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Determinants of black carbon, particle mass and number concentrations in London transport microenvironments

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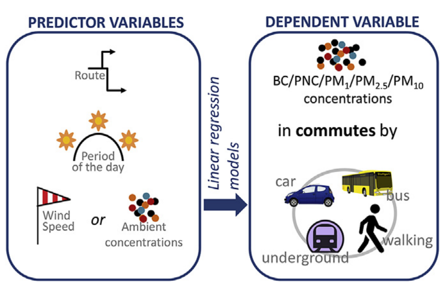
HIGHLIGHTS

- Wind speed and ambient concentrations were main predictors in above-ground modes.
- Ambient PM concentrations explained more variability than wind speed.
- Wind speed had a strongest effect on bus concentrations compared to car and walking.
- Underground line and train's type of windows explained 90% of the variation in PM.
- PM in bus was better explained by ambient concentrations at high spatial resolution.

GRAPHICAL ABSTRACT



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ABSTRACT

We investigated the determinants of personal exposure concentrations of commuters' to black carbon (BC), ultrafine particle number concentrations (PNC), and particulate matter (PM_1, $PM_{2.5}$ and PM_{10}) in different travel modes. We quantified the contribution of key factors that explain the variation of the previous pollutants in four commuting routes in London, each covered by four transport modes (car, bus, walk and underground). Models were performed for each pollutant, separately to assess the effect of meteorology (wind speed) or ambient concentrations (with either high spatial or temporal resolution). Concentration variations were mainly explained by wind speed or ambient concentrations and to a lesser extent by route and period of the day. In multivariate models with wind speed, the wind speed was the common significant predictor for all the pollutants in the above-ground modes (i.e., car, bus, walk); and the only predictor variable for the PM fractions. Wind speed had the strongest effect on PM during the bus trips, with an increase in 1 m s^{-1} leading to a decrease in 2.25, 2.90 and 4.98 μg m^{-3} of PM_1, $PM_{2.5}$ and PM_{10}, respectively. $PM_{2.5}$ and PM_{10} concentrations in car trips were better explained by ambient

Abbreviations: ANCOVA, analysis of covariance; BC, black carbon; CERC, Cambridge Environmental Research Consultants; GLM, Generalised Linear Models; GPS, Global Positioning System; IDW, Inverse Distance Weighting; IMD, Index of Multiple Deprivation; LSOA, Lower Layer Super Output Areas; NOW, non-openable window; OW, openable window; OD, Origin-Destination; PM, particulate matter; PNC, ultrafine particle number concentrations; r, Pearson correlation coefficient; UB, urban background.

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concentrations with high temporal resolution although from a single monitoring station. On the other hand, ambient concentrations with high spatial coverage but lower temporal resolution predicted better the concentrations in bus trips, due to bus routes passing through streets with a high variability of traffic intensity. In the underground models, wind speed was not significant and line and type of windows on the train explained 42% of the variation of PNC and 90% of all PM fractions. Trains in the district line with openable windows had an increase in concentrations of 1 684 cm^{-3} for PNC and 40.69 $\mu g\ m^{-3}$ for $PM_{2.5}$ compared with trains that had non-openable windows. The results from this work can be used to target efforts to reduce personal exposures of London commuters.

1. Introduction

In urban environments, transport emissions frequently cause exceedances of air quality limits, particularly at roadside monitoring locations (EEA, 2016) and the impact of transport-related emissions on public health has been demonstrated (Beelen et al., 2014; HEI, 2010; Hoek et al., 2013). The exposure to ambient particulate matter (PM) is ranked 12th in the Global Burden of Disease and was associated with a global annual estimate of 2.9 million deaths in 2013 (GBD 2013 Risk Factor Collaborators, 2015). In Europe alone, the exposure to $PM_{2.5}$ was responsible for 432,000 premature deaths in 2012 (EEA, 2015).

Traffic-related air pollution is a complex mixture of gaseous compounds and particles that can come directly from the exhaust (Viana et al., 2008); brake and tire wear (Thorpe and Harrison, 2008); resuspension of previously deposited particles on the pavement (road dust; Amato et al., 2009); and formed through physical and chemical processes (e.g. secondary aerosols; Bahreini et al., 2012). Commuters can face higher concentrations in close proximity to traffic (Adams et al., 2001b; Buonanno et al., 2012; Dons et al., 2012; Kumar and Goel, 2016; Kumar et al., 2014; Morawska et al., 2008; Moreno et al., 2015b; Rivas et al., 2016; Zhu et al., 2002; Zuurbier et al., 2010), and especially during morning rush hours (Gómez-Perales et al., 2007; Moreno et al., 2009).

Studies have shown that commuters come in contact with highly variable concentrations of atmospheric pollutants and face short-time extreme peak concentrations during their commutes that result in significant contributions to their total daily exposure (12–32% of daily exposure; Dons et al., 2011; Rivas et al., 2016; Williams and Knibbs, 2016). For instance, Dons et al. (2012) found that 62 individuals from Belgium spent 6% of their day commuting while receiving 21% of their daily-integrated exposure and 30% of their dose of black carbon (BC) during this time. Moreover, Smith et al. (2016) demonstrated how the inclusion of commuting time is crucial for correctly establishing the exposure, with commuters being more highly exposed in comparison to those who stay at home.

There is a growing literature of exposure assessment during commuting (Adams et al., 2001b; Kaur et al., 2005; Kingham et al., 2013; Kumar and Goel, 2016; McNabola et al., 2008; Moreno et al., 2015b; Namdeo et al., 2014; Zuurbier et al., 2010). These studies compare air pollutant concentrations among different transport modes and identify parameters that affect concentrations. Other studies assessed the effect of meteorological variables on personal concentrations in transport microenvironments, such as wind speed or ambient temperature, through correlation analysis (Kingham et al., 1998; Knibbs and de Dear, 2010). However, only a handful of studies, as summarised in Table 1, have attempted to identify and quantify the determinant factors of BC, ultrafine particle number concentrations (PNC), PM_1, $PM_{2.5}$ or PM_{10} concentrations in different transport microenvironments. For example, Adams et al. (2001a) found that wind speed and route choice were determinants of the personal $PM_{2.5}$ concentrations during commutes by bicycle, bus and car in London, while transport mode was not a significant factor. Also in London, Kaur and Nieuwenhuijsen (2009) assessed the effects of traffic intensity, wind speed, temperature and transport mode as determinants of PNC and $PM_{2.5}$ during walking, cycling and trips made by buses, cars and taxis. Their models explained 62% of the variation of PNC, with transport mode, traffic counts, temperature and wind speed being the significant predictor variables; and only 9% of $PM_{2.5}$, with transport mode being the only significant predictor variable and with little effect of traffic intensity. Weichenthal et al. (2008) also observed wind speed and temperature to significantly determine PNC in walking, bus and automobile environments in Montréal. Moreover, the analyses performed by De Nazelle et al. (2012) in a commuting study carried out in Barcelona that included walking, cycling, buses and cars indicated that BC concentrations were explained by transport mode and background concentrations, PNC by transport mode and period of the day and $PM_{2.5}$ only by transport mode.

All these studies contribute to the understanding of factors determining the exposures while travelling in different transport modes. However, much of the observed variation in exposure remains unexplained (Weichenthal et al., 2008) and there are some inconsistencies across different studies. Hence, further research is required to understand the factors that explain the variability in the exposures during commuting. Unravelling the relative roles of determinant factors is key for developing successful strategies for air quality management in transport microenvironments. In addition, models for air pollutants during commuting are required to explore their potential health effects or possible environmental injustices in population-based studies (Sioutas et al., 2005) and can be incorporated into larger models assessing the daily exposure at an individual level.

The overall objective of this work is to assess the determinants of personal concentrations of BC, PNC, PM_1, $PM_{2.5}$ and PM_{10} during commuting through regression modelling. We identified and quantified the contribution of key factors that explain the variation of the aforementioned pollutants in four different commuting routes, each covered by four different transport modes (car, bus, underground and walking) in London. The present work is a comprehensive study, which includes several air pollutants and assesses multiple predictor variables. For instance, no previous study has evaluated the effect of the determinants on personal concentrations of multiple fractions of PM in transport microenvironments.

Table 1
Summary of past studies on the determinants of personal exposure to BC, PNC or PM.

Area of study	Pollutant(s)	Predictor variables explored*	Statistical analysis	Author (year)
London (UK)	$PM_{2.5}$	**Transport mode**, route, **wind speed**, wind direction, precipitation, **temperature**, atmospheric pressure, relative humidity**, traffic counts, fixed monitoring site concentrations**, **bus shell type**	General Linear Model (LM).	Adams et al. (2001a)
Montréal (Canada)	PNC[a]	**Period of the day**, car windows open, **wind speed**, **temperature**	Bayesian Model Averaging (BMA)	Weichenthal et al. (2008)
London (UK)	$PM_{2.5}$, PNC[a]	**Transport mode**, **wind speed**, wind direction, **temperature**, relative humidity, **traffic counts**	General Linear Model (LM)	Kaur and Nieuwenhuijsen (2009)
Barcelona (Spain)	BC, PNC[b], $PM_{2.5}$, CO and CO_2.	**Transport mode**, route, **period of the day, wind speed**, wind direction, temperature, **relative humidity**, **fixed monitoring site concentrations**	ANOVA	De Nazelle et al. (2012)
Beijing (China)	$PM_{2.5}$, CO	**Transport mode**, route, **temperature**, relative humidity, **fixed monitoring site concentrations**	Linear Mixed-Effects Models (LMM)	Huang et al. (2012)
Fort Collins (USA)	$PM_{2.5}$, PNC[c] BC, CO	**Transport mode, route**	Linear Mixed-Effects Models (LMM)	Good et al. (2015)

*Variables in bold are significant predictors for at least one pollutant in one of the transport modes assessed in the study.
[a] Particle size range 20–1 000 nm (P-Trak model 8525, TSI).
[b] Particle size range 10–1 000 nm (CPC model 3007, TSI).
[c] Particle size range 10–700 nm (DiSCmini, Matter Aerosol).

