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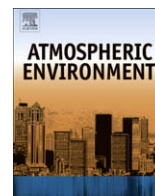
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Technical note

Estimation of particle mass concentration in ambient air using a particle counter

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ABSTRACT

Particle count may have advantage over particle mass concentration for assessing the health effects of airborne particulate matter. However, health effects have mainly been investigated with mass-measuring instruments, so it is important to assess relationships between the variability of particle number, as determined by an optical particle counter, and the variability of particle mass as measured by traditional mass-measuring instruments. We used a light scattering particle counter to monitor the concentration of particulate matter in ambient air in a northern Italian city continuously from August 2005 to July 2006. Six channels were calibrated to count particles in the size range 0.3–10 μm and above. Particles under 0.3 μm cannot be detected by the instrument. The particle counter was placed alongside the mass-measuring instruments of the Environmental Protection Agency of the Region of Piemonte (ARPA). Particle numbers were transformed into masses and compared with PM_{10} and $\text{PM}_{2.5}$ data obtained from the ARPA instruments. Daily average values were compared. The correlation between the two methods was good for both PM_{10} ($R^2 = 0.734$) and $\text{PM}_{2.5}$ ($R^2 = 0.856$); differences between means were significant only for $\text{PM}_{2.5}$. These findings suggest that a light scattering particle counter might be suitable for assessing particulate matter variability in epidemiological studies on effects of air pollution, though further investigations are necessary.

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1. Introduction

Air pollution is an important determinant of human health and the deleterious effects of airborne particulate matter are of major concern. Epidemiological studies have shown that the level of particulate air pollution is associated with adverse short-term (Samet et al., 2000; Jaffe et al., 2003) and long-term health effects (Dockery et al., 1993; Pope et al., 2002; Gauderman et al., 2004).

In all the above-cited studies exposure to particulate matter (PM) was assessed by determining mass concentrations ($\mu\text{g m}^{-3}$) of particles of aerodynamic diameter less

than 10 μm (PM_{10}) or less than 2.5 μm ($\text{PM}_{2.5}$). However, it has been suggested that the number rather than the mass per unit volume of fine particles in air might be more closely correlated with adverse health effects (Wichmann et al., 2000). Particle number is an important indicator of air quality (Gomišček et al., 2004). Ruuskanen et al. (2001) suggested that both particle number and mass concentrations should be measured to provide a comprehensive assessment of urban air quality, as well as to investigate associations between air pollution and adverse health outcomes.

The most important advantages of particle counters are their mobility, low cost, ease of use and their ability to measure particle concentrations over short time intervals (1 s). They can therefore be used to assess spatial and

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temporal variations in particle concentrations (Weijers et al., 2004) and provide good approximations of real exposure in various situations (Gouriou et al., 2004).

However, as health effects have mainly been investigated with mass-measuring instruments, it is important to determine the relationship between particle number, as determined by a particle counter, and particle mass as measured by traditional mass-measuring instruments.

The aim of this study was to assess whether the variability of particle counting correlated satisfactorily with the variability of concentration measurements and whether there were differences in relation to meteorological variables. Our work was also stimulated by suggestions derived from the literature concerning the use of particle counters to assess air pollution concentration (Tuch et al., 1997; Weijers et al., 2004; Gomišček et al., 2004) and to investigate the relationship between particle number and particle mass (Harrison et al., 1999; Yanosky et al., 2002; Hoek et al., 2008).

Such a study is a first step in assessing the validity of counting particles of different sizes, which is expected to be used for studies relating air pollution to health outcomes. The present study was performed in Turin, a city of 902 569 inhabitants in north-west Italy, over the period of one year.

2. Experimental

2.1. The particle counter

The device we used was a six-channel particle counter (model 9012-2, Met One Instruments, Inc., Rowlett, Texas, USA) which employs light scattering technology and a laser diode optical sensor to detect and count particles in six size ranges. The instrument continuously samples air at 2.83 L min^{-1} . The average number of particles counted per litre per minute in each channel is recorded by the data acquisition system.

