Activity Pattern and Personal Exposure to Nitrogen Dioxide in Indoor and Outdoor Microenvironments

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ABSTRACT

People are exposed to air pollution from a range of indoor and outdoor sources. Concentrations of nitrogen dioxide (NO_2), which is hazardous to health, can be significant in both types of environments. This paper reports on the measurement and analysis of indoor and outdoor NO_2 concentrations and their comparison with measured personal exposure in various microenvironments during winter and summer seasons. Furthermore, the relationship between NO_2 personal exposure in various microenvironments and including activities patterns were also studied. Personal, indoor microenvironments and outdoor measurements of NO_2 levels were conducted using Palmes tubes for 60 subjects. The results showed significant differences in indoor and outdoor NO_2 concentrations in winter but not for summer. In winter, indoor NO_2 concentrations were found to be strongly correlated with personal exposure levels. NO_2 concentration in houses using a gas cooker were higher in all rooms than those with an electric cooker during the winter campaign, whereas there was no significant difference were noticed in summer. The average NO_2 levels in kitchens with a gas cooker were twice as high as those with an electric cooker, with no significant difference in the summer period. A time-weighted average personal exposure was calculated and compared with measured personal exposures in various indoor microenvironments (e.g. front doors, bedroom, living room and kitchen); including non-smokers, passive smokers and smoker. The estimated results were closely correlated, but showed some underestimation of the measured personal exposures to NO_2 concentrations. Interestingly, for our particular study higher NO_2 personal exposure levels were found during summer (14.0±1.5) than winter (9.5±2.4).

Key words: nitrogen dioxide, indoor and outdoor sources, gas/electric cooking, personal exposure, smokers, NO_2/ NO_x ratio, time weighted average modelling

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1. INTRODUCTION

Nitrogen dioxide (NO_2) is one of the most common air pollutants in ambient and indoor air (Lai et al., 2006; Hanninen et al., 2004). The major outdoor source of NO_2 concentrations are mobile and stationary combustion sources (Kampa and Castanas, 2008; Lewne et al., 2004), whereas indoor sources includes gas cookers, wood stoves, fireplaces, and environmental tobacco smoke (ETS). NO_2 is formed from the combination of nitrogen and oxygen (O_2) during high temperature combustion processes (Brunekreef, 2001; Baili, et al., 1999). NO_2 and associated compounds can also produce secondary aerosol by the photochemical oxidation (Bencs et al., 2008). In some indoor environments such as industrial workplaces and in homes with gas stoves, peak concentrations may reach 1 to 2 ppm with a 24-h averages NO_2 concentration up to 0.5 ppm (Chan et al., 2007; Monn, 2001).

NO_2 is an irritant gas and can increase susceptibility to airway infections and impair lung function in exposed populations (Kattan et al., 2007; Curtis et al., 2006; Kraft et al., 2005). Several, multi- and single-pollutants time-series studies have also found association between NO_2 and non accidental mortality (Beelen et al., 2008; Brook et al., 2007; Burnett et al., 2004). Table 1 summarizes some of the short-term and long-term health effects of NO_2 exposure over various concentration and exposure time. A review by Latza et al. (2009) also examines some recent studies assessing the health effects of environmental NO_2. The toxicity of NO_2 depends on its oxidative and free radical properties, as well as its ability to form nitric and nitrous acids in aqueous solution on the moist surfaces (Sandström, 1995; Utell, et al., 1991). Its main effect, therefore, on human health is to damage respiratory tract cells such as mucous membranes of the lung (Frampton et al., 2002; Blomberg et al., 1999; Spengler et al. 1983). Hence it is important to study the factors that lead to personal exposure to air pollutants such as NO_2 and how it can be assessed.

The personal exposure to air pollutants from both indoor and outdoor sources has recently received high attention (Krzyzanowski, 2008; Chaloulakou et al., 2008; Mitchell et al., 2007). Personal exposure to pollutants like NO_2 depends on the concentration of NO_2 in microenvironments and the time that one spends in those microenvironments (see for example, Ott, 1982; Monn, 2001; Harrison, et al., 2002). Although high ambient NO_2 concentrations are dangerous to health, indoor NO_2 concentrations can pose a greater health risk due to people spending most of their time indoors. In indoor environments where ventilation is restricted, using wood, solid, liquid and gaseous fuels in a small space in home can lead to high exposure. However, the NO_2 levels may be comparatively lower in newly built houses with proper ventilation (Willers et al., 2006). NO_2 is often found at higher concentrations indoors than outdoors (Lai et al., 2006; Garcıa Algar et al., 2004; Lee, et al., 2002; Bailie, et al., 1999), and houses with gas cookers have been found to have much higher mean 24-hr concentrations than houses with electric cookers (Willers et al., 2006; Hanninen et al., 2004; Berglund, 1993).

This paper examines the relationship between measurements of personal exposure levels of office workers to NO_2 and those measured in microenvironments for an area of Hertfordshire and North London, UK. Although people may be exposed to several different sources during a typical day depending on their activity patterns, this paper focuses on levels measured in the work place, the home and outdoors and how these explain the overall personal exposure of the subjects. This work has implications for air quality monitoring networks and their representativeness of personal levels of exposure to air pollution.

2. METHODOLOGY

2.1 The Study area

The study was carried out in north London and Hertfordshire which consists of several small and medium sized towns (shown in Figure 1). Hertfordshire is a county adjacent to the north of London, which covers an area of 1643sq km and has a population of over 1 million. The county has important transport links, with the A1(M) and M1 motorways for traffic travelling north and south. M25 is a major motorway to the south of the county and encompasses the Greater London area.

2.2 Target population

The target populations of this study were 21 - 60 year old office workers living and working in urban areas in Hertfordshire and north London. For the winter period of 2000, a random sample of 60 office workers were asked to fill in their activities diaries and questionnaires. This number of subjects is in accordance with the WHO guidance of having sample of a minimum of 50 subjects for the sample to be representative of a target population (see for example, EXPOLIS, 1999, WHO 2000). At the same time, weekly average concentrations of NO_2 (personal, bedroom, living room, kitchen, outside front door, office and inside car were measured using two passive Palmes diffusion tubes at each site. Correlations between weekly personal exposures and mean indoor and outdoor concentrations during the same periods were examined. In addition, 30 individuals from winter study participated again in a summer season campaign (2001). The lower number was due to the fact that not all subjects from the winter study were able to participate in this second campaign. The supplementary data (Table S1-S2) shows various detail including the age distribution, male/female ratio, houses with gas cooker, electric cooker, smokers, non-smokers etc.

2.3 Monitoring strategy

During winter 2000 and summer 2001 passive NO_2 diffusion tubes (Palmes, et al. 1976) were used to measure weekly average NO_2 concentrations for fixed indoors microenvironments, an outdoor site and personal average exposures of individuals. The Palmes tube method is simple to use with the tubes having a long shelf live before and after exposure giving both reliable and reproducible results (Bush, et al., 2001). The diffusion tube relies on molecular diffusion of NO_2 through a vertical acrylic tube of known length and cross-sectional area onto a reactive surface or absorbent mesh coated with triethanolamine (TEA) where the molecule is captured by chemical reaction forming a nitrite. After exposure to NO_2 for a seven-day period, the reactive surface is analysed using UV/VIS spectrophotometry at 540 nm and the integrated loading of the reaction product is used to infer the average gas concentration (Palmes et al. 1976). All tubes were prepared and analysed at the University of Hertfordshire laboratory.