2. Methodology

2.1. Route description

The routes between four Origin-Destination (OD) pairs were covered by car, bus and underground in London (Fig. 1), where bus and underground routes included walking segments as the access mode. We selected routes to represent typical commutes for residential areas with different levels of income deprivation measured using the income subscale of the Index of Multiple Deprivation (IMD; Department for Communities and Local Government, 2015). The origin and destination location were selected as zones from a geographical zone system called Lower Layer Super Output Areas (LSOA) used to disseminate aggregate statistics from the UK census, including commuting data. The 2015 IMD also uses the LSOA geography.

The destination was the same for all the routes. We selected the City of London (the central borough of Greater London, which is the financial district) as our destination point since it is the area with the highest employment density (Fig. 1). To select the origin, all LSOA included in Greater London were classified into four different groups according to the Income Score from the IMD: Group 1 to Group 4, from most to least deprived origin area. Although the income deprivation of the area of origin was used to select the routes, this dimension will not be explored in this work because it has been discussed in our previous work (Rivas et al., 2017). We selected an area of origin in each of the four deprivation groups, which was at the average distance for that deprivation group. Table 2 shows the different London boroughs where origin points were located, together with the OD distance and the route specifications for each transport mode (i.e., main roads used for car, and the underground and bus lines). Note that the distance between the origin and destination increased from Group 1 to Group 4. Finally, for each of the four OD pairs, we selected the fastest route for each transport mode to be monitored (Routes 1 to 4; Fig. 1). The fastest travel mode was the underground (43–56 min), followed by car (49–66 min) and then the bus (67–108 min). The same underground lines in opposite direction were monitored for Routes 1 and 3 (Northern line), and for Routes 2 and 4 (District line). In all the underground routes, few sections were not covered (above ground). Round trips (corresponding to two single trips) were monitored for BC, PNC, PM_1, $PM_{2.5}$ and PM_{10}. Sampling was only carried out during weekdays at three different time periods: morning, afternoon and evening hours starting at 07:45, 12:00 and 16:30 h local time, respectively. An extended description of the route selection can be found in Rivas et al. (2017).

2.2. Data collection

2.2.1. Instrumentation

We monitored BC, PNC, PM_1, $PM_{2.5}$ and PM_{10} in different transport microenvironments. BC concentrations were monitored using a MicroAeth AE51 (AethLabs, USA). The MicroAeth provides BC concentrations derived from measurements of the rate of change in absorption of transmitted light at 880 nm due to continuous collection of aerosol deposit on the filter. The effect of

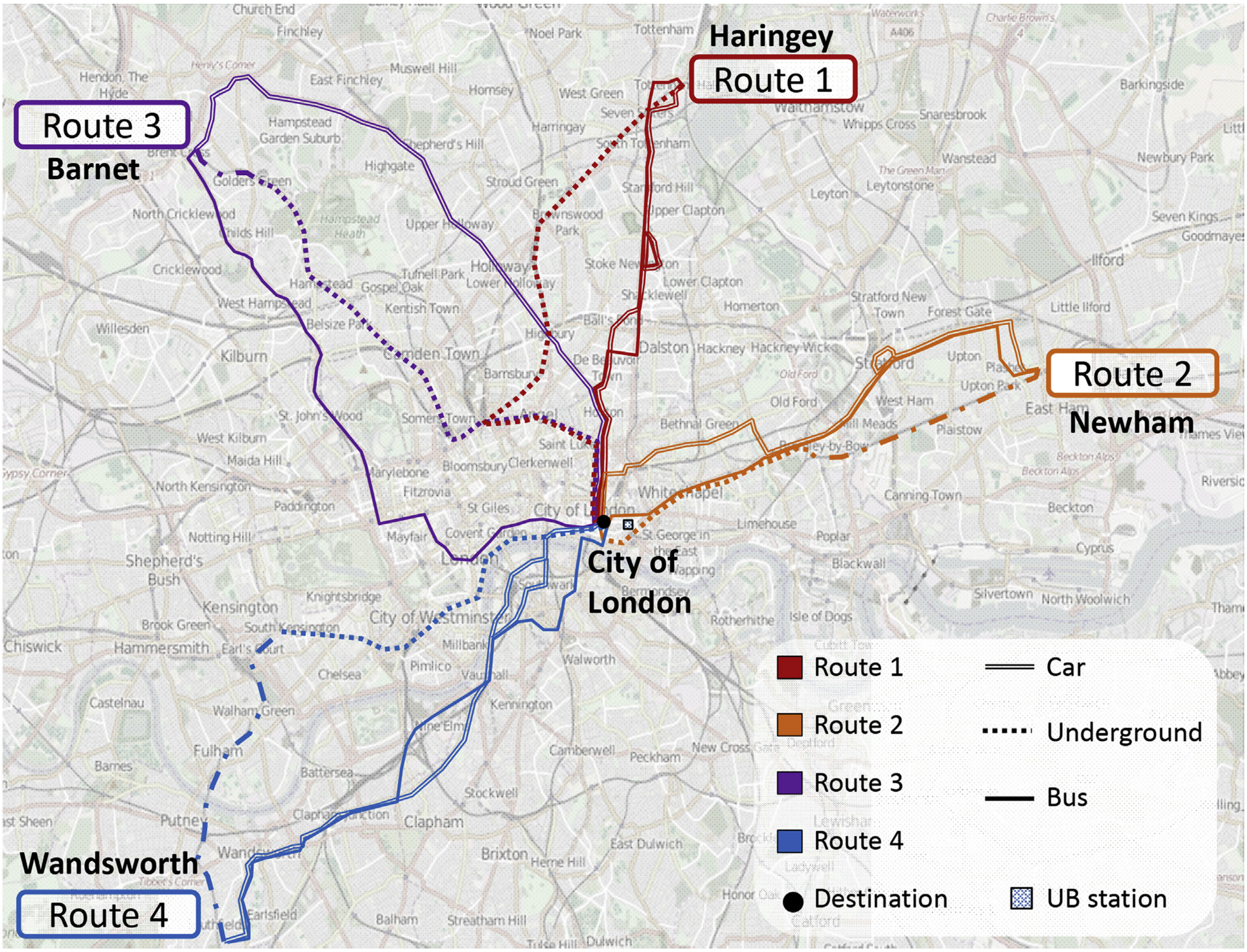

Fig. 1. Location of the routes selected for air pollutant monitoring in different transport modes. UB station is the urban background air quality monitoring station of Sir John Cass School in London.

Table 2
Borough of origin, distance to the destination and main roads (for car routes) or lines (for underground and bus routes) followed for each of the routes and mode of transport. The destination was in the City of London.

Route	Borough of origin	Euclidean distance from origin to destination (km)	ROUTE		
			Car (main road ID)	Underground (line)	Bus (line #)
Route 1	Haringey	7.7	A10 + A501	Victoria + Northern	76
Route 2	Newham	9.4	A11 + A1209	District	325 + 25
Route 3	Barnet	11.5	A1+A1200	Northern	113 + 23
Route 4	Wandsworth	12.2	A3036 + A3200	District	156 + 344

filter loading was kept to a minimum by replacing the filter strip before every trip and setting a flow rate of 100 ml min^{-1}. The time-base was set to 10 s and the original data was post-processed with the Optimised Noise-reduction Averaging algorithm (Hagler et al., 2011). PNC in the 0.02–1 μm size range were measured by means of a P-Trak model 8 525 (TSI Inc., USA) operating at a flow rate of 0.7 l min^{-1} with a time resolution of 10 s. PM_1, $PM_{2.5}$ and PM_{10} mass concentrations were monitored with a GRIMM EDM 107 aerosol spectrometer (GRIMM Technologies Inc., Germany). The instrument provided data every 6 s but these data points were averaged to 10 s afterwards to match the averaging period of the rest of the instruments. The position of the field worker was obtained on a second basis (i.e. 1 Hz) with a Global Positioning System (GPS) Garmin Oregon 350 (Garmin Ltd., USA). For the quality assurance, all the instruments employed for our measurements were rather new or recently factory calibrated to ensure the quality of the collected data. They have been successfully employed in past studies for personal exposure assessment (Azarmi and Kumar, 2016; De Nazelle et al., 2012; Kaur et al., 2005; Rivas et al., 2016). The instruments were carried in a backpack that was specially conditioned and the inlets were positioned at the breathing height (~1.6 m) of the field worker.

2.2.2. Measurements in the different transport modes

Fieldwork was carried out over a total of 40 sampling days between 25 February and 17 June 2016 at 3 different daytime periods. Car measurements were carried out with a petrol-fuelled Peugeot 208 Active 1.2 VTI 82 HP (registered in 2013). The windows of the car were always kept closed and the ventilation was set at 50% fan velocity with no air recirculation. The backpack containing the instruments was placed in the front passenger seat. In the public transport modes (i.e. bus and underground), the backpack was carried on the back while standing and placed on the lap in a way that the inlets were at breathing height if the field worker could have a seat. In buses, the field worker always sat in the first or second row of seats behind the rear door in the lower deck of the bus. If no seats were available, the field worker was standing within the same area. In the underground, the field worker was always at the midpoint carriage of the underground. Since we had no control over ventilation, we registered the information about ventilation conditions and other parameters (e.g. whether the windows in the underground trains were openable) that might influence air pollutant concentrations.

For each trip (in all transport modes), the fieldworker wrote down all the movements in a time-location diary, which allowed to identify the time spent at each part of the route (e.g., walking to the bus/underground station, inside the car, bus or underground train). We monitored a total of 232 single trips, 38% of which were made by car, 41% by underground, and 21% by bus. A total of 225 h of trip monitoring data, involving 10 s averaged 80,720 data points, were available. After discarding the lost data, 98.7% of the data points for PM, 99.4% for BC and 91.9% for PNC remained for analysis. From the 10 s data, we calculated the geometric mean (GM) for each single trip, which was the input for the regression analyses ($n = 82$ for car, $n = 42$ for bus, $n = 87$ for underground, and $n = 130$ for walking). We calculated the GM instead of the arithmetic mean because of the left-skewed distribution of the personal monitoring data.

