Modelling of indoor exposure to nitrogen dioxide in the UK

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Abstract

A dynamic multi-compartment computer model has been developed to describe the physical processes determining indoor pollutant concentrations as a function of outdoor concentrations, indoor emission rates and building characteristics. The model has been parameterised for typical UK homes and workplaces and linked to a time-activity model to calculate exposures for a representative homemaker, schoolchild and office worker, with respect to NO₂. The estimates of population exposures, for selected urban and rural sites, are expressed in terms of annual means and frequency of hours in which air quality standards are exceeded. The annual mean exposures are estimated to fall within the range of 5–21 ppb for homes with no source, and 21–27 ppb for homes with gas cooking, varying across sites and population groups. The contribution of outdoor exposure to annual mean NO₂ exposure varied from 5 to 24%, that of indoor penetration of outdoor air from 17 to 86% and that of gas cooking from 0 to 78%. The frequency of exposure to 1 h mean concentrations above 150 ppb was very low, except for people cooking with gas. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Indoor pollution; Nitrogen dioxide; Microenvironment modelling; Personal exposure modelling

1. Introduction

The real health effects of air pollution depend on the concentrations experienced by people rather than those recorded by stationary air quality monitors located outdoors. Two key features of people which strongly influence their exposures are their mobility and the time spent indoors (Loth and Ashmore, 1994). Recent estimates suggest that the average proportion of time spent indoors by the population in developed countries is about 90%, with considerable variation between individuals (GB Parliament House of Commons Environment Committee, 1991). Although the standards designed to protect the population from exposure to pollutants traditionally derive from ambient concentrations, the relation between these and personal exposure is not well defined. Effective strategies to reduce human exposures to air pollution should consider the contribution from both outdoor and indoor sources. Thus, recent policy encourages more research on the role of indoor pollution and the assessment of the total exposure to pollutants (DoE, 1992, 1995; IEH, 1996).

This paper is focussed on exposure to nitrogen dioxide. Several monitoring studies have measured indoor NO₂ concentrations in order to understand the contribution of indoor sources to human exposure. These studies show that indoor NO₂ concentrations may be substantially higher and more variable in homes with gas cookers, and gas or kerosene space heaters, than in homes without these appliances (e.g. Spengler et al., 1979, 1983; Quackenboss et al., 1982; Marbury et al., 1988; Melia et al., 1990).

In the United States, personal exposures of both individuals and populations have been extensively studied over the past 15 years, and have been supported...
by work in the indoor environment. Personal exposure has been most intensively investigated in a series of large-scale field NO$_2$ exposure studies (Ryan et al., 1988a, b; Spengler et al., 1994). The objectives of these direct studies were to quantify the relative influence of indoor and outdoor concentrations on personal exposure, based on activity patterns, personal and household characteristics and other seasonal and spatial variables.

Direct personal exposure measurements are expensive and technically difficult, and applicable only to samples. An alternative approach to estimating population exposures is to use appropriate computer models. A key concept developed for such modelling work in the US is the microenvironment (ME), defined as a generic location with homogeneous pollutant concentrations where people spend time (i.e. offices with smoking activity or a living room in a house with gas cookers) (Duan, 1982).

To estimate exposures, two types of models are needed: (i) physical models, to predict the pollutant concentrations in different MEs, (ii) exposure models, to simulate the movement of individuals between these MEs through time (NRC, 1991).

Most of the models developed to predict exposures to date use empirical data on indoor/outdoor ratios (I/O) to determine indoor ME concentrations as a function of time (e.g. Sexton et al., 1983; Noy et al., 1986; Lee et al., 1998). Furthermore, the current models do not link activity patterns to mechanistic modelling of ME concentrations. Therefore, they are not able to separate the role of indoor and outdoor sources, or to predict the effects of changing emission rates both indoors and outdoors.

This paper describes a new modelling approach, which incorporates the description of physical processes. In this way, it allows the relative importance of key factors influencing personal exposures to be assessed in different situations. This model can be used to distinguish three types of exposures: (i) outdoor exposure, (ii) indoor exposure resulting from penetration of outdoor air and (iii) indoor exposure resulting from indoor sources.

In the current study, the model predicts the above types of personal exposures, for a representative homemaker, schoolchild and office worker, based on indicative activity patterns. The model predictions for annual mean exposures to NO$_2$, and the proportion of hours in which short-term air quality standards for NO$_2$ are exceeded, are presented and discussed.

