

London Hybrid Exposure Model: Improving Human Exposure Estimates to NO₂ and PM_{2.5} in an Urban Setting

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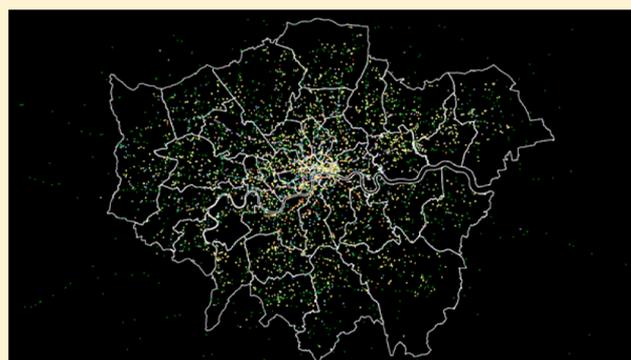
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S Supporting Information

ABSTRACT: Here we describe the development of the London Hybrid Exposure Model (LHEM), which calculates exposure of the Greater London population to outdoor air pollution sources, in-buildings, in-vehicles, and outdoors, using survey data of when and where people spend their time. For comparison and to estimate exposure misclassification we compared Londoners LHEM exposure with exposure at the residential address, a commonly used exposure metric in epidemiological research. In 2011, the mean annual LHEM exposure to outdoor sources was estimated to be 37% lower for PM_{2.5} and 63% lower for NO₂ than at the residential address. These decreased estimates reflect the effects of reduced exposure indoors, the amount of time spent indoors (~95%), and the mode and duration of travel in London. We find that an individual's exposure to PM_{2.5} and NO₂ outside their residential address is highly correlated (Pearson's *R* of 0.9). In contrast, LHEM exposure estimates for PM_{2.5} and NO₂ suggest that the degree of correlation is influenced by their exposure in different transport modes. Further development of the LHEM has the potential to increase the understanding of exposure error and bias in time-series and cohort studies and thus better distinguish the independent effects of NO₂ and PM_{2.5}.



1. INTRODUCTION

Epidemiological research has demonstrated associations between ambient air pollution and a range of health effects especially on the respiratory and cardiovascular systems.^{1–11} Epidemiological methods include estimating PM_{2.5} and NO₂ exposure metrics at a coarse spatial scale, using degrees of longitude/latitude,^{12,13} monitoring stations as proxies,^{2,10,14–16} or by considering the proximity of a subjects address to nearby roads.¹⁷ These studies often ignore the strong spatial concentration gradients and variability that exist within urban areas. More detailed approaches have evolved which estimate pollution concentrations at individual addresses or at the centroid of small areas such as postcodes using geostatistical interpolation,¹⁸ land-use regression,¹⁹ and dispersion models.^{20,21} However, these models do not take account of exposure within the home, at work/school, and in different travel microenvironments, despite significant differences between personal and outdoor exposures demonstrated using comparative measurement studies.^{22–24} Consequently, these methods contribute to random exposure error, which is likely to bias health associations to the null²⁵ and systematic error that

will bias health associations in either direction. Furthermore, the correlations between PM_{2.5} and NO₂ which tend to be observed using such methods make it difficult to investigate the independent associations of these two pollutants with health effects.^{26,27}

Among the first attempts to obtain more detailed exposure estimates was the EXPOLIS study framework by Kousa et al.²⁸ who evaluated the temporal and spatial exposure of the population of Helsinki, Finland in different microenvironments. More recently, Dhondt et al.²⁹ developed a population exposure modeling approach taking into account population mobility in two urban areas in Belgium, concluding that large differences in health impacts occur when assessments neglect population mobility. Recent studies have highlighted the importance of exposure to air pollution during trips in urban areas. De Nazelle et al.³⁰ found that while a number of

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individuals in Barcelona spent 6% of their time in transit, it contributed 11% of their daily exposure to NO₂. Likewise, Houston et al.³¹ concluded that although travel comprised 5% of a participant's day, it represented 27% of their daily exposure to particle-bound polycyclic aromatic hydrocarbons. Other studies have reported that commuters in urban areas receive 12% of their daily PM_{2.5} exposure and up to 30% of their black carbon inhaled dose while traveling.^{32–34} Evidence of this kind has led the Health Effects Institute (HEI) to suggest activity-based or hybrid exposure models, which combine space-time-activity data, personal measurements, and air quality models, come closest to a “best” estimate of human exposure.³⁵

Here we describe the London Hybrid Exposure Model (LHEM), calculating air pollution exposure at population level, while accounting for each individual's movements, during the day. We summarize the population's travel behavior and then compare LHEM exposure estimates with exposure at the resident's address, to assess exposure misclassification. We also assess whether the LHEM provides a better basis for investigating the independent effects of NO₂ and PM_{2.5} on health.