2.3. Variables

2.3.1. Dependent variables

The GM concentrations of BC, PNC, PM_1, $PM_{2.5}$ and PM_{10} per single trip were the dependent variables. We constructed a different model for each transport mode and pollutant, only considering the time spent inside the vehicle. We also calculated the GM of the concentrations faced during the walking time to the bus stop and the underground station to identify the predictor variables while walking. BC was not assessed in the underground trips since concentrations in the underground are overestimated as a result of the interference of Fe in the BC measurements (Gilardoni et al., 2011; Moreno et al., 2015b; Rivas et al., 2017).

2.3.2. Predictor variables

Meteorological data at Heathrow airport (latitude: 51.479, longitude: −0.449; 25 m above the sea level) was provided by the UK MetOffice, at a 1 h time resolution for the following variables: temperature (°C), relative humidity (%) and wind speed (m s^{-1}; Table 3). We selected the Heathrow weather station as it collects the weather information at 25 m above the mean sea level and therefore it keeps the measurements free from the local ground-level turbulence. These characteristics are expected to be representative of the overall London area. The hourly meteorological data were matched up with the averaged trip pollutant concentrations by taking the time-weighted average of the corresponding hours. Meteorological data from Heathrow airport has been employed in other studies regarding air quality modelling in Greater London (Beddows et al., 2015; Gulliver et al., 2011) and in our previous studies (Al-Dabbous and Kumar, 2014; Goel and Kumar, 2016).

We obtained ambient concentrations at a 15 min time resolution for $PM_{2.5}$ and PM_{10} from the Sir John Cass School urban background (UB) station in the City of London (latitude: 51.514, longitude: −0.078). As for the meteorological parameters, the time-weighted average of the corresponding hours was matched up with the measured concentrations in the transport microenvironments.

Moreover, modelled ambient $PM_{2.5}$ and PM_{10} daily average concentrations were provided by the Cambridge Environmental Research Consultants (CERC) for each day of fieldwork at a high spatial resolution (approximately 300 m). The model employed was the ADMS-Urban dispersion model (CERC, 2016; McHugh et al., 1997), which combines the basic ADMS dispersion model (Carruthers et al., 1994; which requires meteorological data) with an integrated street canyon model, a chemistry model and a traffic emissions database (CERC, 2015). The ADMS-Urban has been validated with data from around the world (Hanna et al., 2001; Heist et al., 2013; Jerrett et al., 2005) and is widely used for modelling air quality on scales ranging from large urban areas to street level (Athanassiadou et al., 2010; Gulliver and Briggs, 2005; Smith et al., 2016). The model output files consist of a regular grid of points at approximately 300 m resolution in addition to a large number of near-road receptors that account for the high concentration at street scale. From these files, we generated a raster file (25 m resolution) with the Inverse Distance Weighting (IDW) interpolation for both $PM_{2.5}$ and PM_{10} using ArcGIS 10.1 (Esri Inc.). The GPS data points from each monitored trip made by bus and car were then used to extract the concentrations at each specific location. Finally, we calculated the GM for each trip with valid GPS data (Table 3). In order to match the modelled concentration maps and the actual measured concentrations, for these specific models we discarded the data that lacked an associated GPS position due to a bad satellite signal and then recalculated the measured GM of PM_{10} and $PM_{2.5}$ for each trip. The GPS had no signal in the underground and, therefore, we could not assess the underground concentrations against the modelled ambient concentrations. From the 132 trips made by bus or car, we discarded <10% of the data points for 76% of the trips due to unavailable GPS data. In 8% of the trips, the data discarded was >50%. However, since the trips were long and the data was spatially matched, all the trips were included in the models, with the exception of three trips where more than 80% of the data had to be discarded due to unavailable GPS data.

Other characteristics of the trips were also evaluated: routes, period of the day, and line and type of windows in the underground trains (the latter two combined into a single variable; Table 3). The route is a qualitative variable with four categories that corresponds to the 4 origin-destination routes (Routes 1 to 4; Fig. 1). Period of the day has three categories: morning, afternoon and evening. The line and the type of windows in the underground trains is a qualitative variable including four categories: District line with trains with non-openable windows (NOW), District line with openable windows (OW), Northern Line with OW and Northern + Victoria Line with OW. The justification for combining lines and types of windows into a single variable is that only trains with OW circulate in both the Northern and Victoria line and a single variable makes the discussion more straightforward. The results of the models are identical if the line and type of windows are included as two separate categorical variables.

2.4. Statistical analysis methods

Pearson correlation coefficients (r) and scatterplots were produced to assess the relationship between trip concentration and individual meteorological variables and ambient concentration, as well as to test possible collinearity between the potential predictor

Table 3
Predictors variables explored in the models. The type of variable is indicated and, in case of categorical variables, the different categories are listed and the reference category is indicated.

Variable	Type	Categories	Reference category
Route	Categorical	Route 1 Route 2 Route 3 Route 4	Route 1
Period of day	Categorical	Morning Afternoon Evening	Morning
Line and type of windows[a]	Categorical	District line with non-openable windows (N.O.W.) District line with openable windows (O.W.) Northern line with O.W Northern + Victoria line with O.W.	District with N.O.W.
Temperature (°C)[b]	Continuous		
Relative Humidity (%)[b]	Continuous		
Wind speed ($m\ s^{-1}$)[b]	Continuous		
ADMS ambient concentration ($\mu g\ m^{-3}$)[c]	Continuous		
UB ambient concentration ($\mu g\ m^{-3}$)[d]	Continuous		

[a] Only for the *underground* trips.
[b] From Heathrow, original data at 1 h resolution.
[c] Only $PM_{2.5}$ and PM_{10} and for *bus, car* and *walking* trips with valid GPS data. Matched by route and day. Data at 1-day resolution.
[d] From the UB air quality monitoring station of Sir John Cass School, original data at 15 min resolution.

variables. Correlations were considered statistically significant at a *p*-value <0.05.

The non-normality of the concentration data requires the use of non-parametric models (such as the Generalised Linear Models, GLM; McCullagh and Nelder, 1989) or the decimal log-transformation of the concentrations. Although the distributions of the trip concentrations were slightly skewed (mainly for $PM_{2.5}$ and PM_{10}), we used untransformed concentration to support the interpretations of the results.

We estimated linear regression models with analysis of covariance (ANCOVA), which allows the input of categorical and continuous variables. In a linear regression, the model assumes that the dependent variable (e.g. GM concentration of each trip) is a linear function of the independent variables with a normally distributed random error. Eq. (1) describes the model as follows:

$$Y_i = \alpha + \beta_1 X_{1i} + \beta_2 X_{2i} + \ldots + \beta_n X_{ni} + \varepsilon_i \tag{1}$$

where Y_i is the dependent variable (e.g. GM concentration) for trip i; X_i are the predictor variables for trip i (e.g., wind speed, period of the day); α is the intercept; β is a regression coefficient (slope); and ε_i is the model residual normally distributed with mean 0 and variance σ^2. We excluded those variables violating the assumptions of the linear regression (e.g. collinearity). Moreover, only significant variables (*p*-value <0.05) were included in the final models. Adjusted R^2 were used to evaluate the goodness of fit of the models.

For cross-validation purposes, a subset of a total of 20 trips (8 for car, 8 for underground and 4 for bus) was used for validating the analysis and were, therefore, excluded from the models. Statistical analysis was performed using the R statistical software (v 3.0.2, R Core Team, 2016) and the packages *openair* (Carslaw and Ropkins, 2012) and *car* (Fox and Weisberg, 2016). ArcGIS 10.1 (Esri Inc.) was employed for the extraction and interpolation of the ADMS-Urban modelled concentrations from the *air*TEXT maps corresponding to our routes.

3. Results and discussions

3.1. Correlations with the continuous explanatory variables

Table 4 shows the descriptive statistics for BC, PNC, PM_1, $PM_{2.5}$ and PM_{10} trip measured concentrations, as well as for $PM_{2.5}$ and PM_{10} concentrations extracted from the ADMS maps (ADMS Ambient $PM_{2.5}$ and PM_{10}) and from the UB monitoring station (UB Ambient $PM_{2.5}$ and P_{M10}); and for the meteorological parameters. The means showed in Table 4 were calculated from the previously averaged trip data. Concentrations of all PM fractions were much higher in the underground trains (e.g. $PM_{2.5} = 50.7\ \mu g\ m^{-3}$) than in the above-ground modes (e.g.: $PM_{2.5} = 7.4\ \mu g\ m^{-3}$ for car and $13.2\ \mu g\ m^{-3}$ for bus) which is in accordance with previous studies (Adams et al., 2001b; Cartenì et al., 2015; Martins et al., 2016). The main source of PM in the underground environment is the mechanical abrasion between rails, wheels and brakes, which results in very high concentrations of particulate iron, and the hindered dispersion in such a confined environment (Moreno et al., 2015a; Rivas et al., 2017). PM concentrations while walking were similar to buses with the exception of PM_{10}, which were higher on the buses. On the other hand, the absence of combustion sources such as vehicular emissions in the underground led to lower PNC than in the above-ground modes, as also observed previously (Moreno et al., 2015b). BC concentrations were highest in bus trips, and lowest in walking trips. For the models including ambient concentrations from the ADMS maps, we discarded concentration data when there was no GPS signal. Therefore, the trips with valid GPS data included fewer data points, which can be translated into a shorter period monitored, especially for the bus trips (average monitored time: 70.9 min for bus trips; 54.6 min for the bus trips after the data without GPS information was discarded), but we confirmed that this did not significantly affect average concentrations (Table 4). In a previous study, we identified potential factors affecting the concentrations in the different transport microenvironments (Rivas et al., 2017), while the focus in this work is to identify and quantify the statistically significant predictor variables in order to build an explanatory model of the concentrations faced for travels by car, bus, underground and walking.

