An approach to predict population exposure to ambient air $PM_{2.5}$ concentrations and its dependence on population activity for the megacity London

Vikas Singh[a*#], Ranjeet S Sokhi[b] and Jaakko Kukkonen[c]

a. National Atmospheric Research Laboratory, Gadanki, Andhra Pradesh, INDIA - 517112

b. Centre for Atmospheric and Climate Physics Research (CACP), University of Hertfordshire College Lane, Hatfield, AL10 9AB, UK

c. Finnish Meteorological Institute, Erik Palmenin aukio 1, P.O.Box 503, FI-00101, Helsinki, Finland

*Corresponding author

#Previously at the University of Hertfordshire, UK

Keywords: Particulate matter, population movement, infiltration, OSCAR, microenvironment, population-weighted concentration

ABSTRACT

A comprehensive modelling approach has been developed to predict population exposure to the ambient air $PM_{2.5}$ concentrations in different microenvironments in London. The modelling approach integrates air pollution dispersion and exposure assessment, including treatment of the locations and time activity of the population in three microenvironments, namely, residential, work and transport, based on national demographic information. The approach also includes differences between urban centre and suburban areas of London by taking account of the population movements and the infiltration of $PM_{2.5}$ from outdoor to indoor. The approach is tested comprehensively by modelling ambient air concentrations of $PM_{2.5}$ at street scale for the year 2008, including both regional and urban contributions. Model analysis of the exposure in the three microenvironments shows that most of the total exposure, 85%, occurred at home and work microenvironments and 15% in the transport microenvironment. However, the annual population weighted mean (PWM) concentrations of $PM_{2.5}$ for London in transport microenvironments were almost twice as high (corresponding to 13-20 $\mu g/m^3$) as those for home and work environments (7-12 $\mu g/m^3$). Analysis has shown that the PWM $PM_{2.5}$ concentrations in central London were almost 20% higher than in the surrounding suburban areas. Moreover, the population exposure in the central London per unit area was almost three times higher than that in suburban regions. The exposure resulting from all activities, including outdoor to indoor infiltration, was about 20% higher, when compared with the corresponding value obtained assuming inside home exposure for all times. The exposure assessment methodology used in this study predicted approximately over one quarter (-28%) lower population exposure, compared with using simply outdoor concentrations at residential locations. An important implication of this study is that for estimating population exposure, one needs to consider the population movements, and the infiltration of pollution from outdoors to indoors.

Analysis, based on a modelling approach, demonstrates that it is critical to consider both population movements in key microenvironments and the infiltration of pollution from outdoors to indoors for calculating the total exposure due to the ambient $PM_{2.5}$

1. INTRODUCTION

Most epidemiological studies focusing on health impacts of air pollution are based on relationships between measured pollution concentrations at fixed monitoring sites, or modelled concentrations, and various health indicators (e.g., Pope and Dockery, 2006; Rohr and Wyzga. 2012, de Hoogh et al., 2014). However, such approaches ignore the activity patterns of individuals, i.e., people's day-to-day movements from one location to another and, the infiltration of outdoor air to indoor. Both factors are known to cause significant variations in the predicted exposure (e.g., Beckx et al., 2009; Soares et al., 2014; Kukkonen et al., 2016).

Variations in the individual exposure during the daily activity have been studied by measuring the personal exposure to ambient air concentrations using portable instruments in different microenvironments (Wallace and Ott, 2011, Steinle et al., 2013, 2015; Williams and Knibbs, 2016; Ham et al., 2017; Carvalho et al., 2018). As the studies were based on measurements over relatively short periods, they do not account for the day-to-day and seasonal variations in the exposure to ambient pollutants. To account for the temporal variability, earlier studies (Dockery et al., 1993), estimated the population exposure based on the measured concentrations at the nearest monitoring site, which was then assumed to represent the pollution levels over a fairly wide area. Other studies (Bell, 2006; Brauer et al., 2008) used the concentrations measured at several monitoring sites, to spatially interpolate the pollutant concentrations using inverse distance weighting (IDW) and kriging techniques (Singh et al., 2011) to estimate the exposure. However, such methods do not capture the finer scale spatial heterogeneity in the air pollution across the city. The concentrations of pollutants in urban areas are highly heterogeneous and may vary by an order of magnitude on street scale in different areas due to traffic-originated pollution (e.g., Beevers et al., 2013; Singh et al., 2014; Pattinson et al., 2014; Targino et al., 2016).

Exposure models can vary from simple empirical relationships between health outcomes and outdoor air concentrations up to comprehensive deterministic exposure models (e.g. Kousa et al., 2002; Ashmore and Dimitripoulou, 2009; Soares et al., 2014; Smith et al., 2016). A more refined procedure combines the spatially predicted concentrations, and location and activity of the population, to estimate the spatial and temporal variation of mean exposure in different MEs (e.g., Soares et al., 2014; Kukkonen et al., 2016, Smith et al., 2016). This is particularly important, as accurate exposure estimates are necessary to reliably quantify population health impacts.

Geographical Information Systems based approaches have been used by Jensen (1999) and Gulliver and Briggs (2005) to estimate the exposure from traffic. Considerably more sophisticated Eulerian gridded chemical transport models have been used globally (Lelieveld et al., 2015, Picornell et al., 2019) and at regional scale (Isakov et al., 2007; Borrego et al., 2009; Beckx et al., 2009; Conibear et al., 2018) to estimate the exposure at different grid resolutions. The city scale dispersion models (Carruthers et al., 2000; Sokhi et al., 2008; Singh et al., 2014) and land use regression models (Beelen et al., 2010; Gulliver et al., 2011 and de Hoogh et al., 2014) provide the within-city variations in the concentrations. There are, however, fundamental differences in approach adopted by such methods in terms of the methodology to estimate the concentrations. While dispersion models use a deterministic approach to estimate the pollutant concentrations based on the spatially resolved emissions and meteorology driven dispersion, land use regression models predict the pollutants based on empirical relations between measured pollutant concentrations at a number sites and predictor variables, such as land use, traffic and topography (Beelen et al, 2013; Korek et al., 2016).

Probabilistic models such as EXPOLIS (Hänninen et al., 2003, 2005) and INDAIR (Dimitroulopoulou et al., 2006) provide the frequency distribution of exposure within a population. In order to estimate the spatial distribution of mean exposure, an integrated deterministic modelling approach such as EXPAND (Exposure model for Particulate matter And Nitrogen oxiDes; Soares et al., 2014; Kukkonen et al., 2016) and LHEM (London Hybrid Exposure Model; Smith et al., 2016) has been adopted. These models can be applied for various temporal and urban spatial domains based on the available temporal and spatial resolution of population activity and emission data.

