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Research Article

Ambient PM_{10} Concentration Reconstruction in an Inhabited Area Close to an Industrial Hot Spot by Using Particle Density and Optical Particle Counting Values

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Abstract

Ambient air Particulate Matter (PM) has recently been classified as carcinogenic to humans (Group 1) by International Agency for Research on Cancer; for this reason World Health Organization suggested guideline values in turn endorsed by the European legislation as target values. In some urban areas in Europe these values are often exceeded owing to the combined contributions of different anthropogenic emission sources. The reported case study regarded the PM_{10} concentration monitoring at an urban settlement close to an integrated steel plant in Trieste, a city in northeastern Italy. The monitoring was simultaneously carried out by gravimetric PM_{10} sampling and Optical Particle Counting (OPC) associated with meteorological data collection from January 2014 to April 2014.The aim of this work was to evaluate appropriate correction factors (densities) to be applied to OPC counts to assess gravimetric PM_{10} concentrations. A statistical model has been developed in R software environment by use of in-house scripts. We calculated densities (mean 7.6 g cm^{-3}) close to Fe density for sub-micron PM (0.3, 0.5, 0.7 μm) when the blast furnace was operating, in the same condition we attributed a density of 4.1 g cm^{-3} to the coarsest PM (10 μm), suggesting respectively the contribution of fugitive and stack emissions from the plant. Moreover, taking into account the wind regime variations we could calculate densities related to urban sources, which showed values below 2.0 g cm^{-3} for fine and coarse particles (>1.0 μm) and a mean of 5.1 g cm^{-3} for micron and sub-micron particles (<1.0 μm).

ABBREVIATIONS

BAT: Best Available Techniques; B.F.: Blast Furnace; EEA: European Environment Agency; EU: European Union; GPS: Global Positioning System; IARC: International Agency for Research on Cancer; JRC: Joint Research Center; NE: Northeast; OPC: Optical Particle Counter; PM: Particulate Matter; S.P.: Sampling Point; UNI EN: Italian National Unification European Standard; WHO: World Health Organization; W.S.: Meteorological Station

INTRODUCTION

Air pollution monitoring, control and attenuation are important issues to minimize adverse health effects on population. Among pollutants, particulate matter (PM) has recently been classified as carcinogenic to humans (Group 1) by International Agency for Research on Cancer (IARC), in particular it has been closely associated with increased lung cancer incidence [1].

It can also cause other significant health effects such as cardiovascular and lung diseases, heart attacks and arrhythmias, atherosclerosis, adverse birth outcomes and childhood respiratory diseases. The outcome of these diseases can be premature death [2].

World Health Organization [3] suggested guideline values for PM_{10} and $PM_{2.5}$ to be less than 20 μgm^{-3} and 10 μgm^{-3} on yearly average respectively. Regarding daily means, the limits of 50 μgm-

[3] and 25 µgm^{-3} have been respectively suggested for PM_{10} and $PM_{2.5}$. Nevertheless WHO underlined that these limits are aimed to achieve the lowest concentrations of PM possible to minimize adverse health effects.

The present EU legislation [4] set a short-term limit value for PM_{10} (i.e. not more than 35 days per year with a daily average concentration exceeding 50 µgm^{-3}) and an annual PM_{10} limit value of 40 µgm^{-3}. Furthermore it was set an annual limit value of 25 µgm^{-3} for $PM_{2.5}$ to be met by 1 January 2015 and an exposure concentration obligation of 20 µgm^{-3}based on a three-year average.

The European Environment Agency reported in 2015 [5] an evaluation about pollution levels registered in 2013 in the EU. A total of 17% of the urban population was exposed to PM_{10} levels above the EU daily limit value and 9% above the EU target value for $PM_{2.5}$. Moreover respectively 61% and 87% of the urban population was exposed to concentrations exceeding the annual mean WHO value for PM_{10} and $PM_{2.5}$.

In the document "Air Quality Guidelines for Europe" [6] WHO highlighted the need to pay specific attention to sites affected by defined sources such as traffic and other "hot-spots". In these sites the representativeness should be defined and assessed considering micro-scale conditions, including the buildings around the stations (street canyons), traffic intensity, the height of the sampling point, distances to obstacles and effects of the local sources.

