In-vehicle exposure to NO2 and PM2.5

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In-vehicle exposure to NO_2 and $PM_{2.5}$: A comprehensive assessment of controlling parameters and reduction strategies to minimise personal exposure

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ABSTRACT

Vehicles are the third most occupied microenvironment, other than home and workplace, in developed urban areas. Vehicle cabins are confined spaces where occupants can mitigate their exposure to on-road nitrogen dioxide (NO_2) and fine particulate matter ($PM_{2.5}$) concentrations. Understanding which parameters exert the greatest influence on in-vehicle exposure underpins advice to drivers and vehicle occupants in general. This study assessed the in-vehicle NO_2 and $PM_{2.5}$ levels and developed stepwise general additive mixed models (sGAMM) to investigate comprehensively the combined and individual influences of factors that influence the in-vehicle exposures. The mean in-vehicle levels were 19 ± 18 and 6.4 ± 2.7 μg/m^3 for NO_2 and $PM_{2.5}$, respectively. sGAMM model identified significant factors explaining a large fraction of in-vehicle NO_2 and $PM_{2.5}$ variability, $R^2 = 0.645$ and 0.723, respectively. From the model's explained variability on-road air pollution was the most important predictor accounting for 22.3 and 30 % of NO_2 and $PM_{2.5}$ variability, respectively. Vehicle-based predictors included manufacturing year, cabin size, odometer reading, type of cabin filter, ventilation fan speed power, window setting, and use of air recirculation, and together explained 48.7 % and 61.3 % of NO_2 and $PM_{2.5}$ variability, respectively, with 41.4 % and 51.9 %, related to ventilation preference and type of filtration media, respectively. Driving-based parameters included driving speed, traffic conditions, traffic lights, roundabouts, and following high emitters and accounted for 22 and 7.4 % of in-vehicle NO_2 and $PM_{2.5}$ exposure variability, respectively. Vehicle occupants can significantly reduce their in-vehicle exposure by moderating vehicle ventilation settings and by choosing an appropriate cabin air filter.

1. Introduction

Traffic-related nitrogen dioxide (NO_2) and fine particulate matter ($PM_{2.5}$) are pollutants of significant concern to urban air quality and human exposure. $PM_{2.5}$ is a class 1 carcinogen according to WHO International Agency for Research on Cancer (IARC, 2013) and has been associated with several adverse health effects, including increased morbidity, mortality, and emergency hospital admissions for cardiovascular, cerebrovascular, and ischemic heart disease (Franklin et al., 2008; Zanobetti et al., 2009; Wellenius et al., 2012). NO_2 is also considered to be a leading environmental and adverse health risk factor, since it has been associated with acute threats to public health, increased mortality and increased economic burden (Eum et al., 2019; Fenech and Aquilina, 2020; Kaufman et al., 2016; Renzi et al., 2018; Schwartz et al., 2018). Even though there have been some actions taken to reduce vehicle emissions worldwide both by developing new emission control technologies and by introducing stricter emission standards (Matthaios et al., 2019), the growth of urban populations has introduced greater congestion levels, offsetting some of these benefits.

Over the past decade, in large developed metropolitan areas, there have been significant investments in active transportation infrastructure to promote greener, active commuting (i.e., cycling). However, the emergence of new transportation trends in urban environments (i.e., ride sharing, on-demand transit and self-sufficient vehicles) are likely to increase time spend inside vehicles for a sizable proportion of the population (Harik et al., 2017). Consequently, in many cities, commuters

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spend >1.5-h inside vehicles daily, which may result in elevated exposures to traffic-related NO_2 and $PM_{2.5}$ concentrations (WHO, 2005). This is particularly important for vulnerable groups such as elderly or obese who are most likely to rely on mechanised modes of transportation to achieve their mobility needs (Sivak and Schoettle, 2011; Li et al., 2012; Olabarria et al., 2014). Therefore, since there are not many studies that have investigated the factors influencing vehicle occupant exposure to traffic related pollutants, the challenge of reducing in-vehicle exposure remains as relevant as ever.

According to previous studies, the main determinants which affect in-vehicle air pollution are infiltration of on-road air pollutants and the air exchange (Hudda and Fruin, 2013) between the vehicle microenvironment and the ambient (on-road) environment, which can be either passive (via leaks) or mechanical (through the ventilation system or opening the windows). Air exchange rate, which can be defined as the rate of air turnover between the enclosed environment and its surroundings, plays an important role to the in-vehicle air pollution. By minimising the air exchange rates between in-vehicle and on-road or by using recirculatory conditions with the appropriate filtration media on-road concentrations inside vehicles can be reduced (Hudda et al., 2012; Tong et al., 2019; Matthaios et al., 2020). However, recirculatory conditions or insufficient (low) air exchange rates also lead to in-cabin accumulation of carbon dioxide (CO_2) and bio-effluents exhaled by occupants (Hudda and Fruin, 2018). On-road air quality (that directly affects in-vehicle levels) is influenced by several environmental and traffic related parameters such as regional air pollution, site specific air pollution hotspots, traffic volume and composition, meteorology, urban built environment, vehicle speed, time (rush hour/non-rush hour) and road type (i.e., urban, sub-urban, rural). However, there are several other driving factors and vehicle related factors that affect in-vehicle pollution. For example, fan power affects the air exchange hence the in-vehicle pollution, while driving through tunnels may also affect on-road (hence in-vehicle) levels due to limited pollution dispersion (Yamada et al., 2016; Martin et al., 2016). Other vehicle related parameters that may affect occupant exposure are vehicle cabin size, height of the vehicle's cabin air intake in relation to the vehicles in front, vehicle age, and odometer reading (Chan et al., 1999). Recent studies have also shown that vehicle cabin filters also play an important role in reducing the in-vehicle exposure (Matthaios et al., 2023), while other driving and vehicle related parameters that may affect NO_2, and $PM_{2.5}$ levels such as driving speed, on-road high emitters, traffic lights and vehicle cleaning frequency, number of vents in each car and size of vents, have not been fully investigated.