Fig. 2 presents the scatterplots and the Pearson correlation coefficient for all the potential meteorological predictor variables and pollutants concentrations while Supplementary Information, SI, Fig. S1 shows similar results for the correlations with log-transformed concentrations. The strongest significant (*p*-value <0.05, 2-tailed) linear associations in the car ($r = -0.64$ for BC; $r = -0.59$ for PNC; $r = -0.57$ for PM_1, $PM_{2.5}$ and PM_{10}), bus ($r = -0.71$ for BC; $r = -0.63$ for PNC; $r = -0.70$ for PM_1; $r = -0.71$ for $PM_{2.5;}$ $r = -0.54$ for PM_{10}) and walking ($r = -0.53$ for BC;

Table 4
Descriptive statistics for air pollutants and meteorological parameters for car, bus, underground and walking trips. Car, Bus, Underground and Walking columns show the descriptive statistics for the complete trips. Car GPS, Bus GPS and Walking GPS correspond to the sections of the trips with valid GPS data (if the GPS data was not available for a data point, the data point was discarded). GM = geometric mean; GSD = geometric standard deviation (dimensionless units); AM = arithmetic mean; SD = standard deviation; n = number of trips.

	Complete trips (for models with meteorology and ambient PM data from a single UB station)																Trip sections with valid gps data (for models with spatially modelled PM data)											
	CAR (n = 82)				BUS (n = 42)				UNDERGROUND (n = 87)				WALKING (n = 130)				CAR GPS (n = 82)				BUS GPS (n = 42)				WALKING GPS (n = 43)			
	GM	GSD	Min	Max	GM	GSD	Min	Max	GM	GSD	Min	Max	GM	GSD	Min	Max	GM	GSD	Min	Max	GM	GSD	Min	Max	GM	GSD	Min	Max
BC ($\mu g\ m^{-3}$)	4.4	1.5	1.6	11.2	5.6	1.5	1.8	13.2					2.0	1.8	0.3	7.5	4.4	1.5	1.6	12.2	5.7	1.6	1.8	17.6	3.1	2.1	0.5	16.7
PNC (cm^{-3})	8 879	1	3 932	21,997	9 663	1	3 841	21,067	6 300	1	2 517	12,487	7 331	1	1958	17,335	8 816	1	3 763	25,216	9 976	1	4 068	23,504	8 545	2	1958	21,431
PM_1 ($\mu g\ m^{-3}$)	6.9	1.9	1.8	24.4	9.2	1.6	4.7	20.3	32.9	2.2	5.8	96.9	10.1	1.8	3.8	39.3	6.9	1.9	1.8	24.4	9.2	1.6	4.4	20.4	10.2	1.7	4.3	28.4
$PM_{2.5}$ ($\mu g\ m^{-3}$)	7.4	1.9	2.0	25.0	13.2	1.5	7.2	27.4	50.7	2.5	7.5	158.5	13.6	1.7	6.1	43.4	7.4	1.9	2.0	25.5	12.9	1.5	6.7	26.8	14.0	1.6	6.2	33.0
PM_{10} ($\mu g\ m^{-3}$)	8.3	1.8	2.4	27.0	39.0	1.4	17.9	76.5	95.2	2.6	12.3	289.9	28.5	1.5	11.3	77.4	8.2	1.8	2.4	26.9	37.6	1.4	17.3	74.0	33.1	1.7	11.5	90.3
ADMS Ambient $PM_{2.5}$ ($\mu g\ m^{-3}$)	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	19.8	1.4	10.2	29.9	15.1	1.2	11.4	22.5	15.2	1.3	10.7	22.4
ADMS Ambient PM_{10} ($\mu g\ m^{-3}$)	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	28.3	1.4	14.6	42.5	21.7	1.2	16.4	32.1	21.7	1.2	15.4	31.9
UB Ambient $PM_{2.5}$ ($\mu g\ m^{-3}$)[a]	12.5	1.7	5.0	44.0	13.2	1.6	5.5	33.0	–	–	–	–	12.3	1.7	6.1	36.0	–	–	–	–	–	–	–	–	–	–	–	–
UB Ambient PM_{10} ($\mu g\ m^{-3}$)[b]	28.1	1.7	11.0	89.0	39.0	1.7	10.2	74.8	–	–	–	–	26.0	1.6	11.3	74.5	–	–	–	–	–	–	–	–	–	–	–	–
	AM	SD	Min	Max	AM	SD	Min	Max	AM	SD	Min	Max	AM	SD	Min	Max	AM	SD	Min	Max	AM	SD	Min	Max	AM	SD	Min	Max
Temperature (°C)	10.4	3.0	2.4	16.9	17.2	4.3	9.7	23.9	16.1	3.3	5.7	23.3	16.5	3.7	6.5	24.2	10.4	3.0	2.7	16.9	17.3	4.3	9.7	24.0	17.4	4.3	9.9	24.2
Relative Humidity (%)	66.9	15.6	41.3	98.2	72.6	14.7	34.3	92.6	61.5	20.4	28.8	100.0	65.3	19.5	28.8	100.0	66.7	15.6	41.3	98.2	72.3	14.8	34.3	94.4	72.6	15.1	34.5	98.9
Wind Speed ($m\ s^{-1}$)	4.4	2.3	0.6	12.0	3.6	1.5	1.0	6.4	4.1	1.5	1.5	7.7	3.9	1.5	1.0	7.7	4.4	2.3	0.6	12.1	3.6	1.5	1.0	6.8	3.6	1.6	1.0	7.2
Monitored time (min)	54.6	11.5	37.7	90.2	70.9	15.8	46.7	111.0	28.3	6.3	18.7	44.0	10.1	3.3	3.7	17.7	51.6	9.8	22.8	76.8	54.6	21.8	4.2	101.7	6.5	4.2	0.5	17.5

[a] The number of $PM_{2.5}$ observations (n) available for the UB monitoring station are: 77 for car, 18 for bus and 89 for walking.
[b] The number of PM_{10} observations (n) available for the UB monitoring station are: 78 for car, 41 for bus and 124 for walking.

$r = -0.31$ for PNC; $r = -0.35$ for PM_1; $r = -0.35$ for $PM_{2.5}$; $r = -0.27$ for PM_{10}) were observed for wind speed. Wind speed affects the dilution and transport of vehicular emissions (Kumar et al., 2008) and explains the negative correlation. Previous studies including the assessment of personal exposures in cars, buses and walking have also reported this negative linear association between wind speed and in-transit air pollutants (Briggs et al., 2008; De Nazelle et al., 2012; Kaur and Nieuwenhuijsen, 2009; Knibbs et al., 2011; Weichenthal et al., 2008). Ambient temperature showed a significant positive association for all the pollutants in the buses ($r = 0.37$for BC; $r = 0.55$ for PNC; $r = 0.53$ for PM_1; $r = 0.55$ for $PM_{2.5}$; $r = 0.23$ for PM_{10}) and walking ($r = 0.32$ for BC; $r = 0.27$ for PNC; $r = 0.26$ for PM_1; $r = 0.29$ for $PM_{2.5}$; $r = 0.37$ for PM_{10}) trips, while no linear relationship was observed for car ($r = 0.17$ for BC; $r = 0.20$ for PNC; $r = 0.08$ for PM_1; $r = 0.09$ for $PM_{2.5}$; $r = 0.12$ for PM_{10}) and underground ($r = -0.01$ for BC; $r = 0.01$ for PNC; $r = -0.01$ for PM_1; $r = -0.02$ for $PM_{2.5}$; $r = -0.01$ for PM_{10}) travels. Although the ambient temperature was positively correlated with most of the pollutants in buses and while walking, both temperature and relative humidity were not further explored in the multivariate analysis due to their dependence on the period of the day (morning periods exhibited lower temperatures and higher relative humidity, Fig. S2). The highest concentrations in the morning were more affected by the higher traffic intensity than the ambient temperature and relative humidity conditions. Previous commuting studies uniformly found a negative correlation between temperature and air pollutant concentrations (De Nazelle et al., 2012; Kaur and Nieuwenhuijsen, 2009; Laumbach et al., 2010; Weichenthal et al., 2008).

In the underground, only PNC was significantly associated with wind speed ($r = -0.36$) Fig. 2c) and its correlation was much weaker than those for bus ($r = -0.63$) and car ($r = -0.59$). Wind speed was not significantly correlated with the PM fractions in the underground ($r = -0.16$ for PM_1 and $PM_{2.5}$; $r = -0.19$ for PM_{10}). PM emissions are very high in the underground environment, especially for $PM_{2.5}$ and PM_{10}, and outweigh the contribution of outdoor PM. Thus, a small effect is expected from outdoor wind speed. However, Adams et al. (2001a) found an unexpected strong relationship between $PM_{2.5}$ concentrations in the underground and ambient wind speed ($r = -0.60$), which may be due to enhanced natural ventilation during the windy time spans. To the best of our knowledge, no other study has attempted to evaluate the effect of wind speed on the underground concentrations. BC correlations are presented in the figure for the underground dataset, although we did not assess them due to their overestimation by the Fe interference in underground environment (Section 2.2).