2.4 Siting protocol for passive diffusion tubes

Indoor passive tubes were placed to avoid windows, corners, and heating vents and outdoor passive tubes were located outside homes, approximately 2 m above the ground away from possible localized pollutant sources such as driveways, roads and exhaust vents. All tubes were tracked by individual identification numbers, which were also recorded on their activity diaries and questionnaires. Volunteers were instructed to wear the passive tubes at breathing height by clipping them onto their collar or lapel, to keep them outside of coats and to keep tubes nearby when not wearing them, for example, while sleeping, having a shower or taking a bath.

2.5 Statistical analyses

Statistical analysis was performed with SPSS software. Descriptive data or simple summary statistics (mean, standard deviation, maximum and minimum) were derived to describe the distribution of NO_2 concentrations to which the individuals were exposed. Pair t-test for mean values were performed to find any differences between time weighted average NO_2 exposure values and average personal exposure to NO_2 concentrations. Standard multiple regression analysis was used to assess the importance of indoor NO_2 concentrations measured over the 7-day period. Pearson's correlation coefficient was used to summarise the relationship between personal exposure and the exposure levels measured in microenvironments.

2.6 Calculation of time weighted average micro-environmental exposure

Time weighted average micro-environmental exposure was estimated based on weekly average NO_2 concentrations from home indoor (bedroom, living room and kitchen) and outdoor including in office and car and time activity diaries according to the following equation:

$$E_i = \sum_{j}^{J} C_j t_{ij} \qquad (1)$$

where

- E_i is the NO_2 time weighted average exposure for person i over the specified time period;
- C_j is the NO_2 concentration in microenvironment j;
- t_{ij} is the aggregate time that person i spends in microenvironment j;
- J is the total number of microenvironments that the person i moves through during the specified time period such as indoors at home, indoors at work, indoors in other locations, in transit, and outdoors.

3. RESULTS AND DISCUSSION

3.1 Questionnaires and Time activity diary data

The time activities diaries were filled by 55 subjects (out of a total of 60 volunteers) in winter 2000. Analysis of their activities showed that all volunteers spent more than 80% of their time indoors. The time spent in each microenvironment over the week is shown in Figure 2. Over 50% of the time was spent at homes during winter but less in summer periods, followed by about 30% of the time being spent at the workplaces. The individuals spent 5.5% (in winter) and 4.6% (in summer) of their time in other non-smoking areas such as shopping malls and cinemas, and 2.7% on average in other smoking areas such as in restaurants and public houses. With regard to travelling time, the average total time spent in the traffic was about 45 minutes per day, equivalent to 4.5% (winter) and 4.0% (summer) of the daily activities time. The individuals spent three times (11.9%) of the total daily activities time outdoors during summer in comparison to winter (4%). These results are in agreement with other European studies e.g. Piechocki-Minguy et al., (2006); Harrison, et al., (2002) and EXPOLIS (1999).

3.2 Average personal exposure to NO_2 and average NO_2 concentrations in microenvironments

In winter, average NO_2 concentrations in bedroom, living room and kitchen were significantly higher ($p<0.05$) in houses with gas cookers compared with those with electric cookers, while there was also significant difference in personal exposure. Weekly average NO_2 concentrations in all microenvironments are depicted in Figure 3. The result shows that personal exposure to NO_2 concentrations for all volunteers ranged from 5.7 to 15.4 ppb. This range was 6.3 to 15.4 ppb for volunteers using gas cookers and 5.7 to 11.0 ppb for those using electric cookers. NO_2 concentrations in the bedroom ranged from 3.2 to 15.5 ppb with 6.3 to 15.5 ppb in houses with gas cookers and 3.2 to 11.1 ppb in houses with electric cookers. NO_2 concentrations in the living room ranged from 4.1 to 30.1 ppb with 6.1 to 30.1 in houses with gas cookers and from 4.1 to 11.4 in houses with electric cookers. The results also show that weekly mean NO_2 concentrations in the kitchens with gas cookers ranged from 12.9 to 38.8 ppb (Table 2a). These values were higher than for the kitchens with electric cookers (4.2-9.7 ppb).

In summer, there were no significant differences in NO_2 concentrations in rooms of houses with electric or gas cookers or in personal exposure. Weekly average NO_2 concentrations in all microenvironments are presented in Figure 3. The results shows that personal exposure of volunteers using gas cookers ranged from 12.7 to 18.1 ppb (average 14.6 ppb) and from 11.3 to 15.3 ppb for those volunteers using electric cookers (average 13.3 ppb). Further, it was also noticed that NO_2 levels in kitchens with gas cookers (ranging from 12.8 to 17.7 ppb) were higher than those with electric cookers (ranging from 8.0 to 13.3 ppb). NO_2 concentrations in the bedroom ranged from 12.5 to 17.3 ppb in houses with gas cookers and 10.6 to 14.8 ppb in those with electric cookers (Table 2b). NO_2 concentrations in the living room ranged from 10.8 to 18.2 ppb (from 13.2 to 18.2 in house with gas cookers and from 10.8 to 15.4 in houses with electric cookers).

The highest difference between concentrations in gas cooker and electric cooker houses is observed for kitchens and this would be expected as cooking appliances represent a dominant exposure source. The results also show that indoor contribution to personal exposure to NO_2 is important for winter months especially for those people living in houses with gas cookers. During the summer period, when ventilation is high (such as through open windows), the variation between the microenvironment concentrations is less than for winter (see values of standard deviation in Table 2). Similarly, there is less difference between the personal exposure and microenvironmental levels for the summer period due to the higher ventilation rates and because people tend to spend less time at home.

3.3 Personal exposure to NO_2 and indoor/outdoor concentrations

Outdoors NO_2 concentrations were significantly higher when compared with indoor concentrations for the winter period. Average outdoors NO_2 concentrations ranged from 8.1 to 16.1 ppb, with an average of 12.9 ppb and NO_2 concentrations in offices ranged from 5.6 to 13.5 ppb (average 8.8 ppb) and average NO_2 concentrations in the cars ranged from 4.1 to 11.3 ppb (average 6.8 ppb). In contrast, outdoor NO_2 concentrations were not significantly different from indoor concentrations in summer. The outdoors concentrations ranged from 11.3 to 18.8 ppb (average 14.5 ppb). NO_2 concentrations in offices ranged from 9.2 to 15.2 ppb (average 12.2 ppb) and in cars ranged from 8.7 to 14.4 ppb (average 11.4 ppb). Winter periods are often subject to higher congestion and hence higher traffic emissions of nitrogen oxides. In addition, stable atmospheric conditions are common which restricts the dispersion of air pollution giving rise to higher concentrations especially in cities (see for example, Kukkonen, et. al., 2005). During summer times in contrast, traffic levels can be lower (e.g. due to holiday periods) and greater dispersion which can lead lower ambient levels of pollutants like NO_2.

