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Personal exposure monitoring of $PM_{2.5}$ in indoor and outdoor microenvironments

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HIGHLIGHTS

- Co-location studies with reference instruments confirm low-cost sensor viability.
- Functions to calculate indicative $PM_{2.5}$ mass from measured PNC have been developed.
- The method enables personal exposure monitoring across all main microenvironments.
- Personal measurements provide a wealth of data that needs to be integrated.

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ABSTRACT

Adverse health effects from exposure to air pollution are a global challenge and of widespread concern. Recent high ambient concentration episodes of air pollutants in European cities highlighted the dynamic nature of human exposure and the gaps in data and knowledge about exposure patterns. In order to support health impact assessment it is essential to develop a better understanding of individual exposure pathways in people's everyday lives by taking account of all environments in which people spend time. Here we describe the development, validation and results of an exposure method applied in a study conducted in Scotland.

A low-cost particle counter based on light-scattering technology — the Dylos 1700 was used. Its performance was validated in comparison with equivalent instruments (TEOM-FDMS) at two national monitoring network sites ($R^2 = 0.9$ at a rural background site, $R^2 = 0.7$ at an urban background site). This validation also provided two functions to convert measured PNCs into calculated particle mass concentrations for direct comparison of concentrations with equivalent monitoring instruments and air quality limit values.

This study also used contextual and time-based activity data to define six microenvironments (MEs) to assess everyday exposure of individuals to short-term $PM_{2.5}$ concentrations. The Dylos was combined with a GPS receiver to track movement and exposure of individuals across the MEs. Seventeen volunteers collected 35 profiles. Profiles may have a different overall duration and structure with respect to times spent in different MEs and activities undertaken. Results indicate that due to the substantial variability across and between MEs, it is essential to measure near-complete exposure pathways to allow for a comprehensive assessment of the exposure risk a person encounters on a daily basis. Taking into account the information gained through personal exposure measurements, this work demonstrates the added value of data generated by the application of low-cost monitors.

1. Introduction

Despite recent improvements in air quality in large parts of the world, poor air quality remains a challenge in many urban areas worldwide. The World Health Organization's (WHO) International Agency for Research on Cancer (IARC) has recently identified outdoor

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air pollution as a major cancer agent on a global scale (WHO, 2013a), indicating that health impacts due to exposure to air pollution are still of widespread concern.

Air pollution can affect the respiratory, cardiovascular, cardiopulmonary and reproductive systems and lead to cancer. Epidemiological evidence for these health effects is robust, even though there are still knowledge gaps regarding the exact mechanisms by which air pollutants affect human health (including the effects of pollutant mixtures), and which pollutants should be tackled with priority (EEA, 2013; EPA, 2012; Maudgalya et al., 2008; WHO, 2012, 2013a,b). Controlling air pollution not only directly reduces adverse health effects, but increases general well-being, quality of life, improves public health and can have positive impacts on ecosystem services.

Because of the complex relationships between humans and their environments it is necessary to integrate contextual factors such as environmental, socioeconomic and behavioural, into exposure assessment, which covers all aspects of estimating or measuring exposure to an agent. Investigating variations of individual exposure to pollutants of concern by age, gender, socioeconomic status, neighbourhood characteristics, activity level or ethnicity for instance requires new methods and tools.

Individuals also constantly move in time and space, while the (air) pollution landscape is spatially and temporally highly variable at the same time. This determines, to a large part, individual exposure to air pollution and personal monitoring needs to take account of this. Personal monitoring can provide detailed insight into a person's individual short-term exposure in a specified area. It is substantially different from how traditional methods generate population level exposure estimates, using fixed-site monitoring (FSM) networks and location of residence (Steinle et al., 2013). In addition, personal monitoring provides a more detailed picture of indoor air quality, which is important since people spend a large part of their time in indoor environments (Morawska et al., 2013).

There are wider implications of the shift in paradigm of air pollution monitoring based on a limited number of automatically recording instruments (which are required to prove equivalence to the gravimetric reference methods) at regulatory monitoring network sites to low-cost small and/or portable sensors applied in large quantities (Snyder et al., 2013).

Studies using FSM data often suffer from network sites not adequately representing spatial pollution patterns for personal exposure assessment. A study by Willocks et al. (2012), for instance, fails to identify associations between PM_{10} (particulate matter with an aerodynamic diameter <10 μm) concentrations and cardiovascular disease in Scotland using national air quality monitoring network and hospital admission data. One possible explanation for the difficulty in confirming association presented by the authors is the lack of statistical power due to limited data and day-to-day variation in the available data for both, the health and concentration time series. The authors therefore suggest an alternative cohort study design with more statistical power based on measurements of health and pollution exposure on the individual level. Such a study design also avoids misallocation of exposure while people spend time away from their residence (Setton et al., 2011).

As an alternative and complementary approach to FSM, personal monitoring solutions are emerging, as described by Mead et al. (2013) using monitors developed specifically for the purpose of personal or high density network monitoring. To achieve this, it is necessary to:

1. develop fit-for-purpose instruments and
2. produce them in a commercially-available form at a price appropriate for purchase of large numbers for representative studies.

However, the performance of these low-cost, wearable or portable sensors need to be adequately validated prior to their use in data collection and sharing on a large scale (Snyder et al., 2013).

The general feasibility of personal exposure assessment approaches (e.g. Cole-Hunter et al., 2012; Delgado-Saborit, 2012; Dons et al., 2011) and the utilisation of the large number of smart phones used worldwide for data collection (e.g. de Nazelle et al., 2013; Kingham et al., 2013) have been demonstrated successfully. Most studies apply a simulated study design and focus on certain exposure situations in a specific microenvironment (ME) (Steinle et al., 2013). One crucial issue in these studies is that they often do not take account of all exposure situations a person experiences in their day-to-day life. The reasons for not covering all exposure situations or MEs are varied (e.g. specific focus on sources, no availability of adequate monitoring devices), conclusions about the total exposure for the individual are therefore not possible. It is, however, important to consider not only the heterogeneity of individual exposure in a certain ME but also the diversity of MEs in a person's life. Steinle et al. (2013) introduced a new conceptual model for the assessment of exposure to air pollution which reflects this importance and explicitly takes *context* into account. Including context comprises aspects of a person's lifestyle (e.g. urban or rural environment, type of dwelling, shared accommodation, smoking habit) and specifically integrates them with spatiotemporal and pollution data. This approach can potentially provide that critical additional information which may directly affect individual and public health effects by modifying exposure, or more indirectly, by modifying behaviour, susceptibility and effect.

Following the rationale of this conceptual model the purpose of this paper is to describe the development and validation of a low-cost method to assess personal exposure that is user-friendly and allows a detailed insight into short-term variations of $PM_{2.5}$ (particulate matter with an aerodynamic diameter < 2.5 μm) across a wide-range of MEs. To do so it is necessary to validate the low-cost monitor against equivalent instruments to assess its performance in two different environments. Furthermore, it is necessary to provide a method to convert PNC into $PM_{2.5}$ mass concentrations which allows direct comparison with concentrations measured by equivalent instruments and can be directly related to relevant air quality limit values.

For this study a low-cost particle counter, the Dylos 1700 (Dylos Cooperation, Riverside, California, USA), was identified. In the following the instrument is referred to as the Dylos. This device, which has already been successfully applied in studies assessing second-hand smoke (SHS) exposure (Semple et al., 2012, 2013), has distinct advantages over other small and lightweight equipment typically used in occupational exposure assessments. The Dylos produces low, barely audible, noise levels and the ease of operation reduces the burden on the participants and enables volunteers to carry it without substantially interfering with their daily routine. The unit cost is only a fraction of commercially available particle monitors such as the TSI DustTrak or TSI Sidepak (TSI Inc., Shoreview, Minnesota, USA), which are approximately ten times more expensive, making it possible to source a large number of monitors required for simultaneous personal exposure studies. With the emergence of the concept of Citizen Science (Roy et al., 2012; Tweddle et al., 2012), the need to explore the performance and feasibility of sensors and study designs becomes even more relevant. As Snyder et al. (2013) describe, for many monitoring objectives including those related to Citizen Science, it is not critical to meet the same accuracy requirements of reference or equivalent instruments. Instead, the aim is to achieve known degrees of precision and to assess how the sensors and monitors perform. In this context, using large numbers of low-cost monitors conjointly with equivalent instrumentation can as well improve confidence in the measurements made.

