
	2013	0.014	0.010 - 0.018	
Montcada i Reixac (Spain)	2001/02	0.10	-	(Schuhmacher et al., 2006)
Campdorà, Girona (Spain)	2015	0.015	0.009 - 0.028	Present study
	2016	0.011	0.009 - 0.014	

558

559 Table 5. Total exposure to trace elements ($\text{mg kg}^{-1} \text{ day}^{-1}$) and PCDD/Fs ($\text{ng WHO-TEQ kg}^{-1}$
 560 day^{-1}) and contribution (%) of each exposure pathway.

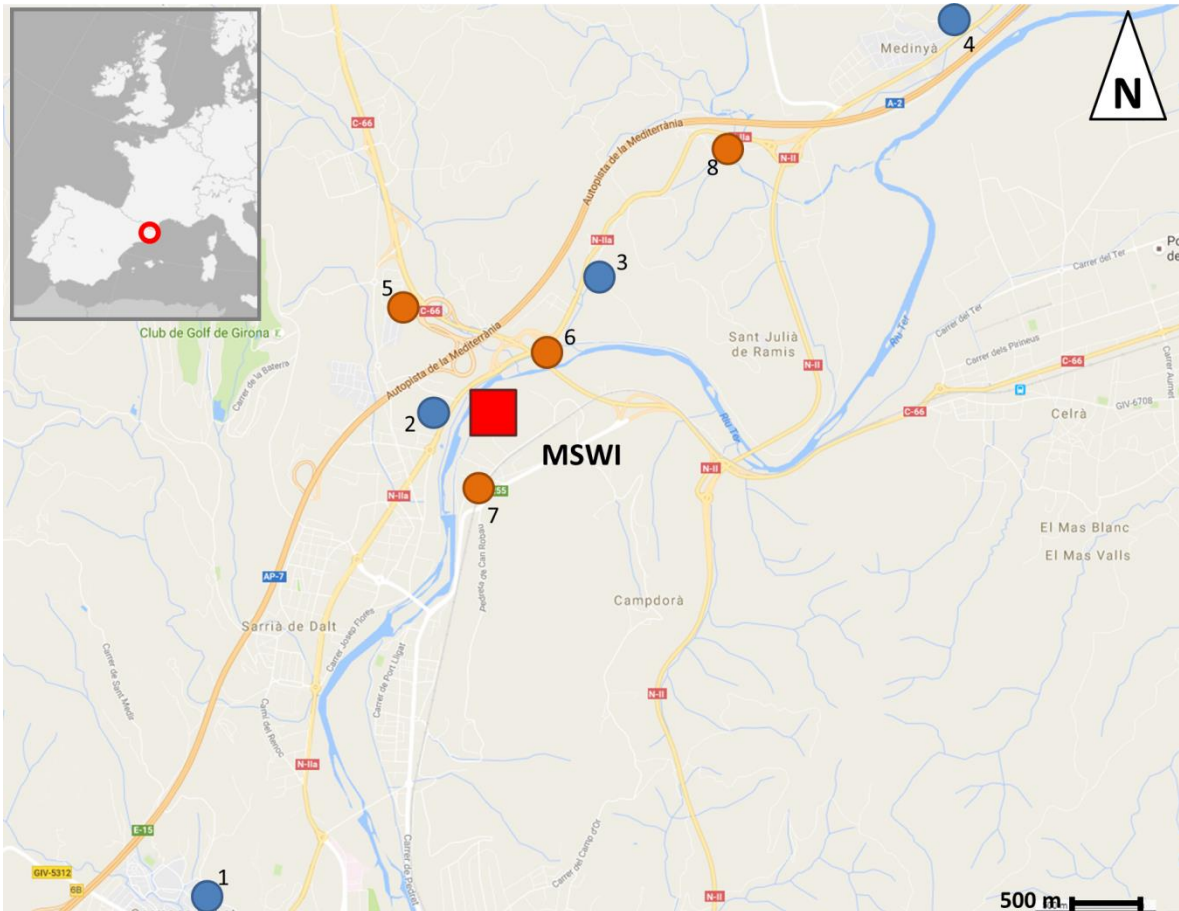
	2015				2016			
	Total	Soil ingestion	Dermal contact	Air inhalation	Total	Soil ingestion	Dermal contact	Air inhalation
As	1.5E-05	48	52	0	1.5E-05	48	51	1
Cd	2.7E-07	94	3	3	3.5E-07	87	3	10
Co	1.0E-05	74	26	0	1.0E-05	73	26	0
Cr	1.6E-05	74	26	NA	2.0E-05	72	26	2
Cu	5.4E-05	59	21	20	1.0E-04	39	14	47
Hg	1.9E-08	74	26	NA	2.5E-08	74	26	NA
Mn	6.7E-04	74	26	0	7.3E-04	74	26	0
Ni	2.3E-05	74	26	0	2.4E-05	74	26	1
Pb	3.4E-05	74	26	1	4.0E-05	71	25	4
Sb	5.9E-08	NA	NA	100	8.0E-07	46	16	38
Sn	2.2E-06	70	25	5	4.0E-06	63	22	15
Tl	1.7E-07	73	26	NA	2.0E-07	72	26	2
V	3.3E-05	74	26	0	4.0E-05	72	26	2
PCDD/Fs	5.3E-06	11	12	77	4.3E-06	13	14	73

