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Management of atmospheric pollutants from waste incineration processes: The case of Bozen

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Abstract

This article presents the case study of a waste incinerator located in a region rich in natural and environmental resources, and close to the city of Bozen, where there are about 100,000 inhabitants. Local authorities paid special attention to the effect of the plant on human health and the surrounding environment. Indeed, among the measures adopted to control the emissions, in 2003 an automatic sampling system was installed specifically to monitor polychlorinated dibenzo-p-dioxin and polychlorinated dibenzofuran (PCDD/F) emissions during the complete operation time of the plant. The continuous sampling system was coupled directly to aerosol spectrometers for the determination of fine and ultra-fine particles in the emissions of the plant. The measurement results suggest that the waste incineration plant of Bozen is not a significant source of PCDD/F, or fine and ultra-fine particles. Immission measurements from other monitoring systems confirmed these results.

Keywords

Incineration, MSW, PCDD/F, PM, sampling, ultra-fine particles, air pollution

Introduction

In the sector of municipal solid waste (MSW) management, incineration options have undergone significant improvements in recent years, as demonstrated in many life cycle analysis studies and research (Consonni et al., 2005; Damgaard et al., 2010; Fruergaard and Astrup, 2011; Larsen and Astrup, 2011; Marculescu, 2012). However, at least for the sector of MSW, incineration still remains the major contributor of polychlorinated dibenzo-p-dioxin and polychlorinated dibenzofuran (PCDD/F) emissions (Rada et al., 2006). In a wider context, metallurgy and uncontrolled burning are the most relevant sources in countries where environmental controls are more carefully operated (Antunes et al., 2012; Lemieux et al., 2004). As the method of release into the atmosphere may significantly change the role of a MSW plant in terms of PCDD/F immissions (Rada et al., 2011), the characterization of the performance of a MSW incinerator should take into account both the assessment of stack emissions and the measurements of pollutants at ground level. In most countries flue gas dispersion modeling takes place in each prevalent weather type, with its concomitant wind direction and speed, and simulates this effect. In this respect a recent article pointed out how the contribution of a modern incinerator, assessed by modeling, can be very low compared with the values that the local monitoring stations can typically measure (Ragazzi and Rada, 2012).

The European stack emission limit for municipal waste incineration plants is 0.1 ng_{TEQ} Nm^{-3}, and similar limits are discussed for other industrial plants. PCDD/F stack gas sampling at stationary sources is usually performed by manual sampling according to Ente Nazionale Italiano di UNIficazione (UNI) norms (UNI, 1999a,1999b,1999c). This allows sampling times between 6 and 8 hours. Only 2–3 characterizations per year are needed to comply with the regulation. Public concern often focuses the PCDD/F emissions during the remaining time. Indeed, the use of a long-term sampling system can enable sampling for a week or even longer (Kahr and Steiner, 2002a; Reinmann, 2002). Thanks to the larger sampling volume detection, statistical variations can be reduced (Tirler et al., 2003).

In addition to PCDD/F, MSW incineration, like many other plants including combustion processes, may be a potential source of other atmospheric pollutants, especially fine particles. As in the case of asbestos, with its long latency (in the order of decades), there are many legitimate concerns about the unknown human health consequences from nano-materials and, specifically, ultra-fine par-

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ticles from specific sources, such as incinerators (Brunner et al., 2006). The need to understand the role of the incinerator of Bozen, Italy, in terms of local contribution to the concentration of ultra-fine particles, motivated the development of specific research activity. The contents of the present article refer to some results from this multi-year research activity, which were taken into account during the local decision process aimed at implementing a new plant (presently under construction) to substitute the one which is the subject of the present case-study. The latter had to be substituted because of concerns about the structure, as its cement structure was built in 1994. However, the need for a new plant was considered as an opportunity to update the adopted technology for reducing pollutant emissions and improving energy generation.

The plant is located in an area renowned for its natural heritage, environmental quality, intensive mountain agriculture and beautiful landscapes. Furthermore, more than 100,000 people live in the urban and suburban areas of the city of Bozen, close to the plant. The incinerator is designed on two parallel lines, with a total treatment capacity of up to 400 t d^{-1} of MSW. The combustion takes place on a roller-type grate with a secondary combustion chamber; energy is recovered through a water tube boiler and a steam turbine, providing electricity and heat for district heating. Flue gas cleaning is performed by a fabric filter and a two-stage wet scrubber, in line with a final selective catalytic reduction unit for nitrogen oxides (NO_x) and trace organics conversion.

