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Personal exposure to $PM_{2.5}$ among high-school students in Milan and background measurements: The EuroLifeNet study

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ABSTRACT

As part of the EuroLifeNet program we measured personal exposure to $PM_{2.5}$ in 90 pupils attending three schools in Milan, over a three-week period spanning November and December 2006, using a portable light-scattering nephelometer. The primary aim was to investigate the relationship between personal exposure to $PM_{2.5}$ and background measurements obtained from a fixed monitoring station. Pearson's correlation coefficient between sampled daily mean exposures and reference values from background station varied from 0.64 to 0.75, with an overall value of 0.63, indicating good agreement. We also estimated that about 40% of the variability in the mean daily personal exposure at the three schools was due to variability in background exposure, the remaining 60% due to between-subject differences in exposures or to other sources of error.

1. Introduction

Epidemiological studies consistently show that exposure to atmospheric particulate matter (PM) has an important adverse effect on human health. Studies on school-age children indicated that PM is associated with adverse effects on respiratory function (Horak et al., 2002; Peacock et al., 2003; Ward and Ayres, 2004; Moon et al., 2009). Population's exposure to PM is generally estimated from fixed monitoring stations which afford standardized measurements of atmospheric pollutant concentrations on a daily or hourly basis. Although pollutant levels measured by monitoring stations in a general way reflect the exposure of individuals to pollutants in the area, they cannot accurately estimate the individual exposure, which has been shown to be profoundly affected by individual circumstances such as time spent outdoors, means of transport used, tobacco smoking, environmental exposure to tobacco smoking, etc. (Roosbroeck et al., 2007; Briggs et al., 2008; Weichenthal et al., 2007; Ashmore and Dimitroulopoulou, 2009).

However, since in epidemiological studies indicators of exposure are usually assessed referring to outdoor concentrations (based on fixed-site monitors measurements or on the distance from major roads), it is important to investigate the relationship between personal exposure and outdoor PM background concentrations. In preceding studies evaluating personal exposure, different levels of association were found, some showing relatively low correlations (Adgate et al., 2003; Brown et al., 2008; Crist et al., 2008), other evidencing higher correlations (Janssen et al., 1999; Wu et al., 2005; Braniš and Kolomazníková, 2010).

The EuroLifeNet program was designed by the Research Centre on Information Technologies and Participatory Democracy (CITIDEP) with the support of the Institute for Environment and Sustainability (IES) of the Joint Research Centre (JRC) of the European Commission (web site http://www.citidep.pt/eurolifenet/, accessed 28 May 2010). Its aims are to measure personal exposure to particulate matter (PM_{10} and $PM_{2.5}$) and raise awareness, particularly among school students, of the hazards of air pollution. In the first pilot project of EuroLifeNet, carried out in Portugal and Italy, school students gathered data on personal exposure to $PM_{2.5}$ using portable nephelometers.

The present study reports data obtained in Italy which were collected by students in three Milan schools. The aim of the study was to investigate the relationship between personal $PM_{2.5}$ exposure obtained from personal portable nephelometers and background concentration, obtained by an air quality monitoring station run by the Lombardy Region Environmental Protection Agency (ARPA). We were also interested in investigating the extent to which personal exposure differed from average background values.

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2. Materials and methods

2.1. Nephelometers and calibration

Personal exposure was monitored using SidePak (TSI Inc., Model AM510, Shoreview, MN, USA) portable (0.46 kg, 106 × 92 × 70 mm) nephelometers that can be mounted on a belt to minimize wearer inconvenience. These instruments are battery-operated laser photometers that express airborne particle mass concentration in mg m^{-3}, and they were used in other similar studies on personal exposure (Muraleedharan and Radojevic, 2000; Chang-Fu et al., 2005; Greaves and Hamers, 2006; Gulliver and Briggs, 2007). The batteries are 2700 mAH rechargeable nickel metal hydride and allow a monitoring time of about 20 h. Each instrument was fitted with a nozzle with a 50% cut-off at 2.5 μm so that larger size particles were mainly filtered out. The instrument takes a measurement once every ten seconds. The data are stored and later downloaded to a computer using Trakpro software and a USB cable.