The objectives of this study were to test a low-cost, user-friendly particle counter for short term exposure assessment. For this purpose, a low-cost particle counter and GPS receiver were applied in a variety of MEs. In addition, to validate the performance of the low-cost particle counter against equivalent instruments (in this case the Tapered Element Oscillating Microbalance Filter Dynamics Measurement System TEOM-FDMS (Thermo Fisher Scientific Inc., USA)), co-location experiments in two different environments (urban background and rural background) were conducted. Finally, an approach was developed based on the validation experiment to convert particle number counts (PNCs) into calculated $PM_{2.5}$ mass for direct comparison with equivalent instrument measurements and air quality limit values, to be used in subsequent exposure assessments.

2. Materials and methods

2.1. Study design and area

The study area is Scotland, a country with highly heterogeneous environments. The climate in Scotland is strongly influenced by the Gulf Stream and the prevailing westerly winds from the Atlantic.

Scotland's population was 5.3 million (estimated) on 30 June 2012 (GROS, 2013). The Central Belt, between the two most populous settlements of the country, Glasgow and Edinburgh, is densely populated with a high level of industrial, road and rail infrastructure. Large parts of the country are, however, rural and have comparatively lower population density and industry (Steinle et al., 2011).

Since this study has a focus on personal exposure monitoring, the definition of a suitable study area depends on where people are most likely to be moving during their monitoring period. This is mainly the area around the Scottish capital, Edinburgh which has a population of 482,640 (estimated population on 30 June 2012 (GROS, 2013)) and the adjacent council areas, East-, Mid- and West Lothian, where most of the volunteers participating in this study live and work. However, there are also profiles covering journeys reaching far up north to the Highlands and the Western Isles, as well as south across the border in to England.

2.2. Study participants

A non-representative group of study participants was recruited from the UK Natural Environment Research Council Centre for Ecology & Hydrology (NERC CEH) located in a rural environment in Midlothian, about 6 km south of the Edinburgh city boundary and 3 km from the town of Penicuik (ca. 15,000 inhabitants (GROS, 2014)). The volunteers all had different daily activity patterns to test the feasibility of the new study design and monitoring approach. This offers the opportunity to monitor exposure for a group of people sharing a common workplace from which their profiles radiate out in different directions to their residences and other activity spaces. A profile is a set of a data (ambient concentration, spatiotemporal information and contextual data) collected by an individual over a period of time, designed to capture everyday activities.

2.3. Portable monitoring solution

The Dylos was used to measure PNCs which were subsequently transformed into $PM_{2.5}$ mass concentration based on co-location experiments. It is a particle counter based on light-scattering technology and has been developed for indoor air quality monitoring for households. In contrast, many other commercially available air quality monitors have been developed primarily for industrial environments and occupational health monitoring applications. The Dylos has been used and evaluated in other studies, both indoors and outdoors, e.g. by Semple et al. (2012, 2013), in the context of exposure to SHS, and by Northcross et al. (2013) and Holstius et al. (2014) who compared the performance of the Dylos against other commercially-available particle monitors (both for chamber and ambient environments).

The Dylos outputs PNC per cubic foot (28.32 L) of air, ensuring constant air flow by a fan channelling air through the measurement chamber. It logs particles in two size classes (with 0.5–2.5 μm "small" and >2.5 μm "large" aerodynamic diameter). The lower particle size detection is stated by the manufacturer to be at 0.5 μm. The upper limit has been determined by Semple et al. (2012) in SHS chamber experiments to be at 65,356 particles per 0.01 cubic foot (when this level is reached the Dylos "rolls over" to zero and records erroneous values) which equates to an equivalent SHS $PM_{2.5}$ of about 1000 $\mu g/m^3$. This limit is unlikely to be exceeded in ambient conditions, unless in a smoking environment or in close proximity to biomass burning or in very heavily polluted traffic environments in large urban areas.

On a full battery charge, the Dylos runs for approximately 6 h. The built-in memory can store approximately one week of data when sampling continuously i.e. one log per minute. Once the memory is full, the Dylos continues to operate, but starts overwriting the oldest data. The Dylos is an easy to operate instrument with one button to switch it on/off and two more buttons to adjust settings. Its low weight (c. 500 g) and relatively small dimensions (12 × 20 × 8 cm) make carrying the instrument easy (Fig. 1). When the Dylos is running on continuous mode (using Firmware V2.5b2), no PNC readings are displayed, but a generic message ("logging data") is shown which has the advantage that volunteers cannot get distracted or adjust their behaviour to influence PNC readings.

A Global Positioning System (GPS) receiver to track the movement of study participants was used in combination with the Dylos to relate observed particle concentrations to time and location. The GPS Trackstick (Telespial Systems Inc., Burbank, California, USA) was selected for this study because of the small form factor (~10 × 3 × 2 cm), low weight (~82 g) and the ease of use with one button operation only (Fig. 1). The GPS Trackstick records date, time, longitude and latitude, altitude, temperature, status (speed), course (N, E, S, W), GPS fix and signal quality approximately every 10 s, depending on signal quality.

A small hiking backpack with elastic cord attachments and side pockets was adapted to carry the instruments (Fig. 1). To avoid accidental pressing of the buttons on the Dylos while it was strapped onto the backpack, a protective plastic cover was fitted over the buttons which still allowed users to switch the device on and off with a pen. To secure the monitor an adjustable Velcro strap was used. The Dylos could be recharged simply by plugging into the mains electricity supply without removal from the backpack. Four monitoring packs were assembled to allow for parallel data collection.

Fig. 1. The monitoring pack — the Dylos monitor is strapped onto the backpack with the inlet (red square) and fan exposed to the air. The GPS is placed in the side mesh pocket.

The design of the monitoring pack and the fact that the Dylos is not water- or splash-proof, yet needs to be worn exposed to ambient air, restricts its outdoor use to dry weather conditions or sheltered/indoor use only. The large surface area of the inlet (Fig. 1) does not allow for an external tube to be attached.

2.4. Contextual data

Detailed contextual and time–activity information was required to allocate time spent to the six MEs defined for this study and for use in time–activity diaries (TADs). Therefore a TAD was created as a web form accessible from any device with internet access. In addition, volunteers were provided with a 24-hour matrix on paper and were encouraged to take their own notes during the day and later transcribe their notes into the web form.

