Intercomparison of five PM10 monitoring devices and the implications for exposure measurement in epidemiological research

Intercomparison of five PM_{10} monitoring devices and the implications for exposure measurement in epidemiological research

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Five different instruments for the determination of the mass concentration of PM_{10} in air were compared side-by-side for up to 33 days in an undisturbed indoor environment: a tripod mounted BGI Inc. PQ100 gravimetric sampler with a US EPA certified Graseby Andersen PM_{10} inlet; an Airmetrics Minivol static gravimetric sampler; a Casella cyclone gravimetric personal sampler; an Institute of Occupational Medicine gravimetric PM_{10} personal sampler; and two TSI Inc. Dustrak real-time optical scattering personal samplers. For 24 h sampling of ambient PM_{10} concentrations around 10 $\mu g\ m^{-3}$, the estimated measurement uncertainty for the two gravimetric personal samplers was larger ($\sim \pm 20\%$) compared with estimated measurement uncertainty for the PQ100/Graseby Andersen sampler ($< \pm 5\%$). Measurement uncertainty for the Dustraks was lower ($\sim \pm 15\%$ on average) but calibration of the optical response against a reference PM_{10} method is essential since the Dustraks systematically over-read PM_{10} determined gravimetrically by a factor ~ 2.2. However, once calibrated, the Dustrak devices demonstrated excellent functionality in terms of ease of portability and real-time data acquisition. Estimated measurement uncertainty for PM_{10} concentrations determined with the Minivol were $\pm 5\%$. The Minivol data correlated well with PQ100/Graseby Andersen data ($r = 0.97$, $n = 18$) but were, on average, 23% greater. The reason for the systematic discrepancy could not be traced. Intercomparison experiments such as these are essential for assessing measurement error and revealing systematic bias. Application of two Dustraks demonstrated the spatial and temporal variability of exposure to PM_{10} in different walking and transport microenvironments in the city of Edinburgh, UK. For example, very large exposures to PM_{10} were identified for the lower deck of a double-decker tour bus compared with the open upper deck of the same vehicle. The variability observed emphasises the need to determine truly personal exposure profiles of PM_{10} for quantifying exposure–response relationships for epidemiological studies.

1. Introduction

Epidemiological studies have consistently shown significant associations between changes in the 24 h average gravimetric airborne particulate matter (PM) concentration and a wide range of adverse health effects in the general population, ranging from increased hospital admissions for respiratory symptoms to all-cause mortality.[1–4] Since exposure–response relationships derived from epidemiological studies are the main source of evidence for the formulation and review of PM air quality standards[5] it is important to minimise uncertainties in the quantification of exposures. The usual estimate for individual exposure to PM in epidemiological studies has tended to be the measurement of PM_{10} at a single outdoor location (where PM_{10} refers to a standard sampling probability curve for which the median particle size sampled has an aerodynamic diameter of 10 μm[6]). However, PM is not spatially uniform, and personal activities within different microenvironments can have a much greater effect on personal exposure than variations in sentinel measurements of ambient concentrations. Watt *et al.*[7] have shown that using a single measurement to represent the exposure of a population may lead to a biased interpretation of the exposure–response relationship.

The city street is a well known example of a heterogeneous microenvironment exhibiting concentration gradients both across and vertically within the street canyon.[8,9] Furthermore, people in temperate developed countries spend, on average, 70–90% of their time indoors[1] where they may be exposed to PM from specific local sources such as environmental tobacco smoke, cooking or resuspension of PM by personal and domestic activities. Seaton *et al.*[10] have recently developed an exposure model based on activity diaries and microenvironments in order to derive better estimates for PM_{10} personal exposure. However, the continued lack of information on the true value of individual exposure is a major shortcoming in epidemiological research and the consequent potential misclassification may conceal a more pronounced relationship between health outcomes and actual exposure.[11]

The most direct way of eliminating this shortcoming is to provide subjects under study with personal monitoring apparatus for PM. The two techniques most commonly employed for sentinel measurement of PM_{10} in ambient air against benchmark standards are: (i) the tapered element oscillating microbalance (TEOM), in which changes to the frequency of vibration of a hollow tapered glass tube are related in real time to the mass of PM that accumulates on the surface of an attached filter (this technique cannot be adapted for personal sampling use), and (ii) gravimetric, in which a known volume of air is drawn through a filter medium and the concentration of PM calculated off-line from the increase in mass of the filter. In both cases, the PM_{10} (or other) fraction is discriminated from a calibrated volumetric air flow (typically 16.7 L min^{-1}) using a standard design single-stage impactor inlet head.[6] Since these requirements necessarily result in bulky, static instrumentation, personal samplers are restricted to

lower sampling flows and alternative inlet discrimination, or measurement based on some property of PM other than mass. A compromise approach is to use static devices in different representative microenvironments to estimate personal exposure from the integration of the product of pollutant concentration and time spent in each microenvironment.