As it measures light transmission, nephelometry does not provide a direct measurement of particulate mass concentration, and conversion of light intensity measurements to mass concentration depends on particle composition, specific weight, and also absorbed humidity; thus, a calibration step is mandatory (Görner et al., 1995). Nevertheless, when airborne particles have a fairly constant size and composition distribution, as can be assumed in urban environments, nephelometric measurements can be readily converted to useful estimates of PM mass concentrations, provided that the nephelometer is frequently calibrated against a recognised standard (Molenar, 2005).

During the study, the instruments were calibrated each weekend by the Institute for Environment Sustainability (IES) of the Joint Research Centre (JRC), Ispra, Italy. This was carried out by placing them together with EU reference gravimetric sampler in the three selected schools in Milan in the respective classrooms with open windows for 48 h and comparing $PM_{2.5}$ readings according to approved methodology (CEN reference method EN 14907). Calibration performed with open windows was an attempt to reproduce the outdoor measurements' conditions.

The gravimetric readings were used to calculate a correction factor for each nephelometer and then applied to the original data of each individual instrument (Chakrabarti et al., 2004; Chen et al., 2007; Gulliver and Briggs, 2004; Chung et al., 2001; Case et al., 2008). Moreover, at the beginning of the study a zero filter was attached to each instrument inlet for the zeroing process.

2.2. Data collection

Ninety pupils from three Milan schools (Feltrinelli, Cremona, Rinascita) were involved in the project which lasted 3 weeks during November and December 2006. Six nephelometers were available for the study, each was used by a different pupil each day. Teachers and pupils were instructed on correct instrument use. The instrument was placed in the classroom at the beginning of each run period (at about 12.00 am) close to the pupil entrusted with it for that day. When pupils moved, in particular when they went home, they took the instrument with them carrying it in a bag mounted on the belt, with the inlet tube placed in the breathing zone, so that the inlet was always exposed. When pupils were at home they were instructed to keep the instrument in the room they mainly occupied, and during the night to place it in their bedroom on a table near their breathing zone. The following morning the instrument was carried to school and remained in the classroom with the pupil during the first hour (until about 9.00 am). So the total monitoring period for each pupil was about 21 h.

The pupils ware also instructed to keep a time-activity diary (TAD) reporting their location and activity every 30 min (Chang-Fu et al., 2005; Fondelli et al., 2008) to make it possible to assess variations in exposure according to the circumstances of exposure, i.e. the means of transport used to and from school, smoking and passive smoking, etc. The one-every-10-second measurements of each nephelometer were averaged over 30 min to match the 30 min diary readings.

All the measurements sampled by the nephelometers were classified as indoor or outdoor according to the corresponding location expressed in TAD. All ambients in school (classroom, laboratory, etc.) and at home were classified as indoor ambients, while all the locations signed during the way from home to school and vice versa (bus, train, car, bike, walking, etc.) were considered outdoor.

Nephelometer measurements were compared with daily (24 h) average $PM_{2.5}$ values obtained by the ARPA background station at Juvara in Milan using a beta-attenuation mass monitor (web site http://ita.arpalombardia.it/ITA/qaria/Home.asp accessed 28 May 2010). Juvara fixed-site monitor (FSM) station is located in a street with a medium traffic density and it is 3.6 km far from Cremona school, 4.1 km far from Feltrinelli school and 6.8 km far from Rinascita school.

All readings obtained in a day by the six nephelometers were corrected using the calibration factors and used to calculate a daily average. As the instruments passed from a student to another during the morning at school (after the battery was recharged), each daily average was calculated using the last sampling period of the first student (from midnight to approximately 9 am) and the period from noon to midnight of the second student.