NA: Not assessed (environmental levels below detection limit).

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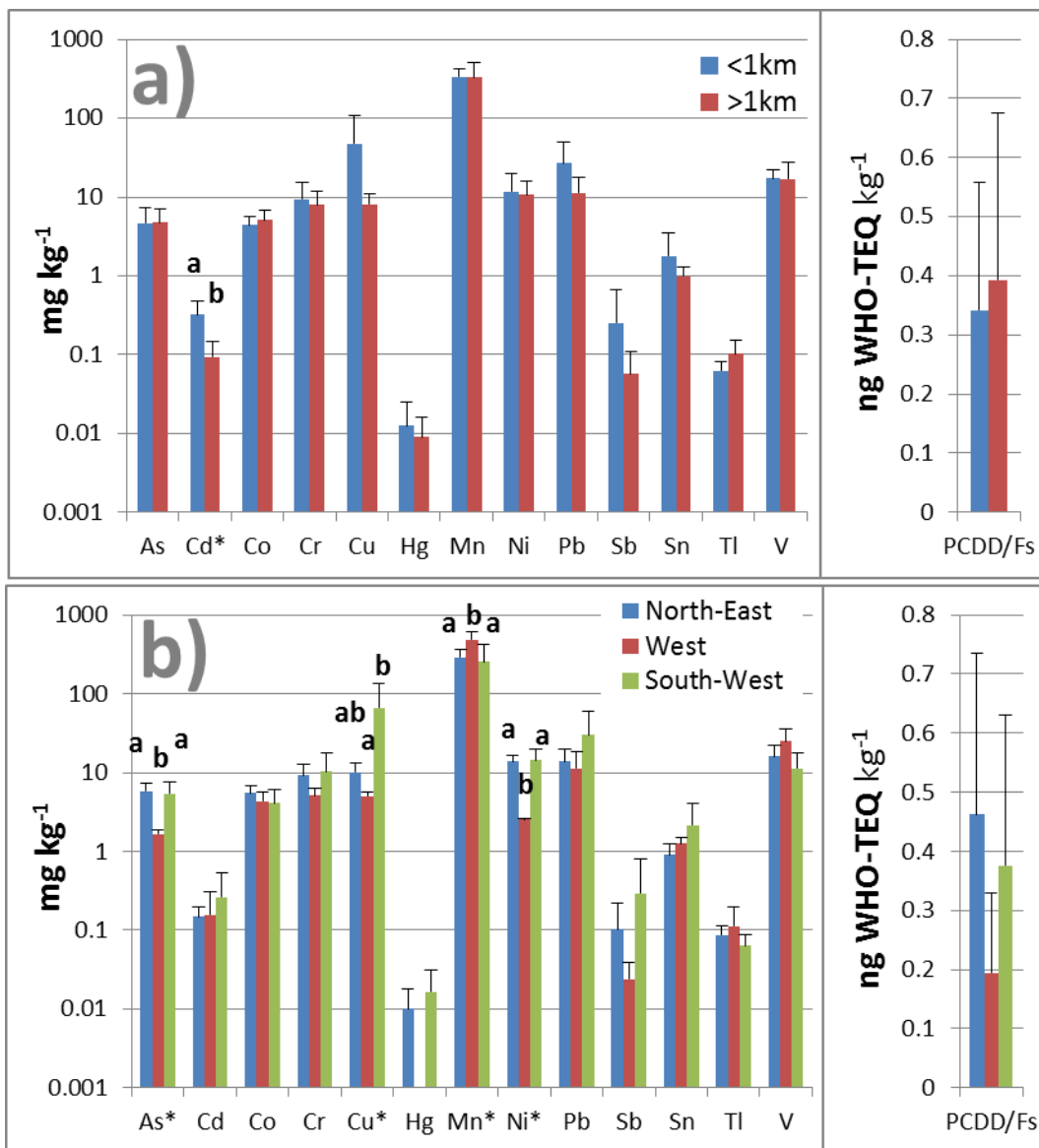
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564