A fabric filter aims to remove most of the particulate matter from the off-gas. The wet scrubber plays a secondary role for this parameter as its main purpose is the removal of acid gases.

Only residual municipal solid waste is treated at the plant. As a consequence, the chlorine content in the input is always far from critical values for hydrogen chloride and PCDD/F generation. Temperatures ranging between 900°C and 1000°C typically occur in the combustion chamber. The oxygen content in the off-gas is kept higher than 7% in order to favor a highly efficient combustion.

Methods

Emission control of PCDD/F

PCDD/F sampling, clean-up and quantification of the incineration plant of Bozen were performed in accordance with the present European Standard Protocols using $^{13}C_{13}$-labeled standards (UNI 1999a, 1999b, 1999c). The flue gas is released through a 50-m high stack, with an inner diameter of 2.5 m. Since 2003, when an automatic sampling system, suitable for monitoring PCDD/F emissions during the complete operation time of the plant, was installed at this incinerator, two probes (2500 mm and 1300 mm long, respectively) allow representative samples to be taken in the relatively large stack.

Short-term (8 h) flue gas sampling was performed by the filter-cooler method and conducted with an automatic, continuous adjusting isokinetic sampling system. Extraction of filter media was done by soxhlet with toluene and extraction of aqueous liquid by liquid/liquid extraction with dicloromethane. An automatic two-column system was used for chromatographic sample clean-up. Pre-packed Teflon® disposable columns containing multilayer silica and alumina were utilized. A long-term sampling system is a mechanically-engineered system that resembles the manual sampling approach for PCDD/F emitted from stationary sources. A small quantity of the flue gas was sampled continuously on a sampling cartridge. Depending on the sampling time—which ranged from 6 hours to 6 weeks—variously representative measurement results were obtained.

Immission control of PCDD/F

The meteorological characteristics linked to release processes concerning the Bozen municipal waste incinerator were examined. The target area lies in the Adige Valley in the Alps, running approximately in a North–South direction. A peculiar consequence of the above conditions is the frequent occurrence of along-valley winds. In particular, the latter typically blow up-valley during daytime and down-valley during night-time in the warm seasons under fair weather conditions (de Franceschi and Zardi, 2009; de Franceschi et al., 2002, 2009; Rampanelli et al., 2004). Accordingly the main deposit sites were identified south and north of the plant respectively, on the basis of a previous numerical modelling assessment (DICA, 2001). The interested reader will find additional information (e.g. dilution factors) in a recently published paper (Ragazzi & Rada, 2012). The first monitoring campaign, carried out in August and September 2006, used two directional air samplers placed to the north and south of the incinerator, each with two cartridges and collecting samples with the wind coming from the north and south respectively.

To obtain more information, a second sampling campaign (December 2006–January 2007) with three directional air samplers was performed. The third sampler was installed in the centre of Bozen, in an area not influenced directly by the incinerator, collecting on two sampling cartridges when the wind came from north or south respectively. The other two samplers still worked in the same place north and south of the incineration plant. These samplers were able to work with three cartridges to collect the air, with the third cartridge also able to work under calm wind conditions. The parameters analyzed were PCDD/F, polycyclic aromatic hydrocarbons and, for the samplers north and south of the incinerator, also particulate matter $(PM)_{10}$, $PM_{2.5}$ and PM_1.

The adopted test standards were EPA 1613 for PCDD/F (EPA, 1994), gravimetric assessment for PMs and gas chromatographic–isotope dilution coupled with high-resolution mass spectrometry for benzo(a)pyrene (BaP).