A number of different static and personal PM sampling instruments are currently available, but a side-by-side comparison of instrumentation for environmental sampling has not been reported. The aim of this work was two-fold: (i) to compare directly three personal PM instruments (two gravimetric, one optical), and one relatively compact static PM_{10} instrument, with a non-portable reference gravimetric instrument fitted with a US EPA approved Graseby Andersen PM_{10} sampling inlet, under controlled conditions; (ii) to assess the use of the optical instrument in an investigation of the short-term variation of PM_{10} concentration within and between different outdoor microenvironments.

2. Methodology

2.1. Instruments compared

The following instruments were compared simultaneously.

(1) A tripod mounted US EPA approved Graseby Andersen PM_{10} sampling inlet with a sampling flow rate of 16.7 L min^{-1} controlled by a US EPA approved BGI Inc. PQ100 mass flow controller (factory calibrated to temperature and pressure conditions of 298 K and 1 atm, respectively). The PM_{10} fraction was discriminated by impaction and was collected on 47 mm diameter quartz filters (QMA, Whatman Ltd., UK).

(2) A gravimetric static sampler for PM_{10} (Airmetrics Minivol). The PM_{10} fraction was discriminated *via* impaction from an air flow of 5 L min^{-1}, and collected on 47 mm quartz filters (QMA, Whatman, UK). Air flow was established using the factory calibrated rotameter (supplied with calibration certificate) and was controlled by an internal pressure transducer.

(3) A gravimetric personal sampling instrument for respirable matter using an industry standard cyclone head (Casella Ltd., UK). This cyclone has a median sampling efficiency at 7 μm.[12] Air was sampled through 37 mm diameter PVC or PTFE filters at a flow rate of 2.2 L min^{-1}. Flow rate was controlled by a rotameter within the pump unit (SKC, UK).

(4) A gravimetric personal sampling instrument for PM_{10} designed by the Institute of Occupational Medicine.[13] The PM_{10} fraction was discriminated from the inhalable size fraction[14] *via* a porous polyester foam plug and was collected on 37 mm diameter PVC or PTFE filters. The sampling flow rate of 2.0 L min^{-1} was controlled by a pressure regulator.

(5) A real-time optical scattering instrument (Dustrak model 8520, TSI Inc., USA). Concentration of PM in the air flow was detected by the extent of forward scattering of an infra-red diode laser beam. Following the manufacturer's recommendation, discrimination for the PM_{10} size fraction was achieved by adjusting the sample flow rate through the inlet orifice. The Dustrak instrument was factory calibrated using standard "Arizona road dust". Measurements were recorded as 1 min averages. Two Dustrak instruments were available.

The volumetric flow rates of the Casella and IOM personal samplers were established using a rotameter calibrated against a UK NAMAS-certified Gilian flowmeter and measured again at the end of each sampling period. Any sample period in which flow rate varied by $>5\%$ would have been regarded as invalid.[12] No samples fell into this category. The Minivol and PQ100 volumetric flow rates were factory pre-calibrated and validated for accuracy against the Gilian flowmeter and a certified Chinook Engineering Streamline orifice flowmeter, respectively.

2.2. Inter-comparison details

Instruments were compared during three periods: 2 July–1 August, 1997; 7 July–17 July, 1998; and 7 October–30 October, 1998. All samples for gravimetric analysis were accumulated over 24 h periods commencing at approximately 10:00 GMT. The 1 min average data from the Dustrak optical instruments were averaged over the same 24 h period. During the July 1997 intercomparison the instruments were located in a room (21.5 m^2 floor area × 4.0 m height) on the second floor of the Edinburgh University Medical School near the centre of the city. One window of the room was kept slightly open (estimated air exchange rate, aer = 1 h^{-1}). In July and October 1998 the instruments were located in a third floor room (34.5 m^2 floor area × 4.1 m height) of the same building. During the July 1998 period the room was ventilated by an inward-blowing fan (aer = 4 h^{-1}) whilst during the October 1998 period the room was unventilated (aer = 0.4 h^{-1}). In all instances the rooms were left entirely undisturbed, other than for the purposes of daily filter changes.