2. MATERIALS AND METHODS

Using the LHEM we estimate exposure to outdoor air pollution for the population of London while indoors, outdoors, and during journeys. The following sections describe the development of each component.

2.1. LHEM Time-Activity Data Set. Space-time-activity data for the LHEM is based upon the London Travel Demand Survey (LTDS),³⁶ provided by Transport for London (TfL) (David Wilby, personal communication) for the period 2005–2010. The LTDS data set is generated through interviews with approximately 8,000 households per annum to ascertain details of each person's daily trips, travel mode, trip purpose, and demographic data, among other factors. Household and person weighting factors, rebased following the 2011 Census³⁷ and calculated by TfL, allow the scaling of the LTDS data set (45,079 people) to represent the population of London (excluding children under 5) - approximately seven million people. For more details see Appendix A of the [Supporting Information \(SI\)](#).

2.2. Trip Route Simulation. The LTDS data set includes start and end coordinates, times of trips, and mode of transport but does not describe the routes taken. These have been simulated in a number of different ways. Before undertaking trip route simulation, the data was cleaned to remove missing or clearly incorrect data, for example, unrealistically quick journeys, missing origin or destinations, and missing journey sections. This removed 9% of the data, leaving a final data set of 45,079 people taking 98,770 trips and a total of 340,754 stages (representing different transport modes within the same trip). To calculate the routes, start and end coordinates, times, and transport mode of each stage were processed in the R Statistical Computing Environment³⁸ using the RPostgreSQL³⁹ extension. Specifically, the start and end points of each stage were formed into URL strings and processed by a number of different routing application programming interfaces (APIs). The Open Route Service API⁴⁰ was used to simulate walking trips (shortest-path), the Project OSRM API⁴¹ to simulate car trips (quickest-path), Google Directions⁴² to simulate cycling (quickest-path), and the TfL Journey Planner⁴³ to simulate public transport trips (overground train, the London Underground, the Docklands Light Railway, and bus). These APIs

returned routes between the start and end locations, which were stored in a PostGIS database, and split into points for each minute of the journey using linear interpolation. For periods between trips, people were assumed to stay indoors at the previous destination point. The resulting data set gave the location and environment (indoor, walking, cycling, driving, etc.) of each of the 45,079 LTDS respondents on a minute-by-minute basis over 24 h.

2.3. Exposure Estimates. **2.3.1. Outdoor Air Pollution Predictions.** Exposure to outdoor air pollution was provided by CMAQ-urban,⁴⁴ which couples the Weather Research and Forecasting (WRF) meteorological model,⁴⁵ the Community Multiscale Air Quality (CMAQ) regional scale model,⁴⁶ and the Atmospheric Dispersion Modeling System (ADMS) roads model.⁴⁷ The CMAQ-urban model output was processed to give annual average hour of day NO_x (NO + NO₂), NO₂, NO, and PM_{2.5} concentrations in 2011, across a 20 m × 20 m grid over the UK (Figure S1, SI). CMAQ-urban was run without bias correction and has previously been submitted to the UK Model Intercomparison exercise run by the UK Government department DEFRA,⁴⁸ performing well against other urban and regional models. In Appendix C and Figure S2 (SI) we summarize the CMAQ-urban model evaluation, which has *r* values of 0.9 (NO_x/NO₂) and 0.77 (PM_{2.5}). The bias for NO₂ and PM_{2.5} is approximately 3% and –10%, respectively.

2.3.2. In-Building Exposure. To calculate indoor exposure to outdoor sources of PM_{2.5} and NO₂ using the LHEM, we apply indoor/outdoor (I/O) ratios for domestic properties to the outdoor CMAQ-urban. As all I/O ratios are less than 1, this means that exposure indoors is less than outdoors at the same location. The modeling methods are described in full in Taylor et al.⁴⁹ but briefly; models were run for 15 building types, derived from the English Housing Survey⁵⁰ and Geo-information Group classifications,⁵¹ which represent 76% of the known London housing stock. The building physics model (Energy Plus 8.0⁵²) estimated dwelling I/O ratios for PM_{2.5} and NO₂ using background air infiltration and exfiltration; indoor and outdoor temperature-dependent window opening, representing realistic occupant behavior; and deposition rates and penetration factors (detailed in Appendix D, SI). Air infiltration and exfiltration were modeled by assigning permeabilities—typical of the age, type, and construction of the buildings, allowing the model to account for infiltration due to wind pressures and buoyancy effects. Buildings were modeled using a Test Reference Year (TRY) weather file for London, representing “typical” weather conditions, and derived from a 30 year baseline, with results output hourly for a year-long simulation. The ratios were summarized by hour of the day and weekday/Saturday/Sunday to match the temporal resolution of the CMAQ-Urban data set and then averaged for each postcode in Greater London. I/O ratios for offices were assumed to have the same value as houses in the same postcode. [Figure 1](#) shows a map of daily average PM_{2.5} ratios (although hourly ratios are used in the model). A similar map of 24 h average I/O ratios for NO₂ is given in [Figure S3 \(SI\)](#). For the postcodes in London where there was insufficient housing stock information to calculate ratios (white areas, [Figure 1](#)), the London average was used (0.31 for NO₂ and 0.56 for PM_{2.5}).