Ultra-fine particle measurements

To measure the emissions of particles with an aerosol spectrometer directly on the stack is usually not possible. Stack gas can contain many compounds and also a relatively high amount of water. Therefore, condensation can occur when the gas is cooled

down during sampling. Condensation will trap most of the particles and bias the obtained results. Condensation of liquid will also damage the measurement devices. So dilution is necessary in the case of MSW incineration plants. In the present case a modified continuous PCDD/F sampling system from monitoring systems was used (Kahr and Steiner, 2002b). The sampler works by the dilution method described in EN-1948 Part 1 (MCERTS, 2010). The sampling equipment permits dilution of flue gas while maintaining automatic isokinetic sampling. In the present case the hot stack gas (160°C) was diluted with pre-cleaned air at a ratio of 1:7. The continuous PCDD/F sampling system was coupled directly to aerosol spectrometer for the determination of fine and ultra-fine particles. The instrument used to detect fine dust was an aerosol spectrometer manufactured by Grimm (model 1.108). The dilution of the flue gas avoids condensation within the aerosol spectrometer. The instrument makes it possible to determine the number of particles, as well as the particle mass. The aerosol spectrometer classifies the particles size in 15 different size channels from 0.25 up to 20 µm. The obtained size distribution spectrum permits qualitative consideration about the origin of PM. Sampling for ultra-fine particles was performed in the same way as for fine particles. For the detection of ultra-fine particles a condensation particle counter from Grimm, Ainring, Germany (model CPC 5403) was used, coupled with a Vienna-type differential mobility analyzer 55706 operating within a range from 5.5 and 350 nm. The granulometric classification of the ultra-fine particles is based on analysis of their electrical mobility.

Results and discussion

Results are presented starting from PCDD/F emission and immission data; PM emission and immission measurements complete this section. In Figure 1 examples of short- and long-term sampling are reported. The short-term PCDD/F concentrations measured at the stack are included between 10% and 80% of the emission limit—the long term average being about a third of it.

Concerning immissions, PCDD/F concentrations determined in the first campaign showed values ≤20 fg_{TEQ} m^{-3}. With northerly winds the immissions resulted in 6 fg_{TEQ} m^{-3} at the northern sampling point (Figure 2, site 3) and 20 fg_{TEQ} m^{-3} at the southern sampling point (Figure 2, site 5). With southerly winds the immissions resulted in 11 fg_{TEQ} m^{-3} at the northern sampling point and 15 fg_{TEQ} m^{-3} at the southern sampling point. These measurements showed values of PCDD/F in the air comparable with the ones of domestic/rural areas in summer.

Results from the second sampling campaign showed that the influence of the incineration plant on the PCDD/F concentration in the air of Bozen is simply not visible, confirming the results of previous preliminary research on the influence of this plant (Caserini et al., 2004): in winter (22 December 2006–18 January 2007) PCDD/F air concentrations ranged from 79 to 90 fg_{TEQ} m^{-3} at the Northern sampling point and from 52 to 84 fg_{TEQ} m^{-3} at the southern sampling point, whilst at the third site (Figure 2, site 6), not exposed to the main contribution of the plant, the values ranged from 155 to 259 fg_{TEQ} m^{-3}. The highest value was measured at the north sampling site under northerly winds. When this air was advected by the wind to the southern sampling site, a lower concentration was measured. The same situation occurred under southerly winds. The northern sampling site showed similar results. Relatively high concentrations were found under calm wind conditions. The above facts provide clear evidence that domestic heating and road traffic are the main sources of persistent organic pollutants (POPs) in the air of the areas analyzed. Concerning the above results, previous modeling confirmed the low significance of the plant contribution in terms of PCDD/F concentrations (DICA, 2001). Indeed, the two areas of highest impact are reported in Figure 2, where the results of the cited modeling are presented for PCDD/F. The maximum values are significantly lower than the ones measured in the ambient air in the present case study: even 2–3 orders of magnitude lower. In Figure 2 six points are marked in order to point out significant locations for unconventional measurements: 1—incineration stack; 2—incineration area close to a highway; 3—northern rural area; 4—suburban area; 5—southern rural area close to the highway; and 6—urban area.