The instrument was calibrated by the manufacturer using traceable polystyrene latex particles, following the method prescribed by the US National Institute of Standards and Technology (web site <http://ts.nist.gov/traceability>, accessed 28 February 2008); the six channels were set to count particles of the following range of diameters:

- (1) $0.3\text{--}0.5 \mu\text{m}$
- (2) $>0.5\text{--}0.7 \mu\text{m}$
- (3) $>0.7\text{--}1.0 \mu\text{m}$
- (4) $>1.0\text{--}2.5 \mu\text{m}$
- (5) $>2.5\text{--}10 \mu\text{m}$
- (6) $>10 \mu\text{m}$

Particles under $0.3 \mu\text{m}$ diameter cannot be detected by the instrument.

The device was installed at the Region of Piemonte Environmental Protection Agency (ARPA) station of

Lingotto, within 1 m of instruments measuring masses of PM_{10} and $\text{PM}_{2.5}$. All instruments were 3 m above the ground. The Lingotto station is situated in a park in a residential area of Turin and is classified as an urban background station.

The particle counter was tested once a month by checking the zero reading while purging the instrument inlet with dust-free nitrogen. Flow rate was measured once a week using a flow meter with the manufacturer's calibration certificate; the pump was adjusted to compensate for small changes in the flow when necessary. To reduce wear on the instrument it collected data for 12 min in each hour, being turned on and off automatically at the beginning and end of this period. The data were divided into 12 one-minute intervals. Measurements were carried out from August 2005 to the end of July 2006.

2.2. Data check and number-mass transformation

The accuracy of hourly estimations using only 12 one-minute measurements was assessed in a pilot study in which we compared the mass estimation using all 60 available one-minute measurements with that obtained using 12 measurements. The agreement between the two was excellent ($R = 0.96$, data not shown in detail), and it is not therefore expected that the smaller number of measurements is likely to introduce a bias in the comparison.

Twelve one-minute values were obtained each hour. The measurements of the first one-minute period were always discarded as they proved not to be reliable. When the measured flow rate differed from the set flow rate of 2.83 L min^{-1} , a correction factor was applied to the data gathered during the preceding week.

Outliers were identified using an algorithm that rejected values five times higher or lower than the mean of the 10 preceding measurements, even if they belonged to the previous twelve-minute period, or the mean of the 10 successive measurements including those in the successive twelve-minute period.

The algorithm used to transform particle numbers to mass assumed particles were spherical (Wittmaack, 2002) and had a density of 1.65 g cm^{-3} , as suggested by Tuch et al. (2000) and Weijers et al. (2004). For each channel, to determine the mass per $\mu\text{g m}^{-3}$ we apply an average

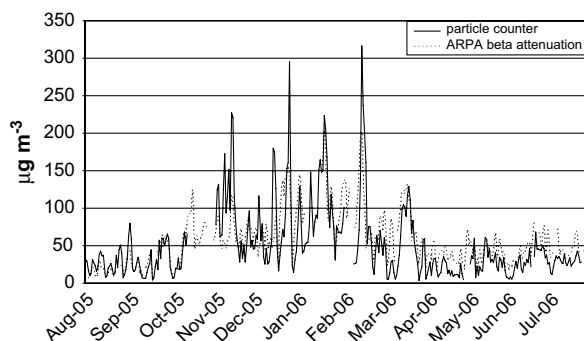


Fig. 1. Daily averages of PM_{10} values determined by the particle counter and particle mass (ARPA) methods over the period from 1 August 2005 to 31 July 2006.