This study aims to assess robustly the most pertinent environmental, vehicle and driving related-parameters that affect in-vehicle exposure to NO_2 and $PM_{2.5}$. It provides an exposure assessment of these concentrations across 33 in-vehicle monitoring trips (of 10 vehicles) and develops a stepwise general additive mixed model (sGAMM) in order to identify important controllable reduction factors that can minimise in-vehicle exposure.

2. Methods

2.1. Experimental, instrumentation and data collection

Measurements of NO_2 and $PM_{2.5}$ were conducted concurrently in-vehicle and on-road using real-time online certified monitors. In-vehicle measurements also included temperature, humidity, and were complemented with global positioning system measurements that also included vehicle speed. The 33 monitoring trips of 10 vehicles took place in Birmingham, UK from summer until winter, on a consistent route during weekdays following the quality assurance recommendations for mobile measurements (Alas et al., 2019; Matthaios et al., 2020). The trips were conducted during normal daytime traffic hours between (10:00–16:00). Each trip lasted approximately 2.5 h, which included multiple loops on the same route, while each loop lasted around 20–22 min (Fig. 1). For every loop we tested one ventilation setting. The three ventilation settings that we tested were: 1) AC: Air condition on, windows closed, recirculation off; 2) RC: Recirculation on, windows closed air condition off; 3) WO: windows open, air condition and recirculation off. Each vehicle was measured three times 1) with the original filter installed (filter already in place for regular use), 2) with a new standard pollen and 3) with a new activated charcoal cabin air filter. After 3 months of use of the activated charcoal filter, three of the vehicles were re-measured again. Table S1 shows the vehicle characteristics.

For in-vehicle/on-road NO_2 measurements we used two certified Thermo Scientific chemiluminescent (EN14211:2012 NO_x) analysers – models 42i-TL (outside) and 42i (inside). For in-vehicle/on-road $PM_{2.5}$ we used two certified TSI3330 instruments. For each trip the instruments were mounted within the cabin of each vehicle, with two equivalent inlets with one protruding into ambient air, and the other into the cabin space in the driver's breathing zone. The instruments were logged via a single laptop and all measurements were averaged to a 1-min time resolution, with offsets adjusted for different inlet residence times (due to instrument flow rate) to ensure consistency between the datasets. The instruments were serviced and calibrated by the manufacturers before the campaign and prior to the start of the campaign they were co-located next to a reference instrument for 2 days at the University of Birmingham Air Quality Supersite for inter-calibration. All instruments were co-located at the same location after the measurement campaign to examine potential shifts in the inter-calibration of measurements. Figs. S1 and S2 in the supplementary shows that the inter-calibrations between the two NO_x and $PM_{2.5}$ monitors and the reference NO_x and $PM_{2.5}$ instrument were similar before and after the campaign ($R^2 > 0.90$ for $PM_{2.5}$ and $R^2 > 0.95$ for NO_2 in both cases) giving confidence in the quality of the measurements.

2.2. Generalised additive model analysis

To assess which parameters affect in-vehicle levels of NO_2, and $PM_{2.5}$, we applied a stepwise generalised additive mixed model (sGAMM) technique (Hastie, 2020). GAMMs allow the modelling of non-linear data while maintaining explainability, since they can be more flexible with the use of smoothing approximations. GAMMs have overall better predictions than linear and generalised linear models since they assume that the relationship between independent and dependent variables are not linear and are especially useful when the relationships between response variables and covariates are not known (Hastie and Tibshirani, 1990). Stepwise GAMM is a step-by-step iterative construction of a GAMM regression model that involves the selection of independent variables based upon comparisons with all possible models that can be created based upon an identified set of predictors. A bidirectional elimination sGAMM was used, which is a combination of forward selection and backward elimination models that test variables that should be included or excluded. In other words, a series of models is fitted, each corresponding to a formula obtained by moving each of the terms one step up or down in its regimen, relative to the formula of the current model. If the current value for any term is at either of the extreme ends of its regimen, only one rather than two steps are considered (Hastie, 1992). In this study, the best model is determined by the Akaike information criterion (AIC). The entire process is repeated until either the maximum number of covariates has been used, or until the AIC criterion cannot be further decreased. The stepwise GAMM model that we used is described in Eq. (1):

$$g(E(Y)) = b + 1_1 + x_1 + log(x_1) + s_1(x_1)\ldots + 1_p + x_p + log(x_p) + s_p(x_p) + Zk \quad (1)$$

where Y is the dependent variable (in-vehicle NO_2 or $PM_{2.5}$), E(Y) denotes the expected value, and g(Y) denotes the *link function* that links the expected value to the predictor variables $x_1 \ldots x_p$. Each variable in the stepwise selection model could be present as not at all, in a linear form,

