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Characterization of the PCDD/F in the Province of Trento

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Abstract

This paper aims to improve the already existing literature data on air concentrations of polychlorinated dibenzo-*p*-dioxin and dibenzofurans (PCDD/Fs). Four monitoring campaigns were carried out between 2002 and 2010 near Trento, a town in the North of Italy. These campaigns showed relatively low PCDD/F concentrations, in line with the values found in other Italian urban and industrial sites. Typical values for agricultural regions were measured in rural areas, with an increase during winter, possibly due to biomass burning. No critical situations were detected along an important highway, in spite of the non-negligible emission factors for traffic reported in the literature.

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1. Introduction

Polychlorinated dibenzo-*p*-dioxin and dibenzofurans (PCDD/Fs) can be found as trace elements in almost all environmental compartments. Human activities, such as some industrial and combustion processes, are responsible for anthropogenic PCDD/F emissions [1-3]. This family of compounds is considered among the most dangerous group of substances for human health due to their bioaccumulation properties and their demonstrated carcinogenicity [4,5]. Populations can be exposed to PCDD/Fs through different pathways: inhalation, ingestion, dermal contact and diet, the latter playing a dominant role, since PCDD/Fs can be accumulated in the fat tissues of animals [6].

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PCDD/F concentrations are typically < 10 fg I-TEQ m^{-3} in remote areas, 20-50 fg I-TEQ m^{-3} in rural areas, 100-400 fg I-TEQ m^{-3} in urban and industrial areas [7]. A ten-year comprehensive study on different sites revealed PCDD/Fs concentration in a range of: 13–357 fg I-TEQ m^{-3} in urban/traffic sites, 10–138 fg I-TEQ m^{-3} in suburban/traffic sites, 42–406 fg I-TEQ m^{-3} in urban/industrial sites, 7–1,196 fg I-TEQ m^{-3} in suburban/industrial sites, and 5–45 fg I-TEQ m^{-3} in rural/industrial sites, whereas concentrations in background areas were 8–28 fg I-TEQ m^{-3} [8].

At European level, sintering plants are one of the most important sources of PCDD/Fs, followed by municipal solid waste incinerators (MSWIs) and lastly by traffic [9,10]. To provide an idea of the role of traffic, if considering the average emission factors [11] and an average driving speed of 100 km h^{-1} on a highway, emissions from cars and trucks could be about two orders of magnitude lower than the emissions from a medium-sized biostabilization plant equipped with a biofilter [12] and about three orders of magnitude lower than the emissions from a MSWI [11]. Overall, in comparison with the 90s, the TEQ of these pollutants have shown a notable decrease due to technological improvements [13-18].

Except for the studies here reported, to the best of our knowledge, a few works have focused on PCDD/F air concentrations near steel complexes and highways. Thus, this study aims at improving the already existing database with the addition of the results of four monitoring campaigns carried out between 2002 and 2010 in the Province of Trento, a region located in the North of Italy.

A first characterization, performed by the Department of Civil and Environmental Engineering (DICA) of the University of Trento, was carried out in Trento in 2002 and 2003 to determine a background reference in view of the construction of a MSWI in the territory of the municipality and to understand the role of a local highway as a potential source of PCDD/Fs. Additional monitoring campaigns were carried out by the Environmental Protection Agency of the Province of Trento (APPA) and the DICA in 2006 and 2007, to find a correlation between potential PCDD/F emission sources and PM_{10} concentrations, measured at fixed air quality stations. This study was repeated in summer 2009 and winter 2010 by the DICA to continue the characterization near Trento. Finally, in 2010, APPA conducted a monitoring campaign to characterize the input of pollutants by industrial activities in a valley (Valsugana) and in the surroundings of Trento. The recent decision of the Province of Trento (late 2013) to base the residual municipal solid waste management on combustion plants out of its territory (also through the production of Solid Recovered Fuel) has important consequences on the choice of the sites that could be characterized steadily in the future to measure the dynamics of PCDD/F pollution in the air of the region. Of course a parallel strategy must be set for the characterization of soils.