With a population of over 8 million in accordance with the 2011 census (ONS, 2012), London is one of the largest cities in the Europe. It serves as an ideal study area, as comprehensive datasets on emissions, air pollutant concentrations and population are available. A few London specific urban high-resolution (from tens of m to a few hundreds of m) dispersion modelling studies have been reported (Beevers et al., 2013, Singh et al., 2014 and Hood et al., 2018). Singh et al., (2014) and Beevers et al., (2013) evaluated dispersion models against annual mean $PM_{2.5}$ measurements and both reported that the regional background was on the average the largest contributor to the total $PM_{2.5}$ concentration. Near busy roads, however, the levels of $PM_{2.5}$ due to vehicular emissions were of similar magnitude as the regional background.

Examining how air pollution distributions are influenced by population activities within a complex urban environment, such as London, it is essential to understand exposure to air pollution. Picornell et al., (2019) highlighted the importance of people’s movements for calculating the exposure using population movement based on mobile phone data. Reis et al., (2018) evaluated the influence of population mobility on exposure in the whole of the UK at a resolution of 1 km×1 km. They reported that taking workday location into account had only a minor influence (0.3%) on the predicted exposure to $PM_{2.5}$, compared with considering simply the residential exposure. However, they did not address the outdoor to indoor infiltration of pollution. The minor effect probably reflects not allowing for the infiltration effects and the fairly coarse resolution.

GLA (2013) provides ambient air PM2.5 concentrations for population weighted exposure calculations over London, but does not allow for different human activities or infiltration of air pollution to indoors. Kaur and Nieuwenhuijsen (2009) and studies of Adams et al. (2001a and 2001b) have examined the personal exposure in London based on the field measurements, including a limited amount of samples. The use of dispersion model combined with space-time-activity data allows the calculation of exposure in detail.

A detailed study by Smith et al., (2016) combines the outdoor pollution concentrations evaluated by the CMAQ-Urban model and space-time-activity data based upon London Travel Demand Survey (LTDS) to estimate the exposure of the Greater London population to the outdoor air concentrations of $PM_{2.5}$ and NO_2 using the LHEM model. They calculated the population average daily exposure in indoor, in-vehicle and outdoor microenvironments and their contribution to the total exposure. Smith et al., (2016) to a large extent focused on the examination of the differences of the exposure values evaluated by the LHEM model, compared with the exposures computed at residential addresses. They also investigated the differences of exposure to $PM_{2.5}$ and NO_2. The present study, in contrast to Smith et al. (2016), also analyzes in detail predicted spatial concentration distribution and population weighted concentrations for $PM_{2.5}$ in main microenvironments (home, work and transport). We considered it important also to investigate the impacts of the spatial heterogeneity of the population and $PM_{2.5}$ concentrations over the whole of London.

In this study, we have extended the previously published development and application of the OSCAR Air Quality Modelling System, which is mainly based on a multiple-source Gaussian dispersion approach. The OSCAR modelled concentrations of $PM_{2.5}$ have been combined with the estimates of the regional background concentrations and population activity based on census data reported by the Office for National Statistics (ONS) in the UK, (ONS, 2012) and population activity from the London Travel Demand Survey (LTDS, 2011 from Transport for London), to predict the population exposure to ambient air concentrations of $PM_{2.5}$ across a megacity of London, UK.

The objectives of this study were to:

(i) Develop and implement a comprehensive approach to analyse and estimate the time activity of the population of London for three microenvironments (home, work and transport);
(ii) Quantify the population exposure to the concentrations of $PM_{2.5}$ in London;
(iii) Examine the relative importance of exposure to ambient $PM_{2.5}$ in terms of key microenvironments, their spatial distributions across Greater London and quantify the difference between central London and surrounding regions; and
(iv) Assess the importance of including the movements of the populations and the infiltration of ambient air pollution indoors to the total exposure of the population, compared, e.g., with using solely the exposure predicted at residential locations.

In order to achieve the research objectives, we have estimated the concentrations and the time-activities of the population, and combined these datasets to examine the exposure of the whole population in London to outdoor concentrations of $PM_{2.5}$. In line with the first objective, we demonstrate a robust methodology that can be applied to quantify spatially resolved population exposures due to air pollution in cities such as London for any time period, without the reliance on excessively detailed population activity data.

2. METHODOLOGY

We present an overview of the methodology, including the modelling of the PM2.5 concentrations and exposure. In addition, we explain the selection and definitions of the microenvironments, and present the data and methods for the assessment of the locations and movements of the population.

2.1 Modelling of the $PM_{2.5}$ concentration in London for 2008

We have used the OSCAR Air Quality Assessment system (Singh et al., 2014; Sokhi et al., 2008) to model the $PM_{2.5}$ concentrations originated from vehicular urban sources in London (Supplementary Figure S1). A detailed description of the modelling domain, road traffic data and model validation can be found in Singh et al., (2014). The OSCAR Air Quality Assessment System consists of an emission model, a meteorological pre-processing model and a road network Gaussian dispersion model (Kukkonen et al., 2001).

The OSCAR modelled concentrations of $PM_{2.5}$ have been combined with the estimates of the regional and urban background concentrations. The annual mean regional and urban background concentrations of PM2.5 at 1 km × 1 km grid resolution were extracted from Grice et al. (2009). The regional and urban background concentration was added to the modelled concentrations originating from the urban vehicular sources by linear interpolation using a geographic information system (GIS). The temporal variability in the annual mean regional and urban background concentrations was derived using the measured hourly time series from a representative urban background station at Camden – Bloomsbury.

The emission model of the OSCAR system is based on the COPERT IV (Gkatzoflias et al., 2012) and Department for Transport (DfT; Boulter et al., 2009) emission functions and factors as used in London Atmospheric Emission Inventory (LAEI¸ GLA, 2010). The $PM_{2.5}$ non-exhaust emissions due to tyre and brake wear were based on the UK National Atmospheric Emission Inventory (NAEI; Dore et al., 2008). The particle resuspension has not been considered because of its relatively small contribution compared with tyre and brake wear (Beevers et al., 2013). Although the OSCAR model does not include a detailed treatment of traffic congestion on emissions, the effects of congestion are allowed for on an average level, via the influence of vehicle travel speed on emissions.