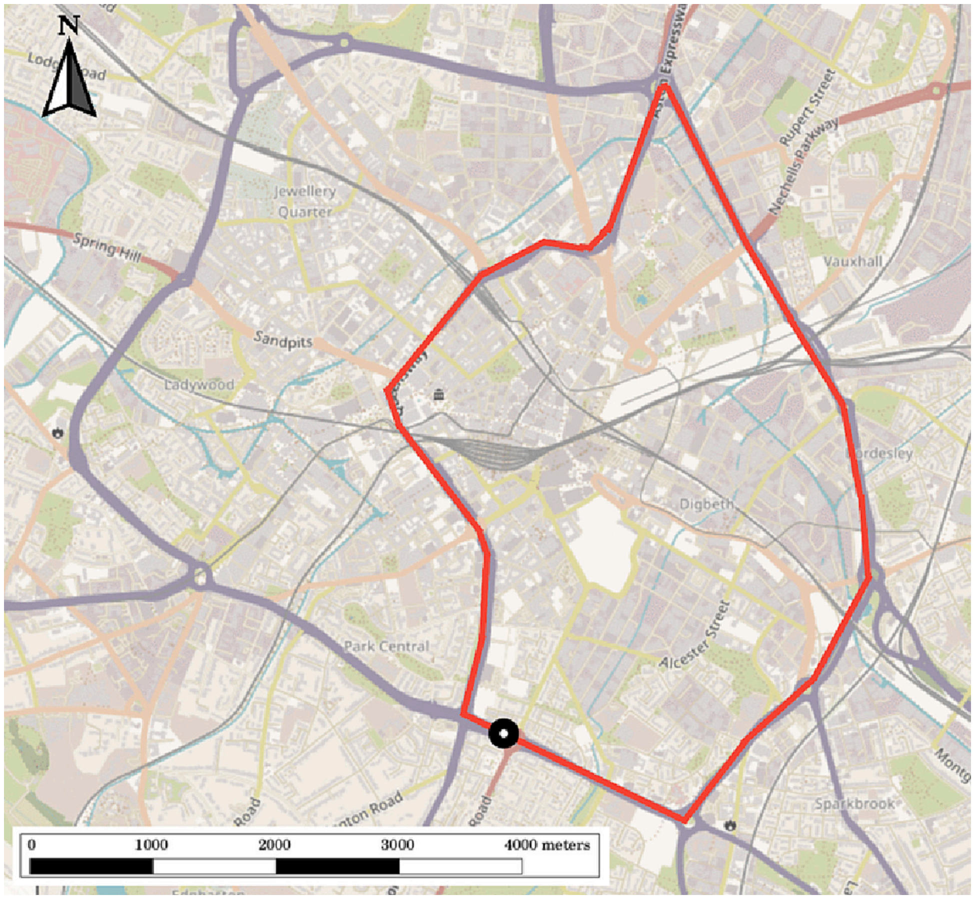

Fig. 1. Map of Birmingham UK. The red line shows the experimental route. The black dot shows the reset point of each loop.

linear in a logarithmic form or nonparametrically. The terms $1_1 \ldots 1_p$ indicate that the covariate is not included in the model, $x_1 \ldots x_p$ denote that the covariates exist linearly in the model, $log(x_1) \ldots log(x_p)$ express covariates that have been included linearly in their logarithmic form, while $s_1(x_1) \ldots s_p(x_p)$ denote covariates as smooth functions that are estimated nonparametrically (Hastie, 2020). The smooth functions are estimated using a variety of techniques, such as splines or penalized regression, and the level of smoothing is controlled by the effective degrees of freedom (edf) associated with each smooth term. Edf represents the amount of flexibility that the smooth term allows in the model, with a lower edf indicating a more constrained (linear) fit and a larger edf indicating a more complex (non-linear) fit. In the model *Zk* represents the random intercept for each car. On-road NO_2, $PM_{2.5}$, NO_2 photolysis rates, temperature, relative humidity and vehicle driving speed were used as numerical variables, while odometer reading, cabin size, number of vents, size of vents, fan speed power, ventilation type, cabin filter type, presence of traffic light, roundabouts, tunnels, leading vehicle high emitters and traffic conditions were used as categorical variables. Roundabouts, traffic lights and tunnels, were defined as the noted times where the driving vehicle was entering and leaving these locations. Traffic conditions were assessed empirically by the volume of vehicles on the road on the driving direction of the vehicle flow. High traffic conditions that led to congestion were defined with 3 while free flow conditions were defined as 0. High emitters were defined empirically by 1) light and heavy-duty vehicles were in front of the car; 2) the colour of the exhaust plume (darker for high emitters); and 3) by noting down the licence plates of these vehicles and checking their age and Euro classes (Euro 4 or earlier were considered as high emitters). Overall, we used 52 variables to select the most significant ones via sGAMM.

To identify the relative importance of individual predictors in the final sGAMM models we fitted alternative GAMM models without each term of the final sGAMM model (Matthaios et al., 2022a), and calculated the reduction in deviance. To assess the predictive power, the generizability and the robustness of sGAMM selection process a cross-validation procedure was performed. The data was split into training and testing data (encompassing 75 % and 25 % of the points respectively) and computed the out of sample prediction accuracy and distribution of selected features. This process was repeated 100 independent times; The distribution of the feature selection (Fig. S3) shows agreement with the main sGAMM findings. 10 fold cross-validation (Table S2) was carried out using gamclass package in R (Maindonald, 2020) and shows good agreement in both R^2 and in root mean square error.

3. Results

3.1. Vehicle and measurement campaign characteristics

The characteristics of the studied vehicles and are listed in Table 1. From the total experiments, (80 %) occurred during June, July, August, and September and 20 % during October, November, and December. The majority of the tested vehicles were >10 years old (60 % manufactured prior to 2011) with half of them being gasoline. Most vehicles had cabins volumes >2.5 m^3 and between 4 and 6 cabin vents, while the interior material was mainly polyester. Most of the tested vehicles had not been cleaned for >3 months and were last serviced between 6 and 12 months since the experiments. Because most experiments were conducted during the summer, the most used ventilation setting was air condition on with windows closed, followed by open windows and thirdly recirculation on with windows closed. The traffic conditions varied during the experiments with 42.2 % of the measurements being

Table 1
Vehicle and experimental characteristics of in-vehicle NO_2 and $PM_{2.5}$ experiments. No of vehicles = 10, No of experiments = 33, No of observations = 4312.