2. Model description

2.1. Summary of modelling approach

The deterministic modelling approach applied here combines two types of models:

(i) A physical model, used to calculate hourly indoor air pollutant concentrations for different microenvironments (MEs), as a function of outdoor concentrations, building characteristics and indoor source emissions.

(ii) An exposure model, used to calculate personal exposures, by combining the movement of typical individuals through a series of microenvironments with the modelled ME concentrations (Ott et al., 1986). The integrated mean daily exposure $E$, of an individual, can be represented as a linear combination of concentrations in $n$ distinct MEs ($C_i$), weighted by the time $t_i$ spent in each of these MEs (Duan, 1982), as follows:

$$E = \sum_{i}^{n} (C_i t_i)/24.$$  (1)

2.2. Physical model – INTAIR

The physical model, entitled INTAIR, is a simple dynamic compartment model, which solves the resulting set of differential equations using a fourth-order Runge–Kutta scheme. The model consists of two compartments. The pollutant concentration in any compartment of the model is defined by the following differential equation (Roed and Goddard, 1990):

$$\frac{dC_i}{dt} = -v_a(A_i/V_i)C_i + \lambda_e fC_o - \lambda_s C_i + \lambda_i(C_j - C_i) + Q_i/V_i,$$  (2)

where $C_i$ is the indoor concentration of the pollutant in ME $i$, $C_j$ is the indoor concentration of the pollutant in ME $j$, $C_o$ is the outdoor concentration of the pollutant, $v_a$ is the deposition velocity of the pollutant, $A_i$ is the surface area of ME $i$, $V_i$ is the volume of ME $i$, $\lambda_e$ is the air exchange rate between indoors and outdoors, $\lambda_s$ is the air exchange rate for transport of pollutants between MEs $i$ and $j$, $f$ is the building fabric filtration factor, and $Q_i$ is the indoor emission rate of the pollutant in ME $i$.

The first term of equation represents the pollutant mass deposited on the internal surfaces of the room, the second term determines the net pollutant mass entering the compartment from outdoors, the third term that leaving the compartment and the fourth term that transported between different compartments. Finally, the fifth term describes pollutant generation from indoor sources. The code is written in Visual Basic for Microsoft EXCEL and the runs have been performed in a PC environment.

2.2.1. Selection of the microenvironments

Five main MEs were identified for this modelling exercise: kitchen and living room (domestic environment – 2 compartments), office and classroom (occupational environment – 1 compartment) and outdoors. For the domestic environment, the two compartments are both linked to outdoors and the inter-room air exchange rate has been set to represent the condition of open doors between the rooms (Fig. 1). For the ME of the bedroom, based on UK field data (Coward and Raw, 1994), NO$_2$...
concentrations are assumed to be 2 ppb lower than in the living room. The five main MEs may further be subdivided into individual microenvironments, according to the type of indoor sources (cooking for the domestic environment – kitchen ME) and ventilation (natural or mechanical for offices).

In addition to the five main MEs, two more were considered, for which there are insufficient data to make representative simulations. These are transport and shops. For the shop ME, the indoor NO\textsubscript{2} concentration is assumed to be equal to that of a naturally ventilated office. For the transport ME, the NO\textsubscript{2} concentrations in cars are assumed to be double those at urban background locations (Tonkelaar, 1983). This is consistent with the ratio of roadside to urban background concentrations, measured along busy roads in Central London (QUARG, 1993).

### 2.2.2. Model parameterisation

Assuming that a constant concentration is maintained inside each ME under steady-state conditions, Eq. (2) can be re-written as

\[
\frac{C_i}{C_o} = \frac{\lambda_v f}{(v_d(A_i/V_i) + \lambda_v)}.
\]

This indoor/outdoor pollutant ratio represents a long-term equilibrium value. Thus, for a ME with no indoor sources, the protection offered against outdoor pollutants is determined by three parameters: (i) the air exchange rate \( \lambda_v \), (ii) the deposition velocity \( v_d \) and (iii) the building fabric filtration factor \( f \). Table 1 summarises the values selected for these parameters as representative of UK buildings, as explained below.

#### (i) Air exchange rate

Air exchange rate (air change h\(^{-1}\) – abbreviated to ACH) or infiltration rate \( \lambda_v \) is the rate at which air passes into a building as a result of its structural leakage or its ventilation system. In a typical British house with windows and other controllable openings closed, this rate lies in the range of 0.3–1.7, with a mean value of 0.7 ACH (Warren and Webb, 1980), which is quite high compared to housing in countries with much more severe winters (e.g. Swedish dwelling mean value: 0.25 ACH (AIVC, technical note 44, 1994)). However, in most cases, the behaviour of residents (e.g. frequency of window opening), in combination with the building characteristics, is an important factor controlling the air exchange rates. In home simulations, we assume a higher air exchange rate in the kitchen than in the living room.

