Atmospheric Environment 45 (2011) 3522–3527

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Measurement of black carbon concentration as an indicator of air quality benefits of traffic restriction policies within the ecopass zone in Milan, Italy

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A R T I C L E I N F O

Article history:
Received 14 January 2011
Received in revised form
27 March 2011
Accepted 4 April 2011

Keywords:
Air pollution
Particulate matter
Aerosols
Traffic pollution
Black carbon
Spatial variability

A B S T R A C T

Traffic restrictions are an unpopular tool to mitigate urban air pollution, and a measurable improvement in air quality is needed to demonstrate the effectiveness of this measure. Previous attempts failed to detect measurable reductions of PM mass pollution within the areas subject to traffic restriction. However black carbon, which is emitted primarily by traffic sources, could be a PM metric more suitable than PM mass to demonstrate pollutant reductions. In this study we report the results of a black carbon monitoring campaign carried out in Milan, Italy, with the aim to detect - and demonstrate more suitably than PM mass - differences in local urban air quality among three zones located very closely with different traffic intensity. The study was carried out in three different days by measuring simultaneously black carbon and PM mass concentrations with fixed monitoring stations located in three main radial roads connecting the outskirts to the city center, each with three segments: 1) an outer one, with no traffic restrictions 2) an intermediate one, subject to the congestion traffic charge called "Ecopass", where a ticket is required to enter for cars equipped with engines prior to Euro 4 standard; 3) the pedestrian zone (no cars admitted) of Duomo Square in the city center, where each of the three main roads ends. The results demonstrated a sharply declining gradient in black carbon levels from the outer zone, without traffic restrictions, to the more central areas, for all of the three radial main roads. The differences in mean black carbon levels in the same day in the different traffic scheme locations were highly significant for each comparison. In contrast to the Black carbon results, mean PM_{10}, $PM_{2.5}$, PM_1 concentrations did not show significant differences among the different traffic zones on the different campaign days. The ratio of black carbon to PM_{10} decreased by 47% and 62% in the Ecopass zone and in the pedestrian zone, respectively, as compared to the no-restriction zone. To the best of our knowledge this is the first study showing that within-city proximal areas with different traffic intensity are associated with different black carbon levels. These data suggest that black carbon is a highly relevant metric of traffic pollution and should be taken into consideration in demonstrating the effectiveness of air quality mitigation measures.

1. Introduction

Atmospheric pollution from suspended particulate matter (PM) represents a risk factor for respiratory and cardiovascular diseases and for cancer, mainly due to the presence of combustion products in the composition of the PM (Dockery et al., 1993; Künzli et al., 2000, 2010; Pope et al., 2002; Biggeri et al., 2004). Traffic emissions are of particular concern in very congested metropolitan cities, since they are associated with overall mortality increase (Hoek et al., 2002), lung cancer risk (Beelen et al., 2008), and worsening of respiratory health in children (Brauer et al., 2002; Van Roosbroeck et al., 2008; Migliore et al., 2009; Rosenlund et al., 2009). In the Milan district of northern Italy, it is estimated that emissions from vehicular traffic amount to about 55% of PM10 (INEMAR, 2010), a regulated pollutant with daily concentrations limit values of 50 $\mu g\ m^{-3}$ (as daily average not to be exceeded for

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doi:10.1016/j.atmosenv.2011.04.008

more than 35 days in one year) according to the EU air quality directive (1999/30/EC, Annex III) and Decreto Ministeriale n. 60, dated 04/02/2002.