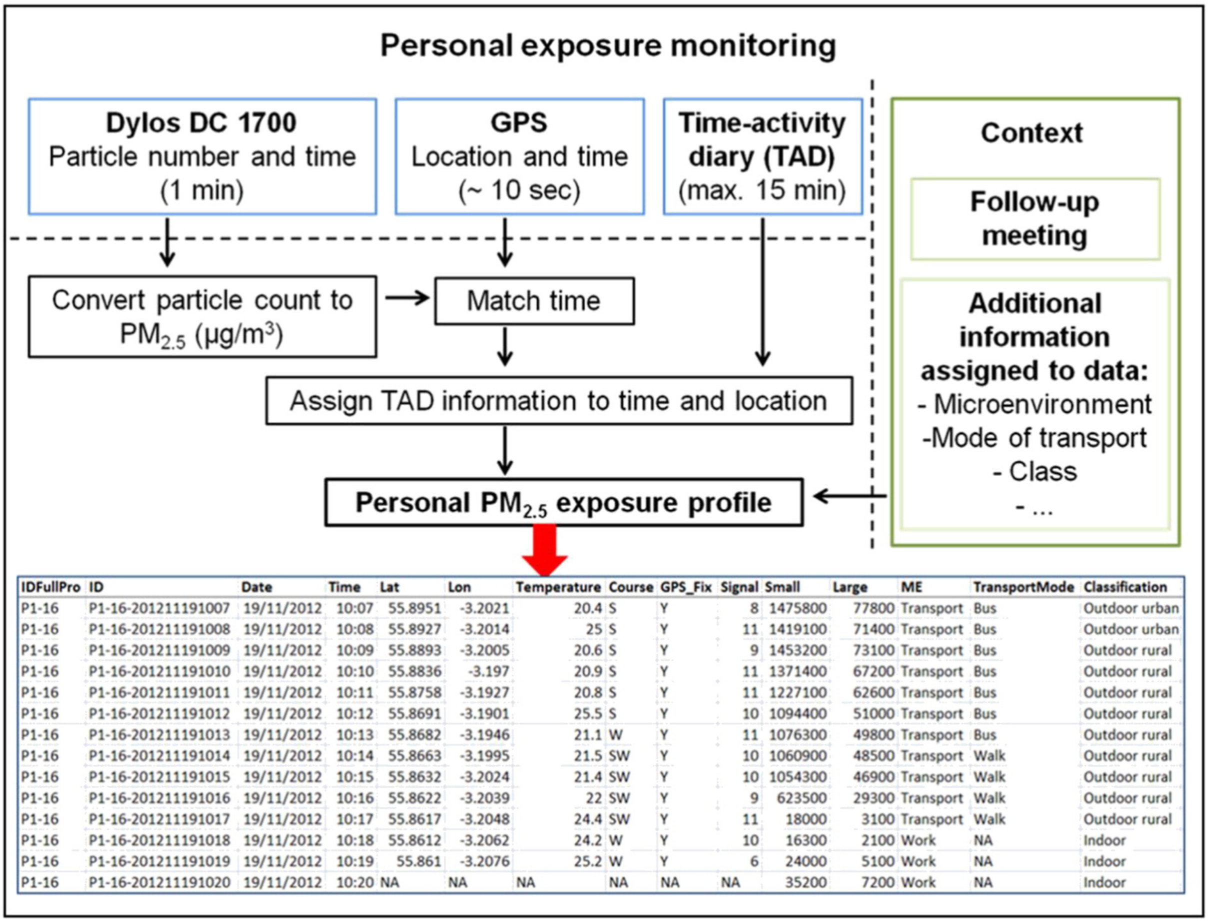
TAD and GPS data were reviewed after return of the monitoring pack and discussed in follow-up meetings with the study participants. Follow-up interviews were mandatory and conducted as informal meetings with the volunteer to talk through their personal data. In those meetings additional temporal and spatial details could be added and ambiguities or gaps in the TAD clarified. This proved to be a vital step as the TAD can only be fairly generic, necessitating the follow-up meetings to explore detailed issues afterwards. These meetings had to take place as soon as possible after the monitoring period while memories were still fresh.

A separate questionnaire was designed as a web form. Volunteers were asked to fill in this questionnaire once during this study. It included questions about the individual's living conditions, the household size and accommodation details, building and neighbourhood characteristics and other contextual factors.

2.5. Data collection, extraction and processing

Data was collected for two periods during November 2012 (P1) and during May 2013 (P2). In total, 17 volunteers collected 35 profiles, which covered a range of activities to highlight the variability of individual exposures.

Fig. 2 describes the process designed to ensure consistency in data analysis and provides an overview of the characteristics of the different datasets. Data processing methods have been developed and MEs allocated based on data collected during November, and then consistently applied to data collected in May. Differences in the logging time steps between the particle measurements (1 min) and the GPS log (ca.10 s) require careful processing of the data. To match the timestamps of both devices a method was developed utilising the Feature Manipulation Engine software (FME) (Safe software Inc., 2014) to match the GPS to the respective Dylos timestamps (at every full minute). The advantage of developing an FME workbench for this process was the self-documenting character of the software tool, and the high degree of re-usability and general applicability to all data collected. Automating the time-stamp matching process substantially reduced the processing effort for each set of data collected by the Dylos and the GPS receiver. Dylos logs without a matching GPS-originated timestamp i.e. indoor logs and where the GPS did not log due to reception problems or battery life, were kept in the dataset without location information.



IDFullPro	ID	Date	Time	Lat	Lon	Temperature	Course	GPS_Fix	Signal	Small	Large	ME	TransportMode	Classification
P1-16	P1-16-201211191007	19/11/2012	10:07	55.8951	-3.2021	20.4	S	Y	8	1475800	77800	Transport	Bus	Outdoor urban
P1-16	P1-16-201211191008	19/11/2012	10:08	55.8927	-3.2014	25	S	Y	11	1419100	71400	Transport	Bus	Outdoor urban
P1-16	P1-16-201211191009	19/11/2012	10:09	55.8893	-3.2005	20.6	S	Y	9	1453200	73100	Transport	Bus	Outdoor rural
P1-16	P1-16-201211191010	19/11/2012	10:10	55.8836	-3.197	20.9	S	Y	11	1371400	67200	Transport	Bus	Outdoor rural
P1-16	P1-16-201211191011	19/11/2012	10:11	55.8758	-3.1927	20.8	S	Y	11	1227100	62600	Transport	Bus	Outdoor rural
P1-16	P1-16-201211191012	19/11/2012	10:12	55.8691	-3.1901	25.5	S	Y	10	1094400	51000	Transport	Bus	Outdoor rural
P1-16	P1-16-201211191013	19/11/2012	10:13	55.8682	-3.1946	21.1	W	Y	11	1076300	49800	Transport	Bus	Outdoor rural
P1-16	P1-16-201211191014	19/11/2012	10:14	55.8663	-3.1995	21.5	SW	Y	10	1060900	48500	Transport	Walk	Outdoor rural
P1-16	P1-16-201211191015	19/11/2012	10:15	55.8632	-3.2024	21.4	SW	Y	10	1054300	46900	Transport	Walk	Outdoor rural
P1-16	P1-16-201211191016	19/11/2012	10:16	55.8622	-3.2039	22	SW	Y	9	623500	29300	Transport	Walk	Outdoor rural
P1-16	P1-16-201211191017	19/11/2012	10:17	55.8617	-3.2048	24.4	SW	Y	11	18000	3100	Transport	Walk	Outdoor rural
P1-16	P1-16-201211191018	19/11/2012	10:18	55.8612	-3.2062	24.2	W	Y	10	16300	2100	Work	NA	Indoor
P1-16	P1-16-201211191019	19/11/2012	10:19	55.861	-3.2076	25.2	W	Y	6	24000	5100	Work	NA	Indoor
P1-16	P1-16-201211191020	19/11/2012	10:20	NA	NA	NA	NA	NA	NA	35200	7200	Work	NA	Indoor

Fig. 2. Flowchart showing the data processing. Individual data sets are merged; additional contextual information is added to enrich personal exposure profiles and to review automatically logged location information. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The matching of "hard" GPS coordinates and timestamps with "soft" real locations and activities as noted in the TADs had to be reviewed manually to ensure that the correct activity was assigned to the logged location. The resulting new dataset, comprising GPS data, PNC and TAD information displayed as Excel graphs with TAD information added as text, was then discussed and confirmed with the volunteer in a follow-up meeting.

Based on the recorded data, additional contextual information such as MEs and classes was added (Fig. 2, green box). The information gained through the TADs and GPS receiver were the basis for developing different MEs. For this study, six key MEs were identified from this analysis (*Home*, *Work*, *Private residential building*, *Public building*, *Transport*, *Outdoor other*) into which all recorded locations were allocated to derive distinct time–activity patterns and exposure profiles.

Each individual data point was assigned to one of the three following classes that are representative of a coarse allocation of personal activity spaces. This classification allows for distinction between different characteristic background pollutant concentrations and the association of observed PNCs with calculated mass concentrations: *Indoor*, *Outdoor rural* and *Outdoor urban*.