The IOM and Casella samplers are designed as personal samplers. The sampling inlets were mounted at a height 1.23 m above the floor, on a vertical board of the same approximate dimensions as a human torso (height 0.49 m, width 0.37 m), in the same manner as worn by a human subject. Recent studies[15,16] have shown that there is no significant difference in the sampling efficiency of a number of personal samplers irrespective of whether the inlets are mounted on a full-size manikin, on a rectangular box of similar dimension to a human torso or are free-lying.

2.3. Determination of filter weights

Filter masses were measured with a Sartorius MC5 microbalance (1 μg readout precision) in an isolated, although not humidity controlled, environment. All filters were equilibrated with ambient humidity before and after each sampling period by leaving them in metal tins, with lids slightly ajar, in the weighing room for 24 h before use. At other times all filters were stored in individual sealed tins. The average of two filter masses (separated by 24 h) was taken before and after each sampling period. A minimum of six control filters of each type was included at even time intervals in the procedure, and the average difference in control filter weights used to correct for systematic changes in filter mass resulting from changes in humidity between the "before" and "after" weighing sessions.[12,17] The balance was calibrated a minimum of six times during each weighing session.

The precision of the weighing method was set by the repeat weighing precision of the balance and the variability of the mass change in the control blanks, and was relatively more important for the personal samplers because of their lower flow rates. Initial problems with filter weighing precision were overcome by use of a corona discharge anti-static device and by changing the filter medium for the personal samplers from PVC to PTFE for the October 1998 experiments. Analysis of variance has been applied to the weighing data from these experiments to derive estimates of precision values directly applicable to the weighing procedure used in this study. The mean standard deviation of mass changes for all sets of six control PTFE filters was 4.7 μg. Hence, the standard deviation in net sample mass on one sample filter, corrected using the mean mass change of six control filters, was 5.0 μg,[17] or 1.7 μg m^{-3} for 24 h sampling at a personal sampler flow rate of 2.0 L min^{-1}. The corresponding standard deviations in net sample mass and volumetric PM_{10} concentration for the quartz filters used in the PQ100/Graseby Andersen sampler were 9.8 μg and 0.4 μg m^{-3}, respectively. These errors are similar to those obtained by Vaughan *et al.*[17] using exactly the same weighing procedure on a balance of the same precision.

2.4. Micro-environment personal exposure measurements

Two series of experiments were conducted with the two Dustrak optical devices to obtain time-resolved data (1 min averages) on personal exposure from outdoor activities.

(1) Two volunteers carried the Dustrak monitors during a series of walking and car driving activities in Edinburgh. The activities alternated approximately every 15 min and followed the pattern of a 15 min walk on a busy road leading out of central Edinburgh, a 15 min drive by car into the suburbs to the south (Liberton suburb) or to the north-west (Queensferry suburb) of the city centre, a 15 min walk on the suburban streets at the opposite end of the car journey, and a 15 min return drive by car to the city centre. The sequence was performed twice in succession by each volunteer and on three separate days (17, 18 and 22 June 1998).

(2) The two Dustrak monitors were carried by two volunteers on a commercial bus company tour of the city of Edinburgh. One Dustrak monitor was carried by a volunteer on the open upper deck of the bus whilst the other was carried inside the closed lower deck of the bus. The trial was repeated on three days (10, 24 and 25 June 1998).

2.5. Statistical analyses

Except where indicated, linear regression relationships were calculated using the reduced major axis (RMA) method[18] which, in contrast to linear least-squares regression, does not assume that the independent variable is error free. Regressions were calculated with no *a priori* assumption of a zero intercept. Frequency distributions of concentrations recorded with the two Dustrak devices during the outdoor exposure experiments were approximately log-normal, as expected for air pollutant concentration data,[19] and were analysed using non-parametric methods.

3. Results

3.1. Indoors instrument inter-comparison

For reasons discussed in Section 2.3, analyses of the gravimetric personal sampler data were restricted to the October 1998 experiment. Least-squares linear relationships between the IOM and Casella cyclone personal sampler measurements and the PQ100/Graseby Andersen PM_{10} measurements are shown in Fig. 1 and 2, respectively. The error bars represent the weighing errors derived in Section 2.3. Both correlations are significant ($r=0.62$ and $r=0.71$, respectively, for $n=15$) and the relationships between personal samplers and the PQ100/Graseby Andersen device do not differ significantly