Long periods of high PM concentrations inside metropolitan cities are common; for example in Milan in January 2010, 16 consecutive days were recorded with a daily average concentrations greater than 50 $\mu g\ m^{-3}$, of which 11 days had daily averages of more than 75 $\mu g\ m^{-3}$ (Invernizzi and Ruprecht, 2010). Traffic restrictions have been introduced by several administrations (London, Stockholm, Singapore, Milan), as a measure to both reduce vehicular congestion and improve air quality within the city centers (Kelly and Kelly, 2009). Where traffic restrictions have been enforced, several benefits are immediately appreciated, such as a decrease in traffic congestion and noise reduction, a decrease in the number of road accidents, and an increase of traffic speed. However a measurable benefit in the air quality marker species measured has not been demonstrated in two cities where such changes have been studied, London and Milan, which adopted very similar measures of traffic restrictions in the city centers, the "*Congestion charging scheme*" (CCS) and the Ecopass, respectively. The London Municipality has been attempting to find a CCS effect on the air quality since 2003, the year of the introduction of the scheme, inside and outside the restricted zone. The analysis of NO_x annual average concentrations, has not shown substantial benefits on the pollution levels, although a larger low emission zone (LEZ) was implemented in 2008 aimed at reducing pollution, whose effectiveness has not yet been assessed (Tonne et al., 2008, 2009). In January 2008, the Municipality of Milan issued a traffic restriction regulation limited to a small area of the historical city center, requiring an entry ticket for the more polluting vehicles. A notable improvement in the air quality was predicted, with an estimated 30% reduction in PM10 concentrations (Milan municipality (a), 2010). The initiative was named Ecopass. The predicted improvements have not been found. A study performed in 2009 (Ruprecht and Invernizzi, 2009) did not demonstrate any difference in PM10 (or PM1 and PM2.5) concentrations between the zones with traffic restriction (Ecopass) and those without them, despite a reduction in the number of vehicles entering the zone with restrictions. Overall traffic emissions were anticipated to show a 19% reduction (Milan Municipality (b), 2010). In a second study performed during January 2010, PM10 concentrations within the Ecopass area were shown to be exceptionally high, similar to no-restriction zones, casting doubts on the efficacy of the restrictions to improve either air quality or to enhance public health (Invernizzi and Ruprecht, 2010). The difficulty to demonstrate a measurable benefit in air quality jeopardizes the future of the Ecopass program.

The failure to find measurable air quality improvements resulting from the Ecopass program could be due the limited dimensions of the Ecopass zone, which represents only 4.5% of the total area of the Municipality (8.2 km^2 out of a total of 181 total km^2), or because of a relatively homogenous distribution of PM concentrations. Since these may be considered to reflect regional pollution sources, they may be not suitable to discern spatial variations of local emissions within cities.

A potentially more suitable marker used to detect pollution differences between different city areas is airborne *black carbon*. Black carbon measurements were used in the recent years to study pollution due to traffic in areas close to freeways (Roorda-Knape et al., 1999; Zhu et al., 2002; Cyrys et al., 2003; Zhang et al., 2004). Black carbon is produced by incomplete combustion of carbonaceous fuels. It is a unique primary tracer for combustion emissions, as it has no non-combustion sources, and is stable once released into the atmosphere. It absorbs light in the visible part of the spectrum, which is the basis of its detection. It is composed of chains of agglomerated graphitic spheres with particles having aerodynamic diameters between 10 and 200 nm. If inhaled it deposits deep in the lungs, and a dose-dependent inverse association between the black carbon content of airway macrophages and lung function in children has been found (Kulkarni et al., 2006). Airborne black carbon exhibits spatial variability, being present at high concentrations in the vicinity of traffic sources, with a lognormal dispersion within 200 m of highways (Zhu et al., 2002). Because of these characteristics, black carbon can be considered an indicator of all primary aerosols from combustion and could be used to detect air quality differences within-city microenvironments with different traffic levels (Clougherty et al., 2008).

The scope of this study was to demonstrate that measurements of black carbon are a more suitable metric than the PM concentrations to identify traffic pollution differences between monitoring sites impacted by different traffic patterns. A pilot personal exposure study was previously conducted by the authors, with black carbon as well as PM10, PM1 and PM2.5 measurements taken while walking back and forth between the no-restriction, Ecopass, and pedestrian zone: the results showed significant differences in black carbon concentrations, indicating that black carbon may be a promising traffic intensity marker (Invernizzi et al., 2010).