2.3.3. In-Vehicle Exposure. For in-vehicle exposure, the pollutant concentration is derived by solving the mass balance equation^{53–55} (1), explained further in Appendix E (SI)

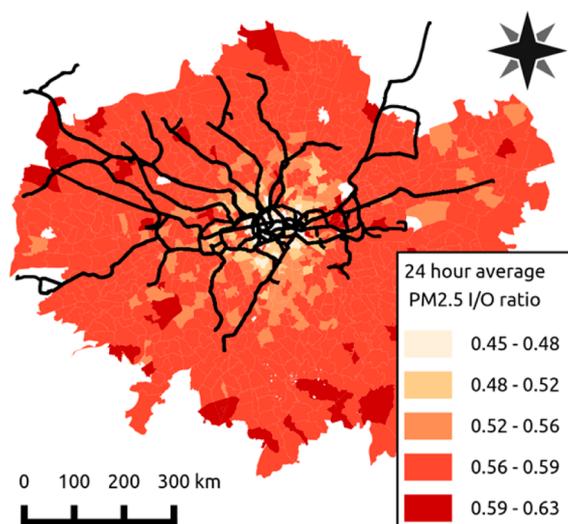


Figure 1. Map of 24 h average indoor/outdoor (I/O) ratios used in the LHEM. The I/O ratios for $\text{PM}_{2.5}$ are smaller in inner London than outer London providing the most protection in areas where the outdoor pollutants are highest. NO_2 (see Figure S3) has a similar pattern between central and outer London but has lower I/O ratios (0.11 to 0.59). Superimposed on the map is the London Underground network to aid orientation.

$$\frac{dC_{\text{in}}}{dt} = \lambda_{\text{win}}(C_{\text{out}} - C_{\text{in}}) - n\lambda_{\text{HVAC}} \cdot C_{\text{in}} - V_g \left(\frac{A^*}{V} \right) \cdot C_{\text{in}} + \frac{Q}{V} \quad (1)$$

where C_{out} is the outdoor concentration surrounding the vehicle, λ_{win} and λ_{HVAC} are the hourly air exchange rates from the windows and mechanical ventilation system, respectively, η is the filter removal efficiency taking values between 0–1, V_g is the deposition velocity in m h^{-1} , A^* is the internal surface area available for deposition, V is the volume of the vehicle, and Q is the in-vehicle particle emission rate in $\mu\text{g h}^{-1}$ (defined as the product of resuspension rate and number of passengers not seated in the vehicle). The total surface area for deposition A^* is calculated by adding the internal surface area of the vehicle A to the surface area of each passenger.⁵³

The ventilation conditions in cars were assumed to be constant throughout the journey with the assumption of no indoor sources, whereas the ventilation conditions were modified by the regular stops of buses and trains, with the model parameters reset at each stop, remaining constant between stops. Resuspension from people's movements was assumed to be the only indoor source of particles in buses and trains with no indoor sources for gaseous pollutants assumed. Details of the parameters used in the in-vehicle mass balance model are provided in Table S2 and Appendix E (SI).

Finally, for the London underground, concentrations were taken to be $94 \mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$ and $51 \mu\text{g m}^{-3}$ for NO_2 . These figures were based upon $\text{PM}_{2.5}$ measurements conducted on London Underground platforms and trains during 22 journeys, over 450 min, using a TSI Sidepak AMS10⁵⁶ (Benjamin Barratt, personal communication) and from NO_2 measurements on the Paris Metro.⁵⁷

2.3.4. Walking and Cycling Exposure. Exposure while walking or cycling was taken from time and location specific CMAQ-urban model concentrations.

By combining these methods, the LHEM calculates exposure minute-by-minute for 45,079 people, representing 6.8m

Londoners. Using these data we calculate the population average daily exposure and the contribution from each indoor, in-vehicle and outdoor microenvironment. LHEM is able to simulate exposure to $\text{PM}_{10/2.5}$, PM components, NO_x/NO_2 , and O_3 , as well as the exposure to separate sources, although here we have limited our analysis to NO_2 and $\text{PM}_{2.5}$.