Outdoor levels of NO_2 could be higher due to increased primary NO_2 emissions from vehicles. Several studies have shown that while NO_x levels show a decreasing trend, the NO_2 levels remain constant in urban centres i.e. higher NO_2/NO_x ratio (Carslaw, 2005; AQEG, 2006; Carslaw et al., 2007; Carslaw and Carslaw, 2007; Grice et al., 2009). These studies also suggest that this increase may be due to an increased NO/NO_2 ration in vehicular exhaust. Furthermore, present vehicular emission control technologies (such as oxidation catalyst, catalytic diesel particulate filter) also contribute to an increased NO_2/NO_x ratio due to increase primary NO_2 emissions. Interestingly an increase in the NO_2/NO_x ratio could also lead to the increased urban ozone levels (Carslaw and Carslaw, 2007).

A summary of personal exposure in different microenvironment is shown in Figure 4 which illustrates that significant exposure occur from indoor sources. This obviously has implications for groups other than office workers, in particular individuals who are elderly or have existing illness and spend most of their time indoors. It also shows the importance of outdoor contributions to the overall exposure for the summer period.

Several microenvironmental models have been proposed to predict indoor NO_2 concentrations as a function of outdoor concentrations, indoor source strength, and key building parameters such as infiltration, ventilation (Sexton et al., 1983; Rijnders et al., 2001; Kulkarni and Patil, 2002; Milner et al., 2005; Dimitroulopoulou et al., 2001, 2006). Sexton et al. (1983) proposed a simple deterministic model to relate exposure to background ambient levels, indoor values, and human activities. The study indicates that indoor NO_2 concentrations vary primarily with outdoor levels and type of cooking fuel, but are also affected by factors such as air-exchange rates and strength of indoor sources. Rijnders et al. (2001) have shown that personal and outdoor NO_2 concentrations are significantly influenced by (a) the degree of urbanization (b) by the traffic density and by (c) distance to a nearby highway. However, considering the above discussion, it would be useful to develop a holistic model that can consider the relative contribution of the outdoor levels to the indoor concentrations, microenvironment designs/structure, influence of personal habits and time activity patterns, including physical properties and cocktail effects of chemicals/reactivity of air pollutants and consequently the implication on personal exposure.

3.4 Average personal exposure of non-smokers, passive smokers and smokers to NO_2

Personal exposure of non-smokers, passive smokers and smokers to NO_2 concentrations and NO_2 concentrations in house microenvironments with electric and gas cookers is shown in Figures 5 and 6. The results from the winter study clearly showed that, average personal exposure to NO_2 of smokers in houses with gas cookers (13.6 ppb) was higher than those non-smokers (10.8 ppb) and passive smokers (10.9 ppb). Furthermore, small but significant differences were noticed for personal exposure to non-smokers, passive smokers and smokers in houses with electric cookers (8.1, 8.7 and 9.4 ppb, respectively). The study also shows that average NO_2 concentrations in kitchens, living rooms and bedroom of smokers using gas cookers were found to be higher than the rooms of smokers, non-smokers and passive smokers with electric cookers. Furthermore various microenvironments have comparatively higher NO_2 levels with smoker and hence it can be suggested that smoking also influence the NO_2 exposure. Further the personal exposure risk may increase during winter, when the windows of a house are kept closed during most of the period.

Results from summer studies showed that average personal exposure to NO_2 of smokers in houses with gas cookers (17.0 ppb) was higher than those non-smokers (14.9 ppb) and passive smokers (13.7 ppb). However, there was no significant difference between personal exposure of non-smokers, passive and smokers in houses with electric cookers (13.1, 13.7 and 13.4 ppb, respectively). Significant differences were found between the average NO_2 concentration in bedrooms and living rooms of smokers using gas cookers and those for rooms of non-smokers and passive smokers. No difference was found for the other areas for non-smokers and passive smokers using gas cookers or electric cookers.

3.5 Time-weighted average personal exposure to NO_2 concentrations

Paired t-test was used to analyse the data for significance. It showed that there was a non-significant difference at the 95% level between time-weighted average NO_2 microenvironment concentrations and average personal exposure to NO_2 concentrations for the winter season. The results show that overall time weighted average ranged from 6.6 to 15.4 ppb (average 10.9 ppb) and the time weighted average of smokers, non-smokers, and passive smokers using electric cookers ranged from 7.0 to 9.3 ppb (mean: 8.6 ppb), 5.4 to 11.7 ppb (mean: 7.6 ppb) and 6.7 to 9.7 ppb (mean: 8.2 ppb) respectively. The time weighted average of smokers, non-smokers, and passive smokers using gas cookers ranged from 11.2 to 12.0 ppb (mean: 11.6 ppb), 7.1 to 14.7 ppb (mean: 10.4 ppb) and 6.7 to 16.5 ppb (mean: 10.5 ppb) respectively**.** The time weighted average gave a good approximation of personal exposure NO_2 levels although there was a small (~6%) underestimation ($y = 0.9433x$, $R^2 = 0.8535$).

In contrast, significant different at the 95% level were found in summer. The results shows that overall time weighted average ranged from 11.0 to 16.3 ppb (average 13.1 ppb). Further, the time weighted average of volunteers using gas cookers ranged from 12.0 to 16.3 (average 12.5 ppb) and from 11.0 to 14.1 (average 13.8 ppb) for those using electric cookers. This is the opposite trend to that observed for winter where the mean of the time-weighted microenvironment concentrations was higher for the cases where gas cookers where used. Higher average time weighted concentrations of non-smokers, passive smokers and smokers using gas cookers (13.6, 13.4 and 15.6 ppb respectively) was observed than those using electric cookers (12.5, 12.9 and 11.6 ppb respectively). The time-weighted average exposure was also plotted against the personal exposure to NO_2 concentrations as shown in Figure 7 which shows that just over 65% of the time weighted average correlates with the direct personal exposure measurements of NO_2 concentrations ($y = 0.5934x + 4.7931$, $R^2 = 0.6533$).

A probable reason for this large unexplained fraction was that the volunteers spent more time outside during the summer campaign and were involved with activities not recorded in present case. In addition, there is an intercept of nearly 5 ppb which indicates there were NO_2 pollution levels to which the person was not exposed. Another complication during summer period is the infiltration of outdoor air into indoors through open windows which was not specifically investigated as part of this study. It should be noted that the number of subjects were significantly less than for the winter campaign and hence the uncertainties in the dataset will be higher.

3.6 Comparison of the current work with other studies

Table 3 also shows a comparative overview of recent studies on NO_2 concentration in various microenvironments and personal exposure. It is interesting to note that none of the studies really covered all the microenvironments as studied in the present case. This highlights the significance of the present study in explaining much of personal exposure assessment of NO_2, especially for the winter case where people spend more time indoor.