Data where the Dylos was carried in backpacks, panniers or handbags or was left in cars or at home while the person was away has been discarded as non-valid, marked accordingly in the "comments" column and excluded from further analysis.

2.6. Equivalent monitoring methods

The Dylos has been applied by other researchers in different settings, both indoors (Semple et al., 2012, 2013) and outdoors (Northcross et al., 2013). In these contexts, it has been evaluated against the TSI SidePak and TSI DustTrak. These studies have confirmed a reliable representation of PNC by the Dylos over its particle size range. For the purpose of this study the Dylos has been applied in mixed indoor and outdoor settings. As this study has been conducted in both rural and urban conditions, it was necessary to validate the Dylos performance against equivalent instruments in both settings. In addition, this validation provided data for the generation of functions to transform PNCs into calculated particle mass concentration in μg/m^3 as typically used in air quality legislation and which is the standard output of equivalent monitoring network instruments.

The reference standard for measuring ambient $PM_{2.5}$ mass concentration is a manual gravimetric method (CEN, 2005) which in practice however, is not able to provide fast, continuous measurements as required for monitoring networks. Hence, the use of automatic instruments designed to provide equivalent results (equivalent instruments) is permitted by EU legislation (CEN, 2013) for the measurement of PM_{10} and $PM_{2.5}$ in a regulatory context (equivalence is defined according to the *Guide to the Demonstration of Equivalence*; European Commission, 2010). Such methods, however, introduce uncertainties to the already complex task of monitoring PM_{10} and $PM_{2.5}$ (Air Quality Expert Group, 2012).

The instrument used most commonly in the UK Automatic Urban and Rural Network (AURN) is the TEOM-FDMS. Its performance has been extensively evaluated in the equivalence programme for monitoring of PM in the UK (Bureau Veritas, 2010). The accurate measurement of PM is a demanding task and notoriously difficult because of factors such as semi-volatile compounds and variations in water-content (Thai et al., 2008). Substantial effort is required to ensure that output data is internally consistent and also comparable with the manual gravimetric reference method. The data ratification process considers values down to -4 μg/m^3 as valid. The TEOM-FDMS is based on complex technology, making it relatively maintenance-intensive and resulting in data gaps due to downtimes. Significant data rejection is not unusual and continuous measurements of PM_{10} and $PM_{2.5}$ remain a challenge (Air Quality Expert Group, 2012).

2.7. Validation approach

For the validation the Dylos was set up for five-day periods in close proximity to TEOM-FDMS instruments at one rural background and one urban background site of the AURN. At Edinburgh St. Leonards (urban), the Dylos was set up on a tripod directly adjacent to the cabin housing the TEOM-FDMS, with the inlet approximately 1 m above ground level, while the TEOM-FDMS inlet is located on top of the cabin at approximately 2.5 m above ground level. At Auchencorth Moss (rural), the Dylos was set up on ground level in a sheltered position next to the monitoring cabin due to the exposed nature of the site and the high winds during the collocation period. Due to the Dylos not being originally designed for outdoor use, the protection of the monitor from direct weather influence determined the location. The two $PM_{2.5}$ monitoring sites available in the study area around Edinburgh, Scotland are:

1. Auchencorth Moss (55.792160 N, −3.242900 W): The Co-operative Programme for Monitoring and Evaluation (EMEP) of the UNECE Convention on Long-range Transboundary Air Pollution Level II Supersite (Torseth et al., 2012) located in a rural environment (*rural background*) approximately 10 km south of Edinburgh on a transitional lowland peat bog.
2. Edinburgh St. Leonards (55.945589 N, −3.182186 W): This UK Automatic Urban monitoring station is located within a small park area in the south side of Edinburgh with the nearest main road being approximately 35 m away. The site is classified as *urban background*, which means that it is located in an urban area away from major sources, broadly representative of city-wide background conditions.

Full details on both sites, pollutants measured, measured data and statistics can be found on the UK-Air website (DEFRA, 2014).

At both sites the equivalent instrument operated was the TEOM-FDMS which was used for the comparison with the Dylos. For additional comparisons there was an *OSIRIS Airborne Particle Monitor* (Turnkey Instruments Ltd., UK) at St. Leonards and a MARGA — *Monitor for Aerosols & Gasses in Ambient Air* (Methrom Applikon B.V., Netherlands), which operates continuously, as part of the United Kingdom Department of Environment Farming and Rural Affairs (Defra) research contract (AQ0647) Eutrophying and Acidifying Pollutants (UKEAP) at Auchencorth Moss.

Each co-location experiment was conducted over a period of five days from 10th to the 15th of April 2013 (Auchencorth Moss) and 30th September to 4th October 2013 (St. Leonards). The duration was chosen to fit the maximum data storage capacity of the Dylos.

3. Validation experiment

3.1. Validating the performance of the Dylos against equivalent methods

The TEOM-FDMS instruments at both sites output data as hourly averages. Hence the Dylos observations were processed to calculate hourly averages from the data collected at 1-min resolution

Fig. 3 displays the scatter plots for the standard major axis regression of the Dylos and TEOM-FDMS hourly data at the two locations. The correlations between the Dylos and TEOM-FDMS at both monitoring sites are good ($R^2 = 0.9$ at Auchencorth Moss and $R^2 = 0.7$ at St. Leonards). A similar result (not displayed here) was found for the MARGA and the Dylos at Auchencorth Moss ($R^2 = 0.8$) in April, while the correlation between the Dylos and the OSIRIS at St. Leonards in October was less strong ($R^2 = 0.3$). The OSIRIS was reported before to underestimate $PM_{2.5}$ in comparison to gravimetric methods (Tasić et al., 2012) and on-going investigations into the OSIRIS dataset suggest that this device may suffer from interferences with relative humidity, which could explain the poor agreement between the OSIRIS and the Dylos.

The TEOM-FDMS data routinely undergoes quality control and has to be ratified before it is officially released to the public.