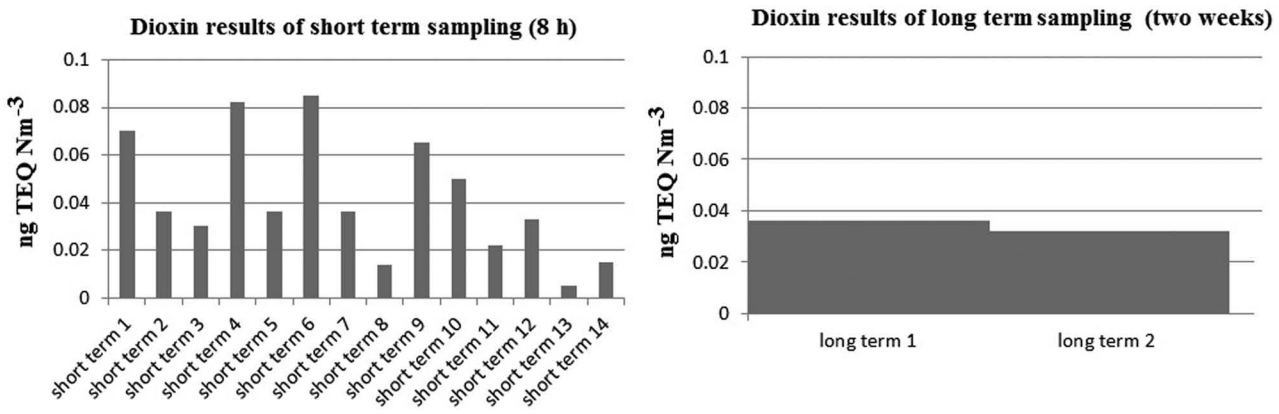

Figure 1. Results of short-term and of continuous, long-term (two weeks) sampling at the stack; results express the average concentration value over the sampling period.

Active air sampling presents a powerful tool in monitoring wind transport of POPs (Tirler et al., 2007a). The measurement results reported in Table 1 show that the waste incineration plant of Bozen is not a significant source of PCDD/F, PAHs or fine particles (the sampling site is the number 5 in Figure 2). Indeed, the concentrations of PCDD/F in ambient air are low compared with other case studies from the European Union (EU) (Vilavert et al., 2012) and non-EU countries (Bakoglu et al., 2005; Xu et al., 2009). In areas influenced by incineration with regulation not as stringent as the one of EU, the ambient air concentration can reach even some $pg_{I\text{-}TEQ}$ m^{-3} (Xu et al., 2009). Regarding the distribution of organic pollutants, approximately 90% could be found in the PM_1 fraction. This fraction presents, in the cited study, only 70% of the PM_{10} fraction. PM_1 has a higher specific surface where organic pollutants can easily be bound on the surface. Data from Table 1 show that when the wind blows from the south the PM concentrations at the site show values higher than the ones when the wind blows from the north; taking into account the location of the incinerator, northern with respect to the site, the conclusion is that the plant is not a significant source of PM.

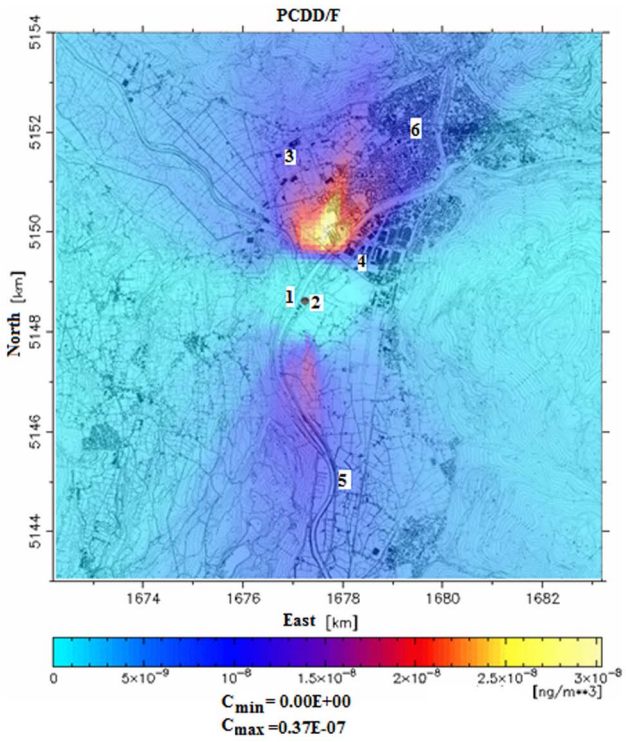

Figure 2. Medium annual concentration at ground level for polychlorinated dibenzo-p-dioxin and polychlorinated dibenzofuran (1: plant location; 2–6: sampling sites).

Measured data showed very low fine dust concentrations (0.25–20 μm) in the flue gas; indeed, the cleaning system of the plant showed a high efficiency in removing fine particles (Tirler et al., 2007b). The levels of some classes were lower than in the air outside of the plant, which is also dependent on the presence of a highway close to the plant.