Compared to other UK studies outdoor NO_2 levels in Hertfordshire falls towards lower range. For example, outdoor level reported include Ashford: 12.4 ppb; Birmingham: 10 ppb; London: 22 ppb; Oxford: 12.4 ppb, Southampton: 27 ppb (Lai et al., 2004; Gracia-Algar et al., 2004; Harrison et al., 2002; Levy et al., 1998; Linaker et al. 1998). Campbell et al., (1994) measured the NO_2 concentration at 243 validated urban sites throughout the UK and the average concentrations varied from 10 ppb (northern Scotland) to 50 ppb (near road side in London). Compared to other European cities (Table 3), outdoor NO_2 levels falls in lower to moderate range in UK. On a global prospective, Asian cities seem to have highest outdoor NO_2 concentrations such as Delhi (36.4±15.6ppb), Hong Kong (38±8 ppb), where vehicular emission seems to be a major contributor (Ravindra et al., 2006, 2003; Chao et al., 2000). European Union aim to achieve an outdoor annual average guide value of 21.3 ppb by 2010.

In addition to the Table 3, various other studies also report the indoor NO_2 levels in European cities such as Kuopio (5.5 ppb), Kjeller (7.8 ppb), Geneva (8.3 ppb), Avon (6.8 ppb), Hamburg (8.8 ppb), Erfurt (9.0 ppb) (Cyrys et al., 2000; Levy et al., 1998; Farrow et al., 1997). However, only few studies report the levels in different microenvironment as depicted in Table 3. It shows that indoor levels are comparable to reported values in UK cities (Lai et al., 2004; Harrison et al., 2002) and also some of the European cities. However, higher indoor levels were also reported for Antwerp, Barcelona, Paris and Prague (Table 3). A study by Breysee et al. (2005), also reports significantly higher levels of indoor NO_2 in Baltimore, USA.

The NO_2 concentration in various microenvironments are related with various factors such as ventilation, electrical/gas cooking or heating, ETS etc (Zota et al., 2005; Gallelli et al., 2002). Algar-Gracia et al. (2004) reported that gas fire increase average NO_2 concentrations by 1.3 fold and gas cooker by 2.1 times. Further, the outdoor NO_2 levels and seasons variability can also influence the levels in these microenvironments (Franklin et al., 2006). People living in small apartments with limited ventilation and lack of local exhaust mechanism have probability to high personal exposure. Gas stoves are typically used for brief periods and combustion by product from these sources are not evenly distributed in an apartment (Zota et al., 2005) and hence NO_2 levels may exceed the maximum hourly limits of NO_2 in kitchen during cooking. However, long term monitoring approaches only provide limited possibilities to study the health effect of short term peak exposure to NO_2. Hence further studies would be needed to address this challenge and to develop a strong database for short term exposure assessment.

In addition to the present study, only few studies have monitored NO_2 levels inside a vehicle (Lewne et al., 2006, Harrison et al., 2002). Interestingly, the NO_2 level seems to be lower in a personal car than a taxi, bus and lorry although this would also be related to the time, traffic intensity and the location in different environments. The NO_2 levels in offices/workplaces/schools seems to be slightly lower than indoor except in some cases; where the they may be situated near an area with dense vehicular activities or near a highway. This aspect, however, was not studied in this work as most work places were away from particular sources such as major roads.

Levy at al. (1998) studied 18 cities in 15 countries around the world and found that personal NO_2 exposures were more strongly correlated with indoor concentrations (r = 0.75) than with outdoor concentrations (r = 0.57) when all countries were considered simultaneously. Linaker et al., (1998) noticed that personal exposure to NO_2 in school children of Southampton and levels ranged from 6 to 137 ppb with a geometric mean of 199 ppb. In contrast to other studies (Monn et al., 1998; Zota et al., 2005; Piechocki-Minguy et al., 2006, see Table 3) personal exposures were found to be higher in summer than winter at Hertfordshire. This could indicate that for this particular cohort of subjects and their micro-environments, outdoor sources were particularly important. Further the personal exposure seems to be strongly dependent on both the levels of NO_2 in indoor and outdoor environments (see Figure 3 and 4). In certain cases, as shown by Lewne et al (2007), specific micro-environments can play an important role in determining the overall exposure level as in the of bus drivers, taxi drivers and tunnel workers.

Although there are ambient air pollution guidelines and threshold values for NO_2, they are limited for indoor air quality (Franchi et al., 2004; 2006; WHO, 2000, 2003). Table 4 shows an overview of outdoor/indoor air quality guide values for NO_2. Kraft et al. (2007) proposed two short-term NO_2 exposure values to protect public health i.e. 53.2 ppb (for 1 hour) and 26.6 ppb (for 24 hours) based on the results of exposure-chamber experiments. A recent study by Pilotto et al., (2004) suggests that a reduction in NO_2 exposure can reduce asthma symptoms. Hence, it would be more useful to propose indoor to supplement the outdoor guidelines and threshold values for NO_2 based on personal exposure in different microenvironments.

4. CONCLUSIONS

Nitrogen dioxide concentration measurements of personal exposure, home and workplace microenvironments (e.g. office, bedroom, living room and kitchen), outdoor and in-car are presented for Hertfordshire and north London during winter and summer. A number of key conclusions can be drawn from this work

(i) The study revealed that average NO_2 concentrations were higher in the various microenvironment of a house with a gas cooker than a house with an electric cooker. Hence, kitchens are a major source of indoor NO_2.

(ii) The concentration in kitchens with gas cooker was noticed to be 2–3 times higher than those with electric cooker. The use of gas cooker in house with poor ventilation significantly increases the risk of high NO_2 personal exposure in indoor microenvironments.

(iii) In comparison to the passive smokers and non-smokers, the highest personal exposures were noticed in smoker's house with the risk of exposure rising further with the use of gas cookers.

(iv) The work shows that where gas cookers are not being used, the outdoor NO_2 could be a major source of indoor NO_2 concentration in various microenvironments.

(v) The high levels of NO_2 were observed in summer but a high correlation was observed between the measured personal exposure and time-weighted microenvironment concentrations during winter.

(vi) A comparison of personal exposure of NO_2 and levels in various microenvironments was also performed with other recent indoor studies. The levels of NO_2 ranged from 3.2 ppb to 30.1 ppb during summer and winter seasons for the various microenvironments.

(vii) The study also supports the conclusion that indoor NO_2 concentrations can better explain personal exposure than outdoor concentrations alone.

(viii) In light of the above points the time weighted average exposure to NO_2 gave a good approximation of personal exposure with some underestimation, when compared with personal exposure to NO_2 concentrations. The mean indoor NO_2 concentrations, especially in bedrooms, have been found to reflect personal exposure closely.

(ix) As we spend most of our time indoor, it is suggested that indoor guide values based on personal exposure in different microenvironments should be developed to support limit values for outdoor levels.