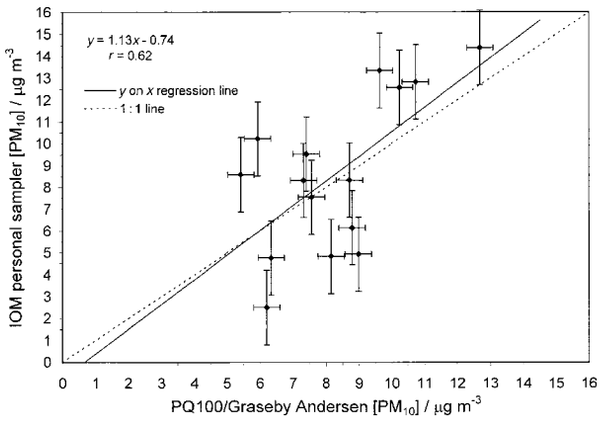

Fig. 1 24 h average gravimetric [PM_{10}] sampled through the PQ100/Graseby Andersen static sampler and the IOM personal sampler, with estimated standard deviation of weighing error. Data are shown for October 1998 only.

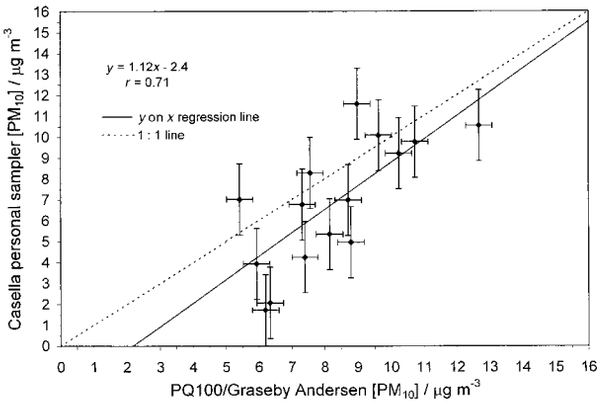

Fig. 2 24 h average gravimetric [PM_{10}] sampled through the PQ100/Graseby Andersen static sampler and the Casella cyclone personal sampler, with estimated standard deviation of weighing error. Data are shown for October 1998 only.

from 1 : 1 (the intercepts are not significant). Nevertheless, there is clearly considerable scatter in the plots.

Fig. 3 shows the RMA linear relationship between the PQ100/Graseby Andersen PM_{10} measurements and 24 h averaged Dustrak 1 data. The Dustrak consistently overreads the PQ100/Graseby Andersen instrument when PM_{10} concentrations exceed about 10 $\mu g\,m^{-3}$, but the correlation between the two is significant. The experimental linear relationship between Dustrak and PQ100/Graseby Andersen PM_{10} measurements (Fig. 3) can be used to transform Dustrak data into values of [PM_{10}] suitable for analysis of personal exposure:

$$PM_{10}/\mu g\,m^{-3} = \frac{\text{Dustrak reading} + 12}{2.2} \qquad (1)$$

For an 88 h period during July 1998 two Dustrak instruments were operated side by side in the undisturbed room and the scatter plot of hourly averaged values is shown in Fig. 4. Despite the excellent correlation between the two instruments ($r=0.99$) there is a significant difference from a 1 : 1 relationship. The observed RMA linear relationship in Fig. 4 was used to convert measurements taken with the second Dustrak into the equivalent reading of the first Dustrak before applying eqn. (1) to calculate corresponding [PM_{10}].

The RMA relationship between 24 h average measurements from the Minivol and PQ100/Graseby Andersen static PM_{10} instruments is shown in Fig. 5. The correlation is significant ($r=0.97$, $n=18$) but Minivol values significantly exceed those of the PQ100/Graseby Andersen. The intercept is not significantly different from zero.

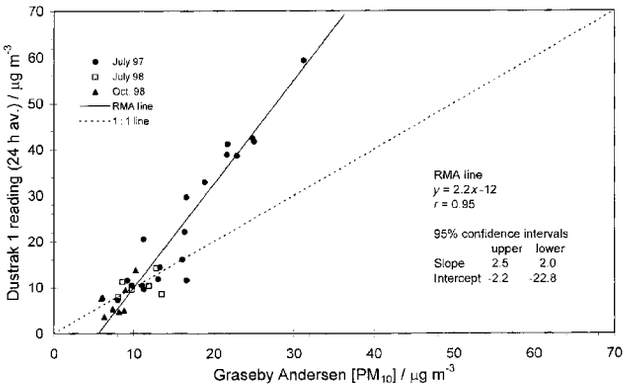

Fig. 3 Scatter plot and reduced major axis relationship between 24 h average PQ100/Graseby Andersen [PM_{10}] and Dustrak PM_{10} measurements.

