A contribution to the evolution of the BAT concept in the steelmaking sector

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Abstract
The sector of steelmaking, in the European Union, is considered today one of the major potential contributors to the atmospheric release and consequent deposition of dioxin. The emissions of a plant depend on many factors: characteristics of the input, process, prevention and removal of dioxin from the gaseous stream. The local impact depends also on the different emission path, namely diffused emissions, secondary emissions and conveyed gases, as each stream can be related to a different way of dilution into the atmosphere. Moreover, the local impact is also related to the meteorological conditions. The high variability of parameters that can affect the local impact from a plant can be analyzed through the deployment of deposimeters, in order to understand the human exposure in the surroundings. The present paper refers to an Italian case-study where deposition was measured using deposimeters placed in sites identified by means of the results of a dispersion model. The role of particulate matter emissions was studied also with an optical particle counter. The obtained values are discussed taking into account seasonality and operation of the plant. Some considerations on the potential evolution of this industrial sector beyond the present concept of best available technology are included too.

Keywords AERMOD dispersion model, deposimeter, optical particle counter, PCDD/F, PM₁₀, steelmaking, steelworks.

INTRODUCTION
In the European Union the steelmaking sector is nowadays considered one of the major potential contributors to the atmospheric release and consequent deposition of PCDD/F. Many factors are involved: level of contamination of the feeding material, process characteristics, prevention and removal of dioxin from the gaseous streams. The local impact of this kind of plants depends also on the emission path, which can be quite different, including diffused sources, secondary emissions and conveyed gases release. The potential release of dust from the plant activities can be significant too. The complexity of the subject suggests the opportunity of adopting integrated and multidisciplinary approaches.

This paper refers to a part of a multi-year research on an Italian case-study concerning an electric arc furnace steelmaking plant. Present depositions were measured by deposimeters and additional information was derived by means of an optical particle counter. The location of these devices was chosen on the basis of dispersion/deposition modeling results. Comparative results between different locations of deposimeters are discussed in order to identify the priority for a further decrease of the plant impact, an improvement that could go beyond the already adopted Best Available Technologies (BATs).

MATERIALS AND METHODS
The adopted dispersion model was AERMOD (USEPA, 2009), an advanced Gaussian plume model able to simulate the dispersion of pollutants in complex terrain. The wind field was derived from local meteorological observations. Emissions were assumed on the basis of the technical available information (APPA, 2012). The model is based on the solution of the stationary advection-diffusion equation and uses a sequence of stationary states with hourly steps. AERMOD can simulate the effect of many sources of different typologies, emitting various pollutants. This model is supported by two pre-processors: one for managing terrain data, the other for managing meteorological...
information. Data from two local meteorological stations were analyzed and the most representative for the study area was selected. A detailed analysis of the emission data from the available technical literature for the studied plant was performed in order to account for the characteristics of the different emission sources (APPA, 2012). Due to time variability in the cost of electricity, the plant often operates at nighttime (from 8 p.m. to 8 a.m.) during workweek and 24 h/d during the weekends.

Vetropyrex Depobulk® deposimeters (Figure 1left) were adopted; they are able to capture the overall deposition of PCDD/F in a standard period of 15-30 days. A special ring avoids interferences from birds excreta. The sampling period presented in this paper concerned many seasons synchronizing the characterization of the deposition with the activity of the plant (stopped for a few long periods, in central summer and at Christmas). An accurate cleaning was planned each time the deposimeters had to operate. Rainy and snow periods were observed in details in order to avoid an overflow of collected water from the deposimeters (data were collected from a local meteorological station). Ambient temperature was checked daily in winter in order to prevent damages to deposimeters from icy events (e.g. glass breakage). The deposition flux of PCDD/F was computed by dividing the collected mass by the surface of the deposimeter (0.0346 m²) and the measurement period.

The adopted optical particle counter (Figure 1right) was the GRIMM portable aerosol spectrometer model n. 1.108. It was adopted for specific comparative characterizations to help to clarify the role of particulate matter (PM) diffused emissions. Measurement sites were selected taking into account also the dispersion modeling results: in this way additional information for a better understanding of the local impact of PM was gathered.