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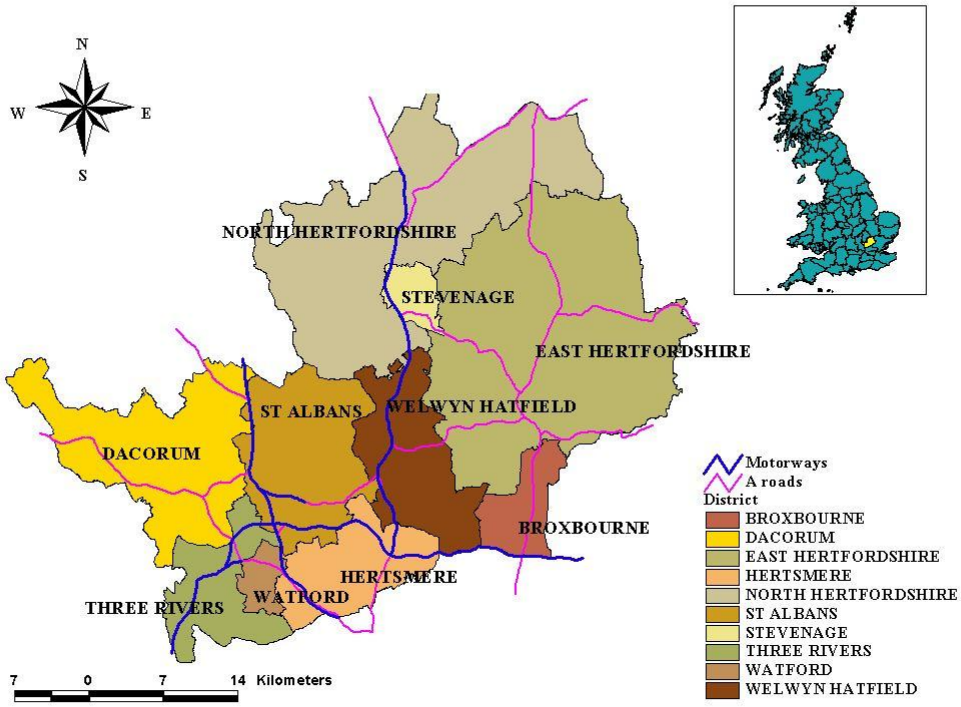
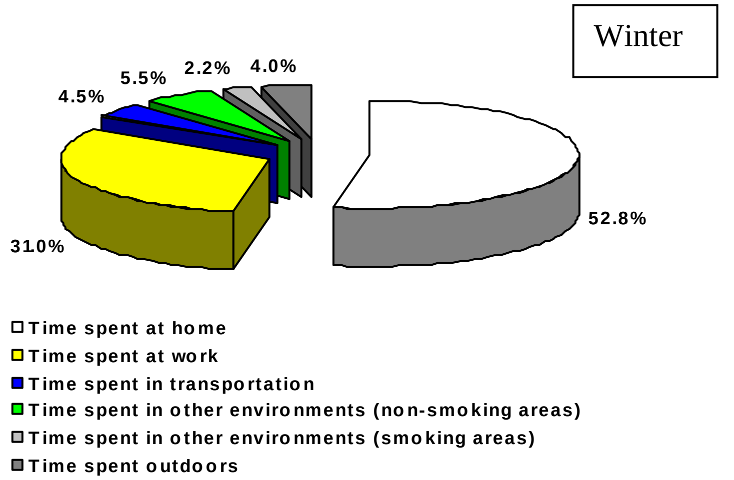


Figure 1: Sketch map of Hertfordshire



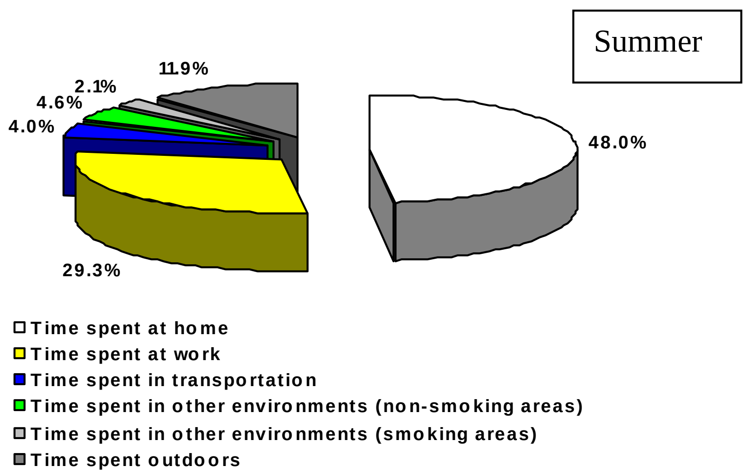


Figure 2: Time spent in microenvironments (house, office, other non-smoking and smoking indoors, outdoors and vehicles) of all volunteers during a 7 day period of exposure in winter and summer

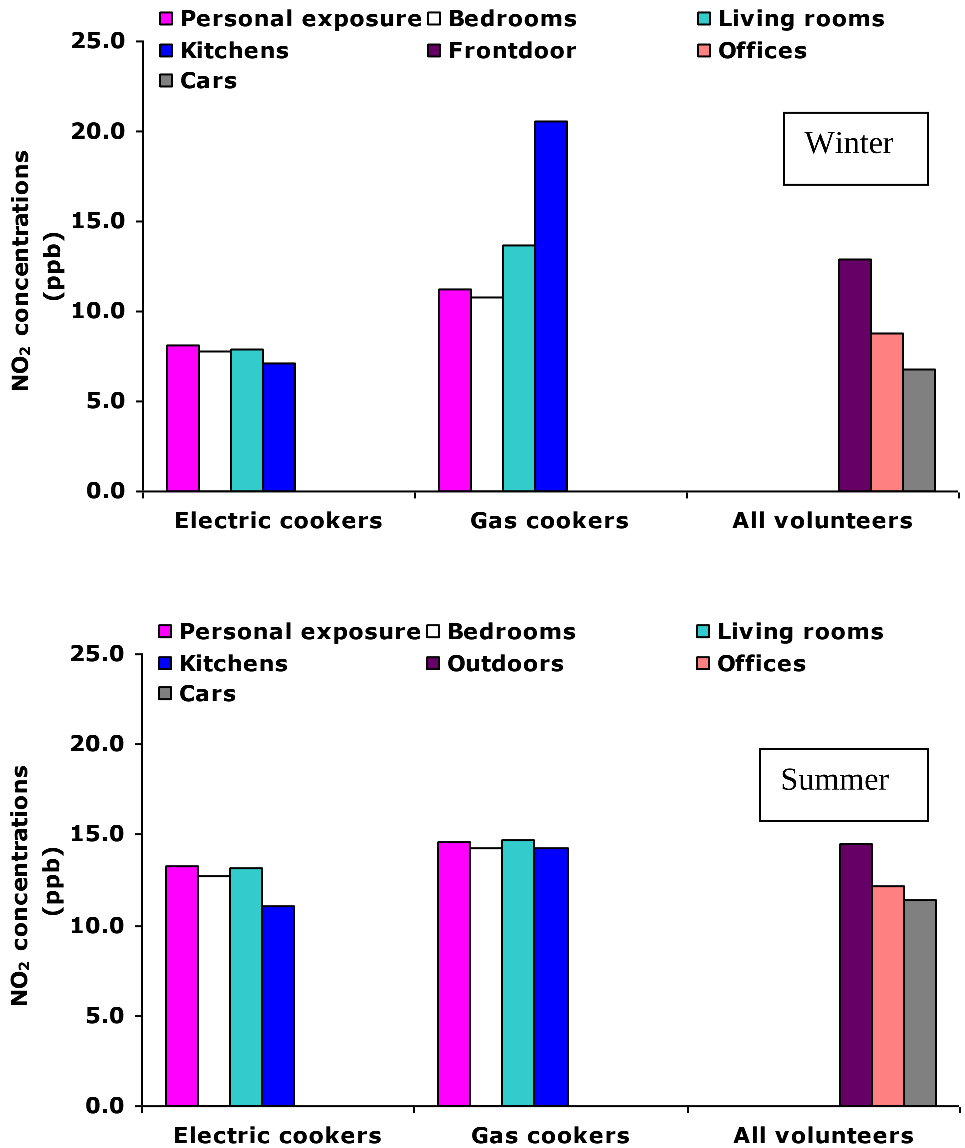


Figure 3: Weekly average NO_2 concentrations in all microenvironments for volunteers using electric and gas cookers for winter of 2000 (upper) and summer of 2001 (lower).