565 Fig. 1. Sampling points location: soil and air sampling points (form1 to 4) are marked in
 566 blue and only soil sampling points (from 5 to 8) are marked in orange. The location of the
 567 MSWI is marked in red square.

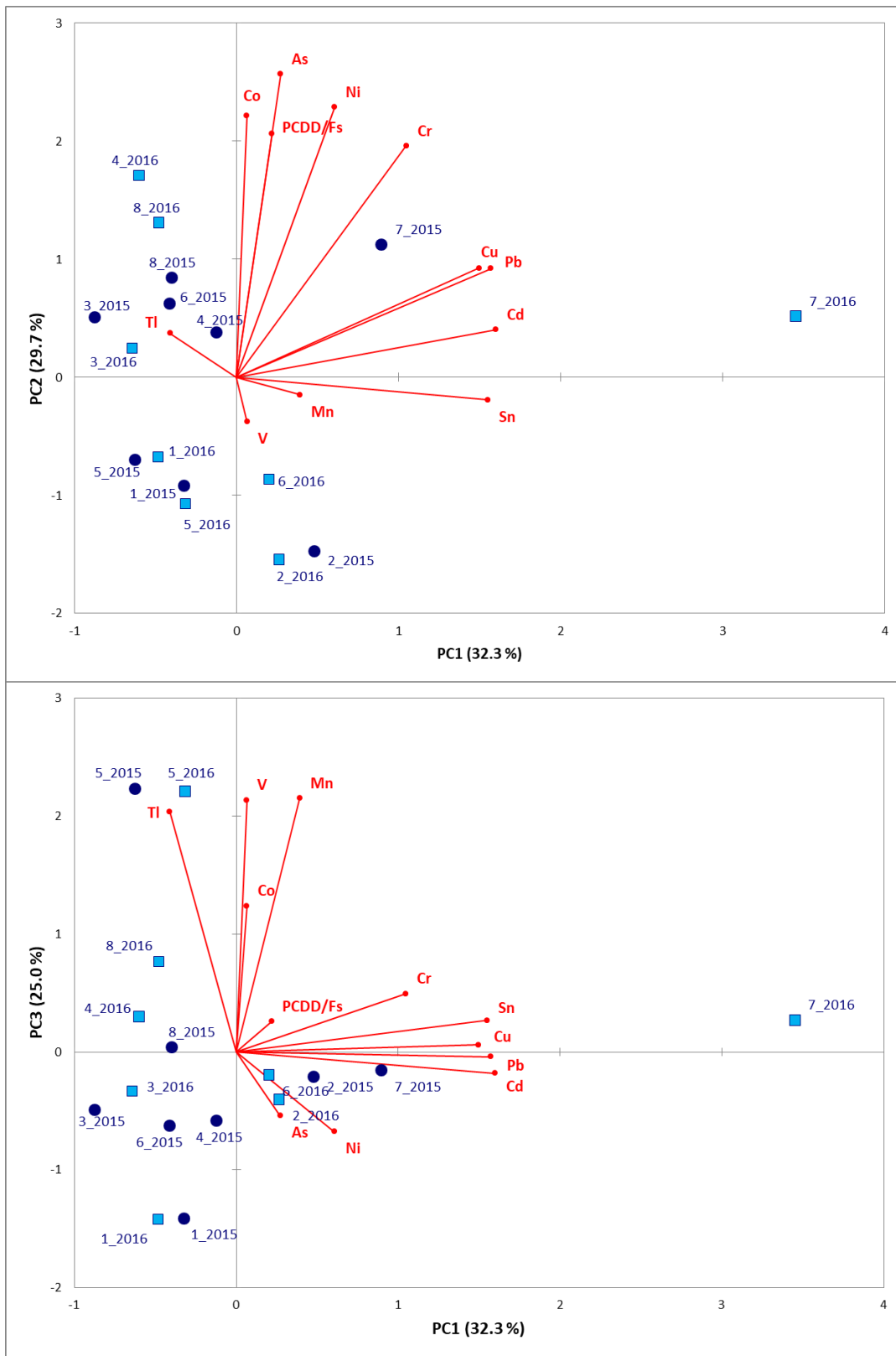
