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Performance of a microenviromental model for estimating personal NO_2 exposure in children

Anna Mölter[a,*], Sarah Lindley[b], Frank de Vocht[a], Raymond Agius[a], Gina Kerry[c], Katy Johnson[c], Mike Ashmore[d], Andrew Terry[d], Sani Dimitroulopoulou[e], Angela Simpson[c]

[a] Centre for Occupational & Environmental Health, Health Sciences Group, School of Community Based Medicine, Manchester Academic Health Sciences Centre, The University of Manchester, Oxford Road, Manchester M13 9PL, UK
[b] School of Environment and Development (Geography), The University of Manchester, Oxford Road, Manchester M13 9PL, UK
[c] The University of Manchester, Manchester Academic Health Science Centre, NIHR Translational Research Facility in Respiratory Medicine, University Hospital of South Manchester NHS Foundation Trust, Wythenshawe Hospital, Southmoor Road, Manchester M23 9LT, UK
[d] Environment Department, University of York, Heslington, York YO10 5DD, UK
[e] Department of Mechanical Engineering, University of Western Macedonia, 50100 Kozani, Greece

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A B S T R A C T

A common problem in epidemiological studies on air pollution is exposure misclassification, because investigators often assume exposure is equivalent to outdoor concentrations at participants' homes or at the nearest urban monitor.

The aims of this study were: (1) to develop a new microenvironmental exposure model (MEEM), combining time-activity data with modelled outdoor and indoor NO_2 concentrations; (2) to evaluate MEEM against data collected with Ogawa™ personal samplers (OPS); (3) to compare its performance against datasets typically used in epidemiological studies.

Schoolchildren wore a personal NO_2 sampler, kept a time-activity diary and completed a questionnaire. This information was used by MEEM to estimate individuals' exposures. These were then compared against concentrations measured by OPS, modelled outdoor concentrations at the children's home (HOME) and concentrations measured at the nearest urban monitoring station (NUM).

The mean exposure predicted by MEEM (mean = 19.6 $\mu g\ m^{-3}$) was slightly lower than the mean exposure measured by OPS (mean = 20.4 $\mu g\ m^{-3}$). The normalised mean bias factor (0.01) and normalised mean absolute error factor (0.25) suggested good agreement. In contrast, the HOME (mean = 31.2 $\mu g\ m^{-3}$) and NUM (mean = 28.6 $\mu g\ m^{-3}$) methods overpredicted exposure and showed systematic errors.

The results indicate that personal exposure can be modelled by MEEM with an acceptable level of agreement, while methods such as HOME and NUM show a poorer performance.

1. Introduction

In the hierarchy of exposure assessment methods, personal measurements are deemed to be the best estimate of actual exposure (Nieuwenhuijsen et al., 2006). However, in many studies personal measurements are not feasible, due to costs or because the exercise would overburden participants. Furthermore, in some studies exposure needs to be estimated retrospectively. As a result, many studies use indirect methods, such as models, to estimate exposure. Ott (1982) proposed a discrete model for exposure assessment, in which a given study area is divided into "boxes" to represent discrete "microenvironments". It is assumed that each microenvironment (ME) has a distinct and homogenously distributed pollutant concentration. It is further assumed that over time

Abbreviations: B_{NMBF}, Normalised mean bias factor $= \overline{\mathrm{Modelled}}/\overline{\mathrm{Measured}} - 1$ if $\overline{\mathrm{Modelled}} \geq \overline{\mathrm{Measured}}$, $(1 - \overline{\mathrm{Modelled}}/\overline{\mathrm{Measured}})$ if $\overline{\mathrm{Modelled}} < \overline{\mathrm{Measured}}$; E_{NMAEF}, Normalised mean absolute error factor $= \sum|\mathrm{Modelled} - \mathrm{Measured}|/\sum \mathrm{Measured}$ if $\overline{\mathrm{Modelled}} \geq \overline{\mathrm{Measured}}$, $\sum|\mathrm{Modelled} - \mathrm{Measured}|/\sum \mathrm{Modelled}$ if $\overline{\mathrm{Modelled}} < \overline{\mathrm{Measured}}$; HOME, Home outdoor modelled exposure; LUR, land use regression; MAAS, Manchester Asthma and Allergy Study; ME, microenvironment; MEEM, microenvironmental exposure model; MPE, Mean prediction error $= 1/N \sum[\mathrm{Modelled} - \mathrm{Measured}]$; NUM, nearest urban monitoring station; OPS, objective measurements using personal sampling.

* Corresponding author. Tel.: +44 0161 2755691; fax: +44 0161 2755595.
E-mail address: anna.molter@postgrad.manchester.ac.uk (A. Mölter).

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a person will move through a continuous series of MEs. The integrated exposure of a person can then be calculated as the sum of concentrations in each ME multiplied by the time spent in this ME (Ott, 1982).

Pollutant concentrations in the microenvironments can be determined in one of three ways: through measurement (Kousa et al., 2002); by assigning a typical value based on existing knowledge (Kim et al., 2002); or through the application of various modelling techniques (e.g., air dispersion, multiple regression, mass balances) (Gulliver and Briggs, 2005). Measuring the pollutant concentration in each ME has only been undertaken in a few studies (Adgate et al., 2004; Morandi et al., 1988; Quackenboss et al., 1986) and in large cohort studies this method is usually not practical. Assigning a typical value to each ME is only possible, if such values are readily available in the literature and are sufficiently reliable. Even then, the population studied would need to exhibit a wide and known variation in time-activity patterns in order to overcome the lack of specificity in ME estimates. Such wide variation is generally not the case for children, who tend to have very similar time-activity patterns (Adgate et al., 2004; Jones et al., 2007).