Variables	No. of observations (%)
Manufactured year	*N* = 10
<2011	6 (60 %)
2011–2018	2 (20 %)
2018–2021	2 (20 %)
Type	N = 10
Gasoline	5 (50 %)
Diesel	2 (20 %)
Hybrid	2 (20 %)
Electric	1 (10 %)
Type of Euro class	N = 10
<Euro 4	1 (10 %)
Euro 4	5 (50 %)
Euro 5	0 (N/A)
Euro 6	3 (30 %)
Engine size (L)	N = 10
≤1.4	(30 %)
>1.4	(60 %)
Cabin volume (m^3)	N = 10
≤2.5	(40 %)
>2.5	(60 %)
Number of vents	N = 10
≤4	2 (20 %)
>4 & ≤6	7 (70 %)
>6 & ≤8	1 (10 %)
Upholstery material	N = 10
Leather	2 (20 %)
Polyester	7 (70 %)
Other	1 (10 %)
Cleaning frequency	N = 10
Every week	1 (10 %)
Every month	1 (10 %)
Every 3 months	3 (30 %)
>3 months	5 (50 %)
Time since last service	N = 10
≤3 months ago	2 (20 %)
>3 & ≤6 months ago	1 (10 %)
>6 & ≤12 months ago	6 (60 %)
>12 months	1 (10 %)
Cabin filter	*N* = 33
Old-activated carbon	12 (36.4 %)
New activated carbon	10 (30.3 %)
New standard pollen	10 (30.3 %)
Ventilation type	*N* = 4312
Windows open	1044 (24 %)
Air condition on	2586 (60 %)
Recirculation on	688 (16 %)
Ventilation fan power (%)	N = 4312
25	382 (8.9 %)
50	794 (18.4 %)
75	282 (6.5 %)
100	1128 (26.2 %)
Air purifier (Y/N)	660 (15.3 %)
Traffic conditions	N = 4312
Low	116 (2.7 %)
Medium	1470 (34.1 %)
Heavy	906 (21 %)
Congested	1820 (42.2 %)
Location	N = 4312
Tunnel	440 (10.2 %)
Traffic light	1009 (23.4 %)
Roundabout	777 (18 %)
High emitters (Y/N)	1462 (33.9 %)

under congested periods.

3.2. *In-vehicle exposure to NO_2 and $PM_{2.5}$*

Fig. 2 shows the mean in-vehicle NO_2, and $PM_{2.5}$ concentrations measured across the tested vehicles. The in-vehicle NO_2 levels (mean ± sd) varied from 11 ± 15 to 24 ± 18 μg/m^3 with an overall mean value of 19 ± 18 μg/m^3 for NO_2. In-vehicle $PM_{2.5}$ experience less variation with an overall mean of 6.4 ± 2.7 μg/m^3 across the vehicles, varying from 5.2 ± 1.8 to 6.9 ± 3.4 μg/m^3. Fig. 3 shows the mean in-vehicle NO_2, and $PM_{2.5}$ concentrations across different ventilation settings and cabin air filters. The data show that there are higher NO_2 values with standard pollen filters than activated carbon filters while the old activated carbon filters also show elevated in-vehicle NO_2 comparing to new activated carbon filters. It is notable that with new activated carbon filter in use, in-vehicle NO_2 concentrations are low and similar when fresh air is coming from outside (AC on: NO_2 = 5.75 ± 5.0 μg/m^3) and when the air is recirculated through the cabin (RC on: NO_2 = 4.24 ± 4.1 μg/m^3) and only increase when the windows are open (Fig. 3A_i). In-vehicle NO_2 levels are substantially elevated with 6–15-month-old-activated carbon filters to 26 ± 16.7 μg/m^3 and 17.3 ± 10.5 μg/m^3 under AC and RC on respectively (Fig. 3A_{iii}). As the activated carbon filter ages its filtration capabilities decrease hence the $PM_{2.5}$ under recirculation increase from 2.5 μg/m^3 (under new pollen & activated carbon filters) to 4.7 μg/m^3 (Fig. 3B_{iii}). Similar behaviour to in-vehicle concentrations described above, is observed to the in-vehicle: on-road (I/O) ratio of NO_2 and $PM_{2.5}$ (Fig. S4).

3.3. *Factors influencing in-vehicle exposure*

Table 2 shows the results from the selected factors of sGAMM model for NO_2 and $PM_{2.5}$ in-vehicle exposures. From the sGAMM predictors listed in Table 2, it is evident that in-vehicle exposure to both examined pollutants was found to be associated with different characteristics related to environmental, vehicle, and driving conditions. Overall, the NO_2 sGAMM model explained 64.7 % of in-vehicle NO_2 variability, showing that the selected parameters reflect the key processes that influence in-vehicle NO_2 levels. From the final NO_2 sGAMM model, the outdoor NO_2 concentration was the most important predictor and explained 22.3 % of the model's explained variability. However, the relationship between in-vehicle NO_2 and on-road NO_2 is non-linear with edf =2.62. The NO_2 photolysis rates explained an additional 7 % of the variability, where in-vehicle NO_2 levels were significantly (p < 0.05) positively associated with greater photolysis rates. Moreover, in-vehicle concentrations were significantly (p < 0.05) positively associated with the use of pollen and old activated carbon filters, while cabin size and use of new activated carbon filters were inversely associated with in-vehicle NO_2 levels. In-vehicle NO_2 levels were also positively associated with windows open and greater fan speed power and negatively associated with the use of air recirculation in mechanical ventilation. The vehicle filter-related factors had a combined explained variability of 23.4 %, while the vehicle ventilation-related factors had a joined variability of 18 %. Odometer reading also affected in-vehicle NO_2 levels in a non-parametric way with a weakly non-linear (1 < edf < 2) relationship and had a variability of 2.5 %. Driving speed and traffic lights were significantly negatively (p < 0.05) associated with in-vehicle NO_2 while road traffic conditions and high leading vehicle emitters had a significant positive association with in-vehicle NO_2 exposures. Traffic conditions were an important predictor, accounting for 13.3 % of the model's explained variance, while the remained driving and road originated factors accounted for 8.7 %. On-road levels might be affected by the driving conditions therefore we added interaction terms in the model between the on-road NO_2 and traffic lights and traffic conditions. The results showed that individually both on-road NO_2 and traffic lights, traffic conditions significantly affect in-vehicle NO_2, however their interactions (combined effect) do not affect in-vehicle NO_2 (Tables S3–S4 model output). In other words, the presence or absence of traffic lights and road traffic conditions does not modify the relationship between on-road NO_2 and the in-vehicle NO_2. The same interaction was applied to $PM_{2.5}$ with similar results (Tables S5–S6 model output).