2. Materials and methods

During the monitoring campaign performed in 2002/2003 (campaign A), air samples were taken at three sites near Trento: a village (site A1), the urban area of Trento (site A2), the peripheral area of Trento (site A3) and the area of maximum impact expected for the MSWI plant (site A4). The samplings were carried out during two different periods of the year to characterize winter and summer meteorological conditions. During the measurement campaign carried out in 2006 (campaign B), several points were identified for a representative air sampling of the interested area; the samples were taken at the following sites:

- 1. Trento North – Two air samples (codes I1 and I2) were taken during a peak period of PM_{10} (average value $> 90 \mu\text{g m}^{-3}$) at two sites located at a distance of about 100 m (site B1) and 550 m (site B2), respectively, from the axis of the A22 highway, which goes through the Adige Valley, where Trento is located. Another sample (code T2) was taken at a site (B3) located at 760 m from the A22 highway during a period with moderate PM_{10} concentrations ($30 \mu\text{g m}^{-3} < \text{average value} < 50 \mu\text{g m}^{-3}$). One more sample (code F6) was taken at 550 m from the axis of the highway (site B7), during a period with high PM_{10} concentrations ($50 \mu\text{g m}^{-3} < \text{average value} < 90 \mu\text{g m}^{-3}$).
- 2. Trento South – Two samples (codes MS1 and MS2) were taken at 60 m (site B4) and 530 m (site B5) from the axis of the highway, respectively, during a period with low PM_{10} concentrations (average value $< 30 \mu\text{g m}^{-3}$); two samples (code MS3 and MS4) were taken at the same locations during a period with high PM_{10} concentrations ($50 \mu\text{g m}^{-3} < \text{average value} < 90 \mu\text{g m}^{-3}$). An additional sample (code MS7) was taken at 240 m from the axis of

the highway (site B6), during another period with high PM₁₀ concentrations ($50 \mu\text{g m}^{-3} < \text{average value} < 90 \mu\text{g m}^{-3}$).

- 3. Gardolo – PCDD/F concentrations (code G2) were measured by an urban traffic air quality station (site B8) during a period with low PM₁₀ concentrations (average value $< 30 \mu\text{g m}^{-3}$).
- 4. S. Chiara Park– PCDD/F concentrations (code SC2) were measured at an urban background air quality station (site B9) during a period with low PM₁₀ concentrations (average value $< 30 \mu\text{g m}^{-3}$).

The PM₁₀ concentrations considered in every campaign were recorded by the fixed air quality stations of APPA. For the campaign of Trento North, the reference air quality station is located in Gardolo (station code GAR), whilst for Trento South the air quality station of S. Chiara Park (station code SCH) was considered, representing the urban background. In 2007, further investigations on PCDD/Fs in air were performed at sites B1 (sample code AD07), B4 (sample code AD06), B6 (sample code AD05), B7 (sample code AD08) and B8 (sample code AD02).

During late summer 2009 and late winter 2010 (campaign C), three sites in the surroundings of Trento were studied: a school in the village of Zambana (site C1), a public park located in the village of Lavis (site C2) and a public park in the village of Terlago (site C3). The sites chosen for campaigns A, B and C are presented in Fig. 1.

Two sites were selected for the samplings carried out in 2010 by APPA in Valsugana (campaign D): the first site (D1) is located at a fish farm, about 900 meters west from the steel making plant, while the second site (D2) is located about 1,500 meters east from the plant, in a yard (Fig. 2). Two monitoring activities were conducted in order to check the presence of PCDD/Fs in the air: the first sampling was performed between 4 January 2010 and 8 January 2010, during a period of suspension of the activity of the steel plant; the second sampling was carried out between 1 February 2010 and 5 February 2010, when the plant was operating.