The meteorological pre-processor GAMMA-MET (Bualert, 2002) was used to process the hourly parameters including wind speed and direction, solar radiation, friction, velocity, temperature, relative humidity and Monin–Obukhov length. The influence of buildings and other obstacles on the dispersion was represented using the roughness length (z_0) (see Seinfeld and Pandis, 2006). Roughness length value equal to 1.5m was used for the central London and a lower value of 0.2m was used for open road environments located in outer London. It should be noted that, in order to retain efficient and reasonable computation run times, complex street canyons were not treated within OSCAR. This may potentially lead to an underestimation of $PM_{2.5}$ concentrations as street canyons would typically reduce dispersion. The model includes dry deposition process for the fine particulate matter originating from the line source (Kukkonen et al., 2001); this has been allowed for in the modelling. However, the chemical transformation processes were not taken into account in the urban scale modelling. Therefore, the particles originating from the urban traffic sources were treated mainly as primary particles, although regional and urban background concentrations used in the model included contributions from secondary particles.

For the sake of brevity, we have not presented any further details on the model and its evaluation against experimental data. For more detailed descriptions, the readers are referred to Singh et al. (2014), Sokhi et al. (2008) and Srimath et al. (2005, 2017).

2.2 Evaluation of population exposure

2.2.1 Definitions of exposure and population weighted concentration

The time averaged population exposure E_i at a given location i (or a computational grid square) and for a given time period t, can be written as (Soares et al., 2014; Reis et al., 2018).

$$E_i = \sum_{j=1}^{N} \sum_{t=1}^{24} C_{ijt} P_{ijt} \qquad (1),$$

where C_{ijt} and P_{ijt} are the pollutant concentration and the number of persons at the location i and microenvironment j at a time period of the day t, and N is the number of the considered microenvironments. Clearly, equation (1) can be defined correspondingly for hourly, daily or annual as in the current case. The use of equation (1) also allows for the modelling of exposure in various microenvironments (MEs), including peoples' movements and the evaluation of outdoor pollution in indoor air.

It is also useful to define a population weighted mean (PWM) concentration to which the population is exposed in different environments. For a time period of 24 hours, this can be defined as (Reis et al., 2018):

$$C_i = \frac{\sum_{j=1}^{N}\sum_{t=1}^{24} C_{ijt}P_{ijt}}{\sum_{t=1}^{24} P_{it}} \qquad (2),$$

where the denominator is the cumulative amount of population within location *i* during 24 hours period. In this current study, we have presented numerical results on the population exposure and population weighted concentration values as annual averages.

2.2.2 Microenvironments

ME is a useful concept when considering movement of people and their resultant exposure to air pollution. It is defined as a location having relatively uniform concentration, such as home or workplace, in which exposure takes place. Three MEs have been considered in this study, namely, home, work and transport. One could also define other, more specific microenvironments. For instance, Soares et al. (2014) considered a microenvironment called 'other environments' that included exposure in recreational activities, such as sports activities, shopping and restaurants. The microenvironments considered in this study are as follows

(i) The home microenvironment includes all the people at home or working at home.
(ii) The work microenvironment includes all the people at workplace. We have assumed, for simplicity, that all the people are working either in offices or inside buildings.
(iii) The transport microenvironment includes exposure of people while travelling in buses, personal cars, trains, pedestrians and cyclists and hence includes all the people travelling by all modes of transport (supplementary Figure S2) to homes, work or to any other location.

As mentioned previously, this study considers only exposure to outdoor air pollution; the effects of indoor air pollution sources in London (Shrubsole et al., 2012) were outside the scope of this study. The infiltration of outdoor air pollutants indoors is dependent on numerous factors, such as, e.g., the structure and ventilation systems of the building, on the particular pollutant, and in case of particulate matter, on its size distribution. As the information on the infiltration coefficients for various buildings and vehicles in London was very scarce, we have used estimates from available literature (Hänninen et al., 2004 and 2011). Further discussion is given in section 2.3.3.

2.3 Evaluation of the location and time-activity of the population

We have analysed the amounts of population at home, at work and in transport microenvironments within London for 2008. The analysis was based on the census population data reported by Office for National Statistics (ONS, 2012). The diurnal variation of population activity has been obtained from the London Travel Demand Survey LTDS (2011). Instead of having individual activity pattern based on the individual trips such as analysed by Smith et al., (2016), our study calculates the population space-time activity that has been estimated by combining the information extracted from ONS (2012) and LTDS (2011). The population space-time activity provides the information on the number of people in a given microenvironment at given time of the day at the census location. This approach allows a population based analysis, as most of the cities have residential as well as work population records based on the census survey.

2.3.1 London population data

The spatial distribution of the London population (supplementary Figure S3) has been taken from the ONS census data. Census of the population is conducted every 10 years in the UK. In census 2011, the data was collected from the 95% household based on the questionnaire that provided the detailed information on the residential and work population. We used the population at the output areas (OA, Census Glossary, 2011) that is the highest available geographical resolution for population allocation published over for all the districts of Greater London. The area of OA is different which varies from 156 m^2 to 12.2 km^2. The median area of OA across London is 0.033 km^2.

The census population information has been reported for the years 2001 and 2011; we have therefore extrapolated the values in 2008, by assuming a linear growth rate of the total population in London from 2001 to 2011. The numbers of residential and workday population were evaluated to be 7.86 and 8.37 million in London in 2008, respectively. The workday population is larger than the resident population due to the population commuting from the outside of London. The growth rates of resident and workday populations were on average 1.39% and 1.33% per annum, during the decade from 2001 to 2011.

The spatial distribution of population during daytime (defined as the period from 7:00 am to 7:00 pm) and nighttime (the other times) have been presented in supplementary Figure S3 (a,b). All the spatial distributions in this study have been presented at the output areas (OA, Census Glossary, 2011) for all the districts of Greater London. As expected, the population density during daytime is clearly higher in central London and in the vicinity of the busiest business districts. The population at night is distributed much more uniformly across the whole area of London.

The percentages of the modes of travel from home to work in London based on ONS (2012) have been presented in supplementary Figure S2. Public transport includes buses, trains and the underground. Public transport accounts for approximately a half of all transportation from home to work. Other vehicular modes of travel, such as private car, taxi and motorcycles are responsible for almost a third of all travels. A fairly small fraction of people, 13% walk or cycle to work.

2.3.2 The London Travel Demand Survey (LTDS) data

LTDS is a continuous household survey of the London area, covering the Greater London area, assessed based on the travel demand. LTDS collects the information on households, people, trips and vehicles. The diurnal and weekly variation of the fractions of people travelling in London obtained from LTDS has been presented in supplementary Figure S4. Clearly, during the weekdays, there are substantial morning and afternoon rush hours peaks.

During the weekends, the amount of people travelling peaks at approximately 11 am, and then slowly decreases at later times of the day.

The percentages of population in the selected microenvironments are presented in Figure 1. Based on ONS (2012) and LTDS (2011) datasets, more than half of the population spent their time at home throughout the day. The data shows that the time spent in both work and in transport environments is distributed fairly evenly during the working hours. However, as expected, there are higher activities associated with the transport environment during the morning and afternoon rush hours.