The third option, modelling pollutant concentrations within a ME based on other known parameters is an alternative approach, which may overcome some of these problems. The advantage of using models to determine ME concentrations is that these can allow improved estimates of the spatial variation between, and temporal variation within each ME, particularly for large scale studies where extensive monitoring campaigns are not feasible. For example, this approach was adopted in a study by Gulliver and Briggs (Gulliver and Briggs, 2005), where PM_{10} concentrations were modelled in the journey ME (based on road networks, meteorology and background monitoring data) and then compared to concentrations measured with portable monitors, to evaluate the model.

A key aspect of any new exposure model is the assessment of the modelled concentrations against monitored data. Prior to being used in an epidemiological setting, the performance of the model has to be evaluated to determine its suitability. The best method to test an exposure model will depend on the specific aims of the model; for example, the ideal way to test a personal air pollution exposure model would be through personal monitoring of the study population.

This study had three aims: (1) To develop a novel microenvironmental exposure model (MEEM), which predicts children's personal exposure to NO_2 and incorporates outdoor and indoor concentrations, as well as time-activity patterns. (2) To evaluate the performance of MEEM by comparing modelled exposures to NO_2 exposures measured with personal monitors worn by schoolchildren. (3) To compare the performance of MEEM against data collected by the nearest urban monitoring station and modelled data for the home outdoor environment, which are commonly used as surrogates for personal exposure in epidemiological studies on effects of air pollution.

2. Methods

2.1. Study population

The personal monitoring campaign was carried out at a secondary school in Greater Manchester, a large conurbation in the Northwest of England, selecting children aged 12–13 years. The study area and age range were selected to resemble the Manchester Asthma and Allergy Study (MAAS) birth cohort (Simpson et al., 2001), as the model is required to retrospectively estimate long term pollution exposure within this cohort. Four school classes of children in Year 7 were invited to take part, one class per season (Table 1). Participating children were asked to wear an Ogawa™ sampler for two consecutive weekdays, with the sampler being exchanged after the first day. This sampling pattern was chosen to capture as much data variability as possible, but without overburdening the children. Children were shown how to attach the sampler to their school blazers and were instructed to wear the sampler as much as possible and to keep it in their bedroom at night. Children were also given a small information card (shown in Figure S1 online supplement) to remind them of the "DOs and DON'Ts" associated with the sampler. In addition to wearing the personal monitors, the children were asked to complete a time-activity diary for the same time period. Parents were asked to give written consent and to complete a questionnaire; children also signed an assent form. Ethical approval to conduct this study was granted by the University of Manchester Research Ethics Committee (Ref. 07268).

2.2. Measurements using Personal Samplers (OPS)

The Ogawa™ sampler is a small passive diffusion sampler with two chambers, each containing a coated collection pad (Ogawa & Co., 2006). The two chamber design enables simultaneous measurement of NO_2 and NO or, as used in this study, two simultaneous measurements of NO_2. On completion of monitoring the NO_2 collection pads were removed from the Ogawa™ samplers, allocated a random number and sent to a commercial laboratory for analysis. The results of the laboratory analysis were accepted, if the coefficient of variation of collection pads from the same sampler was less than 0.25 (Janssen et al., 2001; Jerrett et al., 2007). The personal exposure measured by each sampler was calculated as the average concentration measured by the two collection pads.

Table 1
Summary of study population by measurement group.

		Spring	Summer	Autumn	Winter	Total
	Measurement period	30/04/08–02/05/08	17/06/08–19/06/08	19/11/08–21/11/08	21/01/09–23/01/09	
	No. of participants (invited)	16 (25)	12 (26)	17 (29)	15 (30)	60 (110)
	Gender ratio (boys:girls)	7:9	4:8	11:6	8:7	30:30
Day 1	Personal measurements acceptable[a] (completed)	3 (14)	7 (11)	15 (15)	13 (13)	35 (53)
	Time activity diaries completed	14	10	15	13	52
	Mean measured NO_2 concentration ($\mu g\ m^{-3}$)	–	16.0	14.7	25.9	19.8
Day 2	Personal measurements acceptable[a] (completed)	3 (15)	11 (11)	13 (16)	12 (13)	36 (55)
	Time activity diaries completed	13	12	17	14	56
	Mean measured NO_2 concentration ($\mu g\ m^{-3}$)	–	19.3	18.8	23.8	20.9
	Questionnaires completed	11	8	11	11	41
	Gas cooking appliance in home[b]	9	8	10	8	35
	Smokers in home	1	0	0	2	3

[a] Inclusion criteria: Coefficient of variance <0.25.
[b] Gas cooking appliance = gas hob and/or gas oven.

2.3. Time-activity diary and questionnaire

The time-activity diary consisted of a table with columns showing ten MEs and rows showing 15 min time intervals. For each time interval the children selected the ME that best reflected their predominant location. The ten MEs were: home (kitchen, living room, bedroom, outdoors, other), school (indoors, outdoors), journey (home to school, school to home) and other. In addition, parents of participating children were asked to complete a questionnaire to collect information on their home address, their child's mode of travel between home and school and their home environment. Information on their home environment included questions on the type of kitchen hob and oven used (gas or electric), the cooking duration, the number of cigarettes smoked in the kitchen, living room or child's bedroom, whether windows were open or closed in either of these rooms and the dimensions of these rooms.