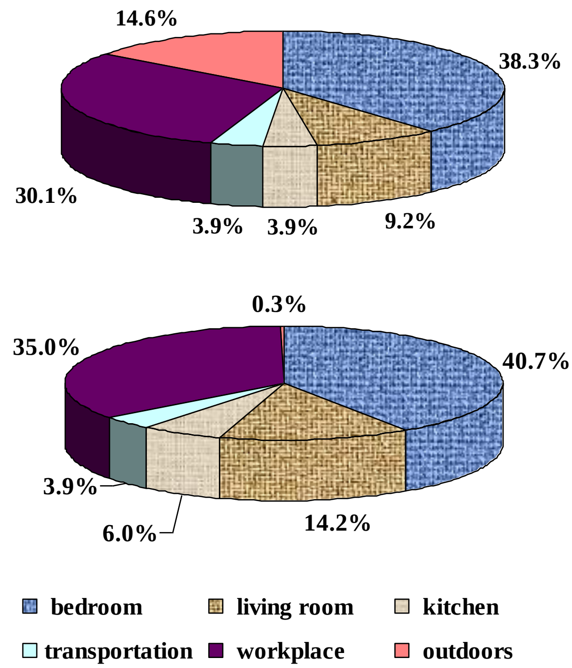


Figure 4: Distribution of personal exposure in different microenvironments during summer (above) and winter (below).

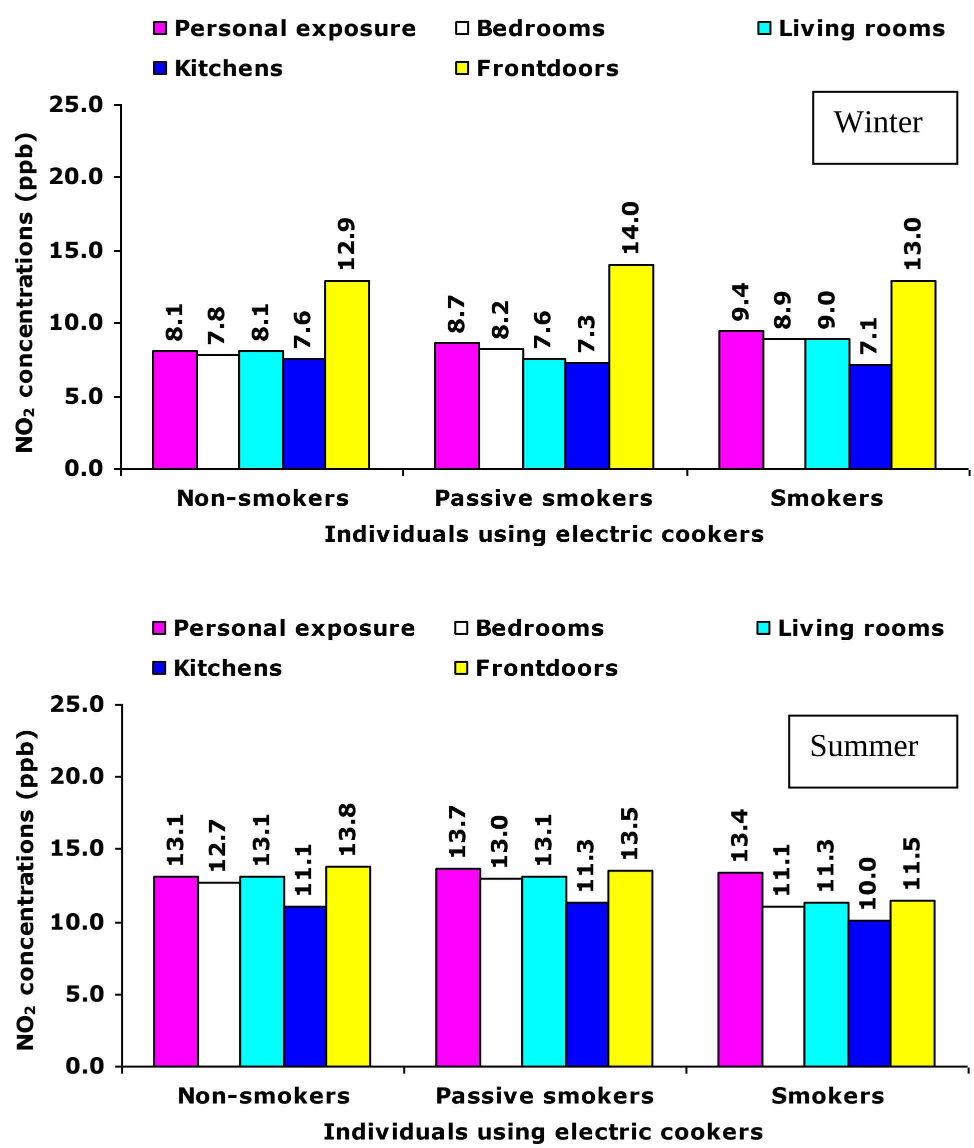


Figure 5: Weekly average personal exposure and NO_2 concentrations in houses of non-smokers, passive smokers and smokers (using electric cookers)