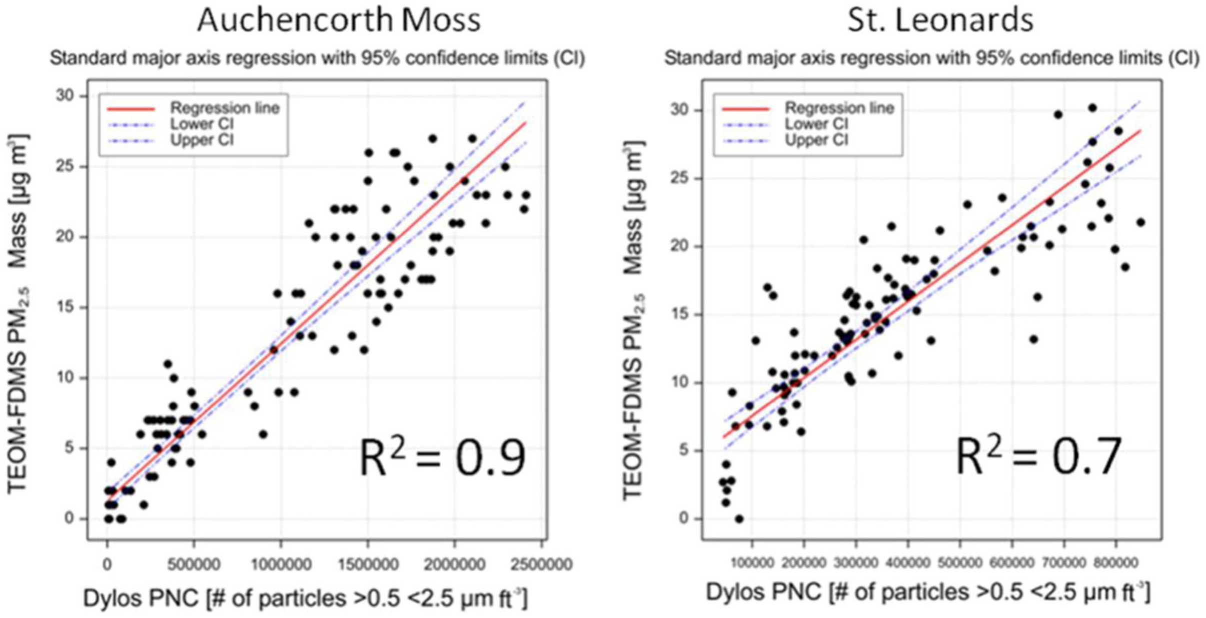


Fig. 3. Comparison of hourly-average PNCs (PNC, # ft^{-3} for particles between 0.5 and 2.5 μm) from the Dylos monitor and $PM_{2.5}$ particle mass (in μg/m^3) from the TEOM-FDMS instruments at Auchencorth Moss (left) and Edinburgh St. Leonards (right). The red line represents the equations from Table 1 used for conversion. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The Dylos performed well during its deployment at Auchencorth Moss, even though there was a substantial observed change in the composition of the $PM_{2.5}$, measured by the MARGA (Twigg et al., in preparation). Both the Dylos and the MARGA observed a decrease in $PM_{2.5}$, as the air masses changed from continental air containing secondary inorganic aerosols (SIA) to cleaner, sea salt dominated air masses originating over the Atlantic Ocean. The change in atmospheric composition appears not to have impacted the correlation between the Dylos and the MARGA, suggesting the Dylos not being sensitive to the composition of the aerosol.

3.2. Deriving calculated $PM_{2.5}$ mass concentration from PNCs

An additional result from the validation process was the development of two functions to derive $PM_{2.5}$ mass concentration from the PNCs monitored with the Dylos, following a similar approach documented by Semple et al. (2012, 2013). Air quality guidelines and health metrics typically refer to particle mass concentrations so it is helpful to provide data as calculated $PM_{2.5}$ mass concentrations.

Based on the good agreement between equivalent instrument (Fig. 6) and the Dylos, two distinct functions were developed to calculate $PM_{2.5}$ mass concentration from the measured PNCs. These functions were applied to all measurements made with the Dylos in outdoor environments. The functions were allocated according to the type of class the person has spent time in. Table 1 displays the functions derived by (a) Semple et al. (2013) which is applied to all *Indoor* environments, as well as the functions derived for (b) *Outdoor rural* (based on the data from Auchencorth Moss) and (c) *Outdoor urban* (based on the data from St. Leonards).

The indoor function was derived from Semple et al. (2013) from over 500,000 min of contemporaneously collected TSI SidePak and Dylos data from 34 smoking or non-smoking homes. Homes with open fire-places were excluded. The measurement instruments were placed in the main living area and data used for the production of a regression equation was randomly selected from the full dataset. The range of 1-minute values for the SHS study was much higher than the range for the outdoor samples in this study. This probably explains the need for a second order equation which holds across a $PM_{2.5}$ range of 0–1000 μg/m^3 while the equations provided in Table 1 for outdoor environments have only been validated over a range of 0–50 μg/m^3. For the outdoor concentration ranges, a linear function provided the best fit.

Fig. 4 displays the trends and absolute values of calculated $PM_{2.5}$ derived from the Dylos PNC and the observed TEOM-FDMS $PM_{2.5}$ data. As expected from using the functions developed based on the correlation of the two instruments, trends and peak timing and magnitude are well captured; however, the Dylos appears to have a higher cut-off concentration i.e. detection limit than the TEOM-FDMS.

In the first part of the data from Auchencorth Moss, the high values (for this site) influenced mainly by long-range transport show a lot of fluctuation which is picked up slightly differently by the Dylos compared to the TEOM-FDMS, while the difference becomes less pronounced towards the end of the co-location period, where local particle sources and sea salt dominated. The calculated Dylos data for St. Leonards shows differences which might be due to the nature of urban background aerosol composition, which is more directly affected by the proximity to highly variable emission sources. For both co-location experiments the inlets of the Dylos and TEOM-FDMS were at different heights which may also have an influence on the calculated $PM_{2.5}$ mass concentration.

In addition to applying both the urban and rural functions to the periods shown in Fig. 4, we compared results for other periods where Dylos instruments were co-located with TEOM-FDMS. For those periods, the calculated $PM_{2.5}$ mass based on Dylos PNC was well

Table 1
Functions to calculate $PM_{2.5}$ (in μg/m^3) from Dylos hourly-average PNC (in particles per cu ft).

Environment	Function	Source
(a) Indoor	$PM_{2.5} = 0.65 + 4.16 \times 10^{-5} \times [PNC] + 1.57 \times 10^{-11} \times [PNC]^2$	Semple et al. (2013)
(b) Outdoor rural	$PM_{2.5} = 1.29 + 1.11 \times 10^{-5} \times [PNC]$	This study
(c) Outdoor urban	$PM_{2.5} = 4.75 + 2.8 \times 10^{-5} \times [PNC]$	This study

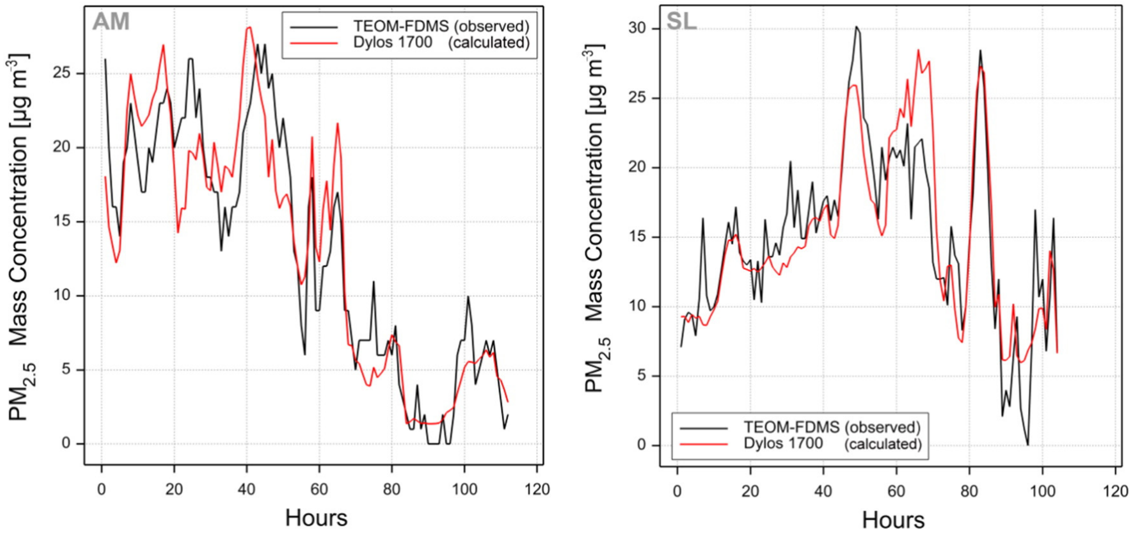


Fig. 4. Comparison of hourly-average $PM_{2.5}$ mass concentrations calculated from the Dylos' linear correlation function with TEOM-FDMS measured $PM_{2.5}$ mass concentrations in µg/m^3 for Auchencorth Moss (*Outdoor rural*, left) and Edinburgh St. Leonards (*Outdoor urban*, right).

comparable with TEOM-FDMS measured particle mass concentrations. For instance, at St. Leonards over a 48 hour period in November 2013, Dylos mean (max) concentrations were 6.2 µg/m^3 (12.2 µg/m^3) compared to the TEOM-FDMS mean being 4.0 µg/m^3 (10.0 µg/m^3). During this period, TEOM-FDMS recorded 5 values at zero µg/m^3 or below, while overall concentrations were rather low.