2.4. Microenvironmental exposure model (MEEM)

The personal exposure of participating children was estimated using a microenvironmental exposure model (MEEM) based on data collected in the time-activity diary, parentally completed questionnaire and exposure estimates from each ME. The MEEM consists of three main environments, home, school and journey, which are further subdivided into indoor or outdoor MEs and in the case of the home ME into kitchen, living room and child's bedroom. This study was restricted to weekday exposures only, as children would spend the majority of their time at home and school. If weekend exposures had been included, a much more complicated and extensive time-activity diary, questionnaire and model would have been necessary, which was beyond the scope of this study.

Fig. 1 shows a flow diagram of how concentrations in each ME were calculated. NO_2 concentrations in all outdoor MEs, i.e., home outdoor, school outdoor and journeys, were estimated through a land use regression (LUR) model specifically developed for the Greater Manchester area (Mölter et al., 2010a, 2010b). The land use regression model provided annual average NO_2 concentrations for 2008 and 2009 at the school and home addresses. For the journeys the shortest route between the children's home and school was calculated using ArcGIS 9.2 Network Analyst (ESRI Inc.). It should be noted that Network Analyst was used to calculate the shortest driving route, not walking route.

In order to provide appropriate outdoor concentrations for each hour of the children's time-activity data, the estimated annual

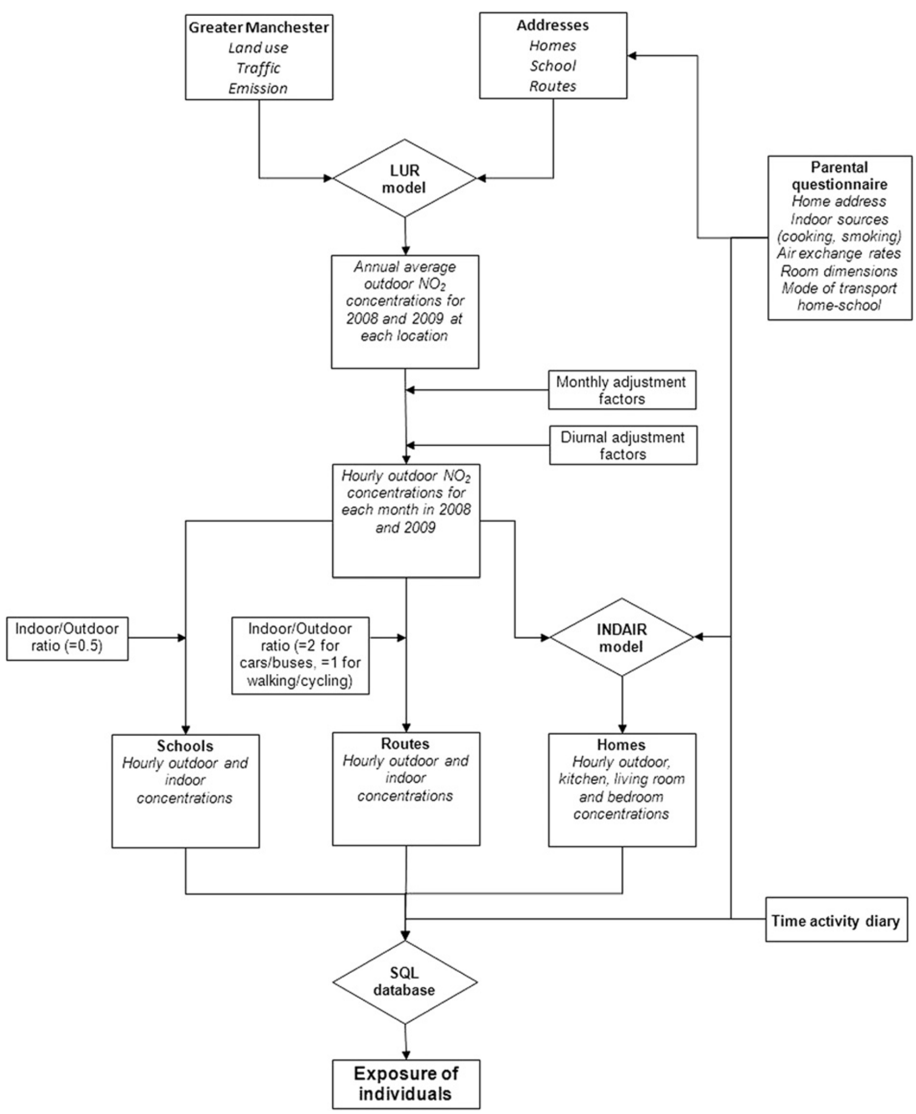

Fig. 1. Flow diagram showing input data, modelling steps, intermediate and final output datasets within MEEM.

average concentrations from the LUR model were first adjusted to give the relevant monthly mean concentration in each year. The adjustment factors were based on the average concentration measured by up to five urban background monitors in Greater Manchester. Then a diurnal profile, also derived from these monitors, was applied to the monthly mean concentrations to obtain hourly concentrations.