Fig. 3 presents the scatterplots and the Pearson correlation coefficient for the measured trip averaged GM concentrations of $PM_{2.5}$ and PM_{10} concentration against (i) the concentrations obtained from a UB station close to the destination at a high time resolution but for a single point in space (trip-averaged); and (ii) the trip-averaged concentrations obtained from the ADMS maps, which are spatially matched through the GPS position of the actual trip but the modelled concentrations are daily averages (thus, low time resolution). Both PM fractions in car, bus and walking trips are strongly correlated with both the UB (car: $r = 0.90$ for $PM_{2.5}$; $r = 0.60$ for PM_{10}, bus: $r = 0.72$ for $PM_{2.5}$; $r = 0.74$ for PM_{10}, car: $r = 0.82$ for $PM_{2.5}$; $r = 0.70$ for PM_{10}) and ADMS ambient (car: $r = 0.54$ for $PM_{2.5}$; $r = 0.52$ for PM_{10}, bus: $r = 0.87$ for $PM_{2.5}$; $r = 0.61$ for PM_{10}, car: $r = 0.82$ for $PM_{2.5}$; $r = 0.57$ for PM_{10}) PM. The indoor/outdoor ratios for in-vehicle (car and bus only) concentrations and ambient ADMS concentrations of $PM_{2.5}$ and PM_{10} are presented in SI Table S1 for each Route. Very low *I/O* ratios (<0.4) were observed in all the routes for the car trips, since the car windows were closed and the filtering system was successful in removing the coarser

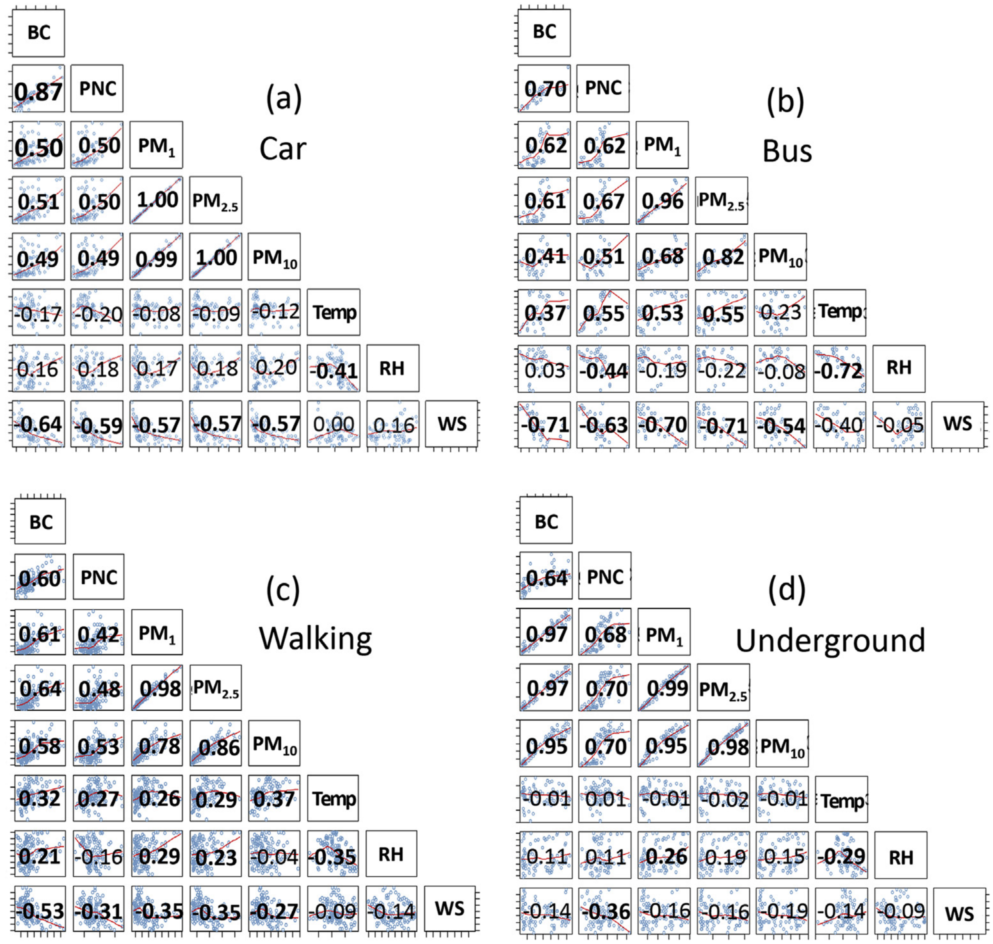

Fig. 2. Scatterplots and Pearson correlation coefficient between pollutant trip-averaged concentration and meteorological variables for (a) car, (b) bus, (c) underground and (d) walking trips. Black numbers indicate significant correlation coefficients at *p*-values <0.05. Temp = Temperature, RH = Relative Humidity, WS = Wind Speed.

particles. The ratios for $PM_{2.5}$ in bus trips were always between 0.7 and 0.8, while the ratios become >1.0 for PM_{10} (between 1.4 and 1.7) indicating the importance of the PM_{10} resuspension. Underground PM concentrations are not significantly correlated (p-value >0.05) with ambient concentrations and therefore have not been further explored in the multivariate regression models.

In the case of car measurements, the correlation coefficients indicate that concentrations in cars are better explained by data with a higher temporal resolution although not spatially matched (data from a single UB monitoring station; $r = 0.90$ for the UB ambient PM versus $r = 0.54$ for the ADMS for $PM_{2.5}$; Fig. 3), while bus trip concentrations are better explained if the spatial variation is taken into account even with a lower time resolution ($r = 0.72$ for the UB ambient PM versus $r = 0.87$ for the ADMS for $PM_{2.5}$; Fig. 3). A possible explanation for this could be that buses take more residential roads, and therefore there is a higher variability among the different sections of the route, which can be only taken into account when being spatially matched. On the other hand, car routes always used main streets and, therefore, higher temporal resolution becomes more important since it accounts for the traffic variation within the day. Correlations for $PM_{2.5}$ during walking are similar for both types (high temporal or high spatial resolution) of ambient $PM_{2.5}$ concentrations ($r = 0.82$ in both cases). For PM_{10}, a higher temporal resolution ($r = 0.70$) seems to catch the PM_{10} variability during walking trip better than a higher spatial resolution ($r = 0.57$).

3.2. Multivariate regression models including meteorology

Concentrations during commuting times might be affected by more than just one variable, therefore we performed multivariate regression models assessing the effect of wind speed together with other possible predictor variables. Table 5 shows the results for the above-ground modes (car, bus and walking) from the linear regression models in which we assessed the predictor variables of route, period, and wind speed (for the equivalent models with log-transformed concentrations see SI Table S2). Only significant variables (p-value <0.05) were included in the models and presented here. SI Table S3 presents the equations describing the models. The route was a significant factor only for BC in car and PNC in buses. Similarly, Good et al. (2015) compared alternative and direct routes by car and found lower BC concentrations in the alternative routes but little differences in PNC and $PM_{2.5}$. For PNC, the selected route for the bus trips was an important factor that explained 30% of the variation (Table 5, Fig. 4). The traffic intensity of the route and the street and built environment characteristics affecting the dispersion of pollutants (and therefore the background concentrations) might be behind the differences between the routes reported in the model results (Ai and Mak, 2015; Amato et al., 2009; Choi et al., 2016; Dons et al., 2013; Goel and Kumar, 2016, 2015a; Weber et al., 2006; Xu et al., 2016). Although PM is influenced by traffic emissions, especially the PM_1 and $PM_{2.5}$ fraction, there are several other sources affecting its concentration and its spatial distribution

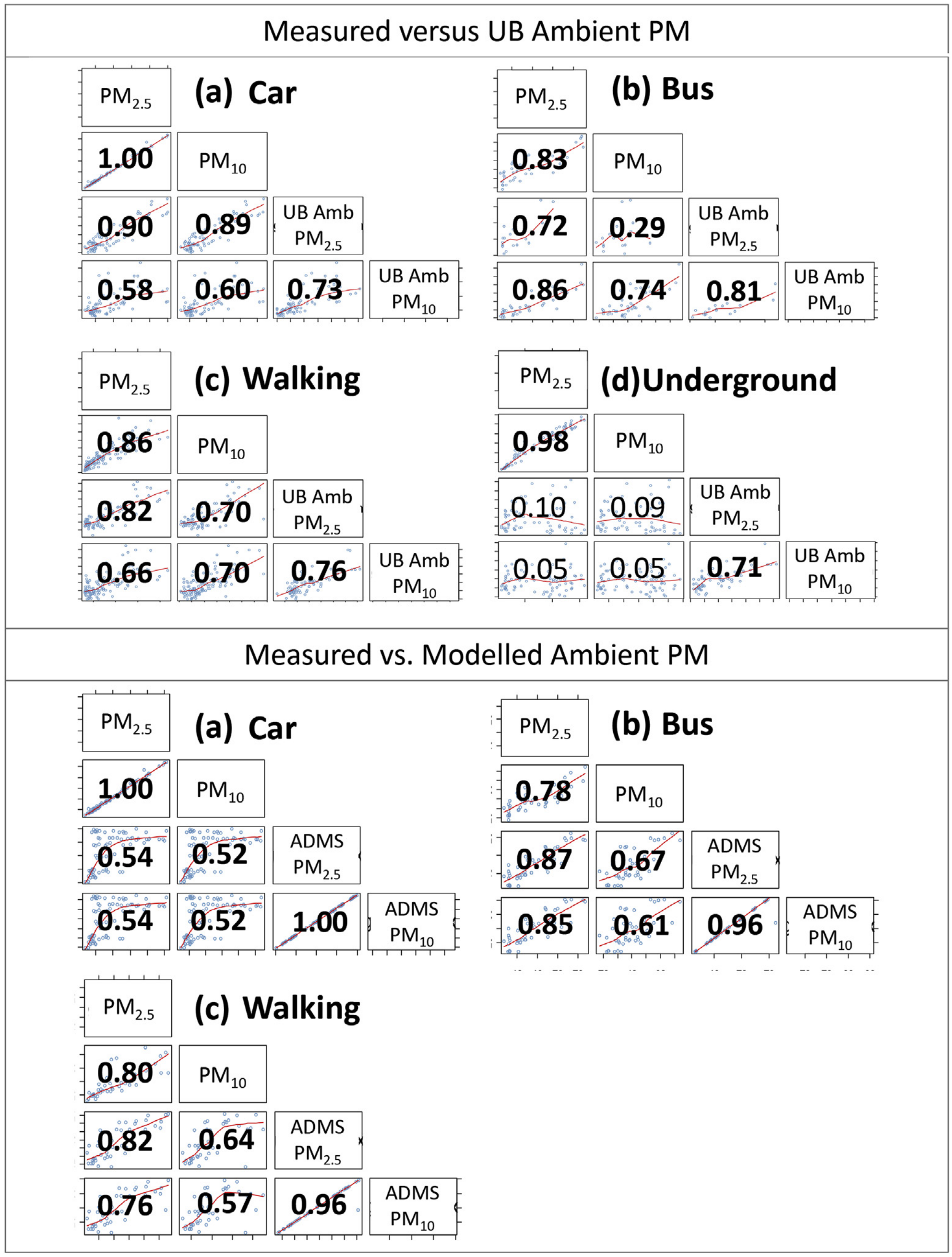

Fig. 3. Scatterplots and Pearson correlation coefficient between in-vehicle, urban background (UB) ambient (high temporal resolution) and modelled (ADMS; high spatial resolution) ambient $PM_{2.5}$ and PM_{10} for (a) car, (b) bus, (c) walking and (d) underground trips. The correlations were made for trip-averaged PM concentrations. Black numbers indicate significant correlation coefficients at p-values <0.05. Temp = Temperature, RH = Relative Humidity, WS = Wind Speed.