2.3.3 Outdoor to indoor infiltration

While indoor sources and sinks were not considered, the contribution of outdoor air pollution to indoor air quality was determined by the use of the efficiency of infiltration, which takes account of outdoor air coming indoors and the ventilation. The infiltration factor is defined to be equal to the fraction of outdoor air pollution that will be infiltrated indoors (e.g., Soares et al., 2014). In this study a mean value of the infiltration factor for $PM_{2.5}$ of 0.60 has been used, based on Hänninen et al., (2004 and 2011), to calculate the concentrations at home and at work microenvironments. Hänninen et al. (2011) presented an overview of a number of European studies that have determined IF's for $PM_{2.5}$ and PM_{10}; the values in the overview ranged from 0.37 to 0.70. Soares et al. (2014) presented an update of part of these values. We have selected the value of 0.60, based on averages of the extensive datasets within these updates, based on the EXPOLIS and ULTRA studies.

Smith et al (2016) have used spatially resolved infiltration factors within a range of from 0.35 to 0.86; however, these have been derived only for domestic buildings. It is not clear, whether these values are representative of commercial areas of London, including the centre.

In the case of the transport microenvironment, the information on the infiltration factors for various modes of transport is not known sufficiently well for a detailed modelling analysis. The mean values and the range of the $PM_{2.5}$ concentrations reported within traffic microenvironments by Smith et al., (2016) suggest that the concentrations within traffic micro-environments are in the range of the ambient concentration, except for the underground environment. The infiltration factor used in this study for all the various transport microenvironments, for all transport modes is therefore assumed to be unity.

3. RESULTS AND DISCUSSION

3.1 Temporal and spatial distribution of PWM concentrations and exposures for different microenvironments

A diurnal variation of the modelled annual average $PM_{2.5}$ concentrations has been presented in supplementary Figure S5. The diurnal profile shows a bimodal distribution. The two broad highs are due to increased urban traffic in the morning, approximately from 7 to 9 am and again in the evening, approximately from 7 to 9 pm. In general, the day time concentrations are higher by 3-4 μg/m^3, as compared with the values at night. The overall $PM_{2.5}$ diurnal profile is of course a resultant of the variations in the emissions as well as meteorology (e.g. changes in boundary layer height) over the day and night hours.

The spatial distribution of the modelled annual mean $PM_{2.5}$ concentrations for 2008 has been previously presented by Singh et al. (2014). The highest concentrations were found near busy roads, motorways, at their junctions, and in the centre of London. For this study, the modelled spatial distributions of PWM concentrations of $PM_{2.5}$ have been presented in Figure 2 for homes and workplaces, transport and for the total of all the microenvironments. All the results have been presented for exposure to outdoor air pollution, including infiltration of outdoor air pollution to indoors.

Across London, our analysis shows that people at workplace and home are exposed to the annual average concentrations ranging from 7 to 11 μg/m^3 of $PM_{2.5}$ with mean value of 8 μg/m^3. However, people in the transport microenvironment are exposed to relatively much higher concentrations, the annual averages ranging from 13 to 20 μg/m^3 with a mean value of 15 μg/m^3.

The analysis of population weighted concentrations has been extended for city-wide mean values. PWM concentrations of $PM_{2.5}$ in the different MEs are presented in Figure 3. People are exposed on average to almost twice as high concentration in the transport microenvironment, compared with the home and workplace environments. However, the total PWM concentration in all the considered MEs is only slightly higher than the corresponding average value in the home and work MEs, due to the large fraction of time that people spend in the home and work environments.

The predicted spatial distribution of population exposures has been presented in Figure 4 for homes and workplaces, transport and for the all combined microenvironments exposure for London in 2008. The highest exposures occurred in the central areas of London, for both the total exposure and for both work and home, and the transport microenvironments. The largest proportion of exposure (85%) takes place at homes and workplaces microenvironments as much of the population spend large amount of the time indoors.

3.2 Spatial difference in concentrations and exposure for central and outer areas of London

A separate analysis has been conducted to understand the differences in exposure for central and outer parts of London. The central parts include Westminster, City of London, Kensington and Chelsea as shown in red colour in supplementary Figure S1 and the remaining area is referred as outer London. The central parts of London have high day time population, due to the working population (supplementary Figure S3). Figure 5 presents PWM concentrations and exposures for Greater London divided into a central part and an outer part. The PWM concentration of $PM_{2.5}$ averaged over the central part of London is 20% higher than the corresponding average concentration in the outer parts of London. However, the population exposure is almost three times higher in central London, compared to outer London. The higher concentrations in central London are mainly caused by traffic originated air pollution. Reis et al., (2018) estimated around 8% differences in annual mean concentrations for $PM_{2.5}$ experienced by individuals living at Mayfair in central London, compared with living at Southfields in outer London. The lower estimate by Reis et al., (2018), compared with the corresponding values in the present study, could be due the exclusion of exposure in the transport microenvironments.

The urban and traffic concentration increment calculations for London by Singh et al., (2014) showed that a major fraction of the total $PM_{2.5}$ concentrations, 73%, was caused by regional background contributions, 19% by urban non-road sources and 8% by the emissions originated by road transport. These percentages provide useful information on the importance of these source categories on a city-wide scale but as indicated above there are spatial differences such as between the central part compared to the outer areas of London.

3.3 Importance of including population activity to quantify exposure to $PM_{2.5}$

The predicted diurnal variations of the population exposure, both including and excluding population activity have been plotted in Figure 6. The figure presents exposures in all the microenvironments, allowing for the influence of the infiltration of outdoor air indoors. The exposure excluding activity has been computed by assuming people spend their time only in the residential (indoor home) environment (but including infiltration effects from outdoor air pollution). The more realistic exposure with activity is where people also spend their time in transport and work environments and hence results in substantially higher population exposure values (by about 20%). This is especially the case during the day time, with higher values during the morning and evening commuting periods. Such a comparison clearly illustrates the importance of increased exposure due to taking account of population activity patterns compared to assuming a static residential population.

3.4 Implication of this study for air pollution exposure and health impact assessments

Air pollutant concentrations at residential locations of the population are commonly used in health impact assessments and epidemiological studies. The implicit assumption in these studies is that the residential exposure is representative of the total exposure of the target population or cohort members. However, this study questions this assumption by showing that exposures in various microenvironments are not the same. As this bias has been present in almost all of the previous larger scale exposure and health assessment studies, it is useful at least to know the magnitude of this uncertainty.