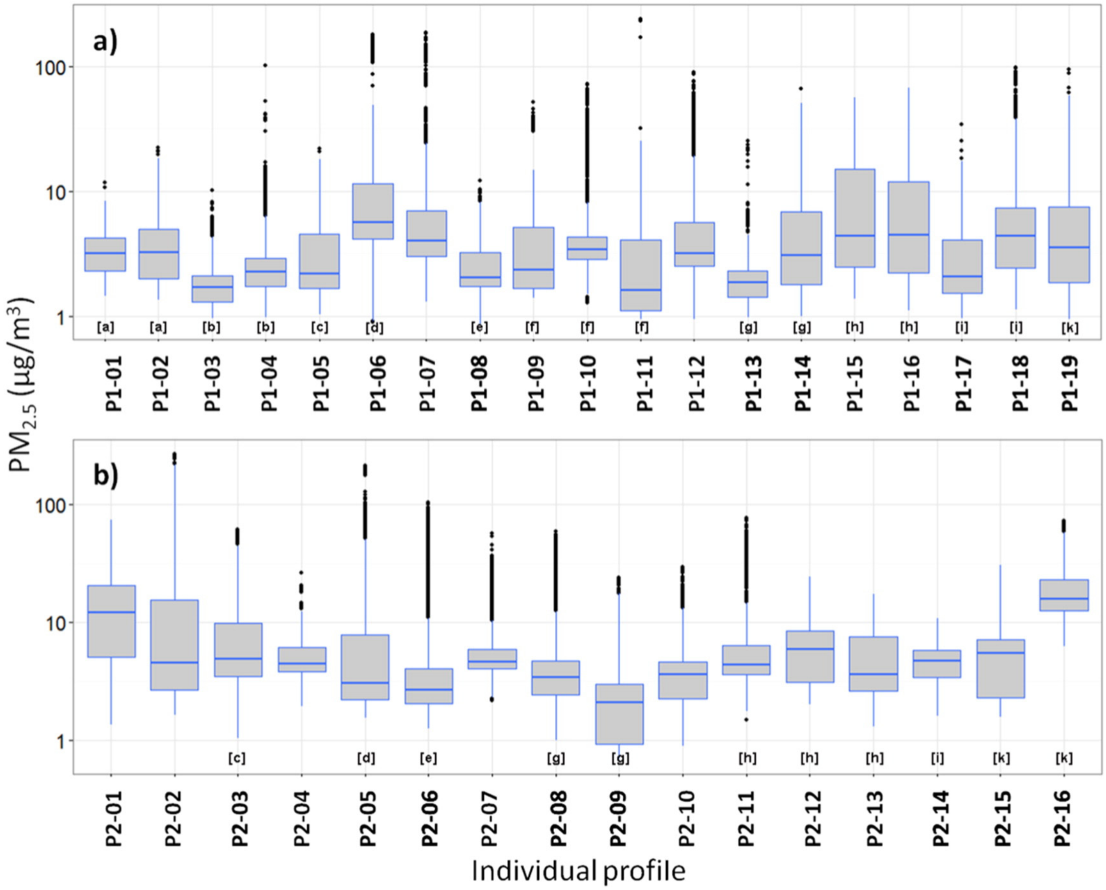


Fig. 5. (a) Calculated $PM_{2.5}$ (µg/m^3) derived for each individual profile from November 2012 (P1) and (b) for May 2013 (P2). Profile data from the same person during each period and between periods are denoted by [a]…[k]. Whisker plots show the 1st quartile (lower end of the box), the 3rd quartile (upper end of the box) and the 2nd quartile/median as blue horizontal line inside the box. The upper whisker extends to the highest value which is within 1.5 times of the inter quartile range (distance between the 1st and 3rd quartiles). The lower whisker to the lowest values respectively. Black dots represent outliers.

Table 2
All data collected within the 35 profiles summarised in average and standard deviation per ME in PNCs and calculated $PM_{2.5}$ mass concentration.

Microenvironment	n	$PM_{2.5}$ (µg m^{-3})		PNC (# ft^{-3})	
ME		Mean	sd	Mean	sd
Home	59,539	8.4	17.3	186,444	415,535
Outdoor other	2157	6.2	6.9	147,444	201,028
Private residential building	2237	10.2	15.2	228,898	366,312
Public building	7468	6.3	8.4	135,632	202,852
Transport	7224	7.0	6.0	151,428	202,423
Work	14,868	3.0	2.2	55,687	52,194

4. Results

All results in the following figures and tables are shown as $PM_{2.5}$ mass concentrations calculated using the functions in Table 1. The *Indoor* function was applied to the MEs *Home*, *Work*, *Public* and *Private residential building*. The functions *Outdoor rural/urban* were applied to time spent in *Outdoor other* or *Transport*.

4.1. Concentration characteristics P1 (November) and P2 (May)

The box and whisker plots in Fig. 5 illustrate the variability of calculated $PM_{2.5}$ concentrations between the individual profiles arising from places visited and activities done by the individual volunteers. Mean concentrations of $PM_{2.5}$ for each profile in the first phase of data (Fig. 5a) collection vary between 1.9 and 10.6 µg/m^3. For the second phase (Fig. 5b) mean concentrations per profile range between 2.5 and 29.6 µg/m^3.

The variability between individual profiles (Fig. 5) is also reflected in the summary statistics for all profiles collected across the six MEs reported in Table 2. Mean and standard deviation (STD) highlight the differences between MEs but also the range of data within each ME. *Work*, which is an office building in a rural environment, has the lowest mean values. The low STD reflects the homogeneity of the data collected in this ME. *Home* and *Private residential buildings* on contrary do not only have the highest mean values but also highest STDs reflecting the variety of activities and sources occurring in those MEs that resulted in concentrations/PNCs over a large range of values.

4.2. Example profiles P1-03 and P1-05

The two profiles shown in Fig. 6 are an example from the dataset collected in November illustrating how concentration, spatiotemporal and contextual data are integrated to analyse an individual exposure profile.

Concentrations are generally low for both profiles in Fig. 6, although a different, individual pattern can be distinguished between the two profiles. In profile P1-03, a maximum of 8.2 µg/m^3 was recorded, whereas for profile P1-05 a maximum of 15.9 µg/m^3 was recorded.

The data gaps in P1-03 (Fig. 6a) in the profile were due to rain when the person was cycling to and from work as well as walking to a destination in the evening. The increase of concentration observed at P1-03 at the *Work* place at ~11:00 can be explained as the result of staff returning to the office after being in a meeting. Ferro et al. (2004) for instance identified indoor sources and human activities to cause events of high concentration but short duration. The short-term increase (concentrations are still <10 µg/m^3) at the beginning and end of the visit to the *Public building* ME are likely due to the volunteer and other people arriving, moving about and getting ready to leave.

Profile P1-05 (Fig. 6b) shows higher levels in the *Home* and *Transport* MEs. The increase at *Home* at 06:00 might be caused by the volunteer's cat moving in the vicinity of the monitor. *Work* is characterised by low concentrations (<2 µg/m^3). This person is situated in an open-plan