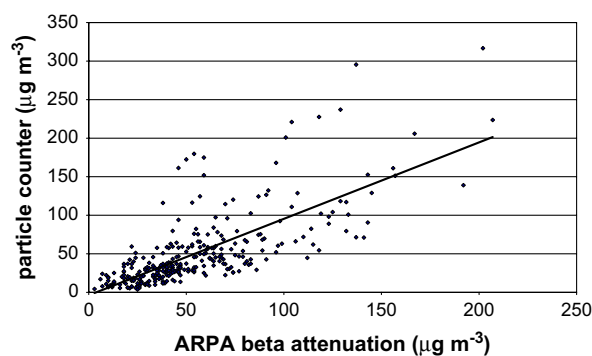


Fig. 2. Correlation between mean daily values of PM_{10} determined by the particle counter and those determined by particle mass (ARPA) method over the period from 1 August 2005 to 31 July 2006.

particle diameter, which was calculated as the arithmetic mean of the stated size interval for the first to fourth channels. For the fifth channel, collecting a wider size range (2.5–10 μm), a value closer to the lower extreme was used (i.e. 4.47). This number had been estimated empirically using a sample of real data, considering that particle number decreases exponentially as size increases.

PM_{10} ($\mu\text{g m}^{-3}$) was calculated as the sum of mass for channels 1–5. Similarly, $PM_{2.5}$ ($\mu\text{g m}^{-3}$) was calculated as the sum of mass for channels 1–4.

2.3. PM mass data

PM_{10} and $PM_{2.5}$ were measured continuously at the ARPA station at Lingotto, using devices that are checked daily. PM_{10} was measured using a beta attenuation SM200 instrument (Opsis, Furulund, Sweden) operating in mass mode. PM_{10} values were determined every 2 h. $PM_{2.5}$ was measured by the European gravimetric reference method using a Charlie HV instrument (TCR Tecora, Corsico, Italy). The $PM_{2.5}$ mass measurements were available as daily values.

2.4. Meteorological data

Hourly values of meteorological variables were supplied by the meteorological station of the Turin Giardini Reali

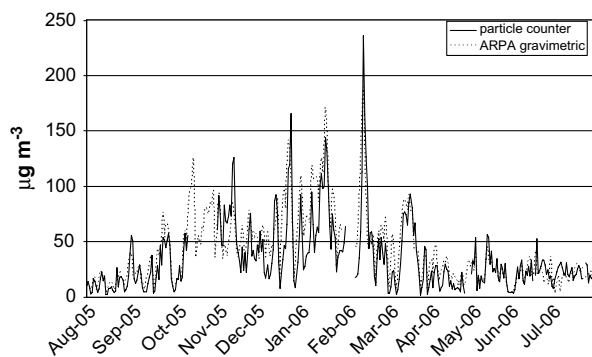


Fig. 3. Daily averages of $PM_{2.5}$ values determined by the particle counter and particle mass (ARPA) methods over the period from 1 August 2005 to 31 July 2006.

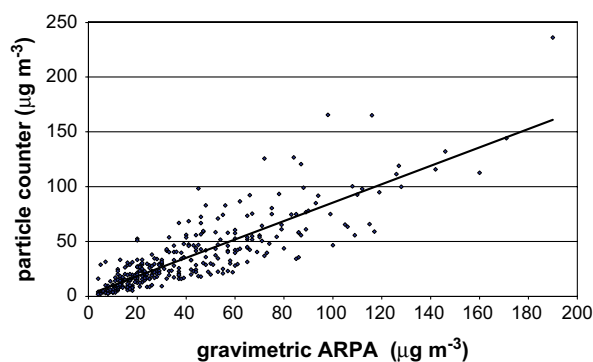


Fig. 4. Correlation between mean daily values of $PM_{2.5}$ determined by the particle counter and those determined by particle mass (ARPA) method over the period from 1 August 2005 to 31 July 2006.

(Royal Gardens, about 6 km from the Lingotto station). Daily averages of the following were used: air temperature ($^{\circ}\text{C}$), relative humidity (%), wind speed (m s^{-1}) and rainfall (mm).