3. RESULTS

3.1. Travel and Exposure to NO_2 and $\text{PM}_{2.5}$ in London.

The results in Table 1 have been split into age categories, totalling 6.8m people. For each of these age categories results have been given for the number of people, the inner/outer London split, the percentage of the day spent in different microenvironments, their trip details during the day, and exposure to NO_2 and $\text{PM}_{2.5}$ in each microenvironment as a percentage of their total daily exposure. Missing data and data “not recorded” represented <0.01% of the total data set.

Results from the LHEM show that on average people spend over 95% of their time indoors, predominantly at home or at work, and that this proportion of time varies by only 2–3% across the age ranges and includes approximately 20% of people who, when surveyed, did not leave their house. Active travel (cycling and walking) takes up about 1–1.7% of the time during a typical day with inactive travel (driving, on the bus, underground, train, motorcycle) taking 1.4 to 3.6%. This translates into the number of trips being approximately 3 for children to almost 7 for adults, with mean journey times and journey lengths ranging between 12.8 min and 3.1 km for children and 26 min and 8 km for adults.

The percentage of daily exposure indoors to outdoor NO_2 and $\text{PM}_{2.5}$ sources ranges between 92% for children and 81% for adults and between 96% for children and 88% for young adults, respectively. Inactive travel in London results in 2.4 times greater exposure to NO_2 than active travel. Inactive travel results in 3.2 times greater exposure to $\text{PM}_{2.5}$ than active travel. People's trips contribute approximately 15% of their daily NO_2 exposure and 9% of their daily $\text{PM}_{2.5}$ exposure.

3.2. NO_2 and $\text{PM}_{2.5}$ Exposure and Exposure Misclassification. Exposure misclassification is an important consideration in estimating exposure to air pollution in health studies. Figure 2 (NO_2 left panel and $\text{PM}_{2.5}$ right panel) shows LHEM exposure estimates compared to the equivalent exposure estimated at the residential address.

For NO_2 , the mean LHEM exposure of $13.0 \mu\text{g m}^{-3}$ (median exposure $12.4 \mu\text{g m}^{-3}$) is approximately 63% lower than the mean NO_2 exposure at the residential address of $34.6 \mu\text{g m}^{-3}$ (median $34.8 \mu\text{g m}^{-3}$). The mean $\text{PM}_{2.5}$ LHEM exposure of $8.5 \mu\text{g m}^{-3}$ (median $8.2 \mu\text{g m}^{-3}$) is approximately 37% lower than the $\text{PM}_{2.5}$ mean exposure at the residential address of $13.5 \mu\text{g m}^{-3}$ (median $13.6 \mu\text{g m}^{-3}$).

Within city variability is important in determining associations with health risks, and for $\text{PM}_{2.5}$, the range in LHEM exposure is between $6.0 \mu\text{g m}^{-3}$ and $32.2 \mu\text{g m}^{-3}$, larger than the range at the residential address ($11.2 \mu\text{g m}^{-3}$ – $20.0 \mu\text{g m}^{-3}$). The LHEM $\text{PM}_{2.5}$ exposure estimates are skewed with a long tail of high exposure concentrations. In contrast, the range in LHEM NO_2 exposures is between $4.3 \mu\text{g m}^{-3}$ and $55.3 \mu\text{g m}^{-3}$, smaller than the range at the residential address ($17.8 \mu\text{g m}^{-3}$ – $88.1 \mu\text{g m}^{-3}$).

To establish more clearly the reasons for the differences in the exposure distributions of the LHEM and residential address methods, scatter plots of $\text{PM}_{2.5}$ and NO_2 are given in Figure 3,

Table 1. Summary of the Time Spent in Each Microenvironment, Trip Details, and the Exposure of London's Population to NO₂ and PM_{2.5} from Outdoor Sources on an Average Day