**Figure 1.** Adopted deposimeter (left) and optical particle counter (right).

**RESULTS AND DISCUSSIONS**

The analysis of wind speed and direction measured in the two meteorological stations close to the plant allowed to derive the wind roses for speed and atmospheric stability classes. Both stations turned out to be characterized by low wind speeds and high percentage of wind calms. Moreover, the neutral and steady classes (D, E and F, according to Pasquill classification) resulted to be dominant, frequently determining unfavourable conditions to the dilution of pollutants. After a comparison of the two stations series, only one was selected for the meteorological pre-processing; the data of the chosen station were characterized by a dominant wind direction (East-West) oriented along the valley axis where the plant is located. The second station is on the contrary influenced from the local orography of the site and it was therefore considered unsuitable to represent the meteorological conditions in the surroundings of the plant. In Figure 2 the results of the meteorological analysis for the selected station are presented.

The yearly PCDD/F deposition map obtained from the model, along with the position of the steel mill and of three measurements sites, are reported in Figure 3. The available measured depositions, scaled to yearly values, are compared with the modeled values: the modeled deposition values result to be smaller than the measured ones (whose data are presented in details below), but they are of the same order of magnitude. Differences between measured and modeled values depend also on the presence of other sources of dioxin, as explained below. Data on the third measurement site are still in progress. The performed modeling allowed the assessment of the contributions of each PCDD/F
source from the plant. Modeling demonstrated that the contribution of the stack to this phenomenon is not significant (when complying with the present regulations). On the contrary, the effect of diffused emissions cannot be neglected.

Figure 2. Wind speed rose and stability rose for the selected station.

Figure 3. Map of modeled yearly deposition with the indication of the values of deposition measured in two locations (values in TEQ).

The most eastern site, showing the lower deposition value, was chosen for the PCDD/F deposition characterization inside the area of a primary school coupled with a kindergarten. The reason for this choice was related to: the location of the school, generally downwind the plant during operation time, the proximity to the emission sources, the sensibility of the receptor (children) and the demographic density of the area. The other measurement site is located in a waste recycling area, managed from the local Municipality and close to the plant. This site was considered more representative of the potential impact of diffused emissions. As expected, the deposition value resulted higher than the one of the first site.

The area of interest is located in a U-shaped valley, where the winter and summer dilution patterns are significantly different. Moreover, winter domestic heating can increase PCDD/F emissions in the area. Deposition values were expected to be variable during the year. The obtained values, reported in Figure 4 and 5, confirm this variability: the periods when the plant was stopped are
pointed out with black margins. Keeping in mind that weekends are related to full-time operation of the plant, their number during each measurement period is reported in the legend.

Figure 4. Comparison of deposition values at the school site (farther site).

Figure 5. Comparison of deposition values at the waste recycling site (closer site).

From data in Figure 4 (school site, farther site) and Figure 5 (waste recycling area, closer site) some considerations can be drawn:

- from August to September the increase of PCDD/F deposition is clear in both the cases related to the farther site; anyway these differences cannot be strictly attributed to the plant (closed and re-opened) because:
  - there is no demonstration that the material used as input of the plant remained the same;
  - the comparable deposition values in Figure 5 (closer site) do not point out this increase;
- the winter deposition peak at the farther site is related to a period during which the plant was not working; this fact may suggest a local presence of additional PCDD/F sources;
- concerning peak intensities, the higher winter peak at the closer site is related to a period during which the plant was re-opened, but the second and third maxima refer to periods when the plant was closed;
the comparison between the values for the two sites show that the proximity of the plant not always determines the higher depositions, even if the overall average value is higher;

all the values remained below the limits proposed in some other countries:
o in the Flanders a range of 3.4 – 14 pg_{TEQ} m^{-2} d^{-1} referred to the maximum annual average deposition is proposed (van Lieshout et al., 2001);
o in Germany a threshold value of 15 pg_{TEQ} m^{-2} d^{-1} is taken into account (LAI, 2000).

the measured values are often similar to the ones measured in rural areas; for example a range of 2.2 – 3.3 pg_{TEQ} m^{-2} d^{-1} was found in rural forest sites in Denmark (Hovmand et al., 2007). These values refer to daily depositions on yearly basis.