Taking into account the information reported above, we focused our study on particulate matter impact of an integrated steel plant in Trieste, a city in northeastern Italy.

Integrated steel making involves a number of processes which generate both stack and fugitive emissions as described in the Best Available Techniques Document for iron and steel production by Joint Research Center (EU) [7].

Because of their economic importance together with environmental issues, integrated steelmaking impacts have been studied in Europe and other continents, focusing both on volatile organic compounds [8-11] and particulate matter [12-16] and its components emissions towards ambient air.

As reported by Almeida et al. [13] the emissions are challenging to be studied because of the presence of both continuous and batch processes, and they concluded that filter-based sample techniques are not suitable to capture short-lived emission events which arise from specific operations.

The use of an optical particle counter (OPC) combined to a weather station appears to be suitable to study the abovementioned impacts because it allows to collect high-frequency data at an easily affordable cost. However, converting OPC channel counts to segregate size mass is not a trivial task [17-19] because a site specific correlation has to be established.

Our approach describes the integration of data recorded by an optical particle counter, by a weather station equipped with an anemometer and, gravimetric data obtained from filters sampled by samplers with PM_{10} impactors. The aim of this study was to evaluate appropriate correction factors (densities) to be applied to OPC counts to assess gravimetric PM_{10} concentrations.

MATERIALS AND METHODS

The site

The sampling site is located in an urban settlement close to an integrated steel plant in Trieste, a city in northeastern Italy (Figure 1). The gravimetric PM_{10} and Optical Particle Counting sampling point (S.P.) was about 180 meters far from the steel plant, which consists of the coke oven batteries (not shown) and the blast furnace (B.F.). The meteorological station (W.S.) located near S.P. on a building roof in order to avoid canyoning phenomena. The blast furnace was shut down at the end of February 2014 due to a change in management, therefore in the present study we report on a eighty days monitoring which covered four months between January 2014 and April 2014 (forty days before blast closing and forty days after it).

PM_{10} sampling and gravimetric measurement

Ambient air PM_{10} concentration has been measured according to the UNI EN 12341:2001 by a HYDRA Dual Sampler (FAI Instruments s.r.l., Italy) equipped with a LowVolume-PM_{10} head impactor at a volumetric flow of 2.3m^{3}h^{-1}. The sampler has been accommodated in a thermostatic cabinet set at 20±4°C so that quartz fiber filters (Ø 47 mm by Pall Corporation, USA) used for automated daily sampling (00:00-24:00) could remain at a constant temperature until they were withdrawn by an operator. Prior to use the filters were heated at 600°C for 2h.

The particle mass was measured by weighting the filters before and after sampling by a balance with a resolution of ±0.1 mg ("Microcrystal 250" by Gibertini Elettronicas.r.l., Italy) after opportune conditioning (temperature of 20±1°C and relative humidity of 50±5%).

Figure 1 The sampling site in the city of Trieste. S.P.: sampling point for OPC and gravimetric PM_{10}; W.S.: weather station; B.F.: blast furnace. The boundary of the steel plant is shown by a black line.

Meteorological data acquisition

Weather conditions have been continuously measured by a control weather station (WeatherStation 150WX by AIRMAR Technology Corporation, USA) for the whole monitoring period. This instrument, equipped with internal compass and GPS, simultaneously measures wind speed and direction, atmospheric pressure, air temperature, relative humidity and dew point. The software WeatherCaster™ of AIRMAR manages the weather station and records data per minute.

Optical particle counting

An optical particle counter with eight channels (model 212 Eight Channel Particle Counter by Met One Instruments Inc., USA) has been used to count the particles in eight size bins (centered in 0.3, 0.5, 0.7, 1.0, 2.0, 3.0, 5.0, 10.0 µm mean diameter respectively). The instrument continuously samples air at 1 $Lmin^{-1}$ and provides data per minute for each channel.

Statistical data elaboration

Statistical data elaboration was performed using R software [20] implemented by open air package [21] to calculate wind roses. Data modeling was performed in R environment by use of in-house scripts as described in the next paragraph.