	child (5–17)	young adult (18–29)	adult (30–59)	elderly (60+)	all ages (≥5)
no. of people	1,620,578	1,156,831	2,932,228	1,124,131	6,833,768
percentage of age group in inner/outer London					
inner London	37%	47%	42%	30%	40%
outer London	63%	53%	58%	70%	60%
percentage of time spent in each microenvironment					
driving	0.8	1.4	2.2	1.5	1.6
indoor	97.7	94.9	94.7	96.4	95.7
walk	0.9	1.7	1.5	1.2	1.3
Underground and DLR	0.1	0.7	0.5	0.2	0.4
bus	0.5	0.9	0.7	0.6	0.7
cycle	0.0	0.1	0.1	0.0	0.1
train	0.0	0.2	0.2	0.1	0.2
motorcycle	0.0	0.1	0.1	0.0	0.0
av number of trips	2.8	5.4	5.6	4.0	4.6
av trip time (mins)	12.8	27.2	26.1	18.4	21.6
av trip length (km)	3.1	7.7	8.1	5.4	6.4
av NO ₂ exposure (μg m ⁻³)	11.9	13.7	13.6	12.2	12.9
percentage of daily NO ₂ exposure by microenvironment ^a					
driving	3.0	5.3	8.5	5.6	6.2
indoor	92.0	82.0	81.2	88.0	85.0
walk	2.6	5.4	4.7	3.3	4.1
Underground and DLR	0.2	2.5	1.7	0.6	1.3
bus	2.0	3.6	2.5	2.2	2.5
cycle	0.1	0.3	0.4	0.1	0.2
train	0.1	0.6	0.5	0.2	0.4
motorcycle	0.0	0.1	0.2	0.0	0.1
av PM _{2.5} exposure (μg m ⁻³)	8.1	8.8	8.7	8.2	8.5
percentage of daily PM _{2.5} exposure by microenvironment ^a					
driving	1.3	2.3	3.8	2.5	2.8
indoor	95.9	87.8	88.6	93.4	91.0
walk	1.4	2.5	2.3	1.7	2.0
Underground and DLR	0.4	5.2	3.6	1.2	2.7
bus	0.9	1.6	1.1	1.0	1.1
cycle	0.0	0.1	0.2	0.0	0.1
train	0.0	0.4	0.3	0.1	0.2
motorcycle	0.0	0.0	0.1	0.0	0.0

^aThe percentages given are the contribution of each activity to total daily exposure for the London population split by age group.

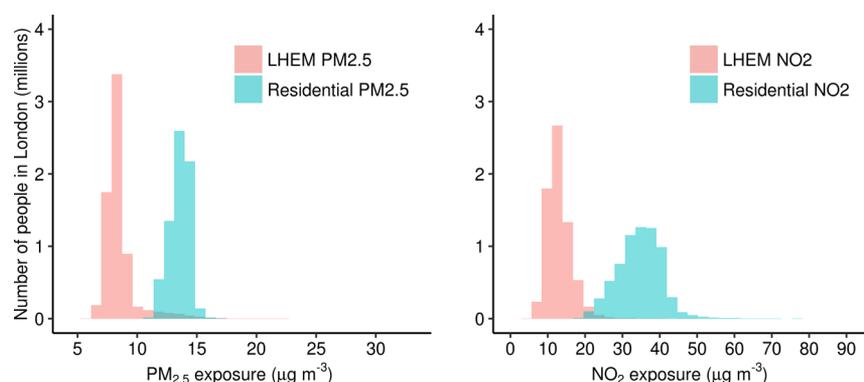


Figure 2. A comparison of the daily exposures of the London population using LHEM and residential address.

divided into active travel, inactive travel, those who remain at home, and all three combined.

The correlation between exposure estimated using the LHEM and at residential address, shown in purple in Figure 3, is relatively weak, having Pearson's *R* values of 0.41 for PM_{2.5} and 0.55 for NO₂. For those who undertake inactive travel,

shown in turquoise, there is also a weak relationship (*R* values of 0.56 for NO₂ and 0.39 for PM_{2.5}) due to the increased time spent in transit while exposed to high PM_{2.5} and NO₂ concentrations. In contrast, there is a strong relationship between LHEM and residential address exposures for those staying at home, shown in red (*R* values of 0.91 for NO₂ and