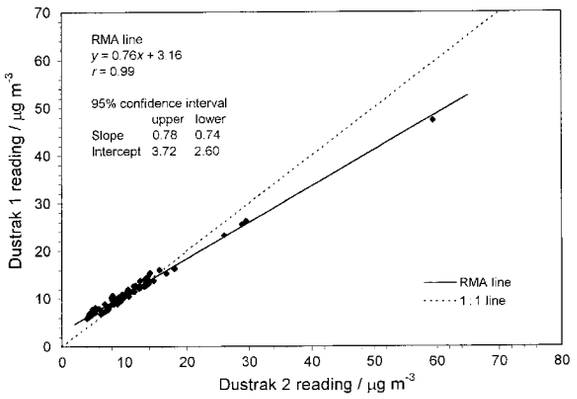

Fig. 4 Scatter plot and reduced major axis relationship between 1 h average PM_{10} measurements of two Dustrak devices.

3.2. Microenvironment sampling

3.2.1. Walking–driving experiments. Two example time series of Dustrak personal sampling data, acquired during the walking and driving experiments of 18th June 1998, and converted to PM_{10} values in $\mu g\ m^{-3}$ using eqn. (1), are shown in Fig. 6. The large peaks in PM concentration at about 10 a.m. on both traces correspond to notations on the time sheets by the volunteers of "diesel bus" (Dustrak 1) and "heavy traffic" (Dustrak 2). The timing is coincidental since at that time the two Dustraks were in separate Edinburgh suburbs over 10 km apart, but the data illustrate the important fact that personal exposure to airborne PM can be highly spatially and temporally variable, the latter on timescales of only a few minutes.

The median walking and driving PM_{10} concentrations measured in each experiment are given in Table 1. The mean PM_{10} concentrations of the four 15 min blocks of walking or driving were compared between walking and driving segments for each volunteer and between the two volunteers on a given day using the non-parametric Mann–Whitney test (Table 1). Data for each day were treated separately because of the confounding day-to-day variation in background PM_{10} concentration. On each of the 3 days of the walking–driving experiments there was no significant difference between PM_{10} concentrations measured by the two volunteers during their walking activities. In contrast, on each of the three days, volunteer 2 (with Dustrak 2) measured significantly smaller PM_{10} concentration during their driving activity than volunteer 1 (with Dustrak 1). In all three experiments, the PM_{10} concentrations recorded by volunteer 1 were not significantly different between walking or driving, whereas for volunteer 2, on two out of three occasions the PM_{10} concentration recorded during driving was significantly smaller than recorded during walking.

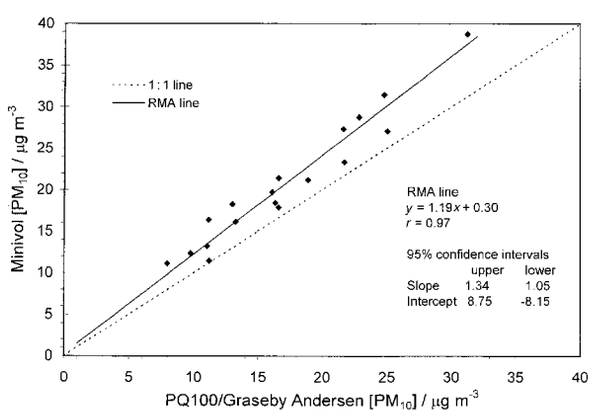

Fig. 5 24 h average gravimetric PM_{10} concentrations sampled through the PQ100/Graseby Andersen sampler and the Minivol sampler.

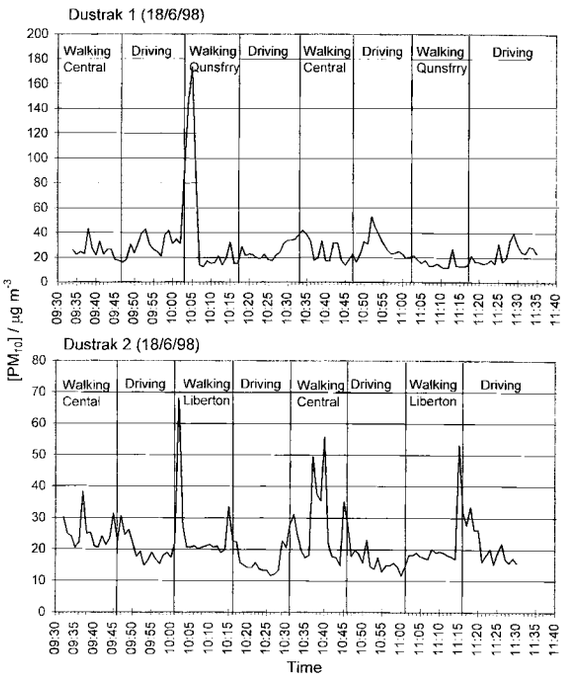

Fig. 6 Time series of alternate walking and driving personal exposures using two Dustrak personal samplers (1 min resolution). Dustrak readings were converted to $[PM_{10}]$ using the calibration relationships in Figs. 3 and 4.