2. Methodology

2.1. Location of monitoring sites

We divided the city area in three sections with different geographical orientation, characterized by three main roads connecting the outskirts with the pedestrian zone of Piazza del Duomo (city center) (see Fig. 1). The monitoring sites were located on the three main roads as follows:

1. Corso Buenos Aires (no-restriction) – Corso Venezia (Ecopass zone) – Piazza del Duomo (pedestrian zone):

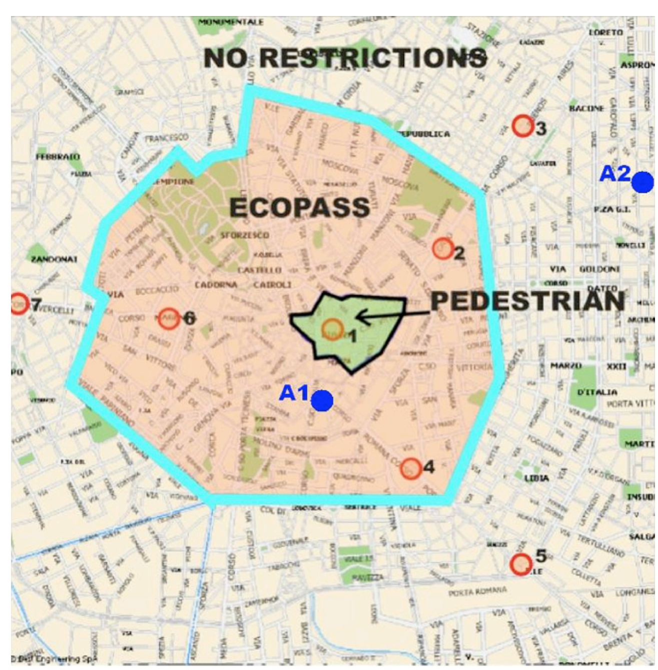

Fig. 1. Map of the three traffic zone in the Milan city center: the inner black line represents the border of the pedestrian zone, the green line indicates the border of the Ecopass zone. Number 1 indicates pedestrian zone monitoring site, numbers #2, #4, and #6 indicate the Ecopass zone sites, numbers #3, #5, #7 indicate the no-restriction zone sites. Positions A1 and A2 indicate the official fixed monitoring sites of the Environmental Protection Agency of Lombardy Region.

a. Corso Buenos Aires (no traffic restrictions): 2 km residential and shopping road with bi-directional traffic on 4 lanes, and sidewalks 3–4 m wide.

   Monitoring site # 3 was at the crossing with Via S. Gregorio.

b. Corso Venezia (Ecopass zone): residential and shopping road, with 4 traffic lanes, and sidewalks 3–4 m wide, sided by a city park in its first 200 m, then restricted to 2 lanes for 1 km until the pedestrian zone, a typical urban street canyon with multi story buildings on both sides of the street.

   Monitoring site # 2 was at the crossing with Via Serbelloni.

c. Piazza del Duomo (pedestrian zone):

   Monitoring site # 1 was at the crossing with Via Santa Radegonda, a square in front of the Milan cathedral.

2. Corso Lodi (no restrictions) - Corso di Porta Romana (Ecopass zone) – Piazza Duomo (pedestrian zone):

d. Corso Lodi (no-restriction): 3 km mainly residential road, with 6 traffic lanes divided by two rows of trees, with sidewalks 2–3 m wide.

   Monitoring site # 5 was at the crossing with Piazza B. Buozzi

e. Corso di Porta Romana (Ecopass zone): 1.5 km mainly residential zone, characterized by two traffic lanes, sidewalks 2–3 m wide, a typical urban street canyon.

   Monitoring site # 4 was at the crossing with Via Orti.

f. Piazza del Duomo (pedestrian zone):

   Monitoring site # 1 was at the crossing with Via Santa Radegonda.