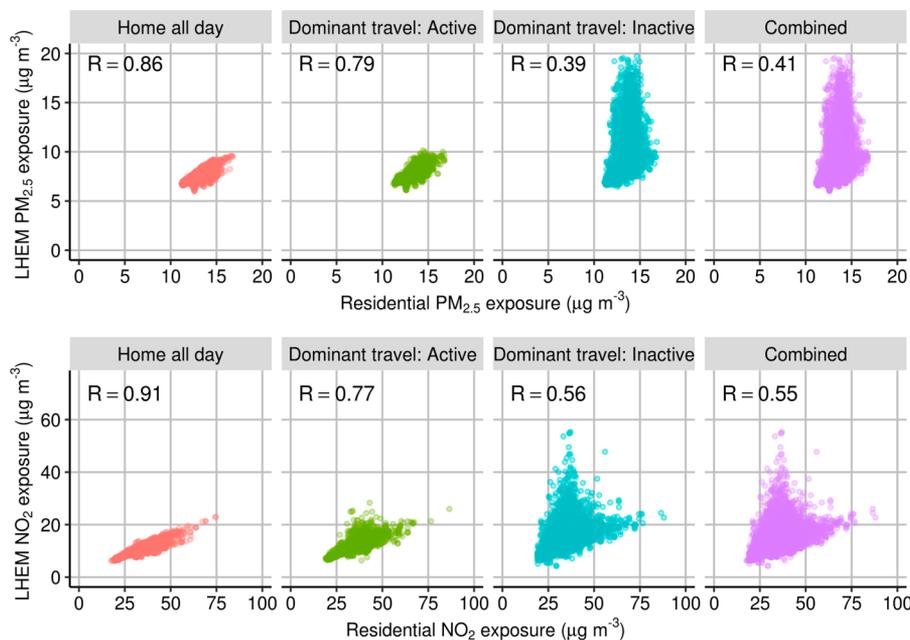


Figure 3. Scatter plots of NO₂ (bottom) and PM_{2.5} (top) LHEM exposure versus exposure at the residential address - demonstrating the relative strength of the relationship between those who undertake active travel (cycle and walk), those that stay at home, and those who undertake inactive travel (car, motorcycle, bus, train, and tube).

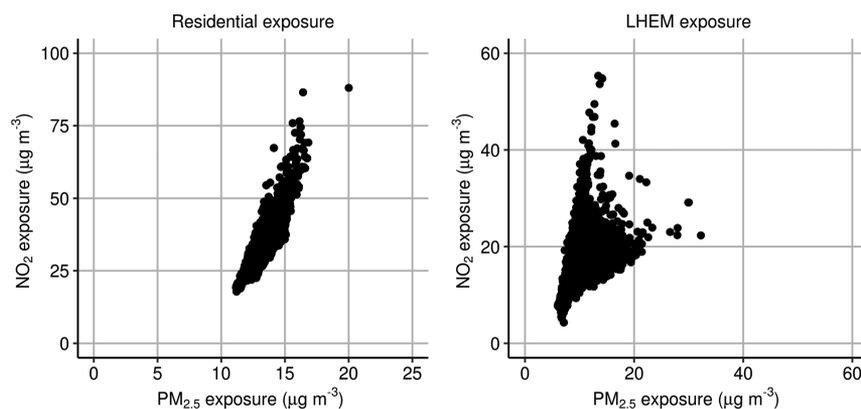


Figure 4. Scatter plots of NO₂ and PM_{2.5} exposure outdoors at the residential address (left) ($R = 0.90$) and using the LHEM (right).

0.86 for PM_{2.5}), and for those undertaking active travel, shown in green (R values of 0.77 for NO₂ and 0.79 for PM_{2.5}). Finally, for individuals whose dominant transport mode is the London Underground/DLR (not shown), there is no correlation between LHEM exposure and residential address exposure to PM_{2.5} (R values of -0.01). In summary, the strength of the relationship between the LHEM and residential address exposure is determined by travel, with inactive travel modes contributing to the highest daily exposure estimates of PM_{2.5} and NO₂ and the largest exposure misclassification (shown in blue, Figure 3).

The LHEM results also suggest that spending long periods of time indoors reduces exposure to outdoor sources of air pollution. I/O ratios (Figure 1 and Figure S3 (SI)) reduce between outer and inner London, due to newer building stock and the number of flats in large buildings, resulting in a small surface area available for infiltration of outdoor air. As a consequence there is greater protection from outdoor air pollution, which is highest in this central part of the city. However, greater air tightness will also increase the impact of

exposure from indoor sources, currently not considered in the LHEM.

3.3. Pollutant Correlations in Exposure Estimates. It is often difficult to establish independent health associations for NO₂ and PM_{2.5} due to their strong correlation outdoors. Scatter plots of NO₂ vs PM_{2.5} outdoor residential exposure results are given in Figure 4 (left panel) and show high correlation ($R = 0.90$). In contrast, the relationship between NO₂ and PM_{2.5} exposure using the LHEM (Figure 4, right panel) shows a much more complicated picture, especially when broken down by dominant travel mode (Figure 3), with the majority of the people who stay at home having the lowest exposures (Figure 3, in red), and those that travel the highest (Figure 3, green and blue/turquoise). Figure 4 (right panel) shows two distinct groups in the population: those who have high NO₂ exposure and low PM_{2.5} exposure, and those who have low NO₂ and high PM_{2.5} exposure. This relationship remains during typical and nontypical days (Appendix F and Figure S4, SI). Further work is needed to better understand these relationships, but when

applied to health studies this contrast has the potential to examine the independent effects of NO₂ and PM_{2.5}.

3.4. LHEM Sensitivity. While the outdoor exposure estimates of CMAQ-urban have been comprehensively evaluated using UK fixed site air quality data (Appendix C1, SI), there is little suitable observed personal exposure data with which to evaluate the LHEM exposure predictions. However, since transport and indoor microenvironments are important for exposure, we have tested the robustness of our results with five sensitivity tests using different input variables in these models. The results are presented as the percentage change in daily PM_{2.5} exposure. Further work on the sensitivity of the PM_{2.5} and NO₂ exposure is planned (see Table 2).