2.5. Data processing and statistical analyses

Particle masses were calculated from the particle counter data as daily averages because the ARPA $PM_{2.5}$ measurements are available as daily averages and because daily averages are generally used in epidemiological studies. The two-hourly ARPA PM_{10} values were also converted to daily averages. A daily average was considered valid only if 75% of the hours were covered.

Logarithmic transformation was performed to render PM distributions normal (Lu and Fang, 2003). Differences between means were evaluated by the two-tailed t -test. Correlations between our PM mass estimates and the PM values supplied by ARPA were assessed by linear regression, considering ARPA values as the independent variable and our mass estimation as the dependent variable.

Correlations between values for each channel, and correlation between the fine ($PM_{2.5}$) and coarse ($PM_{(10-2.5)}$)

Table 1

Influence of meteorological variables on PM_{10} estimated by the particle counter and particle mass (ARPA) methods

| | | No. of days | Mean PC ^a (µg m ⁻³) | Mean ARPA ^b (µg m ⁻³) | <i>p</i> value (<i>t</i> -test) | <i>R</i> ² | β ^c |
|--|-------|----------------|---|--|-------------------------------------|-----------------------|----------------|
| Rain (mm) | 0 | 224 | 52.73 | 58.69 | 0.017 | 0.755 | 0.942 |
| | >0 | 78 | 46.20 | 40.32 | 0.184 | 0.659 | 1.139 |
| Temperature (°C) | <13.5 | 145 | 74.92 | 74.37 | 0.898 | 0.732 | 1.013 |
| | ≥13.5 | 157 | 28.39 | 35.17 | <0.001 | 0.820 | 0.761 |
| Relative humidity (%) ^d | <70 | 129 | 33.83 | 50.43 | <0.001 | 0.798 | 0.698 |
| | ≥70 | 94 | 78.36 | 70.07 | 0.096 | 0.786 | 1.110 |
| Wind speed (m s ⁻¹) | <1.0 | 259 | 46.25 | 48.56 | 0.290 | 0.723 | 0.992 |
| | ≥1.0 | 7 | 32.28 | 40.71 | 0.256 | 0.799 | 0.756 |
| All days | | 305 | 51.07 | 53.97 | 0.179 | 0.734 | 0.969 |

^a PM_{10} mass estimated by particle counter.

^b PM_{10} mass measured by ARPA beta attenuation instrument.

^c coefficient of linear regression ($y = \beta x$), with ARPA values as independent variable and PC values as dependent variable.

^d Excluding rainy days.

Table 2

Influence of meteorological variables on PM_{2.5} estimated by the particle counter and particle mass (ARPA) methods

| | | No. of days | Mean PC ^a ($\mu\text{g m}^{-3}$) | Mean ARPA ^b ($\mu\text{g m}^{-3}$) | <i>p</i> value <i>t</i> -test | <i>R</i> ² | β ^c |
|---------------------------------------|-------|-------------|--|--|----------------------------------|-----------------------|----------------------|
| Rain (mm) | 0 | 241 | 37.58 | 44.12 | <0.001 | 0.867 | 0.852 |
| | >0 | 81 | 31.43 | 28.70 | 0.147 | 0.840 | 1.106 |
| Temperature (°C) | <13.5 | 155 | 51.98 | 60.26 | 0.001 | 0.855 | 0.872 |
| | ≥13.5 | 167 | 21.04 | 21.73 | 0.357 | 0.857 | 0.921 |
| Relative humidity (%) ^d | <70 | 137 | 24.37 | 31.45 | <0.001 | 0.843 | 0.749 |
| | ≥70 | 103 | 55.06 | 61.02 | 0.017 | 0.881 | 0.895 |
| Wind speed (m s ⁻¹) | <1.0 | 269 | 31.87 | 33.41 | 0.135 | 0.848 | 0.963 |
| | ≥1.0 | 8 | 22.87 | 25.37 | 0.462 | 0.890 | 0.908 |
| All days | | 325 | 36.06 | 40.28 | 0.001 | 0.856 | 0.880 |

^a PM_{2.5} mass estimated by particle counter.