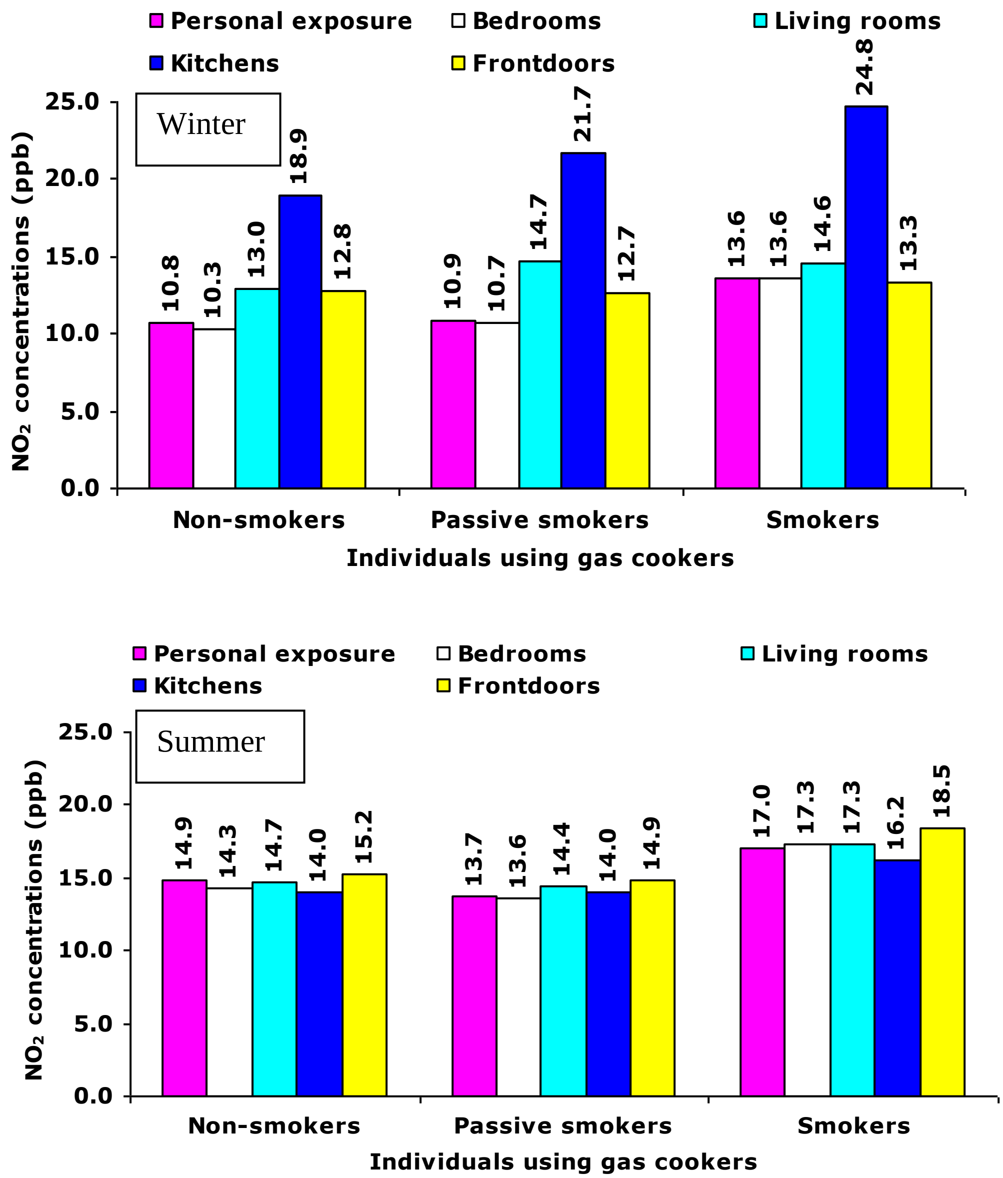
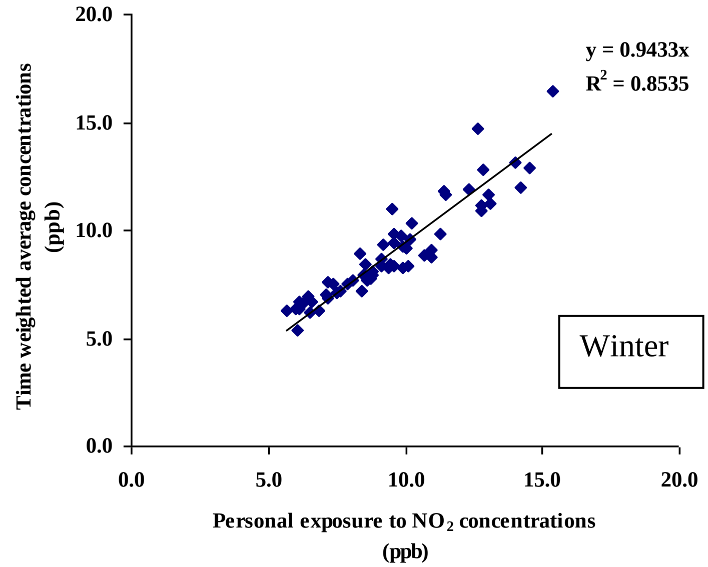


Figure 6: Weekly average personal exposure and NO_2 concentrations in houses of non-smokers, passive smokers and smokers (using gas cookers)



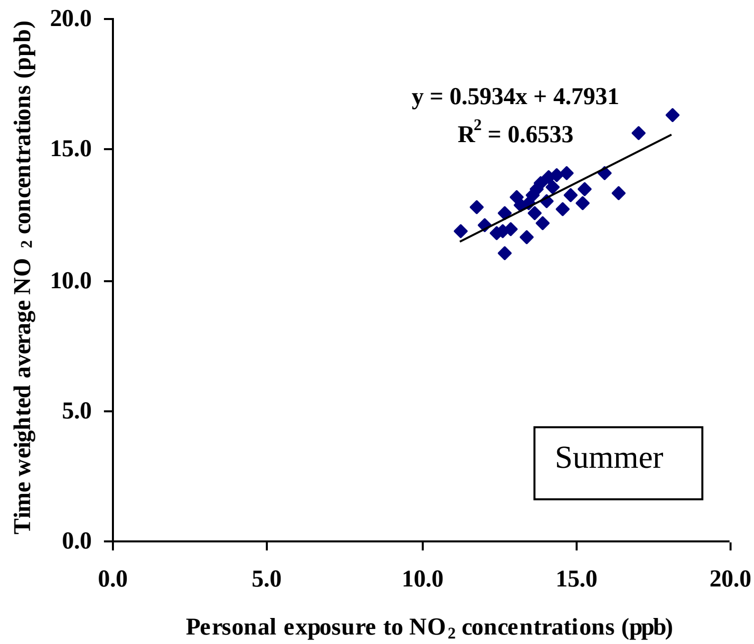


Figure 7: Comparisons between personal exposure to NO_2 concentrations and time weighted average NO_2 concentration

Table 1: Reported exposure levels of NO_2 and associated health effects.

NO_2 Concentration (ppb) †	Exposure Time	Health Effects	References
General			
0.2 - 5		Natural background mean concentration	WHO, 2000
10.6 - 47.9		Outdoor urban annual mean levels	
39.9 - 540		Outdoor urban hourly maxima	
10.6.4		Indoor poorly vented gas combustion appliances (over few days)	
1064		Indoor hourly maxima	
Short-term exposure effects			
2500 – 7500		Very small changes in lung functions	DOE, 1996
200-300		Changes in lung function of sensitive population (e.g. Asthmatic)	DOE, 1996
2000	Healthy non-smoking subjects exposed for 4 hrs on four consecutive days	NO_2 is a proinflammatory air pollutants under condition of repeated exposure	Blomberg et al., 1999
1000	2 hours exposure	No significant affects	WHO, 2000; WHO 2003
2500	2 hours exposure	Pronounced decrements in pulmonary function	
4000	1.25 hours exposure	No affects on Asthmatics	
300	10 min exposure	Decrease in forced expiratory volume in 1 s (FEV_1)	
300		Slight affects on chromic obstructive pulmonary disease	
500 - 15000	3 hours exposure	Healthy subjects having single exposures (for 3 hours) to NO_2 with exercise can induce (a) mild airway inflammation; (b) mild respiratory symptoms in some subjects; (c) small reductions in hematocrit and hemoglobin; (d) possible small reductions in circulating T lymphocytes; and (e) possible increased susceptibility of airway epithelial cells to injury from exposure to respiratory viruses.	Frampton et al., 2002
4.3 - 180.5	1 hours	Exacerbate severe asthma and can cause death among asthmatics (in association with O_3).	Sunyer et al., 2002
212.8		Review identify demonstrable effects at 212.8 ppb or above level (for patients with light asthma at 106.4 ppb)	Kraft et al., 2005