Pollutant concentrations in the home indoor MEs were calculated through the INDAIR model developed by Dimitroulopoulou et al. (2006) and parameterised for the UK. The INDAIR model is a validated physical mass balance model, which calculates indoor concentrations based on infiltration of pollutants from the outdoor environment, deposition on indoor surfaces and emissions from indoor sources of pollutants (Dimitroulopoulou et al., 2000, 2006, 2001b, 2001a). The input parameters for the INDAIR model were derived from the home outdoor ME, information provided in the questionnaires and constants provided in the literature. Further details of the input parameters are shown in Table S1 in the online supplement.

As there were no known sources of NO_2 in the school indoor ME, an Indoor/Outdoor ratio (I/O) of 0.5 was used to estimate the concentrations (Stranger et al., 2008). For the journey MEs the pollutant concentration was equivalent to the outdoor concentration if a child walked to school, and twice the outdoor concentration if a child travelled by car or bus (International Center For Technology Assessment, 2000).

The modelled concentrations in each ME were combined with data from the time-activity diaries to obtain a time-weighted average concentration for each day that the personal monitors were worn. Figure S2 in the online supplement shows an example of the daily variation in NO_2 in each ME and the child's exposure derived from these concentrations and the time-activity data.

In addition to this methodology, we also calculated the mean exposure for each day the personal monitors were worn by means of two "traditional" methods often used in environmental epidemiology (Bayer-Oglesby et al., 2005; Brauer et al., 2002; Hwang et al., 2006):

(1) Home outdoor modelled exposure (HOME)

The modelled concentration for the home outdoor ME alone, derived from the LUR model, was used as an estimate of the outdoor NO_2 exposure.

(2) Measurements from the nearest urban monitoring station (NUM)

During this study (April 2008 to January 2009) hourly NO_2 concentrations were only available from four urban monitoring stations within the Greater Manchester area (1 Roadside, 1 Urban Centre, 1 Urban Industrial and 1 Suburban). The nearest urban monitor was determined by calculating the straight line distance between each child's home address and the available monitoring stations. If no measured data was available from the nearest monitor, data from the second nearest monitor was used. Fig. 2 shows the school and homes of the children as well as the shortest driving routes and locations of the nearest urban monitors.

2.5. Statistical analysis

The personal exposure measurements from the Ogawa™ samplers (OPS) were compared for each child and day with mean daily NO_2 concentrations estimated through three methods: (1) the integrated Microenvironmental Exposure Model (MEEM); (2) measurements at the Nearest Urban Monitoring station (NUM); (3) LUR based estimates for the home outdoor ME (HOME). The data were analysed using SPSS v16.0 (SPSS Inc.).

To compare the modelled results with the measured data the mean prediction error (MPE) was calculated. The MPE indicates whether overall the model overestimates or underestimates the measured concentrations. In addition, the normalised mean bias factor (B_{NMBF}) and normalised mean absolute error factor (E_{NMAEF}) were calculated, which measure the relative bias and relative absolute error of a model, respectively. These factors were introduced by Yu et al. (2006) to evaluate the performance of air quality models and have been found to be statistically robust measures. As performance criteria of a good model $|B_{NMBF}| \leq 0.25$ and $E_{NMAEF} \leq 0.35$ have been suggested (Yu et al., 2006). The datasets were further compared using Wilcoxon's signed rank test and a graphical analysis following the approach suggested by Bland and Altman (1999). Since for epidemiological purposes an exposure method, which provides ranking of subjects according to exposure levels is important, Spearman's rank correlation coefficients were also calculated. However, it should be noted that whereas correlation coefficients compare trends within the datasets, while the above methods are measures of absolute error. It has been shown that a high correlation coefficient alone does not indicate good agreement between datasets and that it needs to be assessed in combination with absolute measures of error (Bland and Altman, 1999; Yu et al., 2006).

To evaluate the sensitivity of MEEM, the predicted NO_2 concentration in each ME was changed in 10% increments and the MPE was recorded. Furthermore, to determine the influence of each ME on the predicted exposure and the MPE, the time spent in each ME was entered into a bivariate linear regression analysis with the MEEM predicted NO_2 concentration and the MPE.

3. Results

3.1. Participation

Participation of the children in the study is summarised in Table 1. On average, 55% of children invited to the study agreed to take part. Of these children more than 86% completed the monitoring and time-activity diaries, but only 68% returned the parental questionnaires. Overall, the number of boys and girls participating in the study was equal; however, this varied by study period. Table 1 also indicates that not all personal measurements were of acceptable quality. In the spring group 80% of measurements did not meet the inclusion criteria, i.e. coefficient of variation <0.25. It is unclear whether this was due to contamination of the collection pads or problems during the laboratory analysis, but as a consequence all spring results were discarded. Unfortunately, this decreased the final sample size to a smaller number than we had originally anticipated. A power analysis indicated that Wilcoxon's signed rank test would detect a small to medium (0.35–0.5) effect size within this reduced sample with a probability of 0.8–0.95.

Table 1 also shows the number of homes with gas cooking appliances and smokers, which are considered to be two important indoor sources of NO_2. Most children lived in homes with either a gas hob and/or gas oven; however, very few children lived with a smoker.