The sGAMM model for $PM_{2.5}$ explained 72.4 % of in-vehicle $PM_{2.5}$ exposures. In-vehicle $PM_{2.5}$ was significantly positively (p < 0.05) associated with on-road $PM_{2.5}$ and ambient temperature. On-road $PM_{2.5}$ was again the most important factor and accounted for 30 % of the

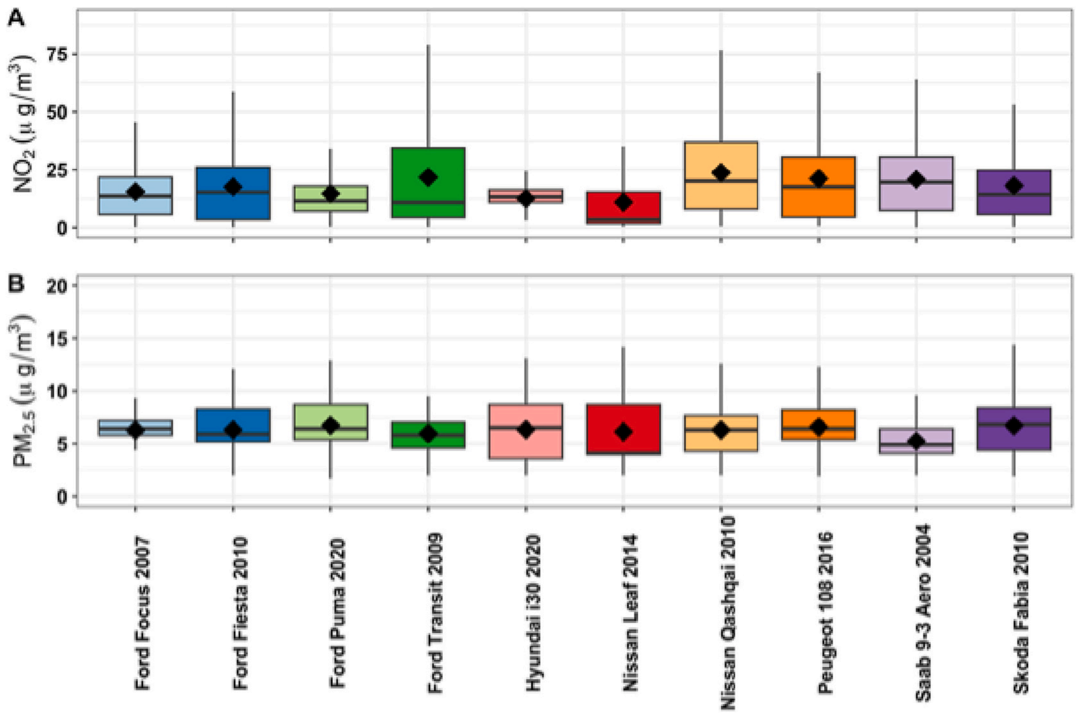

Fig. 2. In-vehicle NO_2 and $PM_{2.5}$ concentrations across the 10 different tested vehicles.

model's explained variability, while ambient temperature explained 1.4 %. $PM_{2.5}$ was significantly negatively associated with vehicle age, cabin size, the use of new activated carbon and pollen filters and the use of air recirculation in mechanical ventilation, and positively associated with periods of opened windows. These vehicle-related parameters together had a variability of 56.9 % with the ventilation related factors accounting for a majority of 40.1 %. The use of indoor air purifiers was negatively associated with $PM_{2.5}$ levels and accounted for another 4.4 %, while driving speed, traffic conditions and roundabouts were all significantly positively associated with in-vehicle $PM_{2.5}$ levels accounting together for 7.4 % of the variance. Fig. S5 shows that both sGAMMs models performed well capturing their respective in-vehicle response variables.

4. Discussion

The present study provided a comprehensive assessment of in-vehicle exposures of vehicle occupants to NO_2 and $PM_{2.5}$ and the factors that influence these exposures in Birmingham UK by including >52 variables for analysis of in-vehicle air quality. The greatest variation (in the 25th–75th percentile) of in-vehicle NO_2 levels found for the Ford Transit Van 2009, probably due to the condition of the car (this is an old converted mobile lab van maintained by the university). The in-vehicle NO_2 levels were generally lower than the 24-h WHO guideline value of 25 µg/m^3 (WHO, 2021) and were one third of those previously reported not only in Birmingham UK (Matthaios et al., 2020), but also in other UK cities (Panchal et al., 2022), and significantly lower than those measured for taxi drivers in London while at work (Bos et al., 2021), and in Sydney (Martin et al., 2016), Tokyo (Yamada et al., 2016), Cairo (Abbass et al., 2020) and Canadian cities (Weichenthal et al., 2015). In-vehicle $PM_{2.5}$ were similar to those reported in Sacramento, USA (Ham et al., 2017) and Leicester, UK (Panchal et al., 2022); however, they were lower than the 24-h WHO guidelines and lower than those reported in Paris, France (Hachem et al., 2021), Xian, China (Qiu et al., 2017) and other cities globally (Kumar et al., 2021). $PM_{2.5}$ exposures under different ventilation settings were also only about half of those for similar studies that reported in-vehicle $PM_{2.5}$ (Alameddine et al., 2016; Ham et al., 2017; Buitrago et al., 2021). Part of these relatively low concentrations for the in-vehicle NO_2 and $PM_{2.5}$ might be explained by the experimental conditions. The measurements occurred in 2021 just after the Birmingham city council implemented a clear air zone in the summer of 2021, and during the same period there was a decrease in of new registered vehicles, as well as a shift in the fleet from diesel to ultra-low emission plug-in hybrid and electric vehicles. (DfT, 2021).