For offices, naturally or mechanically ventilated, the average of the air exchange values measured by Kukadia and Palmer (1996) is adopted for the winter parameterisation. For the summer simulations, the air exchange rate for mechanically ventilated offices is assumed to remain the same, whereas for naturally ventilated offices a value of 1.5 is given; this is considered to be the highest likely value for buildings in the UK with open windows in summer.

Limited measurements of air exchange in schools have been found in the literature. Measurements in France, in two secondary schools during winter, give air exchange rates of 0.3–0.6 and 1.5–2.0 (Rachalet et al., 1994). Generally, it is difficult to estimate the air exchange rates in schools, since they depend on the different habits of children and teachers regarding the opening of the windows. In the current simulations, the air exchange rate is...
assumed to be the same as that of the naturally ventilated office.

(ii) Deposition velocity ($v_d$). The deposition velocity $v_d$ is associated with the loss rate $K$ (h$^{-1}$) of pollutants and a particular surface-to-volume ratio of the building $A/V$, and is given by the expression

$$K = v_d A/V.$$  

For pollutants with a large diffusive component (such as reactive gases), the surface area $A$ represents the total area, which incorporates both wall and floor areas. For NO$_2$, a loss rate of 0.99 h$^{-1}$ (Yamanaka, 1984) was used. Assuming $A/V$ ratios of 1.8 m$^{-1}$ for homes (Mueller et al., 1973; Hayes, 1989; Engelmann, 1992) and 0.9 m$^{-1}$ for offices and classrooms (Sabersky et al., 1973), the parameter $v_d$ then takes the values of $1.5 \times 10^{-4}$ and $3.0 \times 10^{-4}$ m s$^{-1}$, respectively. These values are consistent with those reported in the literature. For instance, measurements in residential rooms with $A/V = 2.0$ m$^{-1}$ revealed that $v_d$ for NO$_2$ varies between 1.0 and $2.0 \times 10^{-4}$ m s$^{-1}$ (Karlsson, 1994).

(iii) Building fabric filtration factor ($f$). This factor represents the ability of a pollutant to penetrate the building envelope. Based on limited measurements, it is widely believed that, in naturally ventilated buildings, the building fabric presents no barrier to particulate pollutants (Thatcher and Layton, 1995). We assume that, if $f = 1$ for particles, gases cannot penetrate less efficiently, and thus $f = 1$ for gases too.

2.2.3. Indoor sources of NO$_2$

The major indoor sources for NO$_2$ are unventilated gas and kerosene heaters and cookers. Tobacco smoking is considered to have negligible emissions compared to gas cooking (Eatough et al., 1989) and consequently it is not considered here as a source of NO$_2$.

The majority of NO$_x$ from gas combustion is emitted as NO and it is estimated that 20–30% of the undiluted products of combustion is NO$_2$ (British Research Gas, 1997). We assume that NO$_2$ represents 25% of the total NO$_x$ emissions; recent field measurements in the UK (Watt, 1999) support this assumption. The emission rate for NO$_x$ from a gas cooker is reported to be 0.125 g kWh$^{-1}$ (Wooders, 1994). Emissions from gas heating were neglected, since personal communication with gas suppliers revealed that the vast majority of appliances are exhausted outdoors.

Given the emission rate for NO$_x$, the gas consumption per household per day in the UK had to be determined. According to the UK Department of Energy (1990), the total gas consumption in the domestic sector was 283611 GWh, of which $\sim 5\%$ (15000 GWh) was consumed in cooking. The percentage of households using gas as a principal fuel for cooking in 1985 was 53%. Based on the National Census of 1981 and 1991, the number of households in 1985 is estimated to be 20 million. The rate of gas consumption for cooking is therefore estimated to be $\sim 1400$ kWh per household using gas per year. British Gas Research estimates (1997) confirm this value, giving the mean annual gas usage per household in cooking as 1465–1758 kWh.