3.2. Measured and modelled concentrations

The NO_2 concentrations obtained through OPS, MEEM, HOME and NUM, together with model performance results, are summarised in Table 2. (Concentration ranges for individual MEs are shown in Table S2 in the online supplement). It can be seen that the mean exposure estimated by MEEM is almost identical to the mean

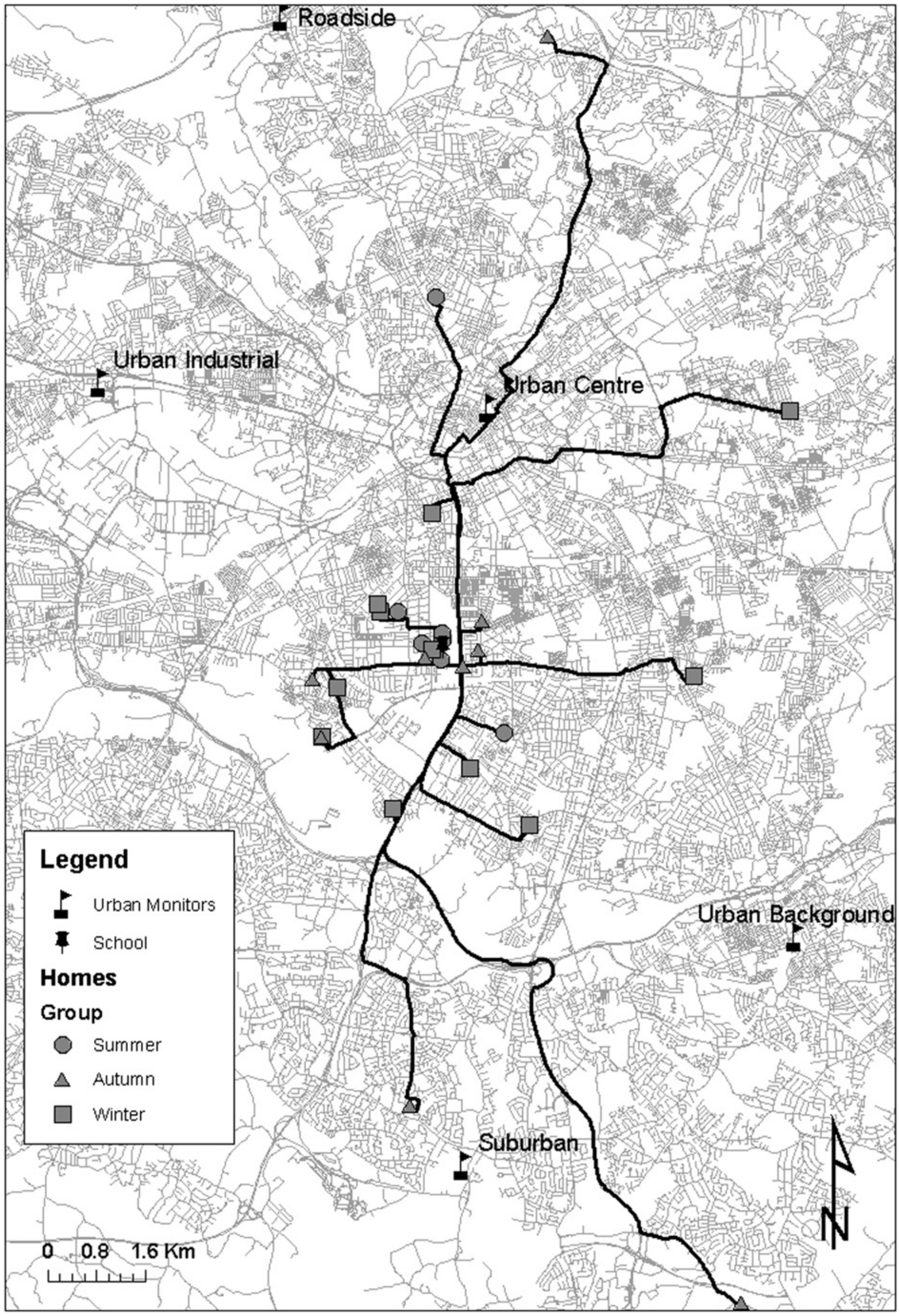

Fig. 2. Spatial distribution of children's homes and the nearest urban monitors.

personal exposure of the children. The range of concentrations for individual children predicted through MEEM was approximately 60% smaller than the range of OPS; however, an outlier in the monitored concentrations greatly increased the range of OPS. The MEEM showed the smallest MPE, B_{NMBF} and E_{NMAEF} of the three methods and it was the only method that met the performance criteria of $|B_{NMBF}| \leq 0.25$ and $E_{NMAEF} \leq 0.35$. Furthermore, the Wilcoxon signed rank test suggested that there was no statistically significant difference between OPS measurements and the MEEM predictions. In contrast, the predictions from the HOME and NUM methods were significantly different to results from the personal samplers and systematically overpredicted exposure. Moreover, the B_{NMBF} and E_{NMAEF} indicated a poor performance of the HOME and NUM methods.

Fig. 3 plots the differences between the personal sampler and the MEEM against the mean of these values, and shows a relationship between the difference and mean, i.e. at higher mean values the MEEM underestimates exposure. Due to this relationship the 95% limits of agreement, shown as dotted lines in Fig. 3, have been based on a method utilising fitted regression and absolute residuals, rather than the mean difference and standard deviations (Bland and Altman, 1999). Most results fall within these limits of agreement. Further details of individual OPS and MEEM results are shown in Figure S3 (Online Supplemental Material).

The rank correlation coefficients (Table 2) showed a moderate correlation between the results from the personal samplers and all three methods. Fig. 4 plots the measured concentrations from the personal samplers against concentrations predicted by the three

Table 2
Summary and comparison of results from different methods.