3. Corso Vercelli (no restrictions) – Corso Magenta (Ecopass zone) – Piazza del Duomo (pedestrian zone):

g. Corso Vercelli (no restrictions): 1 km residential and shopping road, with two traffic lanes and sidewalks 3–4 m wide.

   Monitoring site # 7 was at the crossing with Via Cimarosa.

h. Corso Magenta (Ecopass zone): residential and museum road, 2 traffic lanes, sidewalks 2m wide, a typical urban street canyon.

   Monitoring site # 6 was at the crossing with Via de Togni.

i. Piazza del Duomo (pedestrian zone).

   Monitoring site # 1 was at the crossing with Via Santa Radegonda.

The Ecopass zone requires diesel vehicles manufactured according to standards prior EURO 4 tier (and vehicles conforming to EURO 4 tier without a particulate filter) to pay a toll to enter restricted zone between 08:00 a.m. and 08:00 p.m. The configuration of the central portion of Milan is typical of “urban street canyons”, with a width variable from 11 to 40 m and with 5 to 10-story lateral buildings. The instruments at the monitoring sites were placed at a height of 0.8 m from the ground, and on the same side of the road.

2.2. Equipment description

Black carbon concentrations were measured with three aethalometers (Magee Scientific, USA, microAeth model AE51) with data recorded at 5 min intervals, while PM10, PM2.5 and PM1were measured with optical laser-operated particle counters (Aerocet 531, MetOne Instruments, USA) recording data on 2 min periods.

Besides PM10, measurements were extended to PM2.5 and PM1 because they represent smaller particles than PM10, potentially useful for traffic research. In addition, PM2.5 is a mandated measurement parameter in the EU (Directive, 2008/50/EC), while black carbon particles form a part of PM1. PM2.5 and PM1 concentrations, along with black carbon, might be useful for further interpretation of the research.

Each Aerocet was equipped with temperature and relative humidity (RH) sensors, and all PM classes were corrected for RH interference according to Lowenthal (1995). Power was provided by small electrical inverter and a 12 V battery assuring adequate power for the duration of the sampling.

2.3. Instrument calibrations

The three Aethalometers were factory calibrated and the three OPC were gravimetrically calibrated with a certified analyzer (MetOne BAM 1020). The instruments were operated side by side for approximately 5 h in ambient air to establish their comparability. The differences between the recordings of the three different sets of the instruments were found within the limits of instrument specification and accuracy (Figs. 2–3).

2.4. Selection of locations

Locations were selected to reduce the influence of non-traffic sources of PM, including car parking lots, water aerosols sprayed

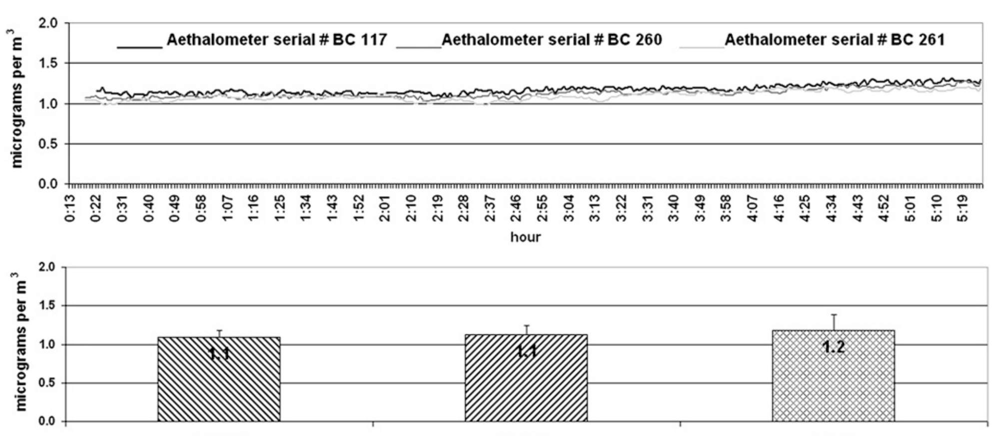

Fig. 2. Comparisons of the three black carbon analyzers. The upper diagram shows 5 h continuous recordings of the instruments operating concurrently in the same environment. The lower bargraph shows the means (SD) of the recordings from each instrument.