is more homogeneous than for traffic tracers (Minguillón et al., 2012), which explain why the route was not a determinant factor for PM. Receptor modelling at an urban background site in London indicated that traffic contributed to 4.5% of total PM_{10} concentrations (non-exhaust emissions excluded) while the traffic source contributed to 45% of PNC (Beddows et al., 2015). At traffic sites, the contribution of traffic emissions is expected to be higher in both PM mass concentrations and PNC. BC is also considered as a better tracer of traffic emission than PM mass (Reche et al., 2011; WHO, 2012).

The period of the day was also significant for PNC (13% of explained variation) and BC (12%) in car trips, but not for bus and walking trips (Fig. 4). Morning trips were used as the reference in the models, with afternoon and evening trips showing negative regression coefficients for both pollutants. This indicates that morning trips will show the highest concentrations, followed by evenings ($\beta = -1.30$ for BC and $\beta = -2\,249$ for PNC; Table 5). Afternoon trips show the lowest concentrations ($\beta = -1.67$ for BC and $\beta = -2\,974$ for PNC). A peak for PNC and BC has been previously observed during the morning in urban environments, exhibiting the impact of traffic emissions during this period (Costabile et al., 2009; Morawska et al., 2008; Pérez et al., 2010; Weichenthal et al., 2008). We would also expect this factor to influence bus and walking concentrations, but they were not statistically

Table 5
Regression models and ANCOVA for car, bus and walking trips. Separate models were performed for each pollutant and transport mode. The regression coefficients (β) for each of the predictor variables included in the model (only significant variables) and the intercept (α) are shown. The Adjusted R^2 for the entire model is presented. n = number of measurements included in the model; p-value(F) = value for the ANCOVA F test, significance set at p-value <0.05; R^2 = ratio of the sum of squared residuals of the corresponding variable to the total sum of squares.

	CAR			BUS			WALKING		
		ANCOVA			ANCOVA			ANCOVA	
	β	p-value(F)	R^2	β	p-value(F)	R^2	β	p-value(F)	R^2
Model for BC	Adjusted R^2 = 0.51, n = 83			Adjusted R^2 = 0.49, n = 42			Adjusted R^2 = 0.27, n = 128		
Route		0.033	0.07						
(reference: *Route 1*)									
Route 2	−1.19								
Route 3	0.00								
Route 4	−0.69								
Period		<0.001	0.12						
(reference: *Morning*)									
Afternoon	−1.67								
Evening	−1.30								
Wind Speed	−0.48	<0.001	0.25	−1.22	<0.001	0.50	−0.49	<0.001	0.28
Intercept	8.47			10.53			4.30		
Model for PNC	Adjusted R^2 = 0.44, n = 74			Adjusted R^2 = 0.56, n = 40			Adjusted R^2 = 0.09, n = 125		
Route					0.001	0.30			
(reference: *Route 1*)									
Route 2				−4 284					
Route 3				−1 121					
Route 4				−1 379					
Period		0.001	0.13						
(reference: *Morning*)									
Afternoon	−2 974								
Evening	−2 249								
Wind Speed	−724	<0.001	0.25	−1 176	0.003	0.16	−580	<0.001	0.09
Intercept	14,538			16,324			10,077		
Model for PM_1	Adjusted R^2 = 0.31, n = 79			Adjusted R^2 = 0.48, n = 42			Adjusted R^2 = 0.12, n = 129		
Wind Speed	−1.50	<0.001	0.32	−2.25	<0.001	0.49	−1.74	<0.001	0.12
Intercept	15.15			18.44			18.77		
Model for $PM_{2.5}$	Adjusted R^2 = 0.32, n = 79			Adjusted R^2 = 0.49, n = 42			Adjusted R^2 = 0.12, n = 129		
Wind Speed	−1.54	<0.001	0.33	−2.90	<0.001	0.50	−1.95	<0.001	0.13
Intercept	15.89			24.87			23.04		
Model for PM_{10}	Adjusted R^2 = 0.31, n = 79			Adjusted R^2 = 0.28, n = 42			Adjusted R^2 = 0.06, n = 129)		
Wind Speed	−1.62	<0.001	0.32	−4.98	<0.001	0.30	−2.45	0.002	0.07
Intercept	17.09			59.22			40.82		

significant and, therefore, finally excluded from the models. In the study from De Nazelle et al. (2012), time of the day was also a significant factor explaining the variation of BC (13%) and PNC (5%) in Barcelona (Spain).

Wind speed was the common significant predictor for car, bus and walking for all pollutants; it was the only predictor variable for the PM fractions. De Nazelle et al. (2012) found wind speed to explain 6% of the variation of BC concentrations but this factor was not important for PNC or $PM_{2.5}$, while in our case it explains between 16% and 50% of the variability in the bus trips, between 25 and 33% in car trips, and between 7 and 28% for walking trips. However, Kaur and Nieuwenhuijsen (2009) and Weichenthal et al. (2008) identified wind speed as a significant determinant of PNC in line with our findings. In all cases, the models indicate a decrease in concentrations with increasing wind speed. Weichenthal et al. (2008) observed that an increase in wind speed of 1 m s^{-1} resulted in a decrease of mean PNC exposures by 223 cm^{-3} and 229 cm^{-3} for bus and car modes, respectively, indicating an identical effect of wind speed in both transport modes. We found a stronger effect, with an increase of 1 m s^{-1} resulting in a decrease of 724 cm^{-3} for car and 1 176 cm^{-3} for buses. Likewise, for BC and PM the effect of wind speed is also stronger in buses than in cars, probably because the diluting effects of wind affect more importantly those vehicles with open windows. There is a consistent increment in the absolute value of the regression coefficient as the particle size increased from PM_1 to PM_{10}, especially during the bus trips: an increase in 1 m s^{-1} in wind speed involves a decrease of 2.25, 2.9 and 4.98 μg m^{-3} for PM_1, $PM_{2.5}$ and PM_{10} respectively (Table 5). Previous studies at roadside traffic monitoring stations observed that for low wind speeds (<2.5 m s^{-1}) there was a decrease in concentrations of the coarse PM fraction ($PM_{2.5\text{-}10}$) with increasing wind speed due to dispersion; on the other hand, $PM_{2.5\text{-}10}$ concentrations increased with higher wind speeds (>4.5 m s^{-1}) due to wind-driven resuspension (Charron and Harrison, 2005; Cheng and Li, 2010; Pérez et al., 2010). This dynamics led to a smooth U-shape trend when plotting $PM_{2.5\text{-}10}$ concentrations against wind speed. Our in-vehicle and walking PM_{10} concentrations do not show this U-shape when plotted against ambient wind speed (Fig. 2), but the fact that wind speed explained much less variation for PM_{10} (Table 4) than for PM_1 and $PM_{2.5}$ might be explained by the wind driven re-suspension of $PM_{2.5\text{-}10}$.

The intercepts for PM in car trips are very similar between the different fractions, while it increases importantly from PM_1 to PM_{10} in bus and walking modes. Windows in the buses were generally open while car measurements were carried out with the windows closed and the outside air getting inside the cabin through the

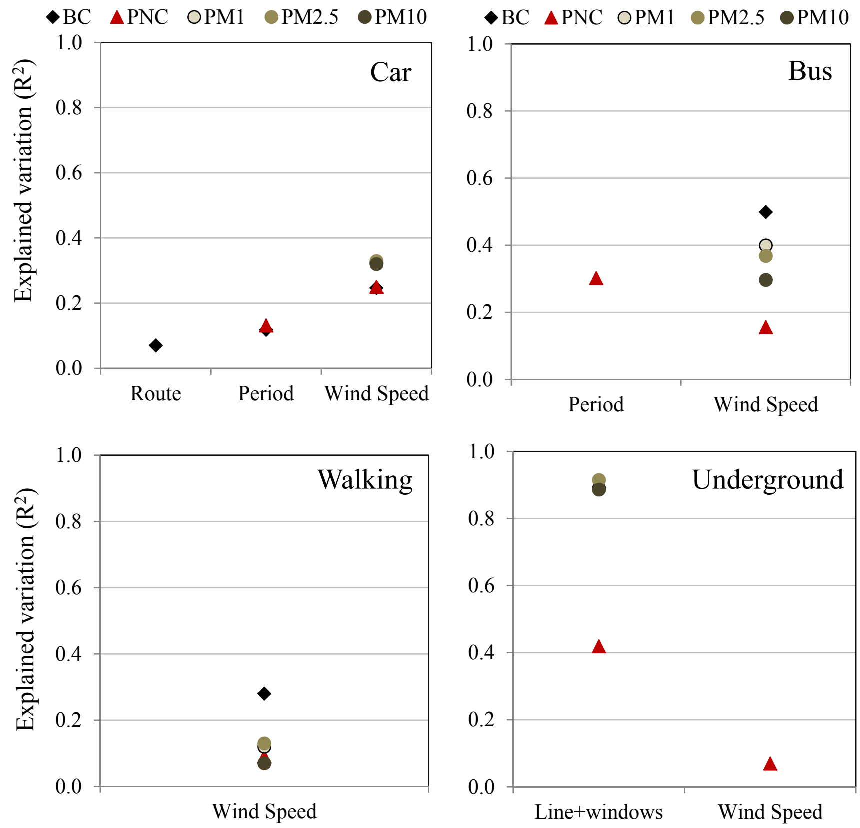

Fig. 4. Explained variation by each factor included in the models for car, bus, walking, and underground.

filtering system. This filtering system is more efficient for coarse particles (Kumar and Goel, 2016), which results in quite similar concentrations in car and bus for PM_1 (as well as for BC and PNC) while being higher for $PM_{2.5}$ and much higher for PM_{10} in buses. Likewise, Huang et al. (2012) reported lower $PM_{2.5}$ concentrations during the commute in taxi compared with buses. Weichenthal et al. (2008) also assessed the determinant factors in buses and cars in Montréal. They did not observe the opening of windows as a significant determinant of PNC exposures, indicating that these pollutants can easily reach the interior of the car and bus cabins regardless of the windows opening (Goel and Kumar, 2016, 2015a, 2015b).