RESULTS AND DISCUSSION

Firstly the sum of counts for each channel and for each day was calculated. Basic statistics for OPC daily counts are reported in table 1. From now on PM data collected by OPC will be named as PM03, PM05, PM07, PM1, PM2, PM3, PM5 and PM10, according to their size.

Assuming that the particle shape is spherical [22] and considering the channel diameter as representative of the mean diameter of the particles in the respective bin, the daily PM_{10} mass can be calculated as follows:

$$PM_{10(\mathrm{OPC})} = \sum_{i=1}^{8} \left(p_i . V_i . d_i \right) (1)$$

where p: particle counts, V: mean particle volume, d: density assigned to the particles, *i*: the *i*-th channel.

Aiming to assign a density for each PM size we built an in-house script which generated density vectors ($d_{vec}=d_1,d_2,\ldots,d_i$) with eight random components to be applied to equation (1). Considering that Fe density is 7.96 gcm^{-3} and that a density of 1.65 gcm^{-3} is considered to be related to urban traffic sources [17,23], the density range 1 to 8 gcm^{-3} was chosen to perform the calculation. Data before and after blast shut down were considered separately.

Fifty thousand density vectors were generated for each run. In order to choose the best suitable density vector a number of parameters were taken into account considering the following equation:

$$PM_{10(\mathrm{grav})\mathrm{vec}} = PM_{10(\mathrm{OPC})\mathrm{vec}} + R_{\mathrm{vec}} (2)$$

where $PM_{10(grav)vec}$ is the vector of the daily gravimetric data, $PM_{10(OPC)vec}$ is the vector of the daily PM_{10} data calculated applying equation (1) to OPC counts and R_{vec} is the vector of residuals.

Mean, median and relative standard deviation (RSD) of R_{vec} were calculated. Moreover there were calculated R Pearson parameter and intercept, slope and R^2 of $PM_{10(grav)vec}$ *vs.* $PM_{10(OPC)vec}$. The best matching results had to show a mean, median, RSD of R_{vec} and intercept which tended towards zero, and a slope, R^2 and R Pearson which tended towards 1. We rated the results according to the abovementioned characteristics and chose the best matching ones, which are reported in (table 2).

In (Figure 2) we reported the time behavior of gravimetric PM_{10} (black line) and the modeled PM_{10}, which is the merge of the two separate model runs (before and after blast shut down, red line). We observed a good fit except for days between 24/1/2014 and 31/1/2014. In that time lapse it occurred a high NE wind episode, typical of Trieste province, reported by a wind rose graph in (Figure 3). Considering that the plant overlooks the sea to the west and the south, NE wind blows from the city to the sea. Focusing on the data relative to that episode (eight days) we could calculate with the abovementioned method a "density fingerprint" which accounted for the urban contribution. The obtained density vector is reported in table 2 and the model fitting is shown by a blue line in (Figure 2).

Considering the blast furnace shut down as discriminating factor, the most significant difference was found in PM05 and PM07 modeled densities. This result, together with the high density of PM03, is congruent with results reported by Mohiuddin

Table 1: Basic statistics for OPC daily counts of PM03, PM05, PM07, PM1, PM2, PM3, PM5, PM10 and gravimetric PM_{10} concentrations in units of μgm^{-3} for the period January 2014 – April 2014.