In the case of ultra-fine particles, the measured flue gas concentrations (Figure 3) were lower than the ones in the area of the plant from where the combustion air is taken. This means that the off-gas treatment line has a high efficiency in removing them. This characteristics were analyzed and quantified recently (Buonanno et al., 2012).

Figure 3 also illustrates some other interesting aspects:

- the presence of ultra-fine particles in the northern rural area is relatively low—at this site the proximity of the area of highest impact to the incinerator does not influence the values; indeed, its contribution is expected to be low (the dilution of ultra-fine particles emitted at the stack makes the role of the plant negligible);
- the contribution of the highway is visible if compared with the rural area, but seems not to be the only significant source; indeed, in the rural site close to the highway (Figure 2, site 5) the ultra-fine concentration is lower than in the urban and suburban sites;
- data in Figure 3 are reported as number of particles in a reference volume; indeed, in case of ultra-fine particles the mass

Table 1. Polychlorinated dibenzo-p-dioxin and polychlorinated dibenzofuran (PCDD/F)and benzo(a)pyrene (BaP) in the different particular matter (PM) fractions in the southern sampling site.

	PM_{10}		$PM_{2.5}$		PM_1	
		PCDD/F		PCDD/F		PCDD/F
	μg m^{-3}	fg_{TEQ} m^{-3}	μg m^{-3}	fg_{TEQ} m^{-3}	μg m^{-3}	fg_{TEQ} m^{-3}
Wind from north	44	59	38	56	33	53
Wind from south	78	52	70	50	63	47
Calm of wind	45	84	39	82	34	77
		BaP		BaP		BaP
	μg m^{-3}	ng m^{-3}	μg m^{-3}	ng m^{-3}	μg m^{-3}	ng m^{-3}
Wind from north	44	2.2	38	2.1	33	1.9
Wind from south	78	2.6	70	2.5	63	2.3
Calm of wind	45	3.1	39	3.0	34	2.8

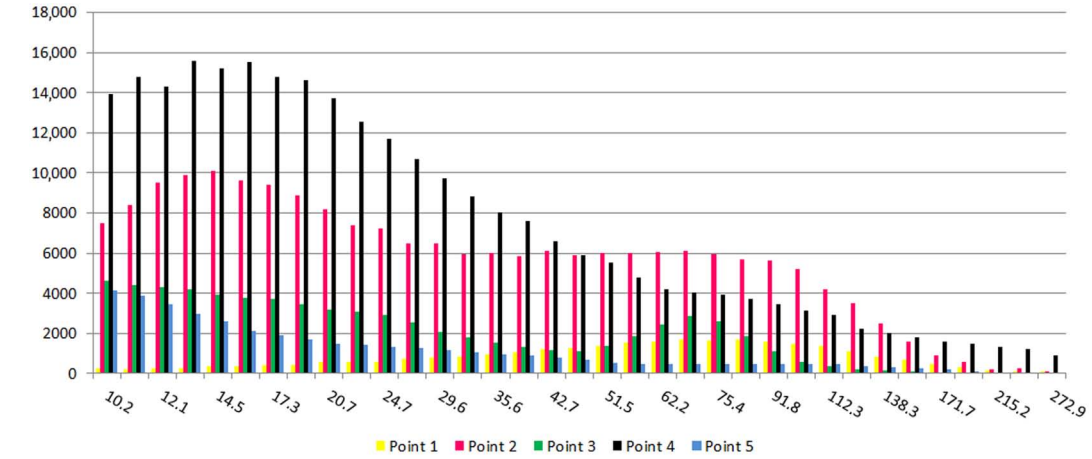

Figure 3. Ultrafine particles at the stack (sampling point 1) and in 4 additional points (sites 2–5).