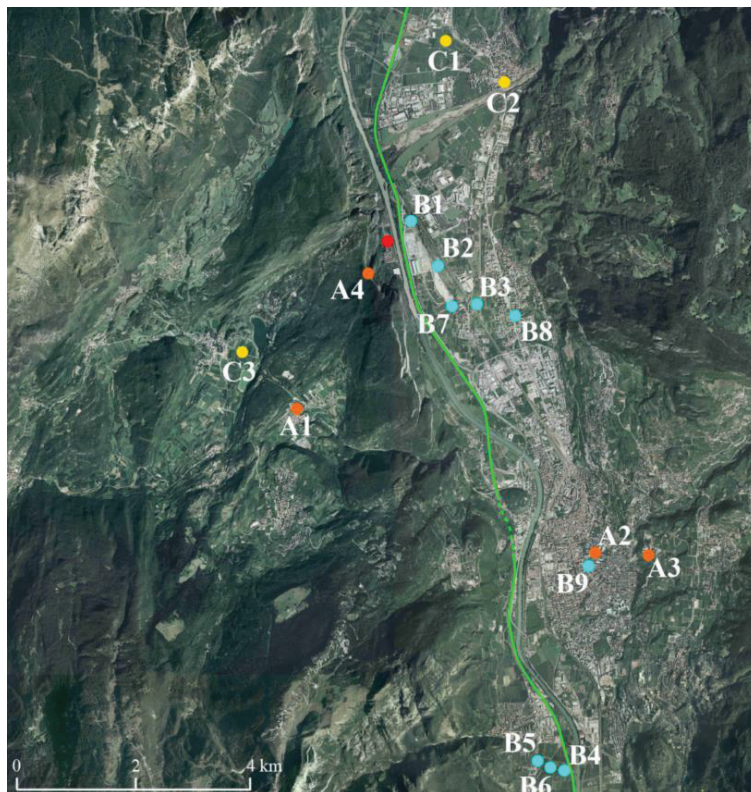


Fig. 1. Route of the A22 highway (in green), location of the expected site for the MSWI (red circle) and location of the sampling sites during the monitoring campaigns A (orange circles), B (light blue circles) and C (yellow circles).



Fig. 2. Route of the A22 highway (in green), location of the expected site for the MSWI (red circle) and location of the sampling sites during the monitoring campaigns A (orange circles), B (light blue circles) and C (yellow circles).

The air samples were taken in accordance with the USEPA specifications, Compendium Method TO-9A [19]. This method is intended for the quantitative determination of polyhalogenated PCDD/Fs in ambient air, which include PCDD/Fs, polybrominated PCDD/Fs, and bromo/chloro PCDD/Fs. The method uses a high volume air sampler equipped with a quartz-fiber filter and polyurethane foam (PUF) adsorbent for sampling 325 to 400 m³ of ambient air in a 24 hour sampling period. The samples are analyzed using high resolution gas chromatography connected with high resolution mass spectrometry.

To relate the PCDD/F concentration to its toxicity, the International Toxic Equivalency (I-TEQ) scheme was used [20]. Given the importance of PCDD/Fs in terms of carcinogenic potential, the total I-TEQ concentrations measured during the monitoring campaigns here presented will be reported together with the related inhalation cancer risk. For this case study, the estimation of a cancer risk is possible only for inhalation, due to the absence of deposition measurements. To calculate the inhalation cancer risk, the total I-TEQ concentrations will be divided by the so-called Inhalation Unit Risk (IUR), defined as the upper-bound excess lifetime cancer risk deriving from continuous exposure to 1 µg m⁻³ of a carcinogenic compound in air [21]. In the case of 2,3,7,8-TCDD, the most toxic PCDD/F congener, the IUR is 1.75E+01 [22].

3. Results and discussion

The background levels of PCDD/Fs acquired during campaign A are generally mild (Table 1). PCDD/F concentrations in terms of I-TEQ were between 11 and 110 fg I-TEQ m⁻³. The results do not show significant differences related to the localization of the sampling sites and mirror the substantial uniformity of the emissivity of the area under investigation.

Campaign B showed concentrations between 10 and 75 fg I-TEQ m⁻³ (Table 2). Two exceptions were detected for the first sample (code I1), which shows a value of 115 fg I-TEQ m⁻³, and for the measurement performed at the air quality station of Gardolo (code AD02), with a value of 138 fg I-TEQ m⁻³. The fact that these values are not confirmed by other samplings can be explained by the presence of particularly unfavorable environmental conditions, as confirmed by the values of PM₁₀ detected by the air quality station of Gardolo in the same period (96 and 86 µg m⁻³ respectively).