Under activated carbon cabin filter and mechanical ventilation settings both NO_2 and $PM_{2.5}$ increased as the cabin air filter in use got older. After the new activated filter has aged (3 months old or more) it is observed that when the air is coming from outside through the air conditioning system, the in-vehicle NO_2 concentrations is similar to those outside, which is also similar to when the windows are open. These concentrations only decrease (due to decrease of air exchange) when the air is recirculated inside the cabin. Similar behaviour to the old activated carbon filter is observed when the new pollen filter is in use. $PM_{2.5}$ exposures, on the contrary, are more ventilation dependent. $PM_{2.5}$ is high in general when fresh air is coming into the cabin either via mechanical ventilation (AC on) or when the windows are open and is reduced when the air is recirculated inside the cabin. This behaviour is observed with both new activated carbon filters and new pollen filters; however, The aging of activated carbon filters resulted in increases of 78 and a 76 % in the in-vehicle NO_2 exposures in AC on and RC on ventilation settings, respectively. These results agree with recent studies that showed significant reductions 80–90 % to the in-vehicle NO_2 under new activated carbon filters compared to 44.2–8.6 % under aged activated carbon filters (Matthaios et al., 2023; Pöhler et al., 2018; Moldanova et al., 2019).

The sGAMM comprehensive variable selection analysis showed that on-road concentrations were a significant factor that influences in-vehicle exposure, which agrees with previous studies (Leavey et al., 2017). On-road levels were highly non-linearly associated to in-vehicle NO_2 revealing the complexity of using only a simple infiltration factor for the pollutant as proxy of its exposure that studies used in the past (Taylor et al., 2019; Ferguson et al., 2021). An environmental factor that affects in-vehicle NO_2 is the photolysis frequency for NO_2, which can affect NO_x partitioning. Ambient temperature was also positively

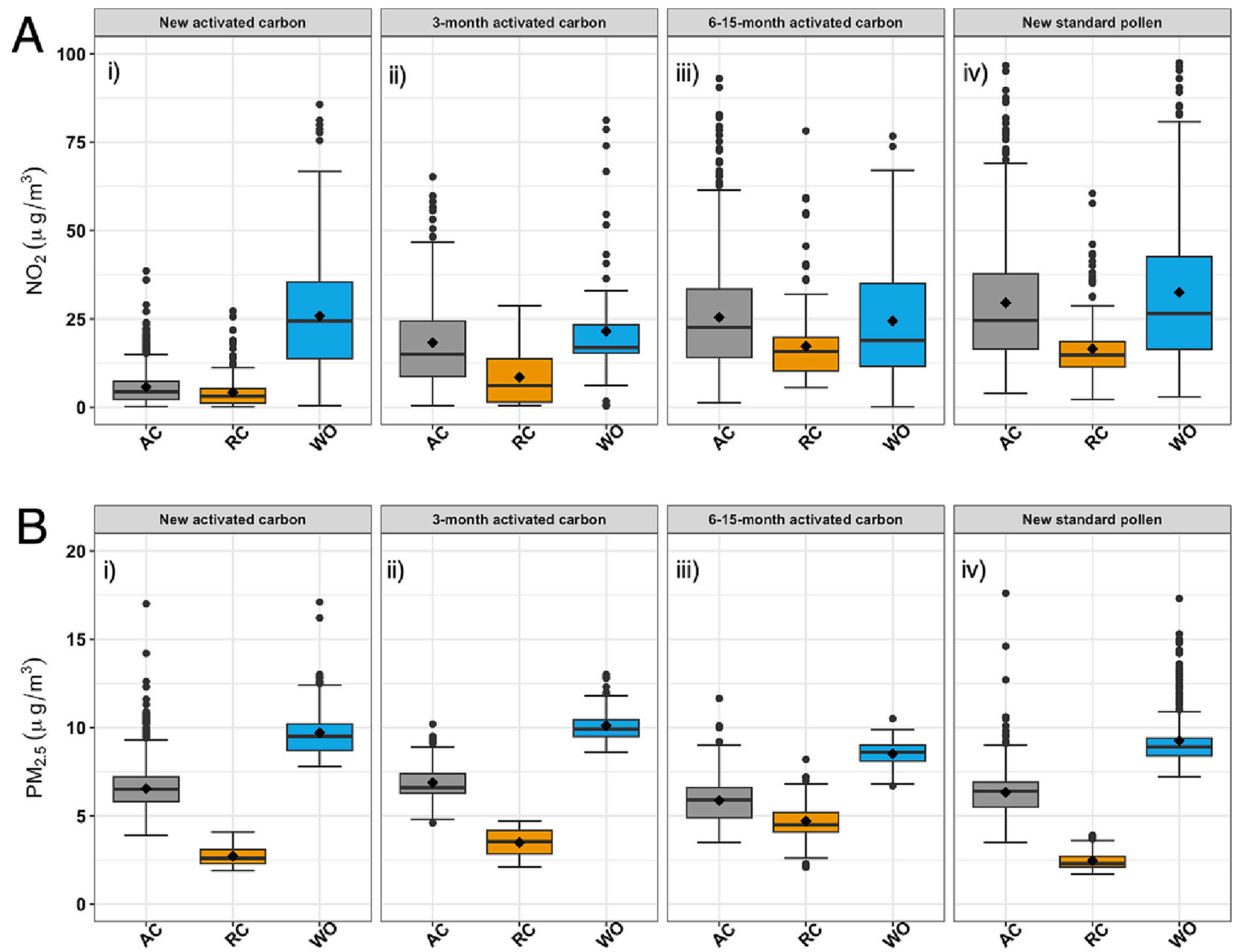

Fig. 3. In-vehicle NO_2 and $PM_{2.5}$ under different ventilation settings and filter types. AC: Air condition on, windows closed, recirculation off; RC: Recirculation on, windows closed air condition off; WO: windows open, air condition and recirculation off.

associated with indoor $PM_{2.5}$ as also reported elsewhere (Matthaios et al., 2022b); however, they explained <3 % of the variance, which is about one third less than other studies (Onat et al., 2019).