^b PM_{2.5} mass measured by ARPA gravimetric instrument.

^c coefficient of linear regression ($y = \beta x$), with ARPA values as independent variable and PC values as dependent variable.

^d Excluding rainy days.

components of particulate matter, estimated by the particle counter, were assessed by Pearson's correlation coefficient (*R*).

Correlation coefficients were calculated for dichotomized values of temperature, relative humidity, rainfall and wind speed. The *t*-test was used to assess the significance of differences between the means of the distributions of each group. All the analyses were performed with Stata/SE version 8.2.

3. Results and discussion

3.1. Particle number and mass

We considered particle count measurements obtained in 365 days from 1 August 2005 to 31 July 2006. A total of 150 335 valid one-minute estimates were available. From these 365 daily averages were calculated, 32 of which were excluded as more than 25% of hourly measurements were missing, leaving 333 valid daily averages. For the PM₁₀ analyses ARPA data were missing for 28 of these days and 305 daily averages were used. For PM_{2.5} analysis, 325 values were used, as only eight corresponding PM_{2.5} values from ARPA were missing.

Over the entire study period, our estimate of the average PM₁₀ was 51.07 $\mu\text{g m}^{-3}$ (standard deviation [SD] 52.25) compared to 53.97 $\mu\text{g m}^{-3}$ (SD 35.49) measured by ARPA. The *t*-test indicated that the difference between the means of the two distributions was not significant ($p = 0.179$).

Daily values were in good agreement, as illustrated in Figs. 1 and 2, with goodness of fit $R^2 = 0.734$.

Fig. 3 shows daily PM_{2.5} values estimated by the particle counter in comparison with those measured by ARPA. Our estimate of average PM_{2.5} over the entire period was 36.06 $\mu\text{g m}^{-3}$ (SD 32.73) compared to 40.28 $\mu\text{g m}^{-3}$ (SD 31.63) measured by ARPA. The correlation was better (Fig. 4) for PM₁₀, with $R^2 = 0.856$. However, the means of the two distributions differed significantly (*t*-test, $p = 0.001$).

The algorithm used to convert number to mass may have had an influence on the mean values we found. Particles are not in general perfectly spherical (Taylor, 2002); furthermore, variations in particle density with size (Wittmaack, 2002) and time of day (Morawska et al., 1999) are also likely. However, using different values for the particle density will change the estimated mass values, but will not affect correlations between the number of particles and the estimated mass.

3.2. Influence of meteorological variables

Meteorological variables were dichotomized to investigate their effects on the estimates of PM₁₀ (Table 1) and PM_{2.5} (Table 2). Median values were used as cut-offs for temperature (13.4 °C) and relative humidity (70%). There were 88 rainy days (≥0.1 mm rain) and 8 windy days (wind speed ≥ 1.0 m s⁻¹) over the study period.

Relative humidity (not analysed on rainy days) had a major influence on the agreement between the two measures: when relative humidity was high, PM₁₀ masses derived from particle counts were higher (though not significantly) than those estimated by ARPA, whereas PM_{2.5} masses from particle counts were significantly lower than those reported by ARPA; when humidity was low, both our PM masses were significantly lower than ARPA values. A likely explanation is that particles are hygroscopic (Wittig et al., 2004) and increase in size on absorbing water resulting in mass overestimation (Wilson and Suh, 1997).

On rainy days the mass estimated by the particle counter was higher and correlation was lower (both for PM₁₀ and PM_{2.5}), presumably also due to water absorption by particles.