From the available data, a significant correlation between the total absolute PCDD/F concentrations and PM₁₀ is not evident. However, a slight correlation can be observed by considering the measurements carried out at the sites sites B1, B2, B3 and B7 and at the air quality station of Gardolo (site B8). By taking into account also the data collected in Trento South, a correlation cannot be observed. This may be due to the fact that the air quality in Trento North (mostly affected by vehicular traffic and industrial activities) differs from the one of Trento South (urban-rural background).

Table 1. Congener-specific concentrations (expressed in fg m⁻³), total PCDD/F concentrations and I-TEQ concentrations [20], measured during the air quality monitoring campaign carried out in 2002/2003 at background sites; in the calculation of the total PCDD/F concentration, values below the detection limit were assumed equal to half the detection limit itself.

Site	A1		A2	A3	A4
Sampling period	2002	Summer 2002	Winter 2002/2003	Summer 2002	Autumn 2002
2378-TCDD	< 10.1	<1.6	<6.5	0.6	0.6
12378-PeCDD	20.4	2.2	16.1	1.0	4.2
123478-HxCDD	16.8	2.3	14.9	1.0	4.4
123678-HxCDD	35.9	47.7	29.4	13.3	8.6
123789-HxCDD	29.5	4.5	27.3	1.5	6.2
1234678-HpCDD	267.3	67.0	261.7	20.5	66.1
OCDD	613.6	576.6	616.0	117.8	197.5
2378-TCDF	80.5	16.6	44.7	9.1	21.3
12378-PeCDF	64.6	9.3	28.4	5.0	9.3
23478-PeCDF	99.7	21.5	56.6	8.0	18.1
123478-HxCDF	51.6	16.6	40.2	6.8	10.8
123678-HxCDF	49.1	13.6	38.1	5.8	11.0
123789-HxCDF	77.9	23.6	50.4	7.9	14.7
234678-HxCDF	30.5	10.5	19.5	3.4	4.1
1234678-HpCDF	112.7	53.0	122.6	24.2	33.9
1234789-HpCDF	36.7	9.7	22.0	5.0	4.9
OCDF	81.1	72.4	65.9	38.9	36.0
Total PCDD/Fs [fg m ⁻³]	1673.0	947.9	1457.1	269.5	451.4
I-TEQ [fg I-TEQ m ⁻³]	110.4	28.6	72.2	10.9	21.6
Cancer risk [-]	5.1E-06	5.0E-07	1.3E-06	1.9E-07	3.8E-07

No correlation can be observed between the distance of the sampling sites from the A22 highway and the total PCDD/F concentrations. Thus, the highway seems not to represent a criticality within the PCDD/F emissive framework, even though road traffic and, especially, diesel vehicles are acknowledged sources of PCDD/Fs. As a consequence, it is reasonable to exclude an important PCDD/F contribution by the highway also outside the urban area, where cultivated lands are present.

The results of the campaign carried out in 2009/2010 (campaign C) are shown in Table 3. The concentrations of PCDD/Fs, expressed in I-TEQ [20], recorded during late summer, show values between 5.60 and 7.74 fg I-TEQ m⁻³, increasing to 10.55-25.62 fg I-TEQ m⁻³ in late winter. For the characterization of PCDD/Fs, the ratio between furans and dioxins (PCDFs/PCDDs) and the profile of homologues are two important parameters. Similar thermal processes show similar fingerprints, due to the analogous mechanisms of formation of some PCDD/F compounds [23]. The typical fingerprints from combustion sources are represented by a ratio PCDFs/PCDDs greater than 1, the increasing distribution of homologous in weight at increasing degree of chlorination for PCDDs, and with a maximum at PeCDF or HxCDF for PCDFs. This may be the case of campaign C, since the measurements confirm the above mentioned characteristics (Fig. 3). PCDDs prevail compared to PCDFs in every site, with a ratio PCDF/PCDD variable between 0.32 and 0.50 in summer, while in winter this ratio assumes a value of about 0.9 for sites C1 and C2. With regard to the ratio between PCDD/Fs and the equivalent toxicity, the values are comparable in every site and vary between 24.45 and 28.19 in late summer and between 17.18 and 29.62 in late winter. Given this variability, a dominant source cannot be properly identified. The measured concentrations may be the effect of a complex interaction between the different sources that influence the air quality at the investigated sites. The choice of the sampling period is important too: in summer, the release of PCDD/Fs into the atmosphere is lower, due both to the absence of important emission sources (e.g., biomass burning for heating purposes) and to the meteorological conditions that facilitate the dispersion and the dilution of the pollutants.