The results also show that vehicle-related important factors to exposure are manufacturing year, odometer, and cabin size. The negative association between NO_2 and $PM_{2.5}$ with manufacturing year, verify what previous literature reported that old vehicles are more leaky with less air tightness in the doors and windows (Knibbs et al., 2010); however, they also point out that pollutants such as NO_2 might be influenced less by air tightness characteristics and possibly to some extent by photolysis rates and in-vehicle oxidation chemistry. Cabin size was negatively associated with both pollutants in agreement with Hudda and Fruin (2013). Smaller cabins have greater surface area to volume ratio and probably higher deposition rates (Thatcher et al., 2002; Ott et al., 2008). Vehicles that have higher odometer reading indicates that they have been older and used more frequently, which might affect the air tightness of the vehicle. The use of either new activated carbon or pollen filters was found to reduce $PM_{2.5}$ concentrations inside vehicles, whereas NO_2 was only reduced with new activated carbon filters and increased with new pollen (non-activated carbon) and old activated carbon filters (Matthaios et al., 2023; Pöhler et al., 2018; Moldanova et al., 2019). Window opening increased both NO_2 and $PM_{2.5}$ concentrations inside vehicles, while the use of air recirculation in mechanical ventilation reduced them, with similar findings reported in the literature (Leavey et al., 2017; Abi-Esber and El-Fadel, 2013; Boogard et al., 2009). The use of air purifiers reduced $PM_{2.5}$ levels as also reported elsewhere (Tartakovsky et al., 2013); however, these are expensive devices and not convenient to operate inside cars as they often require power supplies. Their explained variance was 4.4 % comparing to 11.8 % of the implementation of the new filters that also cost only about one fifth as much as the indoor air purifier.

Driving-related parameters important for exposure included road traffic conditions, heavy emitters, traffic lights and roundabouts. Road traffic conditions are positively associated with NO_2 and $PM_{2.5}$, meaning that during these periods both NO_2 and $PM_{2.5}$ were elevated inside the vehicles. Road traffic conditions were the second most important predictor for in-vehicle NO_2 and a modest predictor for in-vehicle $PM_{2.5}$. Fruin et al. (2008) also reported that traffic density was important for both NO_2 and $PM_{2.5}$ for the in-vehicle levels in Los Angeles, while Weichenthal et al. (2015) reported similar percentage changes (around 4 %) for $PM_{2.5}$ in Canadian cities. Following a high emitter also increased in-vehicle NO_2 levels. Campagnolo et al. (2019) reported that the emission standards of the leading vehicles significantly impact air pollution levels inside the vehicle cabins. Traffic lights and roundabouts had small (<4.4 %) explained variability, where traffic lights were negatively associated with in-vehicle NO_2, and roundabouts were positively associated with in-vehicle $PM_{2.5}$. Traffic lights typically increase particle number concentrations due to rapid acceleration (Goel and

Table 2
Determinants of in-vehicle $PM_{2.5}$ and NO_2 levels as reported by the stepwise general additive mixed-effects model. Coeff: Coefficient of predictor; VIF: Variance inflation factor; S.E: standard error of coefficient. * indicates smoothing approximations.

	In-vehicle NO_2 ($N = 4318$)			In-vehicle $PM_{2.5}$ ($N = 4315$)		
	Coeff (S.E)	VIF	Relative importance (%)	Coeff (S.E)	VIF	Relative importance (%)
Environmental related						
On-road concentrations	2.62*	–	22.3	0.67 (1.0e−02)	1.36	30.0
On-road temperature	–	–	–	0.12 (1.8e−02)	4.37	1.4
Photolysis rates	0.53 (3.4e−02)	1.48	7.0	–		
Vehicle related						
Manufacturing year	−0.05 (1.5e−02)	1.23	2.1	−0.06 (9.0e−03)	1.95	2.4
Cabin size	−0.40 (2.3e−01)	1.45	2.7	−0.20 (7.0e−02)	3.17	2.6
Odometer	1.86*	1.14	2.5	–		
Use of activated carbon filter	−0.43 (3.4e−02)	2.95	11.1	−0.81 (5.2e−02)	1.36	5.4
Use of pollen filter	0.72 (3.6e−02)	2.99	6.2	−0.60 (5.3e−02)	1.44	6.4
Use of old activated carbon filter	0.55 (3.6e−02)	3.06	6.1	–		
Windows open	0.56 (3.0e−02)	1.98	10.1	2.17 (5.4e−02)	2.98	16.4
Use of recirculation ventilation	−0.13 (3.3e−02)	1.75	6.3	−3.28 (6.1e−02)	3.06	23.7
Ventilation fan speed power	0.07 (8.1e−03)	2.10	1.6	–		
Use of air purifier	–	–	–	−0.75 (9.5e−02)	2.20	4.4
Driving related						
Driving speed	−0.003 (1.3e−03)	4.28	2.2	0.01 (2.4e−03)	1.16	1.7
Traffic conditions	0.35 (1.9e−02)	3.45	13.3	0.16 (3.8e−02)	1.78	3.8
Traffic lights	−0.08 (3.8e−02)	3.19	4.4	–		
Roundabouts	–		–	0.11 (4.8e−02)	2.53	1.9
Heavy emitters	0.07 (3.0e−02)	2.23	2.1	–		
Adjusted R^2 (% variance explained)	0.645 (64.7)			0.723 (72.4)		

Kumar, 2014), however they can also act as media for traffic decongestion which can explain the negative association with in-vehicle NO_2. Roundabout locations can also have elevated $PM_{2.5}$ concentrations as vehicles may decelerate, make abrupt turns, and accelerate rapidly in order to enter and exit the roundabout. These driving conditions typically contribute to non-exhaust related particle emissions (Harrison et al., 2021), which might explain the positive association of in-vehicle $PM_{2.5}$ and roundabouts.