The number of cooking hours in the UK is estimated to be 5–10 per week, i.e. approximately 1–2 h per day, 5–6 days per week (Coward and Raw, 1996). Assuming that cooking activity takes place 5–6 days per week, it may be concluded that gas consumption per household in cooking is $\sim 5–6$ kWh per day. Combining the above information, the model simulations assume that cooking activity occurs for 1 h per day, with an emission rate of NO$_2$ from a cooker of 0.875 g h$^{-1}$. For the model simulations presented in this paper, the cooking time is assumed to be 15 min in the morning (breakfast preparation, starting at 7:00 a.m.) and 45 min in the afternoon (dinner preparation, from 5:00 to 5:45 p.m.). We also assumed for these simulations that there is no seasonal variation in cooking activity.

2.2.4. Input data for outdoor NO$_2$ concentrations

Outdoor data of NO$_2$ were obtained for the urban background sites of Leeds, London (Bloomsbury), Birmingham Centre and the rural, relatively remote site of Lullington Heath from the UK Network (NETCEN Web Site, 1996), for the years 1993–1996. Diurnal concentration profiles, based on mean concentrations in each hour for these years, were constructed as model inputs for summer (April–September) and winter (October–March). The annual mean concentrations, averaged over the 3-yr period, are 8.3 ppb at Lullington Heath, 24.7 ppb at Birmingham Centre, 27.2 ppb at Leeds and 34.9 ppb at London (Bloomsbury).

2.3. Exposure model

2.3.1. Population groups – activity patterns

Activity profiles have been constructed for representative individuals from three groups:

(i) Homemakers (HM), who move mainly between the home and outdoors.
(ii) School-going children (SC), who move mainly between the home and school.
(iii) Office workers (OW), who move mainly between home and office.

All these groups are assumed to move mainly between the domestic and the occupational ME, and to spend little time in social activities. Office workers are assumed to travel to work by car, whereas the trip to school made by homemakers and children is assumed to involve walking.
### Table 2
Simulated microenvironments and percentage of time spent in each by the three groups, split by season and weekend/weekday

<table>
<thead>
<tr>
<th>Simulated population groups</th>
<th>Simulated MEs</th>
<th>Percentage of time spent in MEs (%)</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Winter weekdays</td>
<td>Winter weekends</td>
<td>Summer weekdays</td>
<td>Summer weekends</td>
<td></td>
</tr>
<tr>
<td>1. Homemakers</td>
<td>Bedroom</td>
<td>31</td>
<td>35</td>
<td>31</td>
<td>35</td>
</tr>
<tr>
<td></td>
<td>Living room</td>
<td>39</td>
<td>32</td>
<td>22</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>Kitchen</td>
<td>19</td>
<td>17</td>
<td>19</td>
<td>17</td>
</tr>
<tr>
<td></td>
<td>Outdoors</td>
<td>6</td>
<td>4</td>
<td>23</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>Shops–restaurants</td>
<td>5</td>
<td>8</td>
<td>5</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>Transport</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>4</td>
</tr>
<tr>
<td>2. Schoolchildren</td>
<td>Bedroom</td>
<td>41</td>
<td>43</td>
<td>41</td>
<td>43</td>
</tr>
<tr>
<td></td>
<td>Living room</td>
<td>20</td>
<td>33</td>
<td>14</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>Kitchen</td>
<td>6</td>
<td>8</td>
<td>6</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>Outdoors</td>
<td>6</td>
<td>4</td>
<td>12</td>
<td>21</td>
</tr>
<tr>
<td></td>
<td>School</td>
<td>23</td>
<td>—</td>
<td>23</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Shops–rest.</td>
<td>4</td>
<td>8</td>
<td>4</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>Transport</td>
<td>—</td>
<td>4</td>
<td>—</td>
<td>4</td>
</tr>
<tr>
<td>3. Office workers</td>
<td>Bedroom</td>
<td>31</td>
<td>38</td>
<td>31</td>
<td>38</td>
</tr>
<tr>
<td></td>
<td>Living room</td>
<td>22</td>
<td>38</td>
<td>14</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>Kitchen</td>
<td>6</td>
<td>8</td>
<td>6</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>Outdoors</td>
<td>2</td>
<td>4</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>Office</td>
<td>29</td>
<td>—</td>
<td>29</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Shops–rest.</td>
<td>6</td>
<td>8</td>
<td>6</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>Transport</td>
<td>4</td>
<td>4</td>
<td>4</td>
<td>4</td>
</tr>
</tbody>
</table>

Activity patterns were based on data in the BBC Survey of Daily Life 1983/1984 (BBC, 1989). The results of this survey of 6000 people in the UK, of 4 yr old and over, record the percentages of people engaged in a defined set of activities at a given time of day. The time resolution is 15 min in the busiest parts of the day and 30 min at night. Separate information is provided for weekdays and weekends, and for summer and winter. Although the survey is outdated in some respects (for example, it was carried out before the advent of Sunday shopping, so that weekend activity patterns may have changed), it is the most comprehensive of its kind available for the UK. The percentages of time spent in the different MEs by the different individuals are summarised in Table 2. The activity profiles are intended to be indicative, and clearly many individuals in each group will deviate from these activity patterns.