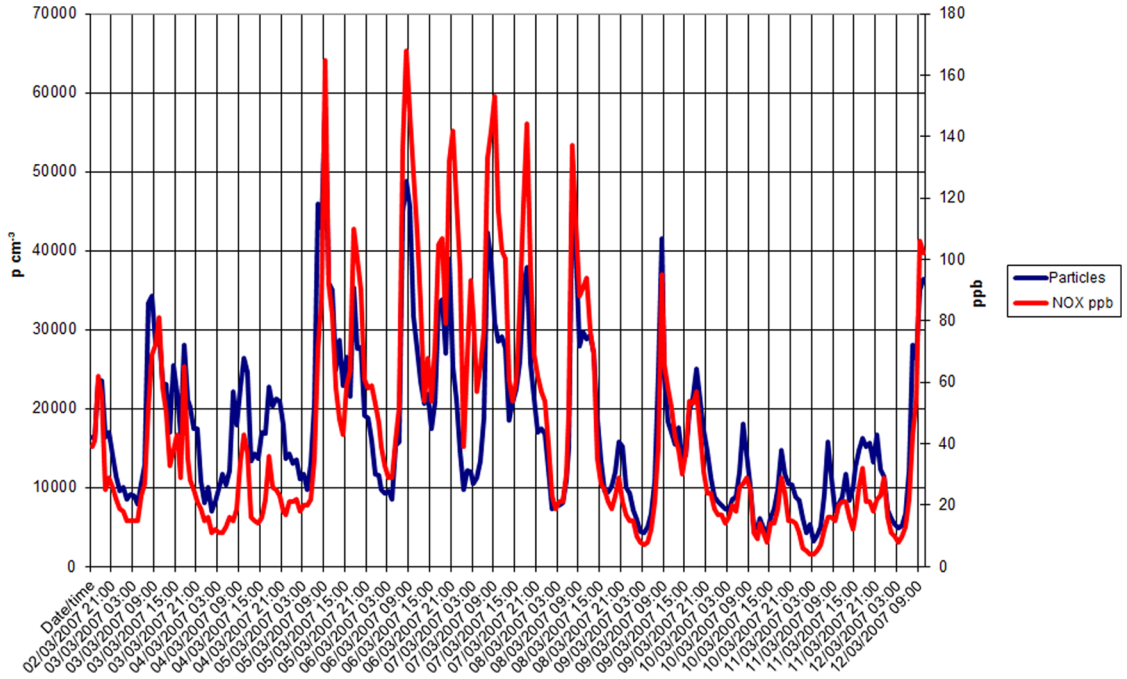

Figure 4. Time trend for ultra-fine particles (from 10 nm to 300 nm) and NO_x in the ambient air in Bolzano.

concentration is not a significant parameter—from this point of view the peaks of ultra-fine particles in the flue gas and in the ambient air are different: the granulometric class showing the highest value in the flue gas is less fine than the one of the peak in the ambient air characterization.

Data concerning the number of particles were also correlated with data taken from the air quality monitoring stations of Bozen center (Figure 2, site 6). There was good correlation as far as the parameters directly linked to vehicle traffic (benzene, toluene, carbon monoxide, nitrogen dioxide, nitric oxide, NO_x and number of vehicles) were concerned. An example is shown in Figure 4. The contribution of NO_x from the plant in the urban area of Bozen (Figure 2, site 6) was demonstrated to be negligible (Ragazzi et al., 2012), thus the NO_x dynamics can be attributed to the traffic. The similarity of the trends of NO_x and ultra-fine particles is clear. Thus, ultra-fine particles in the urban area depends on the traffic dynamics.

Conclusions

From the results described herein, it can be stated that the ultra-fine dust measurements for the town of Bozen could be correlated with traffic: the site close to the incineration stack, not influenced by its emissions, shows high values of ultra-fine particles in an area far from the town, but close to a highway (this highway crosses the town); however, the lower values measured in another site, a rural area close to the highway, demonstrate that the highway is not the only significant source of ultra-fine particles. The fluctuations of NO_x concentration in an urban site show

the same trend of the fluctuations of the number of ultra-fine particles, with peaks that are synchronized with the peaks of local traffic. Other sources of ultra-fine particles seem to be present, as demonstrated by the high values measured in a suburban area far from the highway and main roads, but additional studies should be developed (in particular, the role of domestic combustion of wood could be investigated). Finally, the waste incineration plant of Bozen is not a significant source of fine and ultra-fine particles, nor for PCDD/F. The availability of a detailed pollutant diffusion modeling (in 2001), of a long-term PCDD/F emission characterization (since 2003) and of ultra-fine concentration values (in 2006–2007) allowed the local authorities to manage successfully the pathway of the proposal of the new incineration plant presently under construction.

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