Overall, sGAMM mixed-effects model for $PM_{2.5}$ and NO_2 revealed different influences from factors that are related to both on-road (outdoor) and in-vehicle conditions. On-road and in-vehicle factors are associated with exposures in non-linear ways and further varied with vehicle characteristics, time of the day and location revealing the complexity of the problem and the additional research that is needed to determine the causality of these relationships. The study has a few limitations: The in-vehicle exposures reported here might not be representative for long-term measurements of occupant's exposure inside vehicles, given that they were only during summer and autumn periods in 2021. Since the experiments only occurred during normal daytime hours, they might not be representative for rush-hour periods where the $PM_{2.5}$ burden is often greater (Requia et al., 2018). Our analysis did not incorporate driving behaviour, which may have influenced the results. Instead, we relied solely on an approximation based on the measured driving speed for our analysis. The influences of the identified factors might vary in importance for other vehicles since we only examined a small fraction of vehicles related to the total fleet, however, ventilation settings and appropriate filtration media are expected to be similarly important and effective for every car and location. Variables such as road inclination, height of buildings divided by the width of street and percentage of urbanization and greenness were not included in the study and have been found to be associated with in-vehicle exposures (Lim et al., 2021). Their inclusion could explain some of the variability of these in-vehicle exposures and improve the predictive power of sGAMMs. The inclusion of multiple pollutants would also provide a more comprehensive understanding of in-vehicle air pollution. Even though the results look generalisable (as shown by cross validation) possible biases due to empirical estimates might affect the associations. For example, model driving related parameters of traffic congestion, presence of roundabouts, traffic lights and heavy emitters were estimated empirically. Empirical data can be subject to desirability bias or recall bias, leading to potential under or overestimation of the importance of these factors to in-vehicle exposures. To mitigate this bias, our statistical techniques were adjusted for known driving-related factors such as vehicle speed, geolocation and on-road air pollution level that are associated with traffic congestion and heavy emitters. Sensitivity analyses were performed by varying the number of selective factors distribution assumptions and assessing their impact on our model's findings. The results demonstrated that while the specific model selection estimates might vary, the identified exposure factors and their significance remained consistent. This indicates that our conclusions were robust to potential biases however since these findings might be case specific, further research is needed to confirm and expand upon these findings, particularly in different geographic regions, and with larger and more diverse sample sizes.

5. Implications for exposure

Despite the recent policy directions to promote public transport and active travel as a means of endorsing more sustainable modes of transportation in the UK, the rise of online shopping and on-demand food delivery and services is causing a shift in transportation needs (Piecyk et al., 2021; Allen et al., 2021; Schaller, 2021; Le et al., 2022). According to the Department for Transport in the UK (DfT, 2022), this transportation shift will result in increases in traffic growth, congestion periods and time spent inside vehicles, which are projected to increase over the coming years for both the general population and professional drivers. Therefore, it is of great importance to raise awareness about in-vehicle air quality issues among the general public, car manufacturers, urban planners and policy makers.

The overall NO_2 and $PM_{2.5}$ levels measured inside vehicles were lower than the recommended WHO guidelines (WHO, 2021), however studies have found adverse health effects of $PM_{2.5}$ and NO_2 for short exposure periods (Gaffin et al., 2018) and even below the recommended WHO guidelines (Yazdi et al., 2021; Shi et al., 2021) pinpointing the necessity to identify ways to reduce our exposure to air pollution even further. In a systematic review and meta-analysis, Cepeda et al. (2017)

showed that commuters that used motorised transport lost up to one year of life expectancy compared to cyclists, however they also found that vehicle passengers that used control ventilation settings have less exposure to $PM_{2.5}$ and NO_2 than vehicle occupants under no controlled ventilation. The present study offers novel insights into which factors affect these exposures and to what extent in-vehicle exposure to NO_2 and $PM_{2.5}$ can be reduced by managing environmental, vehicle and driving related parameters.

The study developed stepwise generalised additive mixed models and identified joined and individual factors that affect in-vehicle NO_2 and $PM_{2.5}$ levels. The model identified that the most significant predictor is the on-road air pollution. Furthermore, the model analysis showed that the association of on-road and in-vehicle air pollution is highly non-linear showing the complexity of exposure in one of the most occupied microenvironments globally. Additional improvement and control of the factors determining in-vehicle NO_2 and $PM_{2.5}$ levels that we evaluated in the present study may further improve health for vehicle occupants. Vehicle-based predictors included vehicle age, cabin size, odometer, use of activated carbon and pollen filters, window opening, use of air recirculation mechanical ventilation settings, the ventilation fan speed power and the use of air purifier, which explained 48.7 and 61.3 % of NO_2 and $PM_{2.5}$ respectively. Driving-based predictors included traffic conditions, high emitters, vehicle speed, traffic lights and roundabouts and accounted for 22 and 7.4 % of in-vehicle NO_2 and $PM_{2.5}$ exposure variability. These findings provide key information regarding controllable exposure factors in managing vehicle occupants' exposures and suggest that some factors can be moderated to reduce in-vehicle exposures in urban settings.

An effective way to minimise in-vehicle air pollution is by implementing the "Reduce-Extend-Protect" strategy (Hewitt et al., 2020). The primary focus should be on reducing overall transportation emissions. Increasing the distance between vehicles can also result in improved dispersion and dilution of pollutants, leading to reduced exposure. To protect themselves, vehicle occupants should close their windows and control their mechanical ventilation settings for optimal air exchange, while regularly maintain and replace cabin air filters, which can significantly reduce the impact of on-road air pollution inside the vehicle. These benefits are likely to be amplified for vehicle occupants spending the greatest periods of time within vehicles, including elderly, mobility impaired commuters and professional drivers.

CRediT authorship contribution statement

Vasileios N. Matthaios: Conceptualization, Methodology, Formal analysis, Writing – original draft, Funding acquisition. **Roy M. Harrison:** Validation, Writing – review & editing. **Petros Koutrakis:** Validation, Writing – review & editing. **William J. Bloss:** Validation, Writing – review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The dataset for in-vehicle/on-road $PM_{2.5}$ is available at https://doi.org/10.5281/zenodo.8183017. The dataset from the in-vehicle and on-road NO_2 measurements is available at https://doi.org/10.5281/zenodo.7388363.

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Appendix A. Supplementary data

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