Table 2. Percentage Variation of Total PM_{2.5} Exposure under Five LHEM Model Sensitivity Tests

age/ parameters	test 1: IO ratios	test 2: vehicle ventilation	test 3: vehicle occupancy	test 4: resuspension rate	test 5: deposition velocity
children	+12.2	−0.5	0.0	+0.1	−0.3
young adults	+11.1	−1.1	0.0	+0.1	−0.5
adults	+11.2	−1.9	0.0	+0.1	−0.5
elderly	+11.9	−1.2	0.0	+0.0	−0.4

Test 1: New PM_{2.5} I/O ratios were modeled with a lower particle deposition rate (0.125 h^{−1} instead of 0.19 h^{−1}) and higher building penetration factor (0.82/1 instead of 0.8/1), resulting in ratios which were on average 20% higher, and consistent with those used in other measurement based studies^{58,59} which include indoor sources.

Test 2: The car, taxi, and van ventilation settings for the in-vehicle model were changed from semiopened windows and air-conditioning off (natural ventilation setting) to windows closed and air-conditioning system in use (mechanical ventilation setting), resulting in a lower penetration of outside air into the vehicle.

Test 3: The number of people in buses, trains, and on the Docklands Light Railway (DLR) was increased to the maximum capacity of the vehicles (reflecting rush hour transport conditions). The number of active passengers was also increased (see Table S2, SI).

Test 4: Particle resuspension rates for the in-vehicle model were doubled from the base case value of 0.02 μg/min^{−1} to 0.04 μg/min^{−1} (from Ferro et al.⁶¹).

Test 5: Pollutant deposition rates for the in-vehicle part of the model were increased from the mean values found in INDAIR⁶⁰ to the maximum values found in the same study (Appendix E, SI).

Results of the sensitivity tests show that altering the physical parameters and ventilation settings of the in-vehicle calculations (tests 2–5) had little effect on the overall population exposure with a maximum change in PM_{2.5} exposure being −1.9% due to vehicle ventilation assumptions for adults, although they are likely to be more influential on the results of specific trips/people. The parameter which had the largest effect was changing building I/O ratios, which for all age groups increased their exposure by >11.1%, demonstrating the importance of this microenvironment in exposure calculations.

5. DISCUSSION

Correctly estimating human exposure to air pollution remains an important source of uncertainty in public health research

and one which can push existing associations toward the null, i.e. to incorrectly conclude that no association exists or to affect the precision of the association. The LHEM, introduced here, is the latest attempt^{33,34,60,62} to establish a more comprehensive understanding of human exposure. There are many uncertainties to our model that require further investigation; however, using it, we find that our study population of 6.8m people, representing the population of London (>5 years of age), spends between 94.7 and 97.9% of their time indoors and 2.1–5.3% in transit, depending on age and other variables. These time-activity patterns are broadly typical of other midlatitude urban populations, for example the proportion of the day in transit was 6% in a study in Barcelona³⁰ and 8% for the working population in Helsinki, Finland.²⁸ We find that travel contributes between 8.0 and 18.8% of our population's daily exposure to outdoor NO₂ sources and between 4.1 and 12.2% of outdoor PM_{2.5} sources, with inactive travel contributing more than active travel. That people's trips contribute approximately 15% of their daily NO₂ exposure is similar to findings in Barcelona³⁰ and that approximately 9% of the daily PM_{2.5} exposure comes from people's trips agrees with findings by Fondelli et al.⁶³ in Florence. The concentrations that we model for transport microenvironments (Appendix G and Table S3, SI) tend to have large ranges due to the temporal and spatial variability of the CMAQ-urban model used as input to the in-vehicle model. Results generally agree with studies that include measurements of these environments,^{64–70} although there is a large variation reported in the literature. Time spent in indoor environments contributes the rest of the population's exposure of between 81.2 and 92% for NO₂ and 87.8 to 95.9% for PM_{2.5} from outdoor sources.

When comparing the LHEM method to address-based exposure estimates, we find the average LHEM exposure is 37% lower for PM_{2.5} and 63% lower for NO₂. Results from other similar studies are difficult to find as we combine individual exposures for a large population and do not include indoor emissions. For NO₂ a study by de Nazelle³⁰ of a small number (36) of active volunteers moving around the city found subjects had 27% higher NO₂ exposure (including indoor sources) when a time-activity model was compared to residential exposure. Dhondt²⁹ found ambient NO₂ exposure to be between 5% lower and 15% higher when using dynamic methods with a model which has lower temporal and spatial resolution than the LHEM and a relatively simple transport exposure model. In addition the LHEM incorporates a larger range of activity levels and ages, including a number of participants who do not leave their home, and does not include indoor sources. For PM_{2.5} Burke,⁷¹ when calculating exposure to ambient sources using a stochastic simulation method, found a median exposure to PM_{2.5} of 7 μg m^{−3} compared to our mean of 8.5 μg m^{−3}.