Daily sums of counts		**PM03**	**PM05**	**PM07**	**PM1**	**PM2**	**PM3**	**PM5**	**PM10**	**PM_{10}**
Before	Min	$3.67 \cdot 10^7$	$1.97 \cdot 10^6$	$3.69 \cdot 10^5$	$1.66 \cdot 10^5$	$4.11 \cdot 10^4$	$7.55 \cdot 10^3$	$2.00 \cdot 10^3$	$1.52 \cdot 10^2$	7
(40 days)	Median	$1.17 \cdot 10^8$	$1.45 \cdot 10^7$	$3.28 \cdot 10^6$	$1.47 \cdot 10^6$	$5.39 \cdot 10^5$	$9.11 \cdot 10^4$	$1.91 \cdot 10^4$	$1.20 \cdot 10^3$	27
	Mean	$1.88 \cdot 10^8$	$2.81 \cdot 10^7$	$5.41 \cdot 10^6$	$2.05 \cdot 10^6$	$7.16 \cdot 10^5$	$1.07 \cdot 10^5$	$2.39 \cdot 10^4$	$1.57 \cdot 10^3$	43
	Max	$6.24 \cdot 10^8$	$1.49 \cdot 10^8$	$2.98 \cdot 10^7$	$8.36 \cdot 10^6$	$3.62 \cdot 10^6$	$4.15 \cdot 10^5$	$1.17 \cdot 10^5$	$1.03 \cdot 10^4$	142
After	Min	$1.04 \cdot 10^7$	$1.07 \cdot 10^6$	$3.41 \cdot 10^5$	$1.83 \cdot 10^5$	$7.92 \cdot 10^4$	$1.92 \cdot 10^4$	$5.76 \cdot 10^3$	$1.96 \cdot 10^2$	5
(41 days)	Median	$1.43 \cdot 10^8$	$9.29 \cdot 10^6$	$1.62 \cdot 10^6$	$9.13 \cdot 10^5$	$3.99 \cdot 10^5$	$8.32 \cdot 10^4$	$2.05 \cdot 10^4$	$1.84 \cdot 10^3$	23
	Mean	$1.91 \cdot 10^8$	$1.86 \cdot 10^7$	$2.87 \cdot 10^6$	$1.12 \cdot 10^6$	$4.59 \cdot 10^5$	$9.55 \cdot 10^4$	$2.44 \cdot 10^4$	$2.13 \cdot 10^3$	28
	Max	$6.21 \cdot 10^8$	$1.19 \cdot 10^8$	$1.51 \cdot 10^7$	$3.52 \cdot 10^6$	$1.40 \cdot 10^6$	$2.64 \cdot 10^5$	$6.91 \cdot 10^4$	$5.63 \cdot 10^3$	83

Abbreviations: Min: Minimum; Max: Maximum; PM: Particulate Matter.

Table 2: Best matching results for PM density vectors (g cm^{-3}) before blast shut down, after blast shut down and during high NE wind episode.

Period	PM03 density	PM05 density	PM07 density	PM1 density	PM2 density	PM3 density	PM5 density	PM10 density	Residuals Mean	Residuals Median	Residuals % Std. Deviation	R Pearson	Intercept	Slope	R^2
Before blast shut down	7.98	7.88	6.94	2.20	1.36	4.16	2.40	4.07	0.59	0.36	27	0.976	-0.96	1.01	0.954
After blast shut down	6.95	1.47	1.99	2.53	1.19	3.23	2.91	5.32	-0.12	-0.20	10	0.988	-0.85	1.03	0.976
High NE wind episode	5.58	6.63	4.87	3.18	1.10	1.85	1.10	1.40	0.21	0.23	18	0.966	2.80	0.83	0.933

Abbreviations: PM: Particulate Matter; NE: Northeast.