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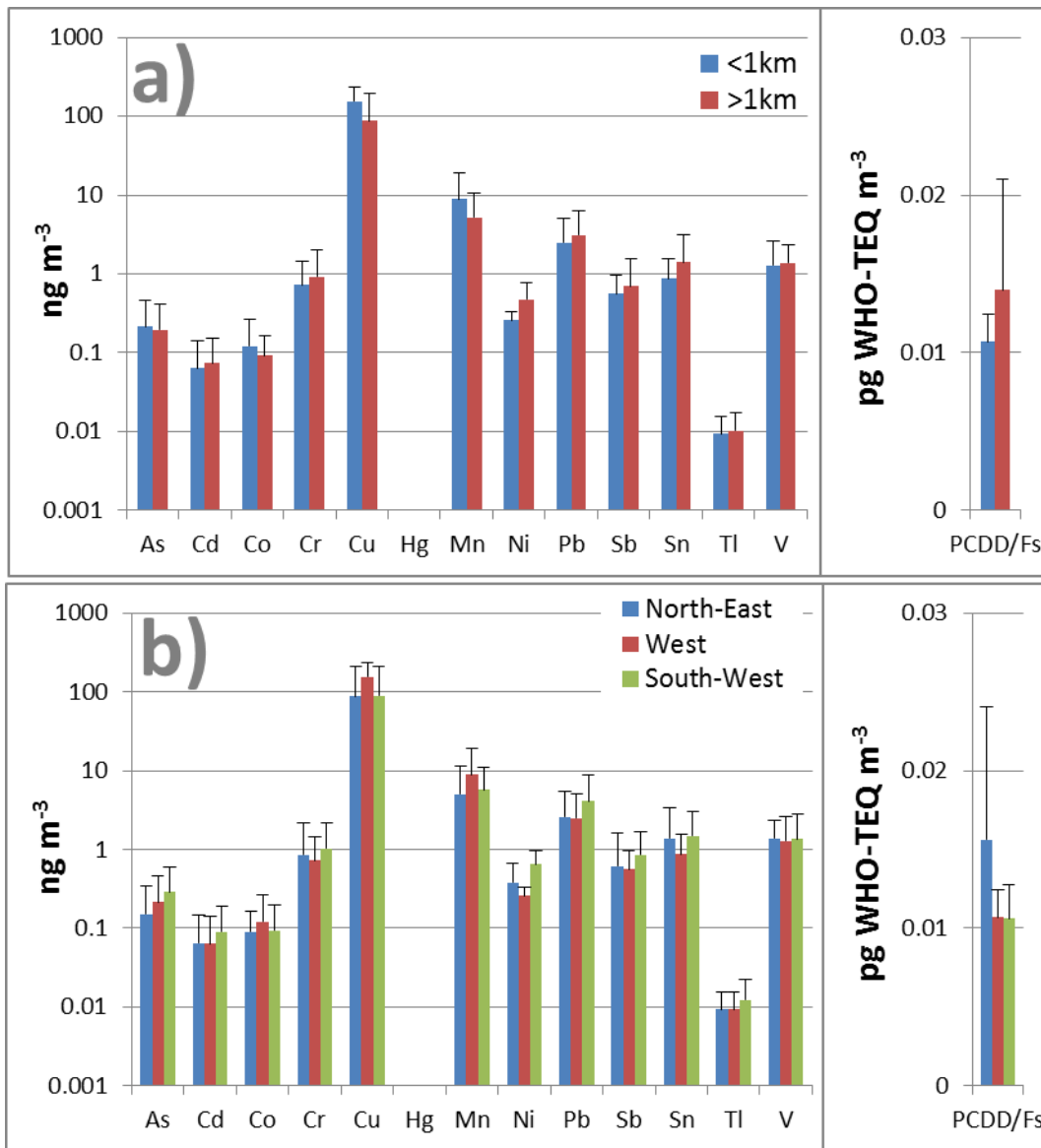
570 Fig. 2. Levels of trace elements and PCDD/Fs in soil samples according to: a) distance and
 571 b) wind direction to Campdorà's MSWI. Asterisks and letters indicates significant
 572 differences at $p < 0.05$.