We have, therefore, evaluated the difference of using only residential coordinates in estimating the total population exposure, compared with using the exposure evaluated separately for the three microenvironments addressed in this study. The exposure assessment methodology used in this study predicted over one quarter (-28%) lower total population exposure, compared with using simply outdoor concentrations at residential locations.

The difference between exposure based on the use of static population exposed to residential ambient concentration and the exposure for a dynamic population moving within three microenvironments is mainly caused by two counteracting factors. (i) The so-called residential exposure in traditional health impact assessments is evaluated based on the assumption that the general population exposure is reflected by the air pollutant concentrations outside the vicinity of their homes. In the present study, we have also allowed for the infiltration effect of the houses and work buildings. The resulting modelled exposure of people indoors affected by a fraction of ambient air pollution that is infiltrated indoors, and the actual exposure inside the homes, is therefore smaller. We have evaluated this exposure reduction to be of the order of 40% (with an infiltration factor assumed to be equal to 0.60) (ii) The exposure in road transport environments is substantially higher than the corresponding exposures at homes. The exposure at workplaces also tends to be slightly higher than that at homes per unit of time (Soares et al, 2014), as the former are more commonly situated near roads with heavier traffic. The resulting predicted exposure is, hence expected to be higher for other microenvironments besides homes. These two factors counterbalance each other to some extent.

It can be shown by simple numerical evaluations that the first mentioned effect (i) is larger than the second effect (ii). The resulting percentage change of the predicted exposure mentioned above (-28%) is therefore negative, but its absolute value is smaller than the above mentioned 40%. The results of detailed computations for the traditional method and the more refined one, both evaluated using the modelling system used in this study, are presented in Figure 7. The population exposure, taking into account all three microenvironments and the infiltration of pollution indoors, was 72% of the corresponding result obtained with the traditional method. The corresponding percentage was slightly lower, 67%, for the population weighted mean concentration, compared with the population exposure.

This percentage has been evaluated for London for 2008; the overall reduction will probably be different for other urban regions and time periods. In particular, the exposure of people spending time near heavy traffic roads, as is the case in central London, will result to higher exposure compared to using residential concentrations.

There is an important implication for exposure and health assessments (e.g., epidemiological studies identifying links between air pollution and health outcomes). The analysis of exposure in this study demonstrates to the importance of taking into account the exposure in various microenvironments and the infiltration of pollution to indoors, instead of using only the residential exposures.

3.5. Underlying assumptions and limitations

The scope of current study has included the exposure to ambient air pollution, both outdoors and indoors; however, we have not considered indoor sources and sinks of air pollution. Various European studies have reported infiltration factors for $PM_{2.5}$ and PM_{10} that range from 0.37 to 0.70 (Hänninen et al., 2011), i.e., substantial fractions of outdoor particulate pollution can be infiltrated to indoor air. The pollution infiltrated from outdoor to indoor air in the western and central European countries may therefore be more important for peoples' health than pollution from the indoor sources, with the exception of tobacco smokers.

We have considered emissions from road transport and most other urban sources; however, we have not addressed contributions from trains. In particular, the metro (underground) microenvironments are outside the scope of this study. The emission modelling allows for the effects of traffic congestion only implicitly, i.e., as a variation of exhaust coefficients as a function of travel speed.

The infiltration factor for the transport microenvironments has been assumed to be unity, due substantial uncertainties of the ranges of these values (Smith et al., 2016). While our analysis was done for 2008, the main findings are relevant also to the present situations, as the temporal changes in $PM_{2.5}$ concentrations for London have been modest during the last decade (e.g., Brook and King, 2017, Font and Fuller., 2016). This study has considered exposure to $PM_{2.5}$, due to its association with serious health impacts (e.g., Rohr and Wyzga. 2012). To retain the focus on exposure, we have not examined health impacts.

In addition, we have not considered explicitly the chemical components of $PM_{2.5}$, or other pollutants, such as NO_2 and O_3, and their resulting health impacts. The eventual goal is to evaluate the exposure and health impacts of all the relevant pollutants, and those for the various chemical components and different properties of particulate matter. However, it is important to understand the spatial and temporal distribution of population exposure to $PM_{2.5}$, before examining the contribution from its chemical constituents. Important questions still remain on how exposure from $PM_{2.5}$ affects the population spatially and in different key microenvironments.

4. CONCLUSIONS

High resolution $PM_{2.5}$ predictions from the OSCAR Air Quality Assessment model for London have been combined with demographic datasets to determine spatial distribution of population exposure for three different microenvironments (home, work and transport). The exposure model includes a treatment of the locations and time use of population and a simple treatment of the infiltration of pollution from outdoor to indoor air. This comprehensive modelling approach has been used to analyse the time activity dependent population exposure for more than eight million inhabitants of London megacity. The annual population exposure to ambient air $PM_{2.5}$ concentrations has been estimated based on hourly time-activities at fine scale for the whole of Greater London.

Numerical results have been presented for time activities, PWM concentrations and the population exposures to $PM_{2.5}$. The computations included the regionally and long-range transported pollution with contributions originating from all urban pollution source categories, including especially those related to vehicular emissions. A number of key conclusions can be drawn from the study.

(i) We have demonstrated the development and applicability of the OSCAR modelling approach for predicting population exposure to $PM_{2.5}$ for a megacity, London, UK. The approach combines high resolution, spatially and temporally resolved concentrations of ambient $PM_{2.5}$ with data on time activity for three main microenvironments. As this approach does not rely on excessively detailed information, it can be utilized for evaluating the impacts of urban and traffic planning and for conducting assessment of adverse health impacts resulting from air pollution exposure as well as for urban air quality research.

(ii) Our analysis shows that on an annual average level, more than half of the population of London is at home throughout the day. The time spent in both work and in transport microenvironments is distributed fairly evenly during the working hours, although expectedly, there were higher activities in the transport ME during the morning and afternoon rush hours. A similar variation of the population activities has been reported by Kousa et al. (2002) and Soares et al. (2014) for the Helsinki Metropolitan Area.

(iii) In terms of microenvironments, people at work and home were exposed to concentrations ranging from 7 to 11 $\mu g/m^3$ of $PM_{2.5}$ on an annual average level, whereas people in transport, were exposed to almost twice as high concentrations, the annual averages ranging from 13 to 20 $\mu g/m^3$.

(iv) Analysis on a city-wide basis in terms of the individual ME and the total population exposures to $PM_{2.5}$ reveals that 85% of the total exposure occurred at home and workplace microenvironments, and 15% in the transport microenvironment. Smith et al., (2014) found in their study that travel was responsible from 4 to 12% of the total population exposure.