2.3. Statistical methods

Descriptive statistics (means, medians, skewness, first and third quartiles) were calculated on 30 min averaged values.

The relationship between arithmetic daily means of nephelometer readings and background station daily average values was evaluated by Pearson's correlation coefficient (R) with 95% confidence intervals.

The SAS software package, version 8.2, was used for the analyses (SAS Institute Inc., 1989).

3. Results

For each nephelometer, at the beginning of the study we tested whether the coarse fraction (>$PM_{2.5}$) was cut off at the inlet by applying an optical particle counter (Tittarelli et al., 2008) to the outgoing air flow. Negligible numbers of particles over 2.5 μm in diameter were counted in all cases. Correction factors derived from comparison between each nephelometer and a reference gravimetric $PM_{2.5}$ sampler and then applied to the original data are shown in Table 1. Each one of the six average calibration factor (last line of Table 1) was applied to the corresponding nephelometer.

The calibration factors calculated comparing each nephelometer with a gravimetric sampler (0.43 on average, as shown in Table 1)

Table 1
Correction factors derived from comparison between each nephelometer and a reference gravimetric $PM_{2.5}$ sampler.

Instrument number	4047	6069	4046	6068	6063	6071
Week-end 1 (11–13/11/2006)	N.A.	N.A.	0.53	0.55	0.35	0.34
Week-end 2 (18–20/11/2006)	0.61	0.40	N.A.	N.A.	0.39	0.38
Week-end 3 (25–27/11/2006)	0.48	0.37	0.36	0.37	0.36	0.35
Calibration factor	**0.55**	**0.39**	**0.45**	**0.46**	**0.37**	**0.36**

Note: N.A. = Not Available.

Table 2
Mean personal exposure of all participating pupils on each day of the study period with median, skewness, first and third quartiles and background reference value.

Day	Mean ($\mu g\,m^{-3}$)	Median ($\mu g\,m^{-3}$)	Skewness	First quartile ($\mu g\,m^{-3}$)	Third quartile ($\mu g\,m^{-3}$)	Background value ($\mu g\,m^{-3}$)
13/11/2006	131.42	99.20	4.49	74.8	118.6	99
14/11/2006	112.71	102.72	4.74	86.4	140	57*
15/11/2006	122.44	118.80	8.06	88.4	148	113
16/11/2006	142.81	120.45	8.55	85	157.8	117
17/11/2006	124.60	115.08	15.82	89.9	150.4	98
18/11/2006	80.47	68.46	7.73	59.7	98.4	77
20/11/2006	56.61	51.24	3.24	41.2	64.3	66
21/11/2006	66.92	52.50	15.73	38.5	74.3	62
22/11/2006	42.78	16.38	4.10	7.14	25.85	25
23/11/2006	49.62	34.10	4.00	25.2	54.7	59
24/11/2006	48.31	46.08	2.68	26	61.3	51
25/11/2006	48.71	49.98	0.52	34.7	61.9	47
27/11/2006	48.85	34.56	11.31	24.5	55.4	43
28/11/2006	64.99	38.50	3.14	27.9	73.7	31
29/11/2006	45.35	29.26	34.77	23.6	50.6	33
30/11/2006	49.41	33.82	18.94	26.0	57.2	54
01/12/2006	48.25	45.08	5.78	34.0	58.1	67*
02/12/2006	54.34	54.60	5.69	42.8	63.4	76*
Overall	75.94	58.08	11.41	32.34	98.88	65.28

Note: the asterisk indicates the background values outside the inter-quartile range.

indicated that nephelometers tend to overestimate PM concentrations, and their calibration is a mandatory step in studies evaluating personal exposure.