573



574

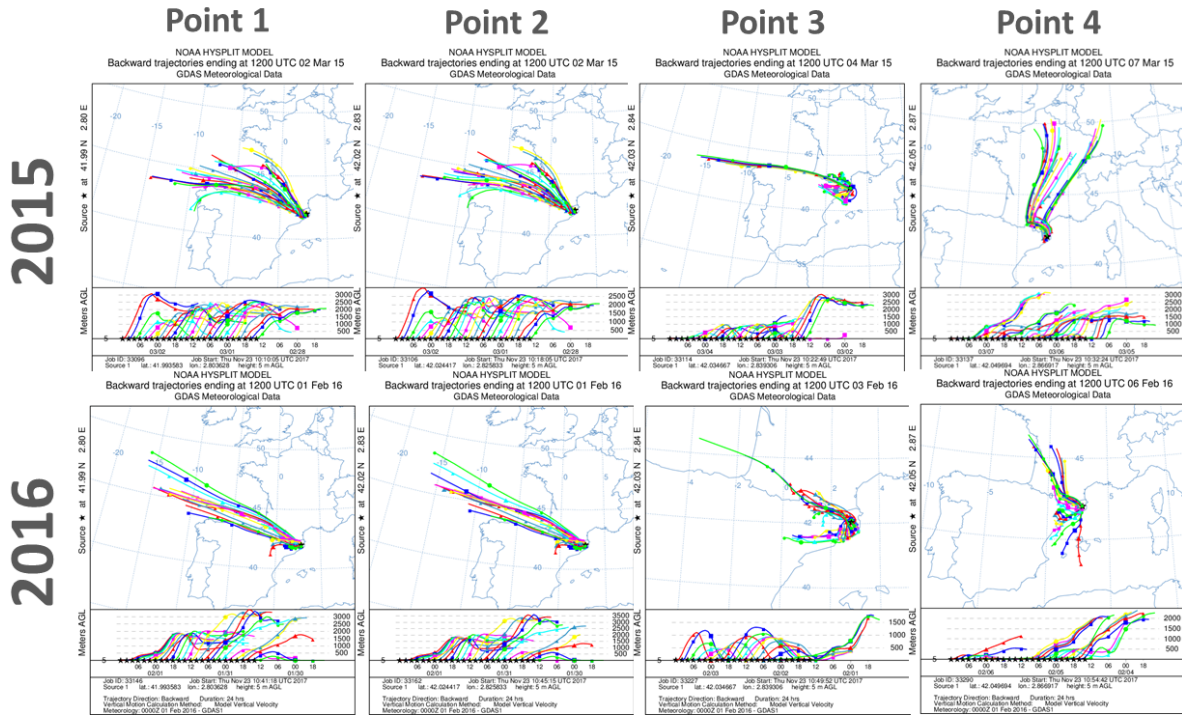
575 Fig. 3. Principal comonents analysis (PC1 vs. PC2 and PC1 vs. PC3) of soil elements and
 576 PCDD/Fs levels collected in both campaigns around to Campdorà's MSWI.