Long-term exposure effects			
8-68.1	2 week average	20% increased risk of respiratory symptom and diseases for each increment of 28.1 ppb	WHO, 2000
6.7-31.1 (mean: 13.6)	1 year	Respiratory infections and symptoms	Brauer et al., 2002
22.4	19 Year time-series analysis	2.25% ncrease in the daily nonaccidental mortality rate	Burnett et al., 2004
10.6-31.9 / 11.7 – 29.3	1 year/ 5 years	Can increase mortality due to cardiopulmonary causes (with PM_{10}).	Gehring et al., 2006
15-100	10 years	20 to 30% decreases in both predicted FEV1 and forced expiratory flow (FEF_{25-75}) between 25 and 75% of forced vital capacity (FVC).	Arbex et al., 2007
0.5 – 480 (median: 29.8)	Over 1 week	Higher levels can increases asthma symptoms in nonatopic children and can decrease peak flow.	Katten et al., 2007

†[NO_2 (ppb) = 0.532 x NO_2 ($\mu g/m^3$)]

Table 2a: Average personal exposure to NO_2 concentrations (of volunteers using electric and gas cookers) and average NO_2 concentrations in house microenvironments measured during winter of 2000

	Average NO_2 concentrations (ppb)							
	Electric cookers				Gas cookers			
	Min.	Max.	Mean	Stdev.	Min.	Max.	Mean	Stdev.
Personal exposure to NO_2	5.7	11.0	8.1	1.8	6.3	15.4	11.2	2.3
Bedrooms	3.2	11.1	7.8	2.0	6.3	15.5	10.8	2.3
Living rooms	4.1	11.4	7.9	2.1	6.1	30.1	13.7	5.5
Kitchens	4.2	9.7	7.1	2.8	12.9	38.8	20.6	6.9

Table 2b: Average personal exposure to NO_2 concentrations (of volunteers using electric and gas cookers) and average NO_2 concentrations in house microenvironments measured during summer of 2001

	Average NO_2 concentrations (ppb)							
	Electric cookers				Gas cookers			
	Min.	Max.	Mean	Stdev.	Min.	Max.	Mean	Stdev.
Personal exposure to NO_2	11.3	15.3	13.3	1.2	12.7	18.1	14.6	1.6
Bedrooms	10.6	14.8	12.7	1.3	12.5	17.3	14.3	1.4
Living rooms	10.8	15.4	13.1	1.5	13.2	18.2	14.7	1.4
Kitchens	8.0	13.3	11.0	1.7	12.8	17.7	14.2	1.3

Table 3: A comparative summary of NO_2 levels in various microenvironments and personal exposure.

Statistics	NO_2 concentration (ppb)							Comments	References
	Indoor					Outdoors	Personal Exposure		
	Bedrooms	Living Room	Kitchen	Vehicle	Office/ workplace				
Average Range	9.1±2.5 3.2-15.5	10.5±4.8 4.1-30.1	13.1±8 8.1-13.4	6.8±1.6 4.1-11.3	8.8±1.8 5.6-13.5	12.9±1.8 8.1-16.1	9.5±2.4 5.7-15.4	Winter	Present Study
Average Range	13.5±1.6 10.6-17.3	13.9±1.6 10.8-18.2	12.6±2.2 8.0-17.7	11.4±1.6 8.7-14.4	12.2±1.5 9.2-15.2	14.5±1.8 11.3-18.8	14.0±1.5 11.3-18.1	Summer	Present Study
Mean				13.1±5.5[‡]		9.5±6.4	16.7	Birmingham	Harrison et al., 2002
Mean Range	11.9±1.0				15.7±0.8	12.4±1	13.6±1.7 11.7-16.0	Oxford, UK	Lai et al., 2004
Median	5.8 23.9 6.1					12.4 27.4 15.5		Ashford, Barcelona Menorca	Garcıa Algar et al., 2004
Mean Range		26±12 4.6-67	33±18 8.1-75			17±7 5.2-29		No-heating period	Zota et al., 2005
Mean Range		43±16 11-78	50±19 10-85			21±5.6 7-31		During heating period	Zota et al., 2005
Average Range	17.6±3.5 6.4-34.6				30.3 7.4-84.6	20.7±3.6 13.3-47.9	22.3±4.0 9.6-41	Antwerp	Stranger et al., 2007
Range	7.4-14.4				15.4-48.9	16-61.2	9.0/20.2	Summer/winter	Piechocki-Minguy et al., 2006
Average	27.1±6.5	28.6±11.2	32.5±11.9			38.2±8.0	24.5	Hong Kong	Chao et al., 2000
Mean Range	18.7±7.3 7.4-45.5				23.9±8.5 7.7-55.3	32±8.1 13.5-58	23.2±6.0 12-45.2	Paris	Mosqueron et al., 2002
Median					8[†]	24	12	School	Van Roosbroeck et al., 2007

Mean				14.9 / 13.8 / 16.0			186±1 17±0.6 49±0.9 22±0.6 28±0.7 24±0.7 25.5 / 31.9 / 36.2	Tunnel worker Outdoor worker Garage- Diesel Garage - Petrol Bus Driver Taxi Driver Taxi / Bus / Lorry	Lewne et al., 07 Lewne et al., 06
Mean	9.6±5.9 14.4±6.9 22.9±12.2				14.4±8.0 19.2±12.8 16.0±9.6	12.8±6.4 19.2±6.9 32.5±10.6		Helsinki Basle Prague	Kousa et al., 2001
			14.1±1.1 11.9±1.3					Gas cooking Electric cooking	Willer et al., 2005
Mean Range	31.6±40.2 4.1-260							Baltimore	Breysse et al., 2005
Mean	13.2±5.2		25±8.8				23.6±5.4 13.2±4.1 21.3±7.1	Workers Students Housewife's	Gallelli et al., 2007
Average	11.2					14.4	16.5	25-40% higher during winter	Monn et al., 1998
Mean	23.6-28.0					25-26		Barcelona	Garcıa Algar et al., 2003
Median Q25-Q75	8.9 6.9-11					11.0/25[††]	11.4 9.2-15	<8°C	Sorensen et al., 2005
Median Q25-Q75	6.6 4.7-10.3					6.6/13.8	9.2 5.7-11.7	>8°C	Sorensen et al., 2005
Mean Range	8.3±1.6 3.3-29.1							High with gas heating house	Gilbert et al., 2006

[†]School; [‡]see details of microenvironment in text; [††]Urban background/Street station; [Q25-Q75]25-75% interquartile range

Table 4: Overview of NO_2 guidelines for outdoor air quality.

	Guideline Value† (ppb)	Average period
Belgium	71.8	1 h
Canada	<53.2	1 h
China	53.2	1 h
Germany	31.9	1 week
	186.2	30 min
Norway	53.2	1 h
UK	21.3	1 h
	106.4	1 year
EPA	53	1 year
WHO	106	1 h
	63.8	8 h
	21	1 year
EU‡	106.4	1 h
	21.3	1 year

† health related concentration/values based on current toxicological and epidemiological knowledge; ‡ to be achieved by 2010