During the study period, $PM_{2.5}$ background daily concentrations varied from a minimum of 25 $\mu g\,m^{-3}$ (on 22 November) to a maximum of 117 $\mu g\,m^{-3}$ (on 16 November), as shown in the last column of Table 2. The mean of this distribution was 65.28 $\mu g\,m^{-3}$, the median was 60.50 $\mu g\,m^{-3}$, the inter-quartile range was 28.75 $\mu g\,m^{-3}$. Similarly, $PM_{2.5}$ daily averages measured by the nephelometers varied from a minimum of 42.78 $\mu g\,m^{-3}$ (on 22 November) to a maximum of 142.81 $\mu g\,m^{-3}$ (on 16 November), as illustrated in the second column of Table 2. The mean of this distribution was 75.94 $\mu g\,m^{-3}$, the median was 58.08 $\mu g\,m^{-3}$, the inter-quartile range was 66.54 $\mu g\,m^{-3}$.

Table 3
Correlation (Pearson's *R* with 95% confidence intervals) between mean nephelometer readings ($\mu g\,m^{-3}$ $PM_{2.5}$) at each school and background values over the study period.

School	Pearson's R [95% C.L.]	Slope [95% C.L.]	Intercept [95% C.L.]
Feltrinelli	0.754 [0.248; 0.927]	1.11 [0.64; 1.58]	8.70 [−29.3; 47.39]
Rinascita	0.739 [0.616; 0.824]	0.82 [0.57; 1.08]	24.82 [5.81; 43.53]
Cremona	0.638 [0.592; 0.679]	0.95 [0.58; 1.32]	2.95 [−24.1; 30.0]
Overall	0.630 [0.628; 0.632]	0.95 [0.73; 1.17]	10.73 [−5.03; 26.5]

The difference between median personal exposure readings and background values over the study period was limited as background readings largely fell into the inter-quartile range of sampled data (see Table 2).

The relationship between the values supplied by the nephelometers and background values, together with regression lines, are shown in Fig. 1, for each single school and for the overall data. The values of the slope and of the intercept of regression lines could be interpreted this way: Cremona and Feltrinelli schools gave measurements generally closer to those supplied by the background station, whereas Rinascita school, which is farer from the station and from the centre of the town, gave exposure values generally lower.

The correlation (Pearson's *R*) between the mean nephelometer readings (six nephelometers each day) and daily background values varied from 0.64 to 0.75; the overall correlation was 0.63 (see Table 3). Daily means of exposures for all pupils are shown in Fig. 1.

Of the total measurements taken by the nephelometers, the majority of the time was spent indoors (67.3%). During the study period the mean of indoor measurements was 79.35 $\mu g\,m^{-3}$, while