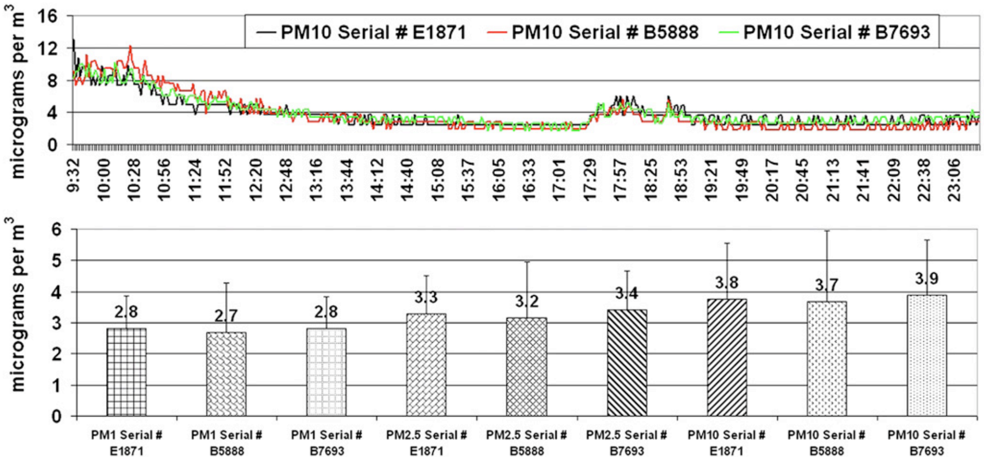

Fig. 3. Comparisons of the three optical particle counters. The upper diagram shows 12 h continuous recordings of the instruments operating concurrently in the same environment. The lower bargraph shows the means (SD) of the recordings from each instrument.

for customer comfort in outdoor gazebo bars (which could produce artificially high reading on the OPCs), road constructions or repair, and wood oven pizzerias. The locations were continuously manned. The sites were located on sidewalks 1 m far from the curb, and the instruments were protected from direct exposure to the sun.

2.5. Measurement time spans

Measurements started each day between 08:00 and 09:00 a.m. at all three monitoring sites, and finished between 07:00 and 08:00 p.m. The recordings on Corso Vercelli and Corso Magenta (July 29th) were interrupted from 10:45 a.m. to 12.15 p.m. due to a brief rainy period which required that the instruments be covered to keep them dry. Data were considered valid only after an instrument warm up period of half an hour.

2.6. PM10 official data

In Milan there are only three official monitoring sites of the Regional Environmental Protection Agency (ARPA Lombardy). The 24 h PM10 concentration mean values were obtained from the validated data from the official site located inside the Ecopass zone (Via Verziere), and from the site of Via Pascal-Città Studi located in the no-restriction zone (site A1 and A2 in Fig. 1, respectively) (http://ita/arpalombardia.it/ITA/qaria/Home.asp). Both of these locations are indeed most representative of urban sites in Milan.

2.7. Data treatment

All data were downloaded from the instruments and processed in spreadsheets (the OPC data were corrected for relative humidity effect, according to Lowenthal (1995)). Data were described as means +/− standard deviation, and the statistical significance of the differences were evaluated by two-tail Student's *t*-test with significance set at $p < 0.05$.