	OPS	MEEM	HOME	NUM
Number	46	46	46	46
Arithmetic Mean in μg m^{-3} (Geometric Mean)	20.4 (19.1)	19.6 (19.0)	31.2 (30.7)	28.6 (24.7)
Range in μg m^{-3}	9.19–54.12	9.70–29.01	23.20–39.28	11.37–52.23
St. dev. in μg m^{-3} (GSD)	7.9 (1.4)	4.7 (1.3)	5.4 (1.2)	15.0 (1.7)
MPE in μg m^{-3}		−0.75	10.8	8.2
B_{NMBF}		−0.04	0.53	0.40
E_{NMAEF}		0.27	0.59	0.64
Wilcoxon's signed rank test		$Z = -0.05$, $p = 0.96$	$Z = -5.25$, $p < 0.01$	$Z = -3.2$, $p < 0.01$
Spearman's rank correlation		$r_s = 0.31$, $p < 0.05$	$r_s = 0.45$, $p < 0.01$	$r_s = 0.44$, $p < 0.01$

OPS = Ogawa Personal Samplers, MEEM = Microenvironmental exposure model, HOME = Modelled Home Outdoor Concentration, NUM = Nearest Urban Monitor.

exposure methods. It can be seen that the HOME method consistently overpredicts personal exposure, whereas the NUM method overpredicts exposure during winter and autumn and underpredicts exposure during summer. In contrast, the MEEM shows no systematic error, but it shows a slightly weaker correlation than the other methods. Furthermore, Fig. 4 shows that NUM provides little variation within groups, due to the limited number of monitors within the study area.

For the purpose of the model evaluation measurements were analysed as individual samples. Further analyses were carried out combining data from day 1 and day 2, i.e. using 48 h average concentrations for individual children, and stratifying the data by day 1 and day 2. The results from these analyses were not materially different to the results shown above (data not included).

3.3. MEEM model sensitivity

Table 3 shows the range of times the children spent in each ME and the associations of these with the NO_2 concentration predicted by MEEM and the MPE. It can be seen that the predicted exposure level significantly decreased when children spent more time in the Home MEs (i.e., kitchen, living room, child's bedroom and outside home). On the opposite, their predicted exposure level significantly increased with time spent in the journey MEs. On an individual basis only time spent in the child's bedroom significantly decreased exposure, none of the other Home MEs had a significant effect. A similar pattern is found in the regression analysis with the MPE; time spent in the Home MEs and Journey MEs are significantly associated with the MPE. While more time spent in the Home MEs is associated with underprediction in MEEM, more time spent in the journey MEs is associated with overprediction of exposure.

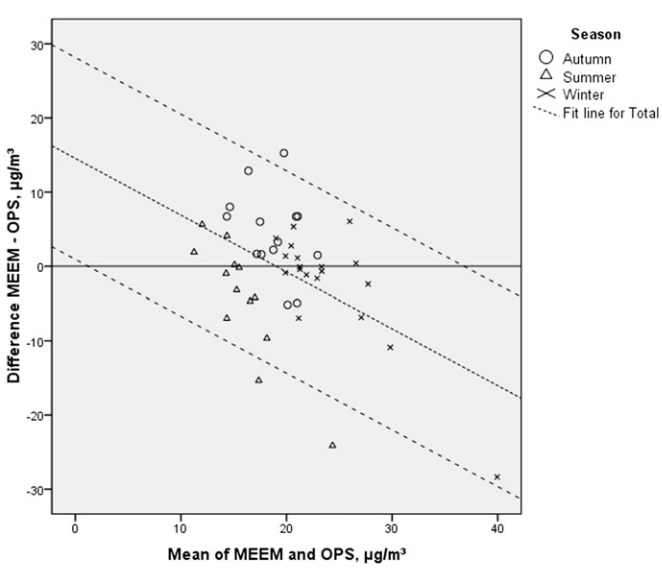

Fig. 3. Difference of OPS and MEEM vs. mean of OPS and MEEM.

Figure S4 (online supplement) shows the change in MPE when the NO_2 concentration in each ME changes in increments of 10%. The largest change in MPE occurred with a change in concentration in the child's bedroom, a 10% change in concentration resulted in a change of ±0.7 μg m^{-3} in the prediction error. The second largest change occurred with a change in the school indoor ME concentration, where the prediction error changed by ±0.4 μg m^{-3}. Changes in concentration in the other MEs led to relatively small changes in the prediction error ranging from 0.04 to 0.2 μg m^{-3}. Overall these results suggest that the main factors influencing the MEEM exposure estimates are the home bedroom ME concentrations, school indoor ME concentrations and the time spent in the journey ME and at home.