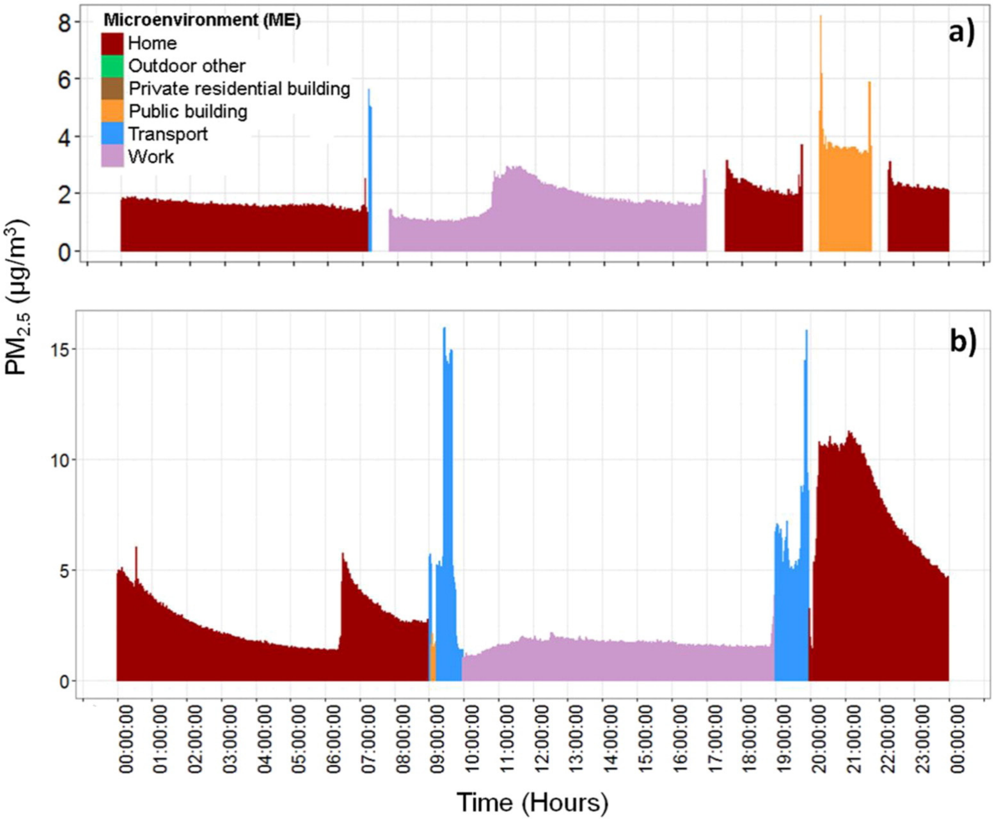


Fig. 6. a) Profile P1-03 showing calculated $PM_{2.5}$ at 1-minute resolution on the 13th of November 2012. b) Profile P1-05 showing calculated $PM_{2.5}$ at 1-minute resolution on the 13th November 2012. The colours indicate which of the defined MEs the person was at which point in time. Note the difference in scale on the y-axis. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

office with people frequently coming and going. Movements in the open plan office do not, however, appear to be picked up by the Dylos which was placed at the person's desk in the far corner of the room, away from the door. In the evening (20:00–21:30) cooking and cleaning increased levels at *Home* to just over 10 μg/m^3 for a short period, slowly decreasing again over night.

Fig. 7 shows the GPS track for P1-05 illustrating the spatial variability of personal exposure. In the morning the person walked to the bus stop, briefly calling in at a post office. The second part of the commute was by bus (western track). Here the highest concentrations of 14 μg/m^3 were recorded during the bus journey (orange dots). The return journey in the evening followed a different route (tracks in east and north) and was done by car, bus and finally by walking. When walking along a main road the highest values of this journey (12–14 μg/m^3) were observed (yellow and orange dots).

Fig. 8 illustrates how the personal exposure measurements using the Dylos (aggregated to hourly average values) compare with the hourly measurements from Edinburgh St. Leonards and Auchencorth Moss for the same time period (profile P1-05) as shown in Fig. 6b. The Dylos shows distinct concentration peaks in the morning and the evening, during which the person spent time in *Transport*, while Edinburgh St Leonards only shows the evening peak. This graph also illustrates the negative concentration values in the ratified data from the TEOM-FDMS instruments, which occur regularly in this dataset during a period of generally low ambient concentrations. Data gaps in this case originate from data having been rejected in the quality control process.

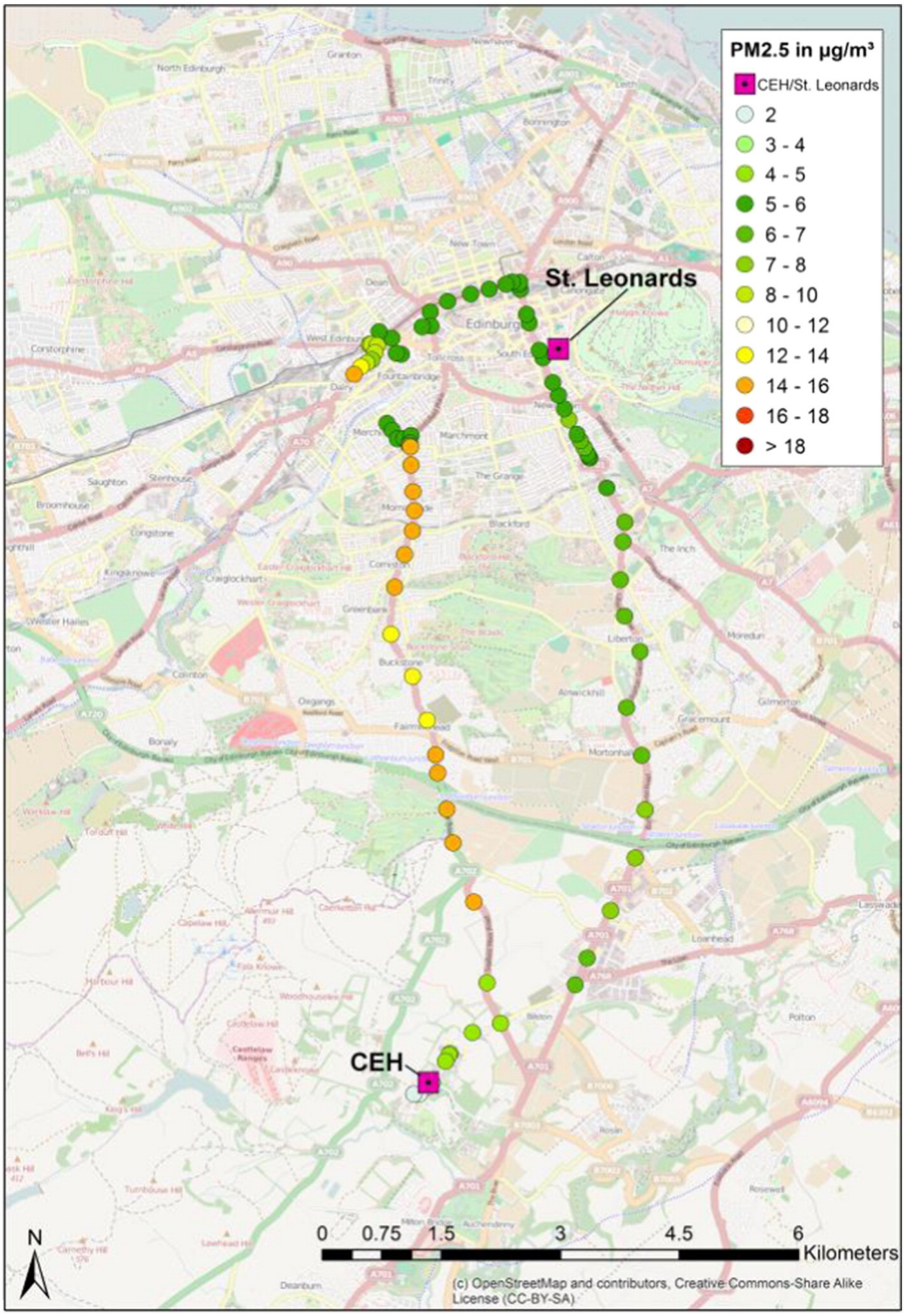


Fig. 7. GPS tracks of P1-05 showing the calculated PM$_{2.5}$ concentration (μg/m^3) logged once per minute. Note that only non-confidential GPS points are shown, and indoor MEs which generally do not have a GPS log are missing. Also shown are the locations of the urban background fixed site station St. Leonards and the location of CEH, the *Work* ME. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

5. Discussion

This study applies a new approach of monitoring personal exposure to PM$_{2.5}$ in a variety of MEs over the course of several days. The novelty of this approach lies in the fact that measurements are taken across the full heterogeneity of places visited and activities conducted to gain as much insight as possible into an individual's total exposure. This is important as individuals are constantly on the move and follow their own activity patterns, which determine their individual exposure. Results for individual profile (Fig. 5) but also the summarised data for all profiles (Table 2) demonstrate the variability of concentrations measured over the course of several days and across the range of the six defined MEs. Looking at profiles in more detail (Figs. 6 and 7) gives an idea of the many different activities and situations (summarised into a respective ME) that influence the personal levels of exposure. Analogous to the concept of the exposome (Wild, 2005, 2012), it is vital to take account of this variability and monitor pollution concentrations in as many situations as possible, providing a comprehensive snapshot of a person's daily exposure to PM$_{2.5}$.