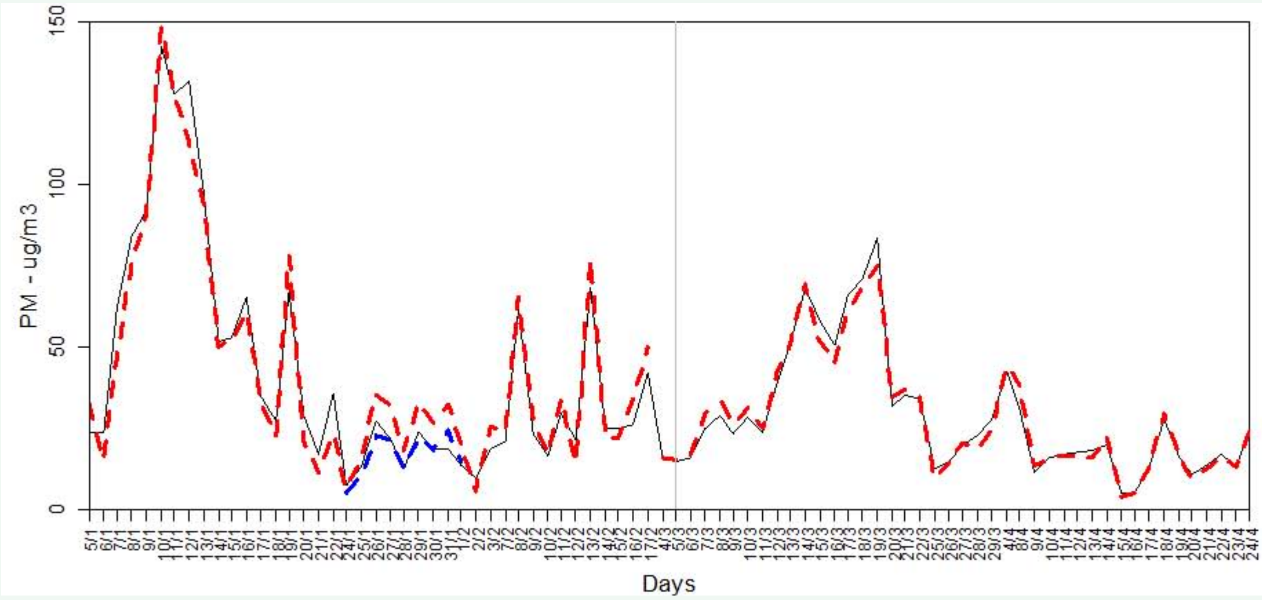

Figure 2 Time behavior of gravimetric PM10 (black line), modeled PM10 (red dashed line) which is a merger of two models: before and after blast shut down (vertical gray line). Blue dashed line represents the model calculated considering only the high NE wind episode.

et al. [12] who estimated that Fe could range up to 95% at the submicron and ultrafine size particles. Almeida et al. [13] found a bimodal distribution for Fe and Mn (density=7.47 gcm^{-3}) rich particles at 0.45 µm and 4 µm, and they related these particles to steel/coke making. This evidence can explain our result for PM3 density. Dall'Osto et al. [16] related the coarse fraction (PM10) to dust suspension from the iron ore stockpiles by wind.

The high density found for PM03 and PM10 fractions after the blast closing can possibly due: the former to an elevated deposition time typical of ultrafine particles and the latter to the blast dismantlement occurred after shut down.

NE wind had influence on fine and coarse fractions, however the ultrafine particles seem to remain affected by site specific peculiarities.

CONCLUSIONS

In this study gravimetric PM_{10} data and OPC data (8 channels) were acquired at an urban site located in the vicinity of a steel plant in Trieste, a city in northeastern Italy. This study led to the determination of a descriptive model capable of reconstructing the concentrations of gravimetric PM_{10} (UNI EN 12341:2001) on the basis of suitable statistical processing (R software) of particle counting by OPC. The model provided the best particulate density assignment for each counted size class (PM03, PM05, PM07, PM1, PM2, PM3, PM5, PM10). A model improvement has been carried out considering the wind regime and the industrial production variability occurred during the four sampling months (January 2014 -April 2014). A larger data set acquisition may allow obtaining a suitable predictive model for the aforementioned

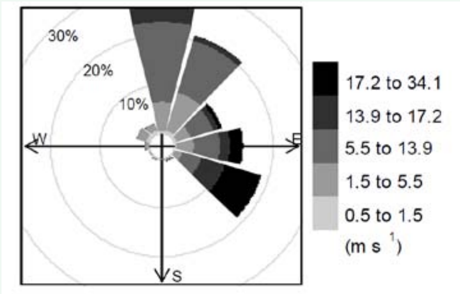

Figure 3 Wind rose of the period between 24/1/2014 and 31/1/2014 (high NE wind episode).

area. In this way, a practical, short time-resolved and rather cheap method can be used to assess the impact of the different pollution sources that insist on the investigated area. This site-specific analytical-statistical approach can be extended to different case studies.

CONFLICT OF INTEREST

Pierluigi Barbieri has been consultant for the Court of Trieste for characterization of air quality in Trieste in relation to possible industrial impacts. The other authors declare that there are no conflicts of interest.

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