Temperature had differing influences on PM₁₀ and on PM_{2.5}: for PM₁₀ the means of the two distributions differed significantly at higher (≥13.5 °C) temperatures, although the correlation was slightly stronger; for PM_{2.5} the two means differed significantly at lower (<13.5 °C) but not higher temperatures; the correlation remained good for both temperature intervals. This could well be a chance

Table 3

Distribution of particle numbers in the five particle size channels, with relative mass transformations

| Channel | Diameter (μm) | Min (10 ⁻³ cm ⁻³) | Max (10 ⁻³ cm ⁻³) | Mean (10 ⁻³ cm ⁻³) | Mean (μg m ⁻³) | % of mass |
|-------------------------|---------------|--|--|---|----------------------------|-----------|
| 1 | 0.3–0.5 | 13 517 | 507 529 | 199 027 | 11.00 | 21.24 |
| 2 | >0.5–0.7 | 1131 | 256 899 | 37 403 | 6.98 | 13.47 |
| 3 | >0.7–1.0 | 247 | 73 111 | 7139 | 3.79 | 7.31 |
| 4 | >1.0–2.5 | 124 | 29 743 | 3054 | 14.14 | 27.29 |
| 5 | >2.5–10 | 9 | 2146 | 206 | 15.93 | 30.68 |
| Total PM _{2.5} | | | | | 35.91 | 69.32 |
| Total PM ₁₀ | | | | | 51.84 | 100.00 |

Table 4
Correlations between channels (Pearson's *R*)

| Channel (μm) | 1 | 2 | 3 | 4 | 5 |
|---------------------------|------|------|------|------|------|
| 1 (0.3–0.5) | 1.00 | 0.74 | 0.57 | 0.48 | 0.40 |
| 2 (>0.5–0.7) | | 1.00 | 0.95 | 0.88 | 0.69 |
| 3 (>0.7–1.0) | | | 1.00 | 0.97 | 0.80 |
| 4 (>1.0–2.5) | | | | 1.00 | 0.90 |
| 5 (>2.5–10) | | | | | 1.00 |

finding, but may be worth investigating at various temperature intervals.

Wind speed had a small effect. On windy days correlations between both PM_{10} and $\text{PM}_{2.5}$ were slightly better than on non-windy days. As expected, PM concentrations by both measurements were considerably lower on windy days (wind speed $\geq 1.0 \text{ m s}^{-1}$) than non-windy days.

3.3. Relationship between particles of different sizes

As shown in Table 3, the number of particles was inversely related to the diameter. Around 199 cm^{-3} particles were counted in the first channel (0.3–0.5 μm) decreasing to around 0.2 cm^{-3} particles in the fifth channel (>2.5–10 μm).

After transformation into mass (last two columns, Table 3), it was found that the fourth plus the fifth channels (>1.0–10 μm) provided the greatest contribution (almost 60%) to total PM_{10} . Considering the first three channels (particles $\leq 1 \mu\text{m}$), the first (0.3–0.5 μm) provided the greatest contribution to PM_{10} (mean 21.2% of total), because of the high number of particles counted in that channel.

Analysis of the correlations (Pearson's *R*) between each of the channels (Table 4) showed that the intermediate channels (second, third and fourth, counting particles from 0.5 to 2.5 μm) correlated most strongly with each other. Values for the first channel (0.3–0.5 μm) correlated well ($R = 0.74$) with those of the second channel (0.5–0.7 μm), but less well with all the others. Channel 5, counting coarser particles (>2.5–10 μm) correlated strongly with channel 4 ($R = 0.90$) and channel 3 ($R = 0.80$).

4. Conclusions

One of the major limitations of the type of particle counter we used is that it cannot detect particles under 0.3 μm diameter, implying the underestimation of total particle number and possible underestimation of mass in our measurements. Such underestimation might contribute to the differences found between the two methods, but would not significantly affect the correlation.

It is important to note that the differences between the two sets of estimates do not constitute a limitation of the particle counting method. They could be due to the optical properties of particles, their chemical composition or their morphology. Particle counting provides information additional to that provided by mass measurement which is likely to be useful for analysing the health effects of particulate matter (Weijers et al., 2004).

Our findings do not suggest that particle counters should substitute conventional instruments, but – though further investigations are necessary – do suggest that they provide valid particle number data of particular importance in epidemiological studies for the evaluation of health effects from particle matter in ambient air.

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