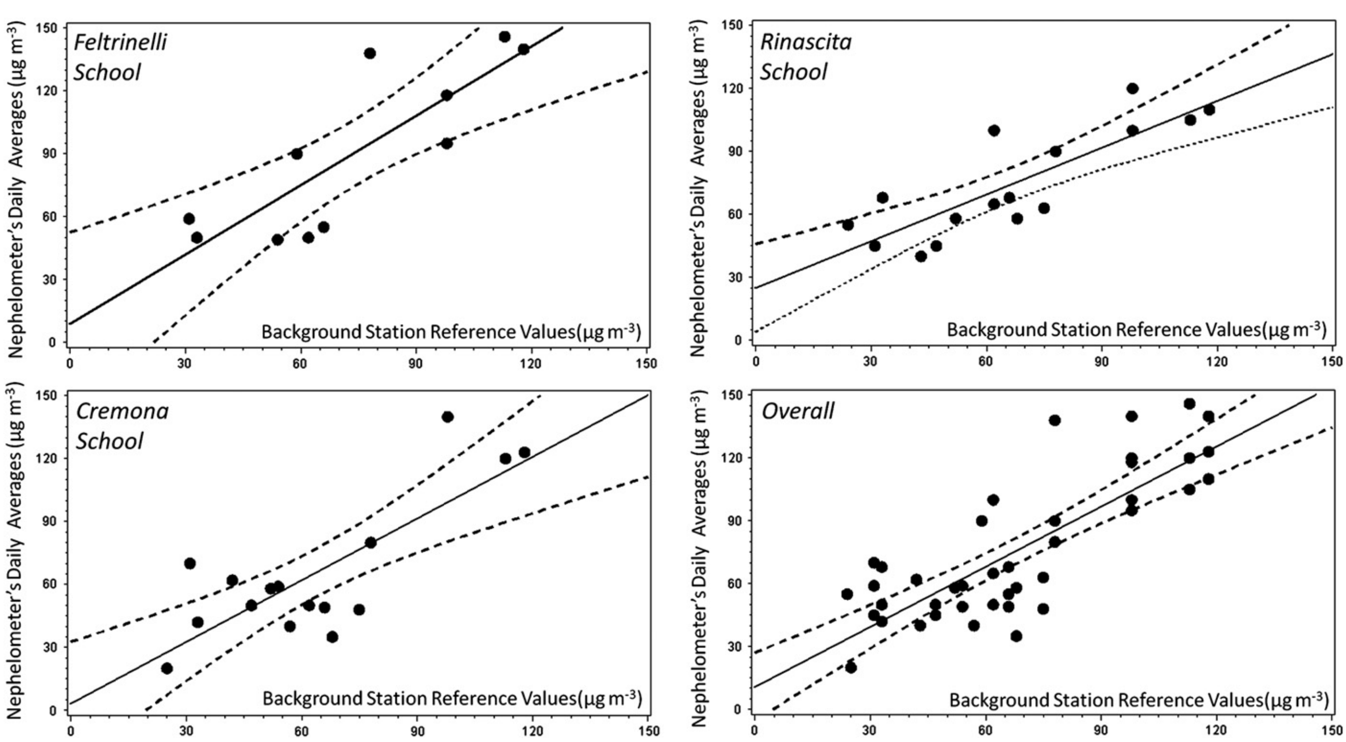

Fig. 1. Correlation between nephelometer's daily averages and background station reference values.

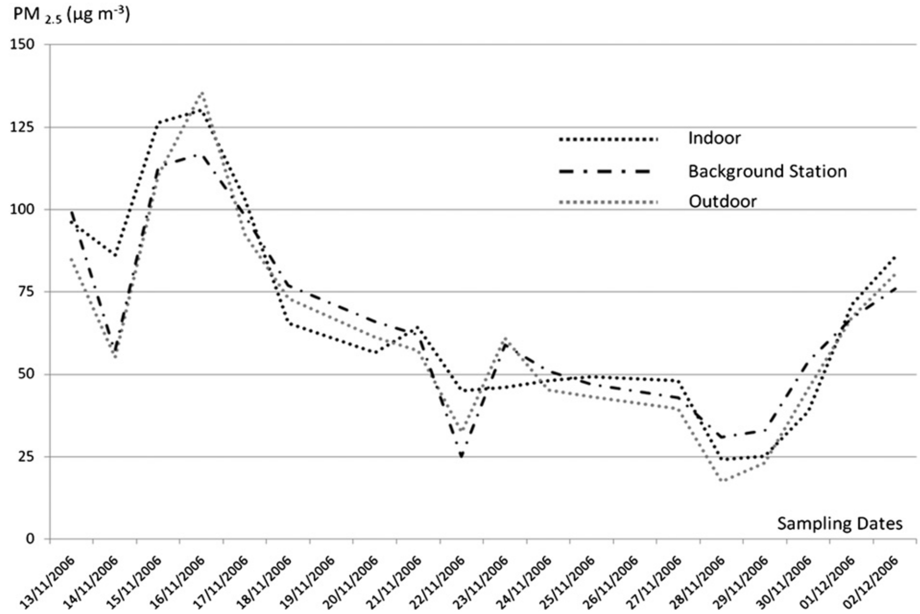

Fig. 2. Daily averages of indoor and outdoor measurements taken by nephelometers compared with background values.

the mean of outdoor measurements was 77.88 μg m^{-3}. Fig. 2 shows the daily averages of indoor and of outdoor measurements, together with background values. A good correlation in temporal pattern between the three distributions was observed.