4. Discussion

By measuring NO_2 using personal samplers in children over a two day period in three seasons, we have demonstrated that modelling based on time spent in microenvironments provides a better estimate of personal exposure, than an outdoor model or the nearest urban monitor. MEEM showed much smaller absolute errors than the HOME and NUM methods, which showed systematic bias. Therefore for the majority of children, MEEM predicted measured personal exposure well. Although overall the MEEM showed a good model performance, in some cases there were large differences between the predicted and measured concentration. There are several possible explanations:

(1) One possible reason could be measurement errors by the personal samplers. Even though personal monitors are considered the best method available to estimate exposure (Nieuwenhuijsen et al., 2006), their results can be erroneous, as was demonstrated by the sampling results obtained in the spring group. By adopting a quality control measure, i.e. coefficient of variation <0.25 between filter pairs, we removed as much unreliable data as possible. Therefore, it is unlikely that the differences between the personal samplers and MEEM were due to measurement errors.
(2) Another explanation for the discrepancies between the personal samplers and MEEM could be uncertainties in the exposure model, such as parameter uncertainty and scenario uncertainty (Fryer et al., 2006; Hertwich et al., 2000; Zou et al., 2009). Unless a large or systematic error was present in the model parameters, parameter uncertainty is unlikely to have had a marked influence. Scenario uncertainty occurs due to misclassification of activities (Morandi et al., 1988; Quackenboss et al., 1986), which for some children may have been present in the journey MEs (Home to School, School to

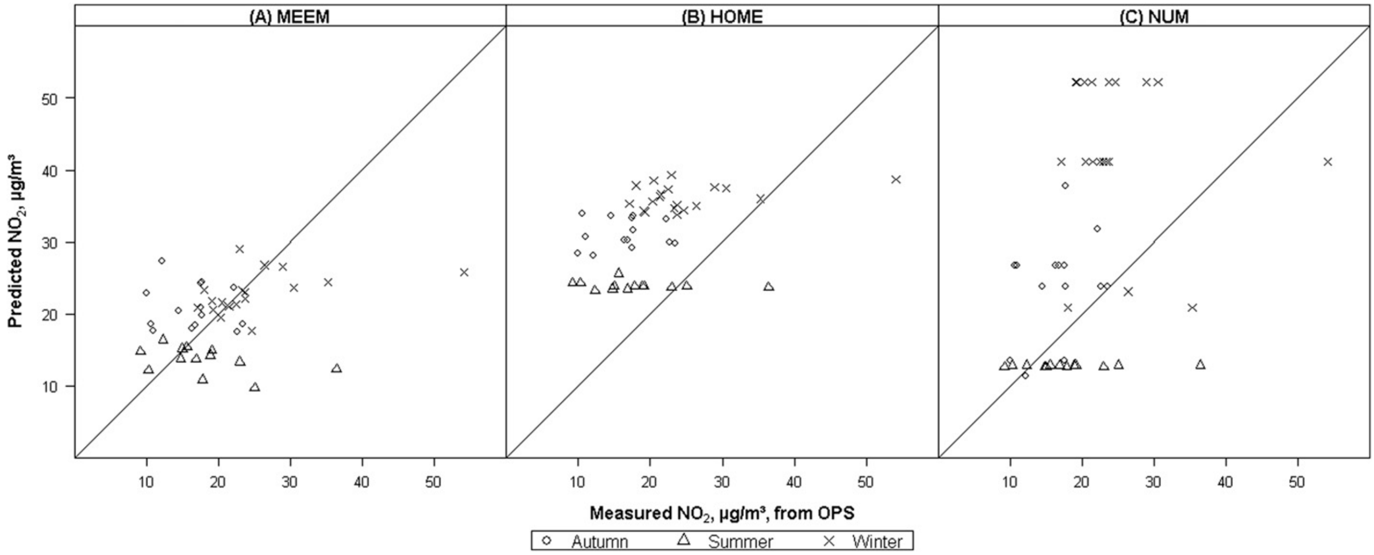

Fig. 4. Results from OPS vs. estimates from MEEM, HOME and NUM.

Home). The journey MEs were based on the shortest driving route calculated through a network analysis. It is unlikely that all children travelled along these routes, particularly those walking to or from school (Gong and Mackett, 2009). The sensitivity analysis showed that the amount of time spent in the journey MEs had a stronger influence than a change in concentration in the journey MEs, therefore a slightly different route would only have a small impact on the overall exposure. However, this also indicates that errors in the children's time-activity diaries may have caused some of the discrepancies between the measured and modelled concentrations. An alternative to the use of time-activity diaries would have been to ask the children to wear GPS trackers as well as their personal samplers. Unfortunately, this was not feasible within the scope of this study.

(3) It is inevitable that there were factors influencing personal exposure of individual children, which were not taken into account in the MEEM. The literature identifies road traffic as the major outdoor source of NO_2 (Colls, 2002) and gas cooking as the major indoor source of NO_2 (Coward and Raw, 1996). Both of these were included in MEEM, but in some cases there may have been additional sources present.

(4) The most likely cause of the largest observed differences between the personal samplers and MEEM is the nature of MEEM and its underlying models. The LUR model is based on the normal distribution and therefore will perform best when predicting concentrations close to the mean. Similarly, the INDAIR model is designed to estimate NO_2 concentrations in a typical home in the UK, and as such is also based on average conditions and parameters. By basing MEEM on the normal distribution and average conditions, the predicted concentrations will apply to the majority of the population. However, a disadvantage of this type of modelling is a poorer prediction of extreme concentrations, as this requires a large deviation from the underlying mean parameters.

Table 3
Associations of time spent in microenvironments with predicted NO_2 and prediction error of MEEM.