3.2.2. Bus tours. Fig. 7 shows the PM_{10} values recorded simultaneously with the two Dustraks on the upper and lower decks of the Edinburgh tour buses for 24 and 25 June 1998. Two features are obvious: (i) there is very good agreement in PM_{10} measurement by the two Dustraks for periods when both volunteers are not on the bus but walking together; (ii) on nearly all occasions PM_{10} concentrations on the enclosed lower deck of the buses greatly exceed PM_{10} concentrations on the open upper deck. Concentrations measured on the upper deck are generally similar to those recorded during the walking periods of the bus tour and the walking–driving experiments.

Table 1 Median values of Dustrak 1 min averages (corrected to $[PM_{10}]$ in $\mu g\ m^{-3}$ using the relationships in Figs. 3 and 4) for walking and driving activities on each of 3 days. Dustraks 1 or 2 were used for exposures including Queensferry or Liberton suburbs, respectively. The means of the four blocks of 15 min walking or driving measurements were compared within and between exposure experiments on the same day using a Mann–Whitney U-test. The evidence for difference (at 95% significance) is indicated

Date	Name of suburb	Walking	Driving	
17.6.98	Queensferry	17.5	24.4	No
	Liberton	17.3	13.7	Yes
		No	Yes	
18.6.98	Queensferry	18.7	26.0	No
	Liberton	20.7	16.9	Yes
		No	Yes	
22.6.98	Queensferry	12.0	14.3	No
	Liberton	12.9	11.3	No
		No	Yes	

4. Discussion

4.1. Instrument intercomparison

Although neither relationship between the Casella or IOM samplers and the PQ100/Graseby Andersen PM_{10} sampler was significantly different from 1 : 1 (Figs. 1 and 2), the scatter in the correlations clearly indicates measurement error. Regardless of any other source of error between personal and PQ100/Graseby Andersen samplers, a major source of error is the measurement of PM mass on the filters. As discussed in Section 2.3, the weighing error in PM concentrations derived from the personal samplers is over four times greater (1.7 $\mu g\,m^{-3}$) than from the PQ100/Graseby Andersen sampler (0.4 $\mu g\,m^{-3}$). These weighing errors contribute $\sim \pm 21\%$ uncertainty to the personal sampler measurements, but only $\sim \pm 5\%$ to the PQ100/Graseby Andersen measurements for 24 h sampling of an average ambient [PM_{10}] of $\sim 8\ \mu g\,m^{-3}$. For this reason, *y*-on-*x*, rather than RMA, linear regressions are used in Figs. 1 and 2. The standard errors of regression in the IOM *versus* PQ100/Graseby Andersen and Casella *versus* PQ100/Graseby Andersen plots (Figs. 1 and 2, respectively) are 2.9 and 2.2 $\mu g\,m^{-3}$. Therefore intrinsic weighing uncertainty accounts for the majority of the uncertainty in the comparison between Casella and PQ100/Graseby Andersen sampler, whereas the IOM and PQ100/Graseby Andersen sampler comparison is subject to additional error. Although it is possible that the Casella cyclone and IOM polyester foam plugs do not sample exactly the same PM distribution as the standard PQ100/Graseby Andersen PM_{10} head, the significant correlations indicate that the sampling characteristics of the personal samplers were consistent with the PM_{10} convention within the uncertainties quoted.

Some previous studies deploying personal samplers on human subjects have reported personal exposures to PM higher than calculated from time-weighted measurements from free-standing samplers.[20,21] The existence of this excess mass near a person (the "personal cloud") is apparently related to the personal activity of the subject and not to the sampling device and is therefore not a factor influencing these intercomparisons.