Table 2. Comparison between the values of PCDD/F and of PM10 during the monitoring campaigns carried out between 2006 and 2007.

Site	B1	B2	B3	B4	B5	B6	B7	B8	B9
Sample code	I1	AD07 I2	T2	MS1 MS3	AD06 MS2	MS4 MS7	AD05 F6	AD08 G2	AD02 SC2
Sampling period [mm.dd]	02.16	02.19 02.23	02.22	03.06 03.14	02.13 03.06	03.13 03.25	02.05 03.23	02.27 04.04	01.18 04.10
Year	2006	2007 2006	2006	2006 2006	2007 2006	2006 2006	2006 2007	2006 2007	2006 2006
Distance from A22 [m]	100	550 760	60		530	240	550	1500	1200
Nearest air quality station	GAR	GAR GAR	SCH		SCH	SCH	GAR	GAR	SCH
Average PM ₁₀ [$\mu\text{g m}^{-3}$]	96	26 35	37	17 61	29 21	59 63	61 54	38 21	86 16
PCDD/Fs [fg m^{-3}]	6640	870 4445	4577	1226 2477	598 3744	2283 1787	1487 2492	375 1014	2745 693
PCDD/Fs [fg I-TEQ m^{-3}]	115	23 57	69	17 33	31 49	33 27	75 32	18 17	138 10
Cancer risk [10^{-6}]	2.0	0.4 1.0	1.2	0.3 0.6	0.5 0.9	0.6 0.5	1.3 0.6	0.3 0.3	2.4 0.2

The results of the campaign conducted in winter 2009-2010 by APPA in Valsugana (campaign D) are shown in Table 4: the values are not critical, but in the period 6-8 January 2010, at site D2, a PCDD/F concentration of 275 fg I-TEQ m^{-3} was found, when the steel plant was not operative. An unexpected very low concentration (0.4 fg I-TEQ m^{-3}) was found in the most remote site (D1). The results of a monitoring campaign carried out in a similar context showed average concentrations between 16.9 and 79.5 fg I-TEQ m^{-3} [24]. With the exception of the two extreme values found, this range mirrors the concentrations measured during campaign D.

In general, the PCDD/F concentrations measured during campaigns A, B and C are in line with the results of other studies carried out in Italy: total PCDD/F concentrations between 1,820 and 2,010 fg m^{-3} were measured in the urban area of Brescia [18]; mean concentrations of 81 and 61 fg I-TEQ m^{-3} were found in the urban areas of Susa and Torino, respectively [25]; the results of a six-month winter monitoring campaign in Rome showed values between 48 and 87 fg I-TEQ m^{-3} [26]; concentrations between 22 and 39 fg I-TEQ m^{-3} were measured in the Po Valley near a MSW plant; a background value of 10 fg I-TEQ m^{-3} was found in a rural area close to Bolzano, whilst a concentration of 221 fg I-TEQ m^{-3} was measured in an industrial area of the Veneto region [27].

Table 3: PCDD/F measurements in air collected during spring 2009 and winter 2010.

Site	C1	C2	C3
Sample code	AD01Z	AD06Z	AD02L AD05L AD03T AD07T
Sampling date [mm.dd]	09.11	03.24	09.21 03.23 09.28 04.08
Year	2009	2010	2009 2010 2009 2010
PCDD [fg m^{-3}]	103.47	233.46	133.35 223.92 138.07 243.35
PCDF [fg m^{-3}]	33.56	206.75	54.26 194.24 68.57 69.19
PCDD/Fs [fg m^{-3}]	137.03	440.21	187.61 418.16 206.64 312.54
PCDF/PCDD	0.32	0.89	0.41 0.87 0.50 0.28
PCDD/Fs [fg I-TEQ m^{-3}]	5.60	25.62	6.66 20.02 7.74 10.55
Cancer risk [10^{-6}]	0.1	0.5	0.1 3.5 01 1.9
PCDD/F/I-TEQ	24.45	17.18	28.19 20.89 26.69 29.62