(v) There is a distinct demarcation of exposure for people spending time in central London compared to other regions. Comparison of the spatial distribution shows that the highest exposures per unit area occurred in the centre of London and in the area of urban business centres. This is the case for both the total exposure and for both work and home, and the transport microenvironments. In terms of population weighted concentration of $PM_{2.5}$, the value averaged over the central part of London is 20% higher than the corresponding average concentration in the outer parts of London. Because of higher $PM_{2.5}$ concentrations due to higher traffic density and high population density, the population exposure per unit area is almost three times higher in central London, compared to outer London.

(vi) The total exposure resulting from all the considered activities, including the outdoor to indoor infiltration compared with indoor home exposure only (inside the homes, considering the infiltration of $PM_{2.5}$ from outdoors to indoors) resulted in about 20% higher exposure to $PM_{2.5}$. This analysis illustrates the importance of allowing for population activity.

(vii) There are important implications also for air quality and health related epidemiological studies that assume that the air pollutant concentrations outside the home place are representative of the total population exposure. These studies also commonly neglect the infiltration of pollutants to indoors. This study shows that the exposures to ambient concentrations of $PM_{2.5}$ can be substantially different in different microenvironments. Results from the current work demonstrate that the total population exposure was over one quarter (-28%) lower on a city-wide average level, compared with using simply outdoor concentrations at residential locations. Smith et al., (2016) have also shown that exposure estimates based on space-time activity and infiltration of $PM_{2.5}$ to indoors is lower; they found a 37% lower value, compared to the outdoor exposure evaluated at residential addresses. However, this proportion will be different for other urban regions and time periods, or when addressing specific population sub-groups. For pollutants that are more dominated by local urban sources (such as, e.g., NO_2), this difference in using only residential exposure could be substantially higher, compared with the corresponding difference in case of $PM_{2.5}$ (Kukkonen et al, 2016).

In exposure and health assessments, therefore, it is important to allow for the movements of the population and for the infiltration of ambient air pollution indoors. The epidemiological studies commonly use outdoor concentrations in the residential areas or at home addresses. The use of more dynamic exposure data in epidemiological studies in the future could substantially improve the accuracy of health impact assessments.

5. AVAILABILITY OF EXECUTABLE MODEL PROGRAM

The executable programs and the datasets used are available as part of a collaboration agreement upon request from the authors.

6. ACKNOWLEDGEMENTS

This work was supported by EU FP7 project TRANSPHORM (Grant agreement: 243406). We also wish to thank for funding of Nordforsk for the project "Understanding the link between Air pollution and Distribution of related Health Impacts and Welfare in the Nordic countries (NordicWelfAir)", and the Academy of Finland to project "Global health risks related to atmospheric composition and weather (GLORIA)".

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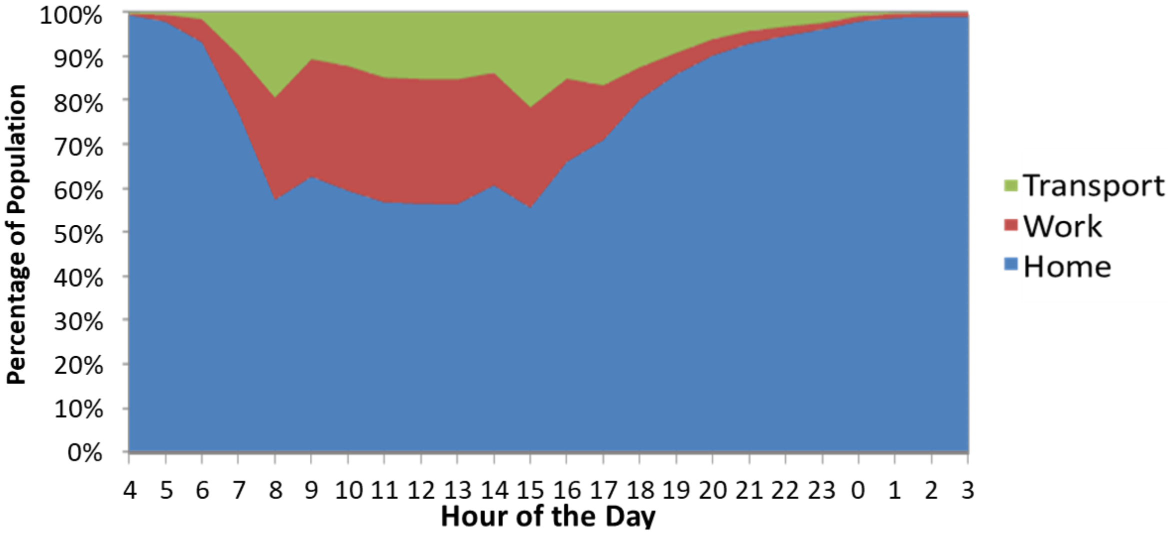

Figure 1. The diurnal variation of the activity of the population in London in three microenvironments in 2008.