In the model simulations, the three individuals are at home at night, but during the day the groups differ in their activity according to their occupations. The main differences in the activity patterns between winter and summer weekdays occur in the afternoon, when homemakers and children are assumed to spend more time outdoors in the summer. From 6:00 to 7:00 p.m., all three individuals are assumed to be in the kitchen for dinner. The homemakers are assumed to spend more time in the kitchen, to prepare dinner and to wash up. Following dinner, people either stay indoors (winter simulations) or move outdoors (summer simulations).

On weekend mornings, people either stay inside the home (winter pattern) or outside (summer pattern). The main meal of the day occurs earlier (from 1:00 p.m. to 2:00 p.m.) and time is allocated in the afternoon for shopping.

#### 2.3.2. Description of the exposure model

The exposure model has been developed on a series of spreadsheets in Microsoft EXCEL. The inputs to this model are:

(i) the indicative activity patterns for each group on a weekday or weekend in winter and summer, in 15-min intervals,
(ii) the pollutant concentrations of the MEs in which this particular group is located throughout the day, as calculated by the INTAIR model, for different indoor sources, in 15 min intervals.

Each 15-min period corresponds to one ME, in which the individual is located. During this period, an individual's exposure equals the concentration in the ME. Based on this information, the personal exposure is calculated, linking the exposure throughout the day to the concentrations of all MEs where the individual is located.
Exposures for the indicative activity patterns of the three groups have been summarised in terms of (i) annual mean exposure, and (ii) proportion of hours in a year with exposures above short-term air quality standards. For NO\textsubscript{2}, this is a 1-h maximum concentration of 150 ppb (EPAQS, 1996).

3. Results

3.1. Physical model

To demonstrate the key findings of the physical model, we present the predicted NO\textsubscript{2} concentrations in the main MEs, using the outdoor data from one site in winter – Leeds (Fig. 2). When there is no indoor source (Fig. 2a), the concentrations in the kitchen are a little higher than those in the living room, due to the higher ventilation rates. In the office simulations (Fig. 2c), the indoor concentration is, as expected, higher for a naturally ventilated office than for a mechanically ventilated office. The classroom simulations, as well as those for the shop ME, produce similar concentration profiles to a naturally ventilated office, since the parameters are assumed to be the same. Gas cooking produces large intermittent increases in the NO\textsubscript{2} levels in the kitchen, with the two peaks shown in Fig. 2b corresponding to breakfast and dinner preparation; the 1 h mean concentration in the kitchen reaches a maximum of 286 ppb. The simulations, which assume that the door between the two rooms is open, also show elevated NO\textsubscript{2} levels in the living room during cooking, with a maximum 1-h mean concentration of 84 ppb.

The seasonal mean NO\textsubscript{2} concentrations in each ME, as well as the mean indoor/outdoor ratio (I/O), calculated for the Leeds data, are presented in Table 3. The simulated winter I/O values are consistent with the pattern shown in Fig. 2; values are in the range 0.44–0.55 in no-source homes, with higher values in naturally ventilated, compared with mechanically ventilated, offices, and in kitchens compared to living rooms. In homes with gas cooking, the seasonal I/O ratio is above 1.0 in the kitchen, but not in the living room. There is a higher I/O value at home in the summer than in the winter simulations (Table 3), both for the no source and the source scenarios. This is due to the increased air exchange rates, which allow pollutants to enter at a faster rate from outdoor air.