2.8. Atmospheric conditions

Continuous data for wind speed and direction, relative humidity and temperature were obtained from the weather station of the

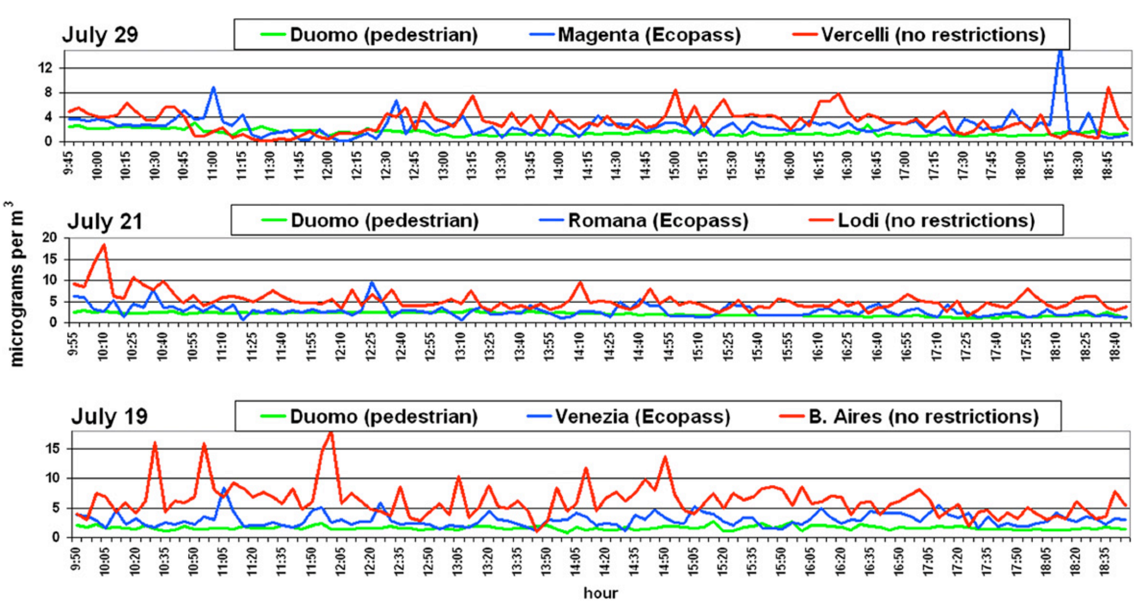

Fig. 4. Continuous black carbon measurements in the three days of Ecopass campaign. No-restriction zone measurements are depicted in red; Ecopass zone sites in blue, pedestrian zone in green.

Table 1
Mean (SD) black carbon concentrations (μg/m^3) measured at the different traffic zones in the different campaign days.

Site locations	July 19th	July 21st	July 29th
Pedestrian zone	1.6 (0.4)	2.0 (0.5)	1.5 (0.5)
Ecopass zone	3.1 (1.7)	2.8 (1.4)	2.6 (1.9)
No restriction zone	6.3 (2.9)	5.2 (2.3)	3.3 (1.9)

$p < 0.0001$ between the three different zones for each day, except for no-restriction *vs* Ecopass zone on July 29th ($p = 0.006$).

Milano–Bresso Airport located to the North of the city, from an internet weather website (http://www.wunderground.com). During the monitoring campaign the weather was typically sunny and warm with temperatures about 30 °C. Prevalent wind direction was SSW, with speed in the range of 3–5 km h^{-1}. An exception occurred during the last day of sampling, when the weather was stormy with a few showers, but the showers did not influence the measurements in any significant manner.

3. Results and discussion

3.1. Black carbon monitoring

Diagrams of continuous recordings of black carbon concentrations in the three days of campaign are reported in Fig. 4: the pedestrian zone of Duomo (site #1) consistently showed lower black carbon concentrations in all of the three campaign days compared to the Ecopass zones (site #2, #4, and #6 in the map), while the sites in the Ecopass zones showed reduced black carbon concentrations as compared to the unrestricted traffic zones (sites # 3, #5 and #7). Mean (SD) black carbon concentrations in the pedestrian-only zone were 1.6 (0.4), 2.0 (0.5) and 1.5 (0.5) μg/m^3 on July 19th, July 21st, and July 29th, respectively, while, in the corresponding days, mean values in the Ecopass locations were 3.1 (1.7) μg/m^3, 2.8 (1.4) μg/m^3, and 2.6 (1.9) μg/m^3, respectively, and in the no-restriction sites black carbon averaged 6.3 (2.9), 5.2 (2.3), 3.3 (1.9) μg/m^3, as shown in Table 1.