The multivariate models for walking trips indicate that only wind speed was a significant predictor for all the pollutants (Table 5). Wind speed explained between 7 and 28% of the variation in the concentrations (Fig. 4). The R^2 of the walking models was lower or much lower than for the other transport modes. This might be due to the shorter duration of the walking trips or because there are other parameters (such as whether the wind direction was blowing perpendicularly from the street towards the commuter position or vice-versa) affecting the concentrations during walking that we have not considered due to unavailability of data.

Table 6 shows the results for the underground dataset with the period of the day, lines and types of windows on the underground train, and wind speed as possible predictor variables (SI Table S4 shows the models with log-transformed concentrations). From all these variables, lines and types of windows were the main and only significant predictor for all pollutants, with the exception of wind speed, which was also significant in the PNC model (although explaining little variation). Lines and types of windows explained 42% of the variation for PNC while a much larger variation for PM_1 (90%), $PM_{2.5}$ (91%) and PM_{10} (89%). District trains with OW had an increase in concentrations of 1 684 cm^{-3} for PNC, 11.52 μg m^{-3} for PM_1, 40.69 μg m^{-3} for $PM_{2.5}$ and 63.33 μg m^{-3} for PM_{10} than trains with NOW. In trains with OW, outside-train pollutants can easily enter inside the carriage. Higher concentrations have also been reported in trains with OW in underground systems from other cities (Cartenì et al., 2015; Martins et al., 2016). Trains circulating in the Northern and Victoria line were always equipped with OW and concentrations in those trains were always higher than trains in the district line. For instance, after subtracting the estimate for District line with OW from the estimate for trains in the Northern line with OW ($\beta_{\text{Northern OW}} - \beta_{\text{District OW}}$) we can observe how trains in the Northern line have an additional 1 287 cm^{-3} PNC, 29.17 μg m^{-3} for PM_1, 60.47 μg m^{-3} for $PM_{2.5}$, and 107.38 μg m^{-3} for PM_{10} in comparison with District trains with OW. Factors that affect concentrations in the underground tunnels, such as the proportion of the sections that go above-ground, the total length of the line tunnel or different ventilation systems in the tunnels, might explain the differences in concentration between the underground lines. In the multivariate analysis, the wind speed was only significant (*p*-value <0.05) for the PNC model and explained only 7% of the variation. The results for the underground could be easily incorporated to other models such as the London Hybrid Exposure Model (Smith et al., 2016) for $PM_{2.5}$ to estimate more accurately exposures on trips in the underground (that particular model uses average concentrations for all underground trips).

3.3. Regression model including ambient concentrations

We performed a linear regression of measured $PM_{2.5}$ and PM_{10} concentrations during car, bus and walking trips against two different sets of ambient $PM_{2.5}$ and PM_{10} data. One of the sets

Table 6
Regression models and ANCOVA for the underground trips. Separate models were performed for each pollutant and transport mode. The regression coefficients (β) for each of the predictor variables included in the model (only significant variables) and the intercept (α) are shown. The Adjusted R^2 for the entire model is presented. n = number of measurements included in the model, p-value(F) = value for the ANCOVA F test, significance set at p-value <0.05; R^2 = ratio of the sum of squared residuals of the corresponding variable to the total sum of squares.

	UNDERGROUND		
		ANCOVA	
	β	p-value(F)	R^2
Model for PNC	Adjusted $R^2 = 0.50$, $n = 80$		
Line (reference: District NOW)		<0.001	0.42
District OW	1 684		
Northern OW	2 971		
Northern + victoria OW	3 045		
Wind Speed	−352	0.002	0.07
Intercept	6 193		
Model for PM_1	Adjusted $R^2 = 0.89$, $n = 86$		
Line (reference: District NOW)		<0.001	0.90
District OW	11.52		
Northern OW	40.69		
Northern + victoria OW	63.33		
Intercept	13.66		
Model for $PM_{2.5}$	Adjusted $R^2 = 0.91$, $n = 86$		
Line (reference: District NOW)		<0.001	0.91
District OW	23.05		
Northern OW	83.52		
Northern + victoria OW	110.91		
Intercept	16.26		
Model for PM_{10}	Adjusted $R^2 = 0.88$, $n = 86$		
Line (reference: District NOW)		<0.001	0.89
District OW	63.75		
Northern OW	171.13		
Northern + victoria OW	193.85		
Intercept	26.93		

corresponds to the PM concentrations measured in a single UB monitoring station close to the destination, which comprises data at a high time resolution (15 min time resolution) but lacks spatial coverage. The other set is the data obtained from the ADMS models with a high spatial resolution, but at low temporal resolution (daily averages). Further information regarding those variables is available in Section 2.3. Models for the underground were not performed, owing to the absence of a linear relationship with ambient concentrations (Fig. 3). We could not assess other pollutants due to the lack of ambient concentration data.

Table 7 shows the results for the above-ground modes (car, bus and walking) from the linear regression and ANCOVA models where the variables route, period, and either UB or ADMS ambient concentration were explored (see SI Table S5 for models with log-transformed concentrations). The route was not a significant explanatory variable for any of the models. The period was sometimes included in the model, although always explaining little of the variation (2–13%), especially for the models with the UB ambient concentration (2–8%).

Ambient concentrations were the main predictor variables, explaining between 26 and 81% of the variation. UB ambient concentrations predicted $PM_{2.5}$ concentrations in car trips much better than location specific ADMS concentrations (adjusted $R^2 = 0.82$ versus 0.32). Our car routes mainly took main and busy roads. This might indicate that for main roads, the variation of concentrations is more importantly affected by the temporal than by the spatial variability. Contrary, $PM_{2.5}$ concentrations for the bus trips were much better predicted by the ADMS than from the single UB monitoring station ($R^2 = 0.80$ vs 0.50) concentrations, although similar R^2 was obtained for PM_{10} for the ADMS ($R^2 = 0.48$) and UB ($R^2 = 0.53$). The routes for the bus lines usually take more residential roads, and the ADMS data would help to capture the within-city variability between the main and the less busy streets. This fact indicates that the variability in concentrations between main roads and small streets is much higher than between different main roads.

Walking trips were better predicted by the UB ambient concentrations ($R^2 = 0.74$ and 0.51 for UB ambient $PM_{2.5}$ and PM_{10}, respectively) although the fit on the models including the ADMS ambient concentrations was also good for $PM_{2.5}$ ($R^2 = 0.67$ and 0.30 for ADMS ambient $PM_{2.5}$ and PM_{10}, respectively).

The models for $PM_{2.5}$ obtained a better fit than for PM_{10}, with the exception of the bus with the UB ambient PM where the adjusted R^2 were very similar. We would expect a lower fit for PM_{10} in the car because the filtering system of the vehicle hinders the entrance of particles in the coarse fraction ($PM_{2.5\text{-}10}$; Kumar and Goel, 2016).

3.4. Extrapolation: validation of the models

A subset of a total of 20 trips (8 for car, 8 for underground and 4 for bus) were not included in the models for subsequent validation. SI Table S6 shows the descriptive statistics for the trips used for validation, which are similar to the ones used to build the models (Table 4). A limitation of this validation is that few samples were extracted from the original dataset to avoid discarding of too much data for constructing robust models. Although the number of samples are relatively lower for validation, this exercise gives reasonably good insight on the performance of the models. Therefore, the following validation is merely informative and should be generalised with caution.

Table 8 shows the equation and the R^2 obtained for the correlations between the measured and the modelled air pollutant concentrations. For the ease of comparison, we included the adjusted R^2 from the multivariate models.

Regarding the models with meteorology, the models for the underground showed the best results when assessing the goodness of fit (Table 8). For the underground models, only one categorical variable was included (except for wind speed in the case of PNC). Just knowing the underground line and if the windows in the train are open allows a good prediction of concentrations. However, this model fails to capture the variability within the line (Fig. S3) that might be affected by other factors that we did not consider (such as the frequency of sweeping the platform surfaces or cleaning the train trails). The correlation for BC concentrations for the car mode is moderate, but weak for PNC and especially for the PM fractions. The validation of the concentration in bus trips is made based on only four observations, and in some cases, it is driven by a simple data point. Moreover, the flat slopes indicate that the model is not able to capture the variability of the actual concentrations. As expected because of the low adjusted R^2 of the models for the walking trips, very weak correlation were observed for this travel mode. Further variables such as the relative direction, parallel or perpendicular, of the wind speed to the street; or the distance to the centre of the street for walking modes, which are not included in this study could be explored to explain the remaining variability.