3.2. Predicted annual mean exposures

The estimated annual mean NO\textsubscript{2} exposures calculated for the individuals in the three groups and the four monitoring stations, for the no source scenario are shown in Fig. 3a. For all groups, the annual mean exposures are lower than the annual mean concentration at the corresponding monitoring site and do not exceed the 21 ppb value adopted as a guideline in the UK National Air Quality Strategy (DoE, 1997), except for the London site. The differences in exposures between groups generally reflect the differences in time spent outdoors or in vehicles. Office workers spend less time than homemakers or children outdoors, but longer in transport, and overall they have very slightly higher modelled exposures. There is also a small difference in exposure due to office ventilation (data not shown), with workers in mechanically ventilated offices having annual mean exposures which
Table 3
Summary of simulated NO$_2$ concentrations (ppb) and indoor/outdoor ratios, using outdoor data from the Leeds site

<table>
<thead>
<tr>
<th>ME</th>
<th>Winter</th>
<th></th>
<th>Summer</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Daily mean (ppb)</td>
<td>I/O</td>
<td>Daily mean (ppb)</td>
<td>I/O</td>
</tr>
<tr>
<td>Outdoor (Leeds)</td>
<td>30.3</td>
<td></td>
<td>24.1</td>
<td></td>
</tr>
<tr>
<td>Home (no source)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kitchen</td>
<td>14.7</td>
<td>0.48</td>
<td>14.6</td>
<td>0.61</td>
</tr>
<tr>
<td>Living room</td>
<td>13.4</td>
<td>0.44</td>
<td>14.6</td>
<td>0.61</td>
</tr>
<tr>
<td>Home (gas cooking)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kitchen</td>
<td>33.3</td>
<td>1.10</td>
<td>29.8</td>
<td>1.24</td>
</tr>
<tr>
<td>Living room</td>
<td>20.4</td>
<td>0.67</td>
<td>18.9</td>
<td>0.78</td>
</tr>
<tr>
<td>Office</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Natural ventilation</td>
<td>16.6</td>
<td>0.55</td>
<td>14.5</td>
<td>0.60</td>
</tr>
<tr>
<td>Mechanical ventilation</td>
<td>13.5</td>
<td>0.44</td>
<td>10.7</td>
<td>0.44</td>
</tr>
<tr>
<td>Classroom</td>
<td>16.6</td>
<td>0.55</td>
<td>14.5</td>
<td>0.60</td>
</tr>
</tbody>
</table>

are 1–2 ppb less than those in naturally ventilated offices which are shown in Fig. 3.

When the same groups are examined for homes with gas cooking (Fig. 3b), the results are very different, and there is an interaction between the groups and site factors. For homemakers, as expected, the annual mean exposures are greatly increased, by over 15 ppb, and in all cases exceed the concentrations at the corresponding monitoring sites. Homemakers at all locations experience annual mean exposures above 21 ppb. However, the size of the difference between their exposure and the monitoring site value is much greater at the rural site, which has

![Annual Mean Exposure to NO$_2$ (No source scenario)](image1)

![Annual Mean Exposure to NO$_2$ (Gas cooking scenario)](image2)

Fig. 3. Modelled annual mean exposures to NO$_2$ for four sites for homes (a) without gas cooking and (b) with gas cooking, for homemakers (HM), schoolchildren (SC), and office workers in naturally ventilated offices (OW).
Table 4
Estimated contribution (%) of penetration of outdoor air (In), indoor sources (Ck) and outdoor air (Out) to annual mean NO\textsubscript{2} exposure, for different sites and groups

<table>
<thead>
<tr>
<th></th>
<th>Lullington Heath</th>
<th>Birmingham</th>
<th>Leeds</th>
<th>London</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>In</td>
<td>Ck</td>
<td>Out</td>
<td>In</td>
</tr>
<tr>
<td>No source</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Homemaker</td>
<td>76</td>
<td>24</td>
<td>76</td>
<td>24</td>
</tr>
<tr>
<td>Schoolchild</td>
<td>79</td>
<td>21</td>
<td>79</td>
<td>21</td>
</tr>
<tr>
<td>Office worker</td>
<td>86</td>
<td>14</td>
<td>86</td>
<td>14</td>
</tr>
<tr>
<td>Gas cooking</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Homemaker</td>
<td>17</td>
<td>78</td>
<td>5</td>
<td>35</td>
</tr>
<tr>
<td>Schoolchild</td>
<td>36</td>
<td>54</td>
<td>10</td>
<td>56</td>
</tr>
<tr>
<td>Office worker</td>
<td>38</td>
<td>55</td>
<td>7</td>
<td>61</td>
</tr>
</tbody>
</table>

significantly lower outdoor concentrations. This reflects the balance between the increase in exposure during cooking activities, which is independent of location, and the exposure due to penetration of outdoor air, which depends on location and outdoor concentrations.