Of the three personal sampler devices investigated, the Dustrak personal sampler data correlates best with the PQ100/Graseby Andersen data, albeit with significant over-estimation (Fig. 3). As discussed above, weighing error for the PQ100/Graseby Andersen measurements is <5 % (for concentrations $> \sim 8\ \mu g\,m^{-3}$) so the majority of uncertainty in the Dustrak/Graseby Andersen relationship presumably derives from the Dustrak values. The non-unity regression slope also reflects systematic calibration differences between the Dustrak and PQ100/Graseby Andersen devices. This is not surprising since the Dustrak does not use a reference sampling inlet and the mass of PM_{10} is inferred from an indirect measurement. The Dustrak response is dependent on the optical properties of the sampled PM and requires calibration. The factory pre-set calibration uses Arizona road dust, which will have different size distribution, shape and reflectance properties to that of UK airborne PM_{10} and will produce different scattering responses for identical masses of PM passing through the instrument. Therefore the calibration in Fig. 3 is more appropriate to the present study. Although the calibration is based on an indoor intercomparison there was remarkably good agreement between ventilated (undisturbed) indoor and outdoor concentrations during these periods[22] so it is reasonable to assume that the nature of the PM sampled by the Dustrak during the intercomparisons was representative of the outdoor PM. More recent indoor experiments have shown that the Dustrak to gravimetric PM_{10} calibration relationship remains the same, [PM_{10}] = Dustrak/2.1 (*cf.* eqn. (1)) for concentrations exceeding several thousand $\mu g\,m^{-3}$.[22] The latter study also shows that in comparisons where [PM_{10}] exceeds 50 $\mu g\,m^{-3}$ the intercept apparent in Fig. 3 is not significant. Overall, these data provide confidence in the Dustrak as a real-time PM_{10} sampler (subject to the appropriate calibration relationship) over several orders of magnitude of particulate mass concentration.

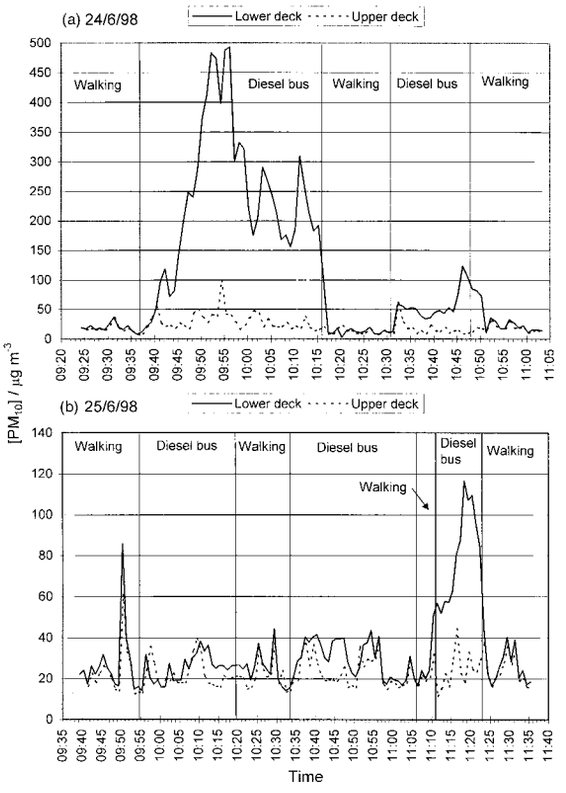

Fig. 7 Time series of simultaneous personal exposure measurements on the open upper and enclosed lower decks of an Edinburgh tour bus. Dustrak readings were converted to [PM_{10}] using the calibration relationships in Figs. 3 and 4.

The necessity for independent calibration of the Dustrak is demonstrated further by the comparison of two Dustrak instruments (Fig. 4). Although the measurements of the two instruments correlate well, the responses differ significantly. The systematic discrepancy may arise from variation in the factory calibration of the optical and electronic response, or in the volumetric sampling rate (again factory pre-calibrated), or in the exact sampling characteristic of the individual orifices.

Systematic difference is also apparent in the comparison of Minivol and PQ100/Graseby Andersen samplers. Despite the high degree of linear correlation through the origin, Minivol concentrations systematically exceed PQ100/Graseby Andersen concentrations by 19% on average (Fig. 5). The Minivol volumetric flow rate was 4.2% lower than expected when measured with a UK NAMAS-certified Gilian bubble flowmeter (after appropriate pressure and temperature corrections). The flow rate of the PQ100 mass flow controller was within the US EPA recommended $\pm 2\%$ tolerance. The systematically lower flow through the Minivol slightly increases the discrepancy shown in Fig. 5 for PM_{10} concentrations derived from the two samplers. This discrepancy probably reflects a difference in PM discrimination at the sampler inlets. Unlike the Graseby Andersen inlet, the Minivol inlet is not US EPA certified as a reference method for PM_{10} sampling. The efficiency of particle sampling by impaction is a function of volumetric flow rate, and the aerodynamic diameter below which 50% of particles impact increases as volumetric flow rate decreases.[23] Thus the Minivol inlet may have been sampling a PM distribution of greater median aerodynamic diameter than the Graseby Andersen inlet (and thus of a distribution of greater mass), although this factor is unlikely to account for all the systematic discrepancy.