Using the LHEM we conclude that the indoor environment is protective of exposure to outdoor emissions sources and that this is reflected in much lower exposure estimates than would be found using exposure outdoors at the residential address. The degree to which buildings are protective varies geographically in London and is pollutant and housing specific. Average NO₂ I/O ratios in the EXPOLIS and SAPALDIA studies^{72,73} are found to be 0.76 and 0.545 respectively, and are higher than our range of 0.11 to 0.59 due to the influence of indoor sources. For PM_{2.5}, Taylor et al. compared the ratios we use (range of 0.35 to 0.86) to existing studies and found good agreement. Furthermore, sensitivity tests demonstrated that

even with increased I/O ratios, the differences between LHEM and residential exposure are still present. Finally a further limitation of the LHEM is the assumption that I/O ratios for commercial buildings are the same as domestic buildings of similar ages, morphologies, and in the same postcode.

In addition, we have shown that transport microenvironments are important in correctly establishing exposure, with those who stay at home having an exposure which is closely correlated with outdoor residential concentrations and those who travel, in particular by car, bus, and by the London Underground being poorly correlated with outdoor concentrations at the residential address. Members of the population that travel by car, bus, and the London Underground are some of the most highly exposed, with small differences in the exposure across the four age groups used in this study.

There is also a more complex relationship between $PM_{2.5}$ and NO_2 LHEM exposure estimates compared to the correlated exposures at residential addresses. Within health studies this may have the potential to examine the independent health effects of NO_2 and $PM_{2.5}$ through the assessment of the differential degree of misclassification of personal exposure to each air pollutant. In future cohort studies in London, participants could complete questionnaires similar to those in the LTDS, which could then be combined with the LHEM to assess personal exposure, and have this linked to their health records. The LHEM could be used as a template for other cities, as it provides population exposure, not possible using alternatives such as personal monitoring. For time-series studies, Zeger et al.⁷⁴ illustrate how estimates of daily personal exposure in the population can be used to adjust the relative risk obtained from time-series studies, using daily ambient concentrations, for measurement error and to predict the likely bias occurring in multipollutant models. For health impact assessments, policies designed to reduce air pollution exposure by changing travel behavior, or by reducing specific micro-environment concentrations, are currently difficult to assess as the relevant exposure metric does not match those currently used in epidemiological studies. This situation can be improved by learning more about the relationships between different exposure metrics using the LHEM or by deriving concentration–response functions from future studies using the LHEM or an analogous model. As the LHEM is based upon CMAQ-urban predictions it is possible to predict exposure to components of PM, such as elemental, organic carbon, nitrate, and sulfate, rather than just PM mass, and also ozone.

The LTDS samples an individual's activity on a single day. The LTDS survey population has coverage of weekday and weekend activities throughout the year, and by combining all individuals and using weighting factors provided by TfL, we obtained an average daily NO_2 and $PM_{2.5}$ exposure for the London population in 2011.

The next stage of the LHEM is better predicted exposure indoors and outdoors from all sources, rather than to outdoor sources as we have here. To do this requires the inclusion of an indoor air pollution model with indoor sources, which given the large amount of time that people spend indoors is likely to alter LHEM exposure predictions. Second, a more comprehensive evaluation of LHEM is needed using personal exposure data, which have advanced rapidly in recent years, including the development of postprocessing methods to ensure high data quality. However, robust evaluation of models like LHEM is still limited due to the cost and size of monitors, as well as the

practicality of obtaining large enough cohorts for the data to be reliable.

To date, we have not found an equivalent LTDS data set outside London; however, there is undoubtedly equivalent data in other cities throughout the world, enabling models such as LHEM to be developed. Although we recognize the significant ethical hurdles that this approach would entail, it is hoped that the LHEM, as the latest in this new breed of exposure models, will lead to linking public health records with personal exposure estimates, to better understand human exposure to air pollution and the associated health effects. Finally, the LHEM can play a part in testing air quality policy scenarios aimed at reducing exposure rather than lowering ambient concentrations, for investigating the exposure of different subpopulations and for assessing exposure indoors and outside.

■ ASSOCIATED CONTENT

📄 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b01817.

Weighting factors for the LTDS, model video summary, CMAQ-urban predictions and evaluation, indoor/outdoor ratios, in-vehicle model details, exposure correlations by number of trips, and microenvironments (PDF)

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Notes

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