The explorative approach applied in this study has been evaluated with respect to its feasibility for personal monitoring and demonstrates issues with monitoring in certain environments due as much to practicality reasons as to the actual design of the tools and devices. All measurement methods however, operate with a certain degree of generalisation and are not able to replicate all aspects of every actual exposure situation.

The Dylos is not specifically designed for non-stationary measurements and its form factor, albeit lightweight and small, could be improved to achieve a less intrusive design and reduce burden for the volunteers. The benefits of using the Dylos include its portability and low-cost, as well as its good agreement with equivalent instruments as demonstrated in this study. While it is not an equivalent instrument and also cannot easily be worn within the breathing zone of the subject, it is suitable for mobile and stationary measurements in the direct vicinity of the subject adding spatial and temporal detail to routine measurements. Time, logistics and costs restrict personal monitoring approaches often to pilot studies; the application and validation of low cost monitoring solutions is thus crucial to up-scale to large-scale studies.

The personal exposure data shown here presents a snapshot of a person's lifelong exposure and thus is not comparable to a person's total exposure. Measurements, such as those presented in this paper, could, however, have significant value as educational tool to raise awareness of people's activities and habits and the resulting potential exposures. More importantly, they can inform and provide data for personal exposure modelling approaches, which naturally complement personal exposure studies and allow for a generalisation of results to larger populations.

To our knowledge, this is the first study to validate the performance of the Dylos in two different outdoor environments against equivalent instruments of the UK national automatic monitoring network for PM$_{2.5}$. The validation experiments in urban and rural Scotland demonstrate the viability of using the Dylos monitor as a low-cost alternative to other commercially available instruments for exposure studies in different MEs. The Dylos well reproduced temporal trends in concentrations when compared to TEOM-FDMS in both outdoor rural and urban environments. One possible caveat of the approach presented here is that the validation is limited to two 5-day periods over comparatively low concentration ranges. The validation experiment demonstrated that the Dylos provides a robust representation of relative changes in particulate matter concentrations. However, for future validation and to extend the use of transfer functions from PNC to PM mass to other environmental conditions (e.g. different transport modes) further co-location experiments will need to be conducted over longer periods of time and at different locations.

Our approach also allows for the distinction between urban and rural environments by applying respective functions to transform PNCs into particle mass, accounting for the different particulate matter components typically found in these environments. This approach adds a level of detail to the personal monitoring approach similar to the principle of having urban and rural national network stations providing detailed insight into different environmental compartments.

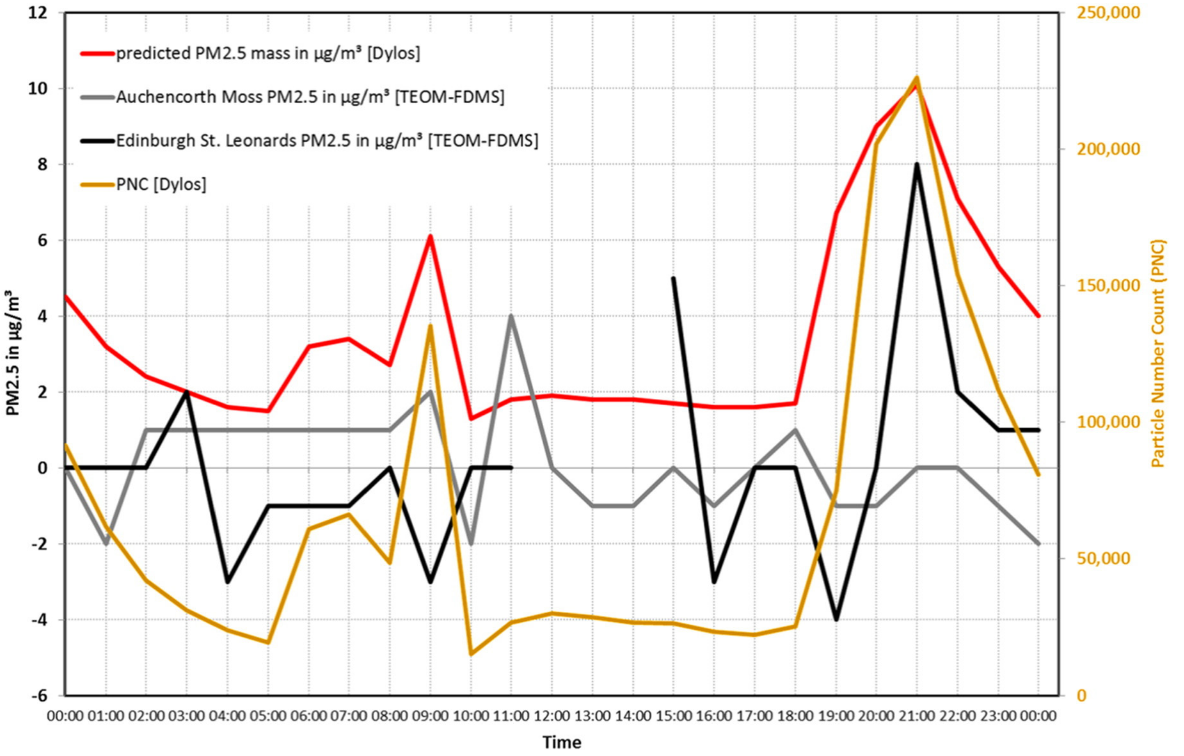


Fig. 8. Hourly PNCs (PNC, # ft^{-3}) measured for profile P1-05 (see Fig. 6b), calculated PM$_{2.5}$ mass concentrations based on Dylos PNC and PM$_{2.5}$ mass TEOM-FDMS from both AURN monitoring sites for the 13th of November 2012.

While the focus is currently on PM mass concentrations, the emerging concern about ultrafine particles (UFP) in exposure research may create a need for other metrics to assess ambient PM levels. The development of exposure–response functions based for instance on particle numbers or other related metrics may require rethinking current (particle mass based) air quality limit values and enable the direct use of PNC as a viable indicator for exposure and, ultimately, health risk.

6. Conclusions

This pilot study has demonstrated that personal exposure monitoring is a viable method for improving knowledge about individual level exposure to environmental stressors. As a methodology it is grounded on a compromise between instrument precision and information content and is restricted by feasibility and privacy issues.

By exploring the feasibility of the method in everyday situations and across the full heterogeneity of microenvironments this study has taken a step in this direction. Furthermore it has shown that the application of a low-cost monitoring solution in combination with other available monitoring data and assessment methods provides reliable exposure information. However, it should be kept in mind that using any low-cost air pollution monitors does not claim to deliver the same precision as reference or equivalent methods for measuring PM, but rather offers a low-cost solution to provide an indication of exposure to particulate matter with improved spatial and temporal variability.

The potential for a more comprehensive integration, e.g. including additional datasets such as modelled or measured meteorological and pollution data into the analysis can increase the scope and strength of this integrated approach. While the degree of detail of the information gained by personal monitoring is very high, the scope is so far limited by small sample size and limited spatial coverage. Using the approach elaborated in this paper for Citizen Science applications could increase both the quality and quantity of data collected and thus improve the characterisation of exposure patterns. This would further benefit the development of exposure models, in particular supporting the development of up-scaling parameters to larger populations and inform the design of representative personal exposure studies.

Further research needs to focus on improvements regarding devices and, in conjunction, study design with the aim to allow for a seamless coverage of all MEs and activities, while reducing the burden to study participants.

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