This study demonstrates the importance of instrument intercomparisons to determine systematic error and the magnitude of random error for given devices. The uncertainty associated with measurement of $\sim 10\ \mu g\ m^{-3}$ ambient PM_{10} concentration in a 24 h period, are $\pm 20\%$ for the IOM and Casella personal samplers, $\pm 15\%$ for the Dustrak sampler, $\pm 5\%$ for the Minivol sampler, and $< \pm 5\%$ for the PQ100/ Graseby Andersen sampler. These error ranges exclude potential sources of systematic error such as flow rate and/or optical calibration. Flow rate calibration is required for accurate conversion of PM_{10} mass to volumetric concentration and to ensure that inlets sample the PM distribution specified. It is important that data derived from personal samplers deployed in an environmental, rather than occupational, context are interpreted within realistic bounds of uncertainty such as revealed in this study.

4.2. Exposure in outdoor microenvironments

Comparatively few published data exist on exposure to particles within vehicles, in contrast to investigations of in-vehicle exposure to other traffic-related pollutants such as carbon monoxide, oxides of nitrogen, benzene and volatile organic carbons.[24–27] Many of these studies report higher concentrations of pollutant inside the vehicle than outside, but whilst there is some evidence that inside concentrations correlate to engine type or surrounding traffic density, it is also evident that inherent vehicle-to-vehicle variability is a dominant factor. The exposure measurements reported here corroborate this for PM_{10}. The significant difference in concentration of PM_{10} between the two volunteers whilst driving but not whilst walking emphasises the intrinsic dependence of an individual's exposure to PM and their immediate microenvironment, and will reflect inherent differences between the volunteers' cars and their personal preferences for in-car ventilation. The contrast in exposure between two passengers sitting at different places on the same bus is particularly striking (Fig. 7). (*N.B.* the buses operate a no-smoking policy so there is no direct influence from environmental tobacco smoke.) Gee and Raper[28] have recently reported similarly widely variable concentrations of PM_4 in buses in the centre of the city of Manchester, UK. Concentrations were extremely variable ranging from less than $20\ \mu g\ m^{-3}$ to almost $1000\ \mu g\ m^{-3}$. Their average in-bus concentrations of $\sim 250\ \mu g\ m^{-3}$ were considerably higher than average background PM_{10} concentrations recorded at the static city-centre monitoring site, while average exposure of cyclists to PM_4 was not much greater than the average PM_{10} background concentration. Widely varying exposures to particle number from different microenvironment activity and modes of transport has also been reported recently by Brauer *et al.*[29] using an optical size-fractionated particle counter.

The variability in PM exposure is most likely due to two factors: (i) particle number and size distributions in exhaust emissions vary as much within as between engine-types for both diesel and gasoline engines;[30,31] (ii) air exchange rates for individual vehicles vary considerably (between 1 and $47.5\ h^{-1}$ for a stationary car[32]) depending on ventilation used.

The large peaks of very short-term exposure to PM observed in these experiments raise the very important issue of the appropriate timescale for assessing health effects of exposure to ambient PM. The current UK PM_{10} standard of $50\ \mu g\ m^{-3}$ applies to an averaging period of 24 h and has been set by consideration of a large number of epidemiological studies that used 24 h values as the PM_{10} metric.[33] Salvi *et al.*[34] have recently demonstrated marked systemic and pulmonary inflammatory responses in acute exposures of healthy volunteers to diesel exhaust for 1 h. Furthermore, it has been suggested that the major detrimental health impact of PM may be towards susceptible sub-groups of the population[35] but data are insufficient to address whether the appropriate timescale of exposure for these subgroups differs from the population as a whole.

Finally, it is inevitable that the composition of PM will differ between microenvironments (*e.g.* in-car *versus* indoors) and this may also have an impact on health outcome. There may well even be differences in PM composition between the upper and lower decks of the tour buses (Fig. 7) with contributions on the lower deck from poorly dispersed PM from furniture, clothing and human skin. More data on the spatial and temporal variability of personal exposure to PM are urgently required.

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