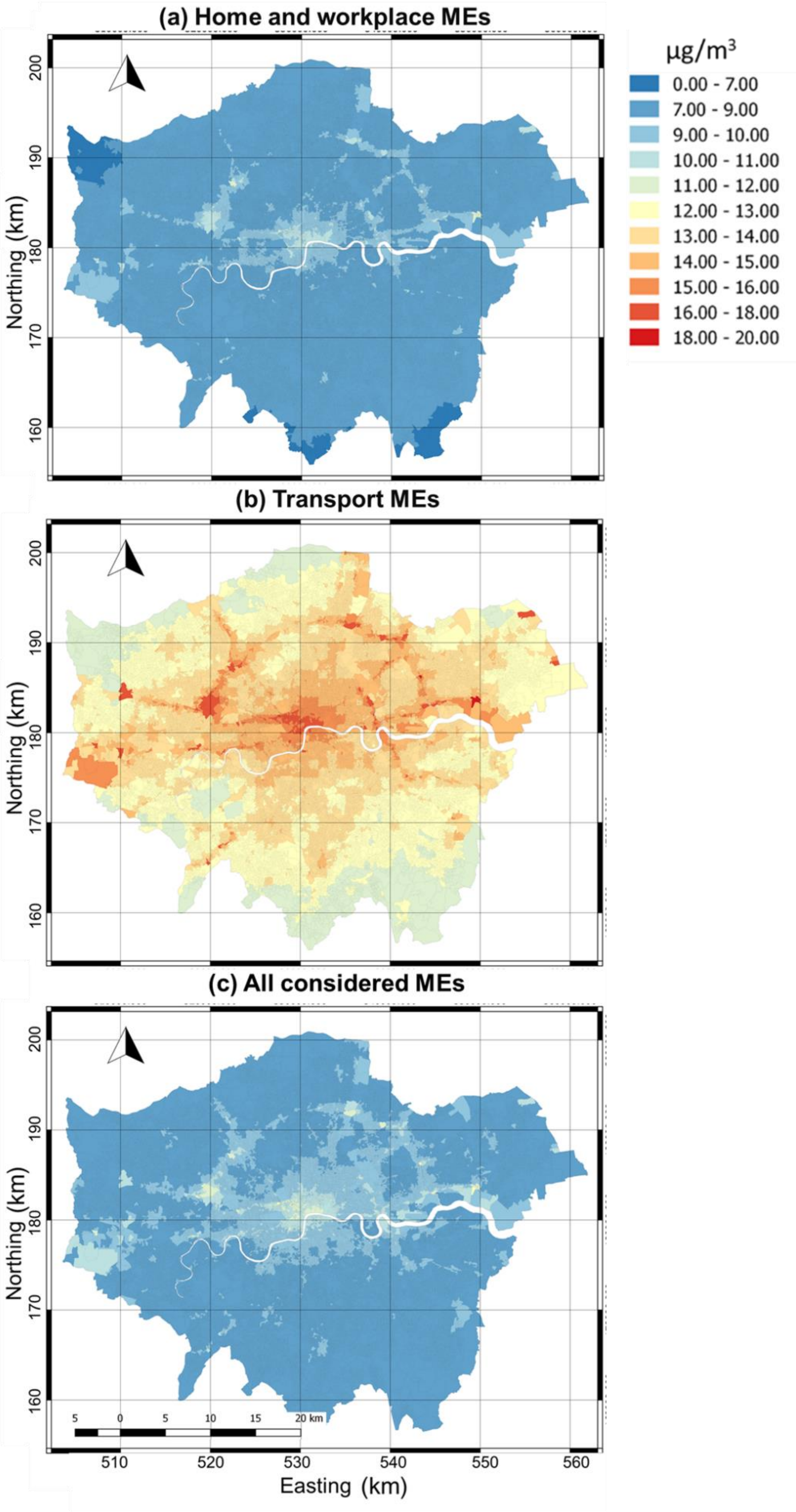

Figure 2 a-c. Population weighted mean concentrations of $PM_{2.5}$ in London (a) at homes and workplaces, (b) in traffic and (c) in the three considered microenvironments in 2008 (µg/m^3).

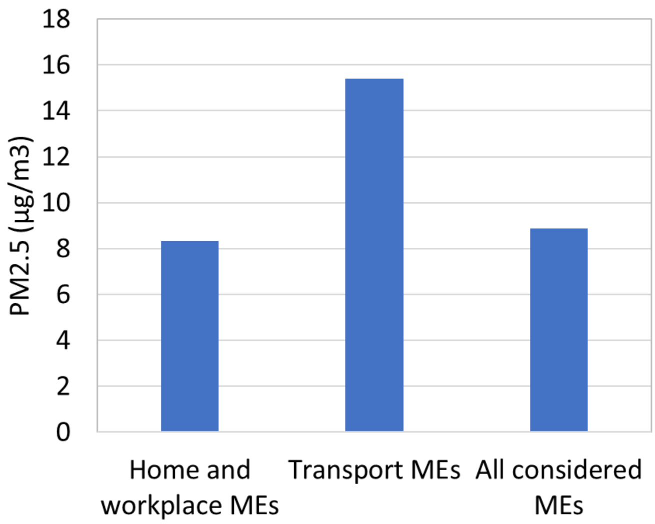

Figure 3. Population weighted mean concentration of $PM_{2.5}$ in combined home and workplace microenvironments, in transport microenvironments, and in all of these microenvironments combined in London in 2008.

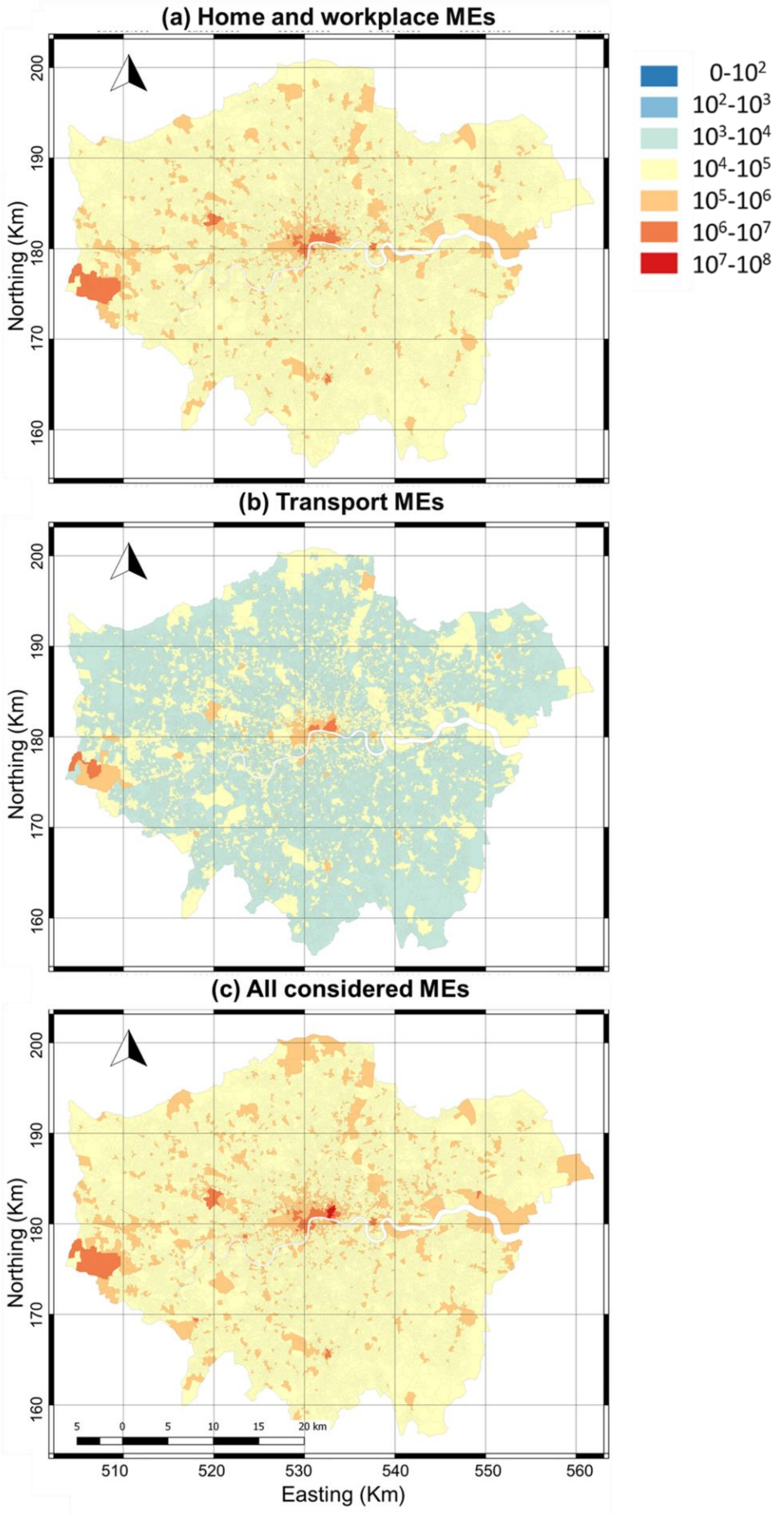

Figure 4 a-c. The predicted population exposures (µg/m^3 × number of people) to $PM_{2.5}$ (a) in homes and workplaces, (b) in transport, and (c) in all the considered microenvironments combined in 2008.