The increase in annual mean exposures for the other groups was, as expected, considerably smaller (about 6 ppb), as they experience elevated NO\textsubscript{2} from indoor sources primarily during time spent in the living room rather than the kitchen. The increment is slightly lower for children than for the adult groups, possibly because of the lower time spent in the living room in the evening. With the exception of the rural site, the annual mean concentrations for these groups range from slightly below, to slightly above 20 ppb, while all groups in London experience annual mean exposures above 25 ppb. The other differences between the groups largely reflect those observed in the no source scenario.

Table 4 summarises the estimated proportion of the annual mean exposure for each group which can be attributed to penetration of outdoor air, indoor sources and outdoor exposure. The relative contribution does not vary between the sites in the no home source scenario, but where a source is present, there are, as expected, differences between sites, with the contribution of indoor sources being greater at sites with lower ambient concentrations.

The contribution of outdoor exposures to annual mean exposures was relatively small. In the gas cooking scenarios, no group has a contribution from outdoor exposure greater than 15% of the annual total exposure. In the no source scenarios, the contribution of outdoor exposure for homemakers and children was above 20%, reflecting the fact that these groups spend more time outdoors than occupational groups, especially in the summer. Likewise, the contribution of penetration of outdoor air is lower at the sites with low ambient concentrations in the gas cooking scenario.

Finally, there is a large difference between monitoring sites in the contribution of indoor sources, for all groups. Homemakers have the greatest contribution to exposure from gas cookers, which provide between 45 and 78% of their total annual exposure, depending on the site. The contribution of this source for all other groups in urban areas is equal to, or below, 30%; however, even for children and office workers, it exceeds 50% at the rural location.

3.3. Frequency of peak exposures

The percentage of hours in which the UK 1 h mean standard for NO\textsubscript{2} of 150 ppb was exceeded differed dramatically between the groups. At each site, homemakers, cooking with gas for the periods assumed in the activity profiles, would experience 1-h mean exposures above 150 ppb for at least 1 h on every day of the year, i.e. for 4.2% of hours. In practice, cooking activity will vary from day to day, and from person to person, and hence this exposure will be quite variable. However, modelling of this would require much more detailed information on cooking activity patterns.

In contrast, all the other groups have very low frequencies of exposure to concentrations above 150 ppb, with the greatest predicted frequencies being no more than 0.02% (0–2 h over the year) for the most polluted site in central London. Thus, for all groups except homemakers, indoor sources of NO\textsubscript{2} did not increase dramatically the frequency of exposure to NO\textsubscript{2} concentrations above 150 ppb. The highest 1-h concentrations for these other groups were generally experienced outdoors or when using transport, or in the living room, during gas cooking. This finding again highlights the significance of the assumed activity patterns for the findings of the study. Similarly, the assumptions about the penetration of NO\textsubscript{2} generated in the kitchen into the living room will be critical, and will depend on the specific house geometry.
4. Discussion

This study has provided, for the first time, modelled estimates of exposure to NO\textsubscript{2} in the UK, which include indoor exposures. A modelling approach has been adopted, which links a physical microenvironmental model with a time-activity model. The value of our approach is that allows the effects of different physical features of the indoor microenvironments and different activity patterns to be quantitatively compared. Furthermore, it allows the separation of indoor exposure into that due to indoor sources and that due to penetration of outdoor air. However, the use of mean values for the parameters and a single indicative activity profile for each group in our study means that the expected variability in personal exposures is not represented in the model predictions, and the results of this study can only be indicative of NO\textsubscript{2} exposure patterns in the UK population.

Nevertheless, the model predictions clearly demonstrate the large effect of indoor activities on personal exposures, which differ greatly from those monitored at the corresponding fixed site monitors, and the large contrasts between exposures, which arise from differences in activity patterns and exposure to indoor sources. Therefore, although the model predictions can only be regarded as indicative of population exposures, this type of preliminary modelling work is of considerable value. It identifies key factors which influence indoor concentrations and personal exposures in the UK, providing a first estimate of the relative importance of pollutant sources for chronic and acute health effects and evaluating potential implications for policy on air pollution control both outdoors and indoors.

Due to the limited data on the indoor environment in the UK, many of the parameters of the physical model can only be estimated, or based on studies carried out elsewhere where building design and climate may differ from the UK. The key uncertainties in the physical model are as follows:

(i) the fact that the two-compartment approach does not adequately represent the actual homes in which people live;
(ii) the limited UK data available to quantify the key parameters of the model. Current cooking and activity patterns may differ substantially from those in the 1980s and may vary seasonally more than the current simulations allowed. For source strength, especially, there is a great uncertainty regarding the current applicability of 1985 data on gas usage, given socio-demographic changes over the last 15 years;
(iii) the limited representivity of the monitoring sites, especially given the variation in locations used by individuals.