In the case of the models including ambient concentrations, the R^2 for the car, bus and walk were moderate to good (0.30–0.80). $PM_{2.5}$ and PM_{10} concentrations in transport microenvironments

Table 7
Regression models and ANCOVA for car, bus and walking trips. Separate models were performed for each pollutant and transport mode. The regression coefficients (β) for each of the predictor variables included in the model (only significant variables) and the intercept (α) are shown. The Adjusted R^2 for the entire model is presented. n = number of measurements included in the model; p-value(F) = value for the ANCOVA F test, significance set at p-value <0.05; R^2 = ratio of the sum of squared residuals of the corresponding variable to the total sum of squares.

	CAR			BUS			WALKING		
		ANCOVA			ANCOVA			ANCOVA	
	β	p-value(F)	R^2	β	p-value(F)	R^2	β	p-value(F)	R^2
Measured vs. UB ambient PM									
Model for $PM_{2.5}$	Adjusted $R^2 = 0.82$, $n = 77$			Adjusted $R^2 = 0.50$, $n = 18$			Adjusted $R^2 = 0.74$, $n = 89$		
Period (ref: *Morning*)		0.018	0.02					<0.001	0.08
Afternoon	−2.02						−5.68		
Evening	−1.65						−5.10		
UB Ambient $PM_{2.5}$	0.68	<0.001	0.80	0.65	<0.001	0.52	1.00	<0.001	0.67
Intercept	0.76			4.16			5.21		
Model for PM_{10}	Adjusted $R^2 = 0.35$, $n = 78$			Adjusted $R^2 = 0.53$, $n = 41$			Adjusted $R^2 = 0.51$, $n = 124$		
Period (ref: *Morning*)								0.025	0.03
Afternoon							−5.98		
Evening							−2.02		
UB Ambient PM_{10}	0.2	<0.001	0.36	0.7	<0.001	0.54	0.7	<0.001	0.47
Intercept	2.8			22.4			12.7		
Measured vs. ADMS ambient PM									
Model for $PM_{2.5}$	Adjusted $R^2 = 0.32$, $n = 80$			Adjusted $R^2 = 0.80$, $n = 40$			Adjusted $R^2 = 0.67$, $n = 43$		
Period (ref: *Morning*)		0.039	0.06						
Afternoon	−2.56								
Evening	−3.58								
ADMS Ambient $PM_{2.5}$	0.48	<0.001	0.28	1.16	<0.001	0.81	1.58	<0.001	0.68
Intercept	1.08			−6.16			−11.33		
Model for PM_{10}	Adjusted $R^2 = 0.31$, $n = 80$			Adjusted $R^2 = 0.48$, $n = 40$			Adjusted $R^2 = 0.30$, $n = 43$		
Period (ref: *Morning*)		0.039	0.06		0.015	0.13			
Afternoon	−2.63			−3.47					
Evening	−3.87			−11.15					
ADMS Ambient PM_{10}	0.35	<0.001	0.26	1.39	<0.001	0.40	2.1	<0.001	0.32
Intercept	1.65			10.54			−12.7		

were by far better predicted by ambient concentrations than by wind speed.

4. Summary, conclusions and future work

It is known that commuters can face very high and variable concentrations but the determinants of these high concentrations are still not fully understood. In this work, we identified and quantified key drivers of the exposure during commuting in four different routes in London. We performed linear regression models with ANCOVA to assess the determinants of the mean concentration faced while travelling by car, bus, underground or walking. We developed separate multivariate models for the dependent variables of wind speed and ambient concentrations to avoid collinearity. We could not assess other meteorological parameters such as ambient temperature due to their dependence on the period of the day.

Wind speed was a significant predictor variable of all pollutants for the above-ground modes (car, bus and walk) but not for the underground. A decrease in concentration was observed with increasing wind speed, for example, an increase of 1 m s^{-1} resulted in a decrease in PNC of 724 cm^{-3} for the car and 1 176 cm^{-3} for the buses, probably because the diluting effects of wind affect more importantly those vehicles with open windows. For PM, the absolute value of the wind speed coefficients increased together with particle size, especially for the bus trips; models predicted a decrease of 2.25, 2.90 and 4.98 μg m^{-3} for PM_1, $PM_{2.5}$ and PM_{10}, respectively, for each unit of increase in wind speed (m s^{-1}). Wind speed explained between 7 and 50% of the variability and was the only significant predictor for PM concentrations. On the other hand, the route was a significant predictor variable for BC in car (explaining 7% of the variability) and for PNC in buses (30% of the variability). Different traffic intensity, type of fleet and speed of the traffic flow in each of the routes and the effect of built environment on pollutant dispersion might affect on-road concentrations of traffic-related pollutants such as BC and PNC. Time of the day was also a significant predictor variable for BC and PNC in car trips.

Surprisingly, our results showed that around 90% of the concentration variations in the PM fractions (42% in PNC) in the underground trips were explained by the underground line and by the type of windows. District trains with openable windows had an increase in concentrations of 1 684 cm^{-3} for PNC, 11.52 μg m^{-3} for PM_1, 40.69 μg m^{-3} for $PM_{2.5}$ and 63.33 μg m^{-3} for PM_{10} than trains with non-openable windows. Wind speed explained only 7% of the PNC in the underground.

Ambient concentrations were predictor variables for $PM_{2.5}$ and PM_{10} trip concentrations, explaining between 26 and 81% of the variation in the above-ground modes (car, bus and walking). Ambient concentrations were a better predictor of in-transit $PM_{2.5}$ and PM_{10} concentrations than wind speed (e.g., for $PM_{2.5}$: $r = 0.90$, $r = 0.72$, $r = 0.82$ for correlations with ambient concentrations; $r = -0.57$, $r = -0.71$, $r = -0.35$ for correlations with wind speed; car, bus and walking respectively). Ambient PM concentrations at a high temporal resolution from a single monitoring station explained a higher variation of concentrations in car trips than daily averages spatially matched, while the contrary was observed for bus trips. This is due to the bus routes running through streets with a high variety of traffic intensities, which is captured only by

Table 8
Equation and R^2 from the correlation between the measured pollutant concentration and the modelled concentrations obtained by using the models including meteorology and ambient concentrations (models detailed in Tables 6–8). The adjusted R^2 from the models is also shown for comparison. In bold are the values with $R^2 \geq 0.30$.

	Correlation modelled vs measured			Model
	n	Equation (y = modelled; x = measured)	R^2	Adjusted R^2
For the models with meteorology (shown in Tables 6 and 7)				
Car				
BC	8	y = 0.35x + 3.33	**0.31**	**0.51**
PNC	8	y = 0.21x + 8 028.4	0.12	**0.44**
PM_1	8	y = 0.14x + 7.40	0.04	**0.31**
$PM_{2.5}$	8	y = 0.13x + 7.92	0.03	**0.32**
PM_{10}	8	y = 0.10x + 8.88	0.02	**0.31**
Bus				
BC	4	y = −0.05x + 7.37	**0.50**	**0.49**
PNC	4	y = 0.25x + 9 080	0.21	**0.56**
PM_1	4	y = −0.02x + 12.29	0.05	**0.48**
$PM_{2.5}$	4	y = −0.02x + 16.89	0.02	**0.49**
PM_{10}	4	y = −0.10x + 4 934	**0.34**	0.28
Walking				
BC	12	y = 0.22x + 2.08	0.21	0.27
PNC	11	y = −0.02x + 8 170	0.01	0.09
PM_1	12	y = −0.01x + 12.76	0.00	0.12
$PM_{2.5}$	12	y = 0.00x + 16.12	0.00	0.12
PM_{10}	12	y = 0.03x + 31.24	0.02	0.06
Underground				
PNC	7	y = 0.52x + 2 684	**0.45**	**0.50**
PM_1	8	y = 1.07× − 6.00	**0.93**	**0.89**
$PM_{2.5}$	8	y = 1.08× − 9.75	**0.96**	**0.91**
PM_{10}	8	y = 1.07× − 20.43	**0.95**	**0.88**
For the models with UB ambient concentrations (shown in Table 7)				
Car				
$PM_{2.5}$	8	y = 1.59x + 5.88	**0.49**	**0.82**
PM_{10}	8	y = 0.67x + 2.24	**0.57**	**0.35**
Bus				
$PM_{2.5}$	2	–	–	**0.50**
PM_{10}	4	y = 0.04x + 37.69	0.00	**0.53**
Walking				
$PM_{2.5}$	8	y = 0.53x + 4.36	**0.85**	**0.74**
PM_{10}	11	y = 0.49x + 13.47	**0.52**	**0.51**
For the models with ADMS concentrations (shown in Table 7)				
Car				
$PM_{2.5}$	8	y = 0.24x + 6.55	0.05	**0.32**
PM_{10}	8	y = 0.22x + 7.35	0.04	**0.31**
Bus				
$PM_{2.5}$	4	y = 0.20x + 10.20	0.27	**0.80**
PM_{10}	4	y = 2.12× − 49.78	**0.46**	**0.48**
Walking				
$PM_{2.5}$	4	y = 0.34x + 11.39	**0.34**	**0.67**
PM_{10}	4	y = 0.70x + 18.02	**0.54**	**0.30**

location-specific concentrations (high spatial resolution).

Further possible explanatory variables (such as the relative direction – parallel or perpendicular – of the wind speed to the street; the distance to the centre of the street for walking modes; traffic intensity and composition, travel speeds, road size, type of fuels) could also be included in future studies in order to explain the remaining unexplained component of the variation of concentrations in commuting exposures and to better assess if a the most deprived are also the most exposed while commuting. Nevertheless, in some cases, the models were able to explain a large component of the variation in concentrations (especially in the underground trips), and this information can be used to reduce personal exposures of London commuters as well as to extrapolate the estimates of exposure at a population level for other investigations such as epidemiological studies. Furthermore, understanding the variation in concentration could contribute to more efficient and inclusive air quality policies in urban centres and better urban planning. Finally, a policy relevant addition to this study would be to measure the personal exposure of cyclists for different routes, given Transport for London's ambitions to develop cycling as an integral part of London's transport system.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2017.05.004.

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