3.2. PM monitoring

Mean (SD) PM10, PM2.5 and PM1 concentrations did not show significant differences among the different traffic zones for all of the three days of monitoring (Fig. 5). Although the comparison between our 10 h data and the 24 h official averages is not feasible, our results were still in line with the PM10 concentration from the two fixed stations of ARPA (A1 and A2 in the map, Fig. 1), which again did not show significant differences between the two zones with different traffic intensity: on July 19th, July 21st, and July 29th, in the no-restriction zone 24 h PM10 concentrations were 20 μg m^{-3}, 32 μg m^{-3}, and 16 μg m^{-3}, while in the Ecopass location 20 μg m^{-3}, 34 μg m^{-3}, and 18 μg m^{-3}, respectively.

3.3. Black carbon/PM_{10} ratios

In all the three days of monitoring, the ratios of black carbon/PM10 for each location showed a decreasing gradient from the no-restriction > Ecopass zone > pedestrian zone. The means (SD) black carbon/PM_{10} ratios of the three campaign days were 22.6% (4.0), 11.8% (1.7), and 8.5% (1.1) in no-restriction, Ecopass and pedestrian zone, respectively. On average, the black carbon contribution to PM10 decreased by 47% and 62% in the Ecopass zone and the pedestrian zone, respectively, as compared to no-restriction zone.

3.4. Limitations and advantages

This study was a preliminary demonstration, conducted over a limited number of campaign days in one season (summer) only. Nevertheless, we recorded near-continuous data on each day for both black carbon and PM measurements, and the analysis confers solidity to the conceptual study design. In addition, personal exposure data showed a similar gradient in black carbon concentrations (Invernizzi et al., 2010). Summer may be a particularly suitable season to conduct traffic pollution campaigns in Milan because of the lack of interferences of PM emissions from residential and office heating. During July, commercial and industrial activities are still at their normal levels, whereas a substantial drop in traffic and population occurs typically in early August. Particulate matter PM10 is regulated with a concentration limit that is allowed to be exceeded only 35 times in a calendar year. Typically, the exceedances occur in winter. While the calendar year includes two halves of two different winter periods, this metrics still depends heavily on the weather and temperature characteristics of the two winter periods in question. A mild winter will typically result in a lesser number of exceedances than a cold winter. In Europe, most of the last few winters, except the winter 2009/2010, have been characterized with a low number of cold periods. We demonstrate in the present study a metric that is less susceptible to such seasonal influences. The proposed parameter is the spatial gradient