Nevertheless, the model does provide results which are broadly consistent with mean I/O ratios reported for the UK. The most comprehensive study in UK homes, covering 174 homes in the Avon area (Coward and Raw, 1994), showed mean values of 0.6–0.7 over a year in homes without gas cooking, while shorter-term measurements gave values for London of 0.6 ± 0.3 and for Watford of 0.8 ± 0.4 (Spengler et al., 1996). Mean I/O ratios for homes with gas cooking in the study of Coward and Raw (1994) were approximately 1.35 for kitchens and 0.9 for living rooms, again values which are slightly higher than those predicted in our simulations for Leeds (Table 3); however, in this case the value of I/O ratio will depend on the outdoor NO\textsubscript{2} concentration. Furthermore, a recent BRE study of personal exposures in Hertfordshire reveals I/O ratios of 0.84 for the office environment, 1.52 for kitchen with gas cooker, 0.67 for kitchen with electric cooker, 0.8 for living rooms and 0.6 for bedrooms (Crump et al., 1998). The peak concentrations associated with gas cooking are also consistent with other data reported in the literature. For instance, Ross (1996) found maximum 1-h mean levels of 233 ± 144 ppb (range 53–593 ppb) in kitchens during gas cooking and 113 ± 78 ppb (range 37–295 ppb) in living rooms, in a survey of 10 UK homes. These values compare well with the 1 h maximum concentrations of 286 ppb in the kitchen and 84 ppb in the living room, predicted by our model.

The results from the exposure model (Table 4, Fig. 3) can be compared with the results from a recent BRE study involving 30 office workers in Hertfordshire (Crump et al., 1998). Fixed site concentrations and personal exposures were measured by diffusive samplers over a 14-day period. The personal exposure to NO\textsubscript{2} was 11.6 ± 4.2 ppb (12.7 ± 4.1, for those with gas cookers, 9.9 ± 4.1, for those with electric cookers), while the outdoor concentration outside the work was 16.2 ± 4.2 ppb and outside home 15.2 ± 5.2 ppb. Referring to Fig. 3, our model would predict, for these outdoor concentrations, which are intermediate between Lullington Heath and Birmingham, an annual mean exposure for office workers of about 10 ppb in no source homes about 15 ppb in gas cooking homes, which are broadly consistent with the measured data. The greatest contribution to measured exposures was due to the time spent in buildings (mean 85%), with the outdoors accounting for 11%, and the time in vehicles for 3% of total exposure, again consistent with our model predictions (cf. Table 4). From the above comparisons, it is clear that, while a comprehensive validation of the model with UK data was not possible, both the physical model and the exposure model can predict values which are consistent with, or slightly underestimate, measured I/O ratios, indoor concentrations and personal exposures for NO\textsubscript{2} in the UK. The underestimation of indoor concentrations and I/O ratios is most likely to be due to our model assuming a value of
air exchange rate which is lower than that in the homes used in these UK field studies. A formal Monte Carlo analysis, which was beyond the scope of this initial study, would allow the effect of uncertainties in model parameterisation on the predicted indoor concentrations and personal exposures to be examined in more detail.

The exposure model predictions identify the homemakers using gas cookers as the group receiving particularly high exposures to NO\textsubscript{2}. This group consistently experienced annual mean exposures greater than those at the corresponding monitoring site; in the case of the rural site the increase was particularly marked. It is noteworthy that, using the outdoor data for London, all four groups experienced annual mean exposures above 25 ppb when they had homes with gas cooking. It is clear that the annual mean exposure of these groups is critically dependent on the assumptions made about the amount of cooking and their location during cooking activities. The extent to which cooking increases NO\textsubscript{2} concentrations in other rooms may also be an important factor; for example, Colucci (1997) found higher concentrations in living rooms and bedrooms of small flats than in larger houses.

The fact that the estimates of population exposure made in this study are very different from those which would have been made based on outdoor concentrations indicates the importance of research on population exposures to major air pollutants indoors. As this study has shown, the exposure indoors is critically dependent on physical features of the indoor environment which are, as yet, poorly characterised in the UK, including air exchange rates, deposition rates, and indoor source strengths. The ways in which these and other parameters are influenced by building design, meteorological conditions, ventilation characteristics and indoor design, among other parameters, need to be urgently explored. Finally, the interactions between these factors and the range of different activity patterns within the UK population need to be examined in more detail.

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