In Figure 6 and 7 the ratios between total PCDD/F and TEQ are reported for each sampling period. The variability of the values could be explained by the absence, in the area, of a source of PCDD/F dominating all over the year.

**Figure 6.** Comparison of the ratios PCDD/F / TEQ at the school site (farther site).

**Figure 7.** Comparison of the ratios PCDD/F / TEQ at the waste recycling site (closer site).
The obtained deposition values, even in the hypothesis of attributing them entirely to the plant, show that by complying with recent regulations (a limit of 0.5 ngTEQ/Nm³ has been recently imposed to the plant), dioxin deposition can be lower than some environmental management targets. The same consideration cannot be applied to past depositions, due to less stringent limits over the past decades. Moreover, a variation in the characteristics of the plant feed and process management could increase the deposition values although, during this study, the PCDD/F emissions of the plant remained significantly lower than the limit (APPA, 2012).

Deposition of heavy metals was contextually measured in the area close to the plant (results of these measurements not reported in this paper). Nevertheless, high values of heavy metals depositions did not correspond to high values of PCDD/F depositions. One possible explanation could be a low temperature stripping of part of the PCDD/F present in the slag cooled outdoor. Indeed the PCDD/F stripping phenomenon was recently pointed out in unexpected sectors (Rada et al, 2007).

From these observations a priority for going beyond the present BAT concept may be represented by a different management of slag, presently performed outdoor with uncontrolled emissions. The adoption of an on-line PCDD/F sampling system at the stack makes sense only after an extreme minimization of the PCDD/F diffused emissions, that could be a second priority.

Among all the hypotheses for the quantification of the diffused emissions (APPA, 2012), the choice of the most reasonable estimate of diffused emissions of PM was made also taking into account the values of the Air Quality station present in the area (located very close to the school site). The highest potential impact, as estimated by the dispersion model, resulted to be about 100 m South of the plant, in an area with low urbanization. The concentrations induced in the most populated area have been assessed to be smaller than 10 µg/m³ on yearly basis.

![Figure 8](image.png)

**Figure 8.** Expected ground level PM₁₀ concentrations (yearly averages) according to the most realistic diffusive scenario.

As the PM concentrations strongly depend on the assumptions on the amount of diffused emissions, the results of a few measurements by optical particle counters were used for a deeper understanding of the local impact of the plant. In this paper, data recorded in two sites (Figure 9) are reported. Measurements in Figure 10 and 11 were made on the evening/night of July 25th 2011 (4 sequential measurements, in eastern and western sites, with plant off and on) and on August 31st 2011, when the plant was closed for the summer stop (2 measurements, in eastern and western sites, with plant off).
Figure 9. GRIMM measurement sites (red points from left to right: West side and East side)

Figure 10. GRIMM measurements of PM$_{10}$.

Figure 11. GRIMM measurements of PM$_{2.5}$.
Data measured by GRIMM particle counters could not allow detecting the specific contribution of the plant, which seemed to participate as one of the PM sources without a dominant role. Anyway, the variability of the operating conditions of the plant could change the scenario detected by these measurements. The presence of the AQ station in the area is therefore strategic.

CONCLUSIONS
The present paper deals with the results of a part of a wider research on the impact of a steel mill in northern Italy. A part of the characterization of two sites significant for proximity and population presence has been shown.

The obtained deposition values, even in the hypothesis of attributing them entirely to the plant, show that, by complying with recent regulations, PCDD/F deposition can be kept lower than a few environmental limits considered as a reference of the sector.

Modeling results showed the effect of the terrain morphology on the behavior of the concentration and deposition (PCDD/F and PM$_{10}$), depending on the release height. The role of diffused emissions resulted potentially significant, pointing out that an evolution of the present BATs available for this sector should take into account this aspect as a priority.

A critical aspect can be related to the slag management. A part of the measured deposition values, relatively high, showed a high PCDD/F deposition in an area close to the plant even if parallel deposition values of heavy metals (not shown in this paper) were low. One possible explanation can be a low temperature stripping of a part of the PCDD/F present in the slag cooled outdoor.

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