	Range of time spent in ME (minutes)	Association with predicted NO_2 exposure		Association with mean prediction error	
		B	*p*	*B*	*p*
Main types of MEs					
Home	705–1035	−0.035	<0.01	−0.042	0.01
School	120–675	0.000	0.95	−0.009	0.29
Journey	15–180	0.075	<0.01	0.077	0.01
Individual MEs					
Home – Kitchen	0–150	0.020	0.22	−0.012	0.67
Home – Living Room	0–390	0.006	0.44	0.005	0.66
Home – Bedroom	450–930	−0.022	<0.01	−0.022	0.09
Home – Outdoor	0–180	−0.030	0.10	0.030	0.34
Home – Other	0–195	−0.015	0.34	−0.042	0.10
School – Indoor	120–615	−0.001	0.91	−0.009	0.34
School – Outdoor	0–135	0.003	0.87	−0.019	0.52
Journey – Home to School	15–90	0.125	<0.01	0.103	0.08
Journey – School to Home	0–105	0.115	<0.01	0.137	0.01
Other	0–210	0.015	0.27	0.008	0.73

A limitation of this study was the relatively small sample size. As described above one of the seasonal datasets had to be discarded for reasons beyond our control, which resulted in a smaller sample size than expected. Although personal monitoring studies of children are often based on small sample sizes (Liard et al., 1999; Van Roosbroeck et al., 2006; Zipprich et al., 2002), a larger sample size would have strengthened the findings of this evaluation study.

A recent review has shown that research projects, which recruit children through their school, typically yield participation rates of 30–60% (Wolfenden et al., 2009). Therefore, the average participation rate of 55% obtained in this study is within the normal range found for school based studies. Children were introduced to this study as a group during a school lesson, rather than individually or through their parents, which may have resulted in a slightly higher participation rate. The benefit of approaching the children during a school lesson was that it provided them with a good understanding of the study topic and purpose, as well as giving them ultimate control over their participation. The disadvantage of this approach was that only children with an active interest in research decided to take part. However, it is also likely that children with an active interest in the research followed instructions more carefully than would have been the case for children "told" to participate by their parents.

The mean personal exposure measured in this study was lower than the mean personal exposure measured in previous studies involving children of a similar age (Liard et al., 1999; Linaker et al., 1996; Van Roosbroeck et al., 2006). However, there are a number of differences that may explain the lower mean concentration measured in this study: the above studies were carried out 5–14 years earlier than this study and it is well documented that annual average NO_2 concentrations in the UK have decreased since 1997 (Bower et al., 2009). In addition, the above studies were carried out over 3–6 months and unlike this study did not aim to capture seasonal variation. Although the proportion of children with gas cooking appliances in their home was slightly lower (60–75%) in the above studies, the proportion of children exposed to cigarette smoke was much higher (40–60%) in the above studies. Finally, the above studies were carried out in different geographical settings.

Previous studies have shown that indirect exposure estimates derived from time spent and concentrations in MEs can be a good surrogate of personal exposure (Kousa et al., 2001) However, most previous studies based their ME concentrations on measured concentrations and few studies used models to predict concentrations in MEs (Dimitroulopoulou et al., 2006; Gulliver and Briggs, 2005).

It was found in previous studies that fixed urban monitoring sites are relatively poor predictors of personal exposure (Kousa et al., 2001; Ott, 1982). The present results confirm these findings. In this study the nearest operating urban monitor was on average about 4 km away from the child's home. Several studies have shown that intra-urban variation in air pollution can occur over short distances (Luginaah et al., 2006; Monn, 2001; Wheeler et al., 2008), therefore relatively large prediction errors would be expected and have indeed been found in this study. Furthermore, data from only 4 urban monitors were available, which meant usually >60% of children were assigned to the same urban monitor. This resulted in a lack of variation between subjects, which would make this exposure data unsuitable for an epidemiological study. It should also be noted that for the summer group no data was available from the nearest monitor and data from the second nearest monitor had to be used. The fact that urban (and personal) monitors rarely provide a complete dataset for the entire study period is often overlooked in the design of exposure studies.

Similarly to the current study, Quackenboss et al. (1986) also found that home outdoor NO_2 concentrations are a worse predictor of personal exposure than estimates derived from a time-weighted ME model. Our results show that with few exceptions the HOME method consistently overpredicted exposure of the children. This was expected as the time-activity diaries showed that the children spent approximately 90% of their time indoors, where NO_2 concentrations are usually lower than outdoors unless indoor sources of NO_2 are present. Previous studies have indicated that the home indoor environment has a large impact on personal exposure (Quackenboss et al., 1986; Zipprich et al., 2002), which is due to the large amount of time spent there. Furthermore, similar to our study Zipprich et al. (2002) found that up to 70% of the variation in personal NO_2 exposure could be explained by NO_2 concentrations in the bedroom and time spent in indoor locations. However, this also suggests that if only one ME is used to predict exposure, the home indoor concentration should be used rather than the outdoor concentration.

5. Conclusion

The prospective birth cohort study is the current gold standard epidemiological tool for studying chronic diseases that originate in early life (Rothman et al., 2008). As it is not possible to measure exposure to pollutants in study participants throughout their life, tools for modelling exposure need to be developed and their performance needs to be evaluated. This study indicates that the MEEM model developed to estimate exposure in children can predict personal exposure with an acceptable level of agreement. Furthermore, this study demonstrated that a model based on time-activity data, indoor and outdoor concentrations provides better exposure estimates in terms of absolute error than the nearest urban monitor or an outdoor pollution model. Therefore, our study clearly indicates that there are good prospects for using the MEEM model in epidemiological studies such as MAAS. However, ideally further evaluations including extending the time frame to holidays and weekends, as well as exploration of the model's performance over a wider range of exposures would be desirable.

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Appendix. Supplementary material

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.atmosenv.2012.01.030.

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