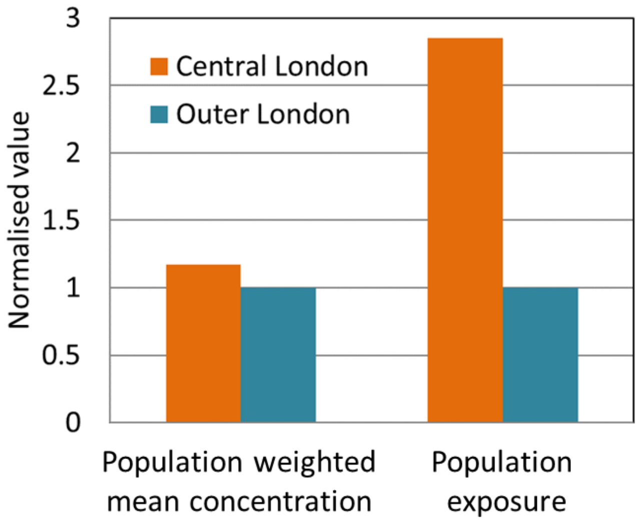

Figure 5. Relative population weighted mean concentrations of $PM_{2.5}$ and population exposure per unit area in central and outer London (Supplementary Figure S1). The values have been normalised to the values of the outer London.

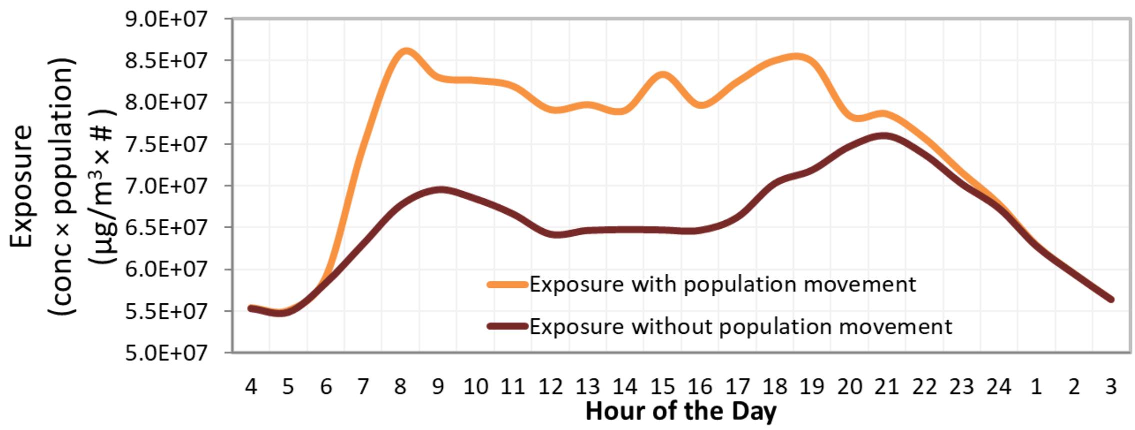

Figure 6. Diurnal variations in population exposure where people spend all the time in a residential (home) indoors environment (Exposure without activity) and combined exposure when people move within the transport and work environments (Exposure with activity), taking into account of infiltration of outdoor air pollution indoors for all the microenvironments. The difference illustrates the influence due to the population activity.

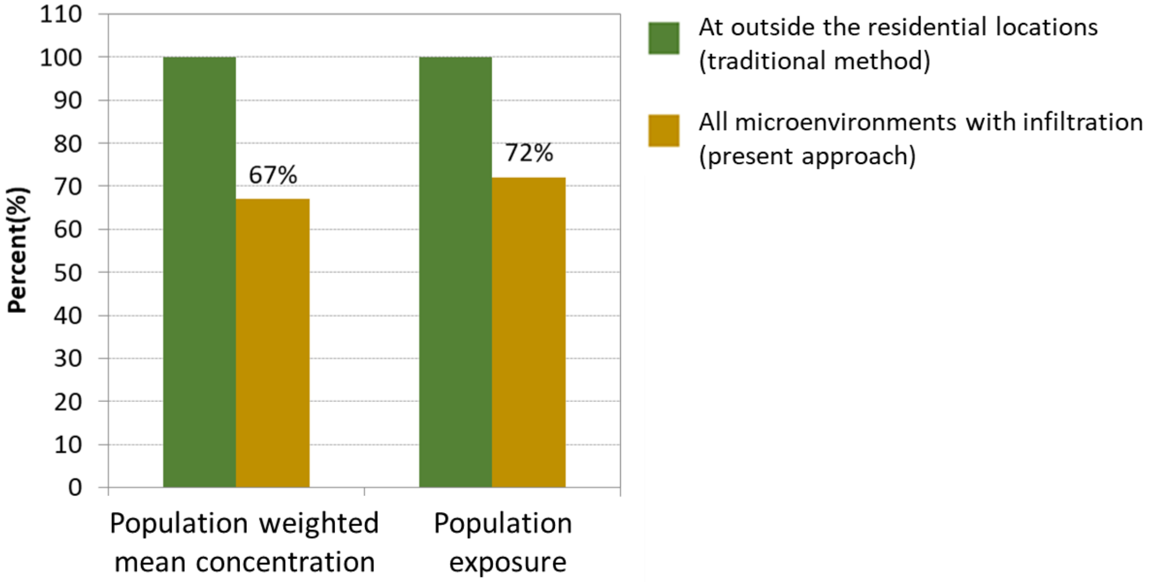

Figure 7. Predicted relative population weighted annual mean $PM_{2.5}$ concentrations and population exposure in London, calculated using the traditional method outside the residential locations (traditional method) and using the approach presented in this work. The approach of this work allows for three microenvironments and infiltration of pollution from outdoor to indoor. The values have been normalised to the values of the traditional method.