577

578 Fig. 4. Levels of trace elements and PCDD/Fs in air samples according to: a) distances and
 579 b) directions to Campdorà's MSWI. Asterisks and letters indicates significant differences at
 580 $p < 0.05$.

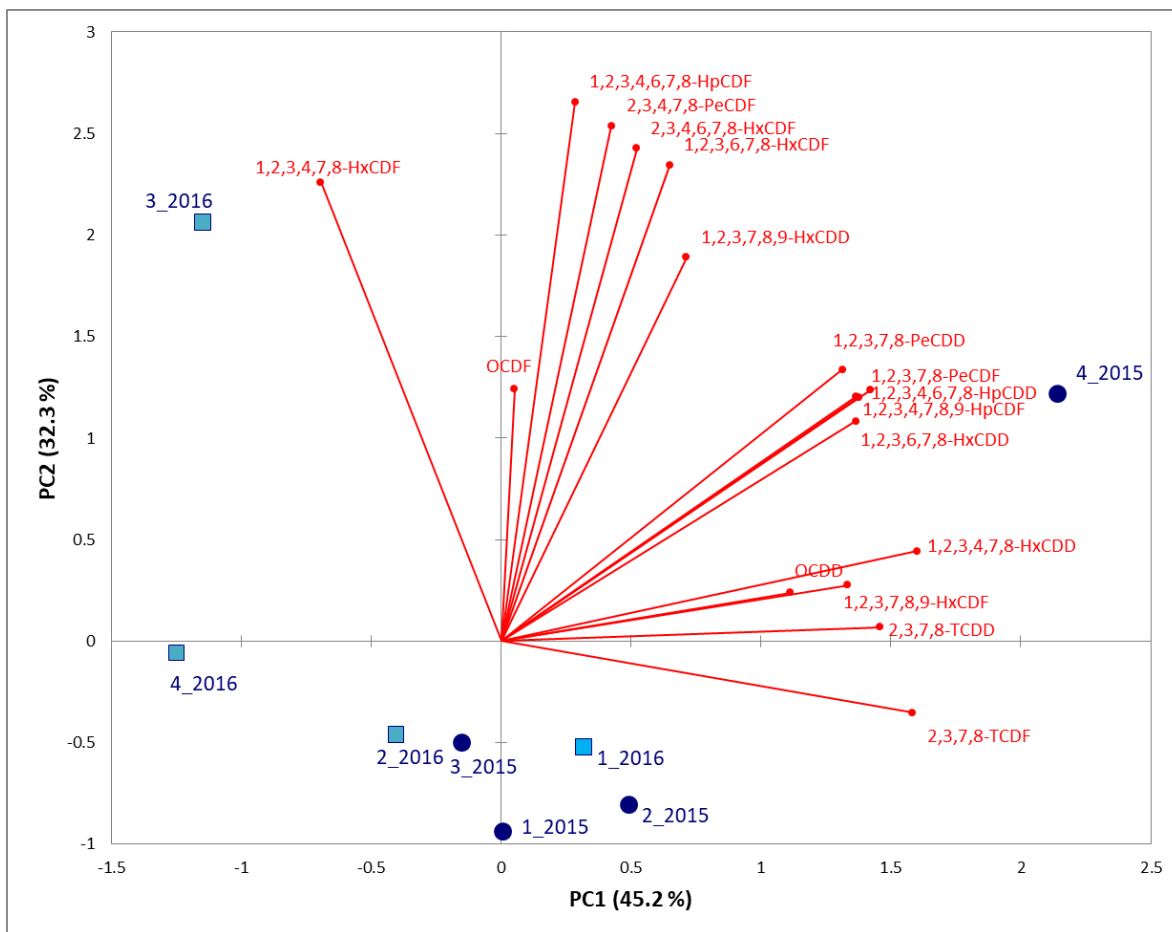
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583 Fig. 5. HYSPLIT model back-trajectories for the air samples collected in Campdorà's
584 MSWI during 2015 and 2016.