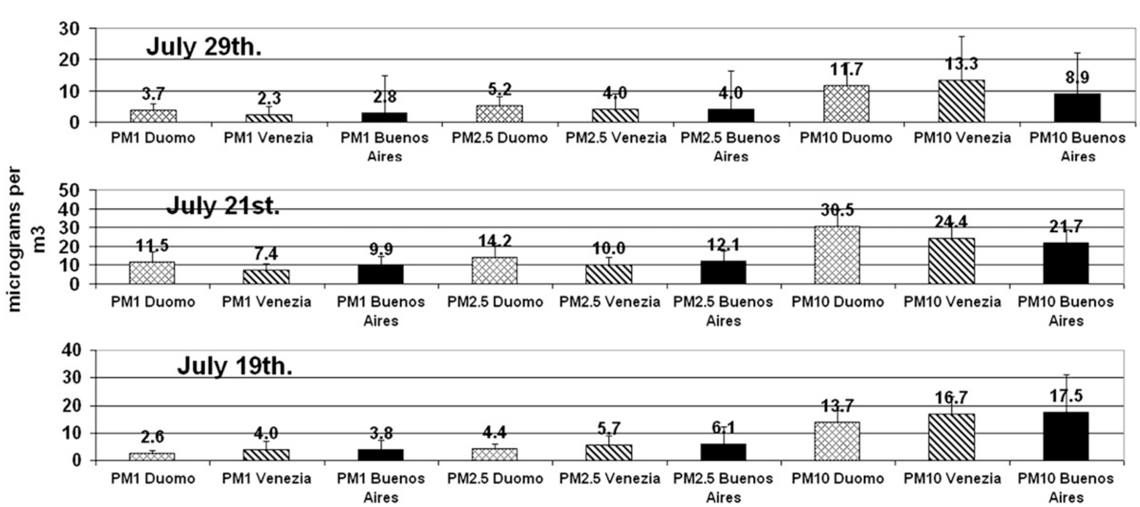

Fig. 5. Mean (SD) PM concentrations measured at the different traffic sites in the three different campaign days.

in concentration, the concentration being measured in the same time period but at different locations, that is parallel measurements in three zones with different levels of traffic restrictions. Measuring a primary pollutant – black carbon, eliminates the uncertainties due to processes that are responsible for formation of secondary aerosols that form a substantial portion of PM10 mass (Jimenez et al., 2009). These also depend on the ambient conditions whereas black carbon is stable in the atmosphere.

4. Conclusions

The "Ecopass" monitoring campaign showed a significant decrease in black carbon concentrations from no-restriction zone > Ecopass zone > pedestrian zone. The data were consistent for the different locations, on three different days. In contrast, the measured PM data (PM10, PM2.5 and PM1) did not show any difference in pollution among the different traffic intensity zones, not even for the pedestrian zone as compared to the no-restriction zone. Accordingly, the official 24 h PM10 official ARPA data were also unable to show an improvement in air quality in the Ecopass zone as compared to no-restriction zone. In the three monitoring days PM10 concentrations in the Ecopass zone were identical or higher as compared to no-restriction zone, in line with a previous analysis (Invernizzi and Ruprecht, 2010).

The efficacy of black carbon measurements as an indicator of the impact of traffic restrictions has been demonstrated by Wang et al. (2008) and Westerdahl et al. (2009): these studies recorded black carbon concentrations at road centers, road sides and in the city of Beijing, aiming to show the effectiveness of air pollution reduction policies during the 2008 Olympic Games. The black carbon measurements in these studies clearly demonstrated the impact of traffic restrictions (limitation of polluting vehicles and alternate circulation based on car registration numbers) on air quality, and even the possibility to characterize and identify the most polluting vehicles.

In this study we demonstrated the ability of black carbon measurements to distinguish different levels of traffic-derived pollution between proximal urban sites located very closely - on average only 1 km from each other - subject to different traffic influences: i.e. no-restriction *vs* Ecopass zone, and Ecopass zone *vs* pedestrian-only zone in the City of Milan. Our data corroborate the use of black carbon measurements as a powerful tool for evaluating traffic pollution impacts on urban areas. We believe that such measurements are particularly helpful to policy and decision makers in demonstrating the effectiveness of road and traffic regulations to the public.

Acknowledgement

This study was funded by SIMG, Società Italiana di Medicina Generale (Italian College GPs). We would like to thank Francesco and Beniamino Mazza for their help in operating the monitoring sites, and Tony Hansen for insightful comments.

GM acknowledges the support of the Slovene Ministry for Higher Education, Science and technology (grant SMER 3211-09-000304) for partially funding the development of the Aethalometers used in the study. GM's participation in the study was also partially funded from the commercial activities of Aerosol d.o.o., a manufacturer of the Aethalometers. GM recognizes that the paper is not intending to promote the sale of the equipment.

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