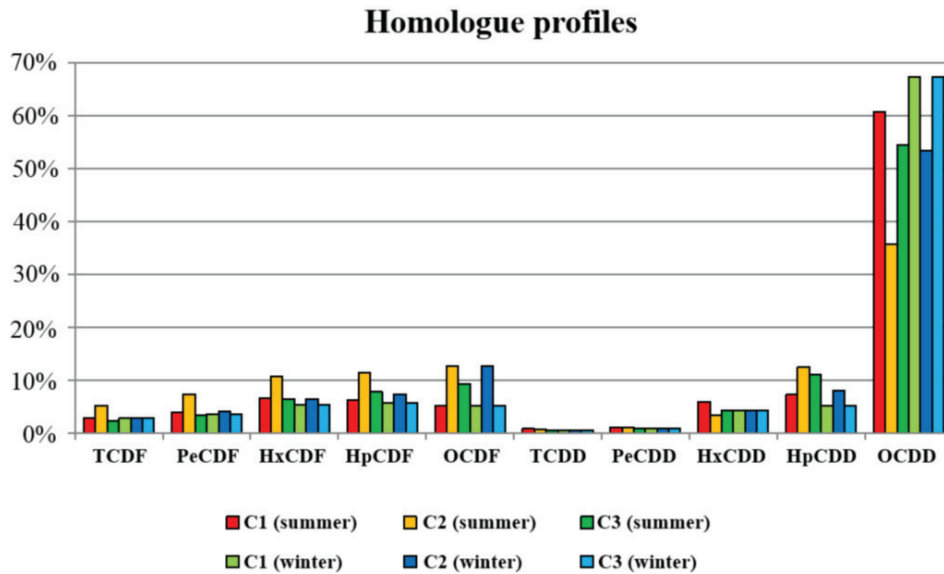


Fig. 3. Homologue profiles for the measurements carried out during campaign C.

The cancer risk related to the exposure to the levels of PCDD/F concentration found during the four monitoring campaigns is acceptable among all the samples, with a maximum of 5.1×10^{-6} at site A1. 10 samples out of 35 showed that the concentrations would imply a cancer risk higher than 1.0×10^{-6} , which is considered the optimal standard value for risk assessments related to one source.

After more than 10 years of PCDD/F characterizations in the air of the region, some considerations can be made about a strategy of sampling for the future. Until 2013 the interest for PCDD/F characterization was related basically to the future role of the planned incinerator and the present role of a steel making plant. Specific interest has been related to the local role of wood combustion and highway presence. The recent decision of modifying the management of municipal solid waste towards energy recovery not based to a local incinerator and the favorable results related to the area of the local steel making plant makes it less interesting the setting of a multi-year strategy of PCDD/F characterization in air in the region. A yearly soil sampling in the area of the steel making plant can be adequate for control purposes.

4. Conclusions

The air quality monitoring campaigns carried out between 2002 and 2010 in the province of Trento show relatively low PCDD/F concentrations, in line with the values found in other Italian urban and industrial sites. Anomalous results were only detected during the monitoring campaign carried out in the surroundings of the steel making plant: in particular, a very low concentration and an unexpected peak when the plant was not operating were found ($0.4 \text{ fg I-TEQ m}^{-3}$ and $275 \text{ fg I-TEQ m}^{-3}$, respectively). Typical values for agricultural regions were measured in rural areas, with an increase during winter, probably due to biomass burning; typical values for urban areas were found in the urban sites, with higher concentrations, compared to rural sites, due to the presence of more combustion sources; however, no critical situations were detected, especially along the A22 highway, in spite of the non-negligible emission factors reported in the literature. About this aspect, the need for up-to-date emission factors is clear, since the most recent data refer to 2004 [28]. In fact, the vehicle fleet is continuously developing, as well as the technology of engines and off-gas treatment systems. Thus, the emission factors present in the literature may overestimate the current emissive contribution from road traffic.

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