585

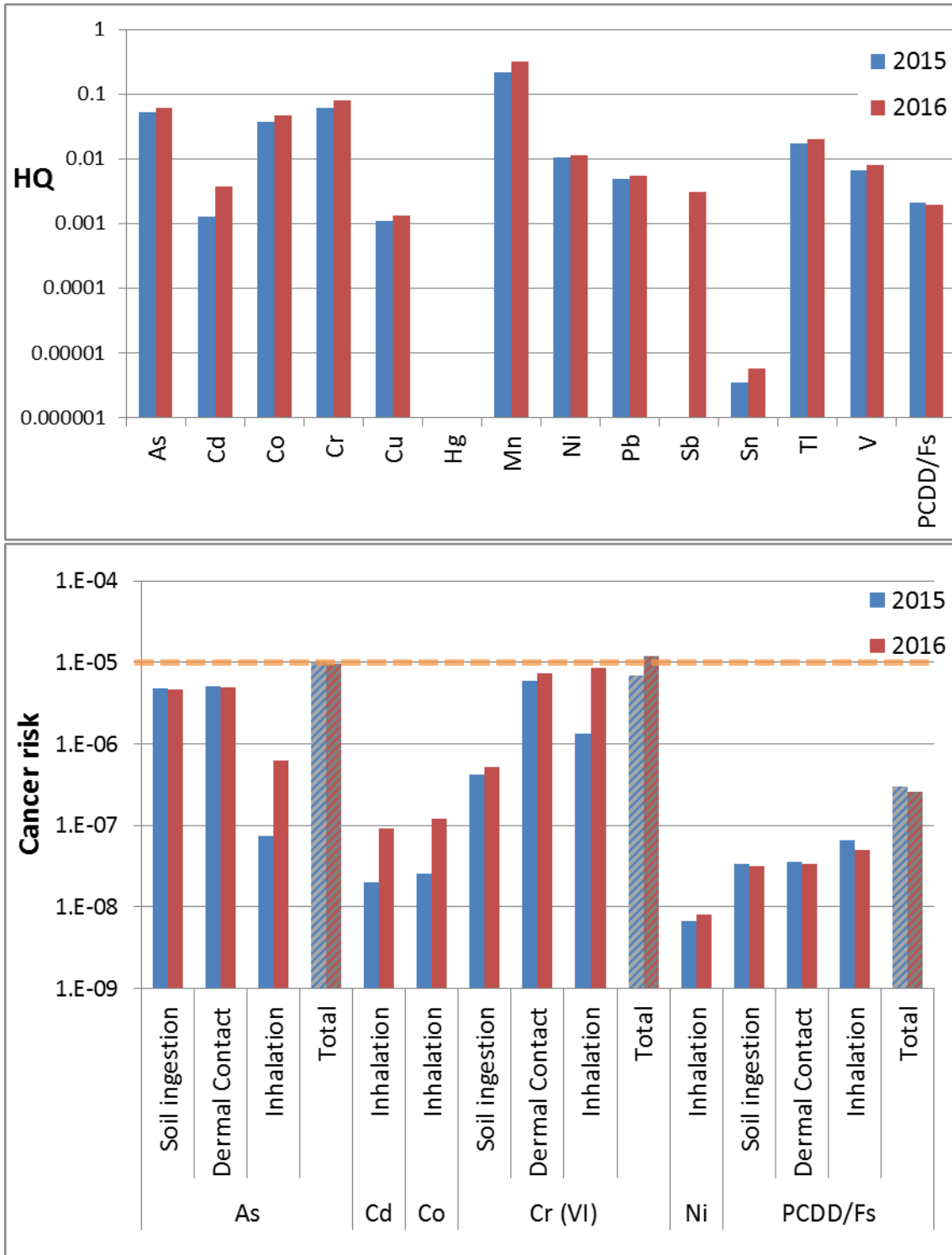


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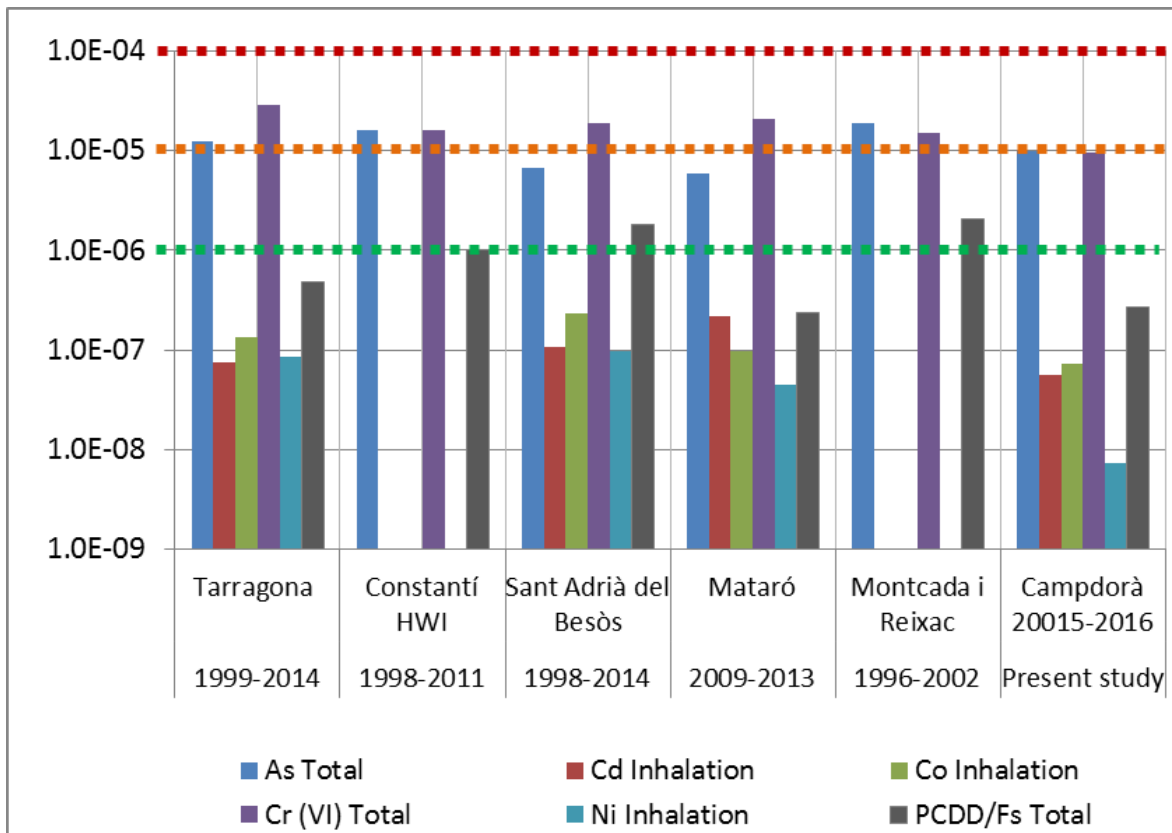
587 Fig. 6. Principal component analysis (PCA) plot of PCDD/F congeners in air samples
 588 collected around Campdorà's MSWI.

589

590



593 Fig. 7. Non-cancer (HQ) and cancer risks derived from trace elements and PCDD/Fs
 594 exposure in the 2015 and 2016 campaigns around the Campdorà's MSWI.



595

596 Fig. 8. Mean cancer risk around various incinerators (MSWI and HWI) located in Catalonia
 597 (Spain). Cancer risks of trace elements around the Constantí-HWI and Montcada i Reixac-
 598 MSWI calculated using soil levels only.

599

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