4. Discussion

Epidemiological studies on the health effects of air pollution usually make use of data from fixed monitoring stations as a proxy of individual exposure as it has often been impracticable to measure the personal exposure of each study participant. However it is important to measure pollutant concentrations in the micro-environments in which people spend their time (indoor and outdoor) because $PM_{2.5}$ concentrations are higher in many micro-environments than those measured by fixed site monitors (Adams et al., 2001).

Nevertheless our analysis revealed good overall concordance between personal $PM_{2.5}$ exposure and exposure measured at the reference site. The overall correlation coefficient R between the nephelometers reading and daily background values was 0.63. The square of this value (R^2) is 0.3963. This coefficient could be read as about 40% of the variability in the mean daily personal exposure at the three schools during the study period due to the variability in background exposure, the remaining 60% to differences in between-subject exposure, but also to some errors due to the instruments used.

The overall correlation found ($R = 0.63$) is comparable with those found in similar studies (Janssen et al., 1997; Noullett et al., 2006). Other studies evidenced generally lower correlations (Kaur et al., 2007) or varying relationships according to different areas (Nerriere et al., 2005). One study monitoring personal exposure of a single person during one year found a strong correlation with fixe-site monitor (Braniš and Kolomazníková, 2010). Other factors that could influence the correlation between personal exposure and fixed-site measurements, such as meteorological variables like wind speed (Adams et al., 2001), relative humidity (Wu et al., 2005; Crist et al., 2008) or inversion conditions (Noullett et al., 2006) were not considered in our study. Anyway, during the sampling period varying meteorological conditions were observed.

As shown in Table 1, we found different calibration factors for the six nephelometers used in this study (from a minimum of 0.36 to a maximum of 0.55). This raised some question about the reliability of the instruments. Anyway, the possible error in measurements could have affected the absolute values of the $PM_{2.5}$ monitored, but not the variability, which was found correlated with the variability of background data.

The total monitoring period for each nephelometer during a day was about 21 h. All those measurements were averaged to compare them with background daily values supplied by FSM. Our opinion is that the three hours missing each day (approximately from 9 am to 12 am, when the nephelometers were charging in the classroom) would not have changed dramatically the average values, because the instruments were placed in the classrooms the whole morning, supplying data for the first and the last hour inside the school.

This study was not adequate to estimate the differences between indoor and outdoor concentrations, as simultaneous measurements with two instruments (one outdoor and one indoor) are needed for this kind of evaluation. Nevertheless, indoor and outdoor measurements both contribute to the daily average values calculated using the data supplied by the nephelometers. Therefore, an attempt to investigate whether their contribution could be different, and at which extent, was given in the manuscript.

In preceding studies on personal exposure, the percentage of time spent indoor varied from a range of 56–66% (Kousa et al., 2002) to 75.6% (Kim et al., 2006), 82.7% (Liu et al., 2003), 84–93% (Sarnat et al., 2006) and 90.5–95.2% (Quintana et al., 2001). The value we calculated (67.3%) is probably an underestimation of the total time spent indoor by the students, because at least for three hours at school the instrument was not active.

The relationship found between personal exposure and reference measurements suggest us that background values represent a proper proxy for personal exposure, though some errors were found in nephelometers' measurements and some caution is necessary using them. Anyway, to identify and assess the major factors affecting individual exposure of $PM_{2.5}$ besides the background concentration could be an interesting issue (Kaur et al., 2005; Greaves et al., 2008; Zhou and Levy, 2008; Chen et al., 2008; Kim et al., 2007).

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