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Impact of Microenvironmental Nitrogen Dioxide Concentrations on Personal Exposures in Australia

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ABSTRACT
Indoor and outdoor NO_2 concentrations were measured and compared with simultaneously measured personal exposures of 57 office workers in Brisbane, Australia. House characteristics and activity patterns were used to determine the impacts of these factors on personal exposure. Indoor NO_2 levels and the presence of a gas range in the home were significantly associated with personal exposure. The time-weighted average of personal exposure was estimated using NO_2 measurements in indoor home, indoor workplace, and outdoor home levels. The estimated personal exposures were closely correlated, but they significantly underestimated the measured personal exposures. Multiple regression analysis using other nonmeasured microenvironments indicated the importance of transportation in personal exposure models. The contribution of transportation to the error of prediction of personal exposure was confirmed in the regression analysis using the multinational study database.

IMPLICATIONS
The significance of indoor sources on personal exposure to NO_2 has been determined in many previous studies. This study demonstrates the relative importance of indoor sources in Australia. A microenvironmental model with three dominant environments (i.e., residential indoor, residential outdoor, and workplace indoor) significantly underestimated personal exposure. Regression analysis showed that transportation was a major contributor of the error in prediction of personal exposure. This was also confirmed in the multinational study database. The finding suggests a need for exposure assessment during transportation to construct accurate personal exposure models.

INTRODUCTION
NO_2 is a byproduct of high-temperature fossil fuel combustion. Abundant indoor and outdoor combustion sources make NO_2 one of the most ubiquitous pollutants in the urban environment. Despite wide distribution of sources, indoor NO_2 concentration is the dominant risk factor of personal exposure. Individuals were found to spend ~90% of their days indoors and about two-thirds of the day inside their homes.[1,2] When personal NO_2 exposures were measured for 568 subjects from 18 cities in 15 countries, indoor concentration was responsible for 75% of the variation in the total personal exposure of office workers.[3] The relation between indoor levels and personal exposures was associated with geographical conditions, indoor sources, and other demographic characteristics.

The presence of a gas range has been identified as one of the major factors contributing to indoor and personal NO_2 exposures. Significantly higher NO_2 concentrations were measured in homes with gas ranges; mean two-day personal NO_2 exposures were 34.8 ppb and 20.5 ppb in homes with and without gas ranges, respectively.[3] The use of a gas range provided a mean indoor/outdoor (I/O) NO_2 ratio of 1.19, compared with 0.69 for those homes without gas ranges. In the multinational study, the association between gas ranges and indoor NO_2 exposure was independent of country-specific parameters.

In addition to residential indoor NO_2 sources, traffic emission has been associated with respiratory health effects.[4] Pollutants from traffic emission include various toxic chemicals, and they can affect not only ambient levels but also indoor levels in homes in heavy traffic areas. The distance between home and traffic road is important, especially for children, due to their limited travel distance from home. For the working population, commuting may be a significant source of personal exposure.

In Australia, personal exposure to NO_2 has not been characterized. Based on a few published studies, large geographical variations were found. In a study in Perth, Western Australia, the indoor mean four-day average level was found to be 28 ppb, slightly higher than outdoor levels of 24 ppb.[5] In the Latrobe Valley, Victoria, the annual mean indoor level was 8 ppb.[6] The Victoria study documented higher NO_2 concentrations in kitchens than in living rooms and bedrooms and reported seasonal variation, with higher levels measured during winter.

In this study, indoor, outdoor, and personal NO_2 concentrations were measured by a protocol identical to that used in the multinational study.[3] An identical questionnaire for housing characteristics and an activity diary also were used, allowing direct comparison between the present study and values reported by Levy et al.[3] In addition, a personal exposure model was used to estimate the impact of commuting in the working population.

METHODS

Residential indoor, residential outdoor, and workplace indoor NO_2 concentrations and personal NO_2 exposures were measured for 57 subjects from six offices located in Brisbane, Queensland, Australia. The NO_2 concentrations were simultaneously measured during a 2-day period in June 1999. Winter was chosen because active combustion in winter tends to elevate both indoor and outdoor NO_2 concentrations. Participants filled out an activity diary and a questionnaire about their homes and their surroundings during the course of the study. The study proposal was identical to the multinational study. Details of the study protocol are explained in a paper by Levy et al.[3]

Each site had a coordinator who recruited up to 10 office workers and provided them with all sampling materials. All NO_2 concentrations were measured by passive filter badges.[7] Quality was assured by blanks from each site to account for differences in sealing quality and lag time before analysis. Once sampling was completed, the NO_2 passive badges were analyzed by a spectrophotometer (Beckman Model DU640).

Personal exposure can be approximated as the time-weighted average of microenvironmental concentrations. Although not all microenvironments were measured in this study, personal NO_2 exposure was estimated using residential indoor exposure, workplace indoor exposure, and residential outdoor exposure according to eq 1

$$P_i = (IH_i \bullet I_i + OH_i \bullet O_i + WI_i \bullet W_i)/T \qquad (1)$$

where P_i is estimated time-weighted average personal NO_2 exposure for participant i, IH_i is number of hours spent inside the home for participant i during the sampling period, OH_i is number of hours spent outside the home for participant i during the sampling period, WI_i is number of hours spent inside the workplace for participant i during the sampling period, I_i is measured average residential indoor NO_2 concentration for participant i, O_i is measured average outside home NO_2 concentration for participant i, W_i is measured average workplace indoor NO_2 concentration for participant i, and T is total exposure time.

Since not all microenvironments were measured, the personal exposures from eq 1 were significantly lower than the measured personal exposures. Differences between the measured personal exposure (P) and the personal exposure estimated by eq 1 (P_i) could be explained by nonmeasured microenvironments. The concentrations at different microenvironments were estimated as the regression coefficient b in eq 2

$$P - P_i = b_{IO} \bullet F_{IO} + b_{(WO+OO)} \bullet F_{(WO+OO)} + b_T \bullet F_T \qquad (2)$$

where P is measured personal NO_2 exposure (ppb), F_{IO} is fraction of hours spent inside other than at home and workplace, $F_{(WO+OO)}$ is fraction of hours spent outside other than near home (including outside workplace and other outdoors), and F_T is fraction of hours spent on transportation.

The impact of transportation in personal exposure was also determined using the database of the multinational study.[3] In the analysis, cities with a sample size of less than 20 (i.e., Bombay, Manila, Zagreb, and Sosnowiec) were excluded. Data from Mexico were excluded due to possible sampling error. The multiregression analysis was conducted after the data were combined into country or area. The data were combined into eight regions: (1) North American cities (Boston and Ottawa), (2) Scandinavian cities (Kjeller and Kuopio), (3) Western European cities (Berlin, Erfurt, and Geneva), (4) United Kingdom (London and Watford), (5) Korea (Seoul and Taejon), (6) Japan (Sapporo and Tokushima), (7) China (Beijing), and (8) Australia (Brisbane).

RESULTS

A total of 58 subjects were recruited from six organizations located in Brisbane, Queensland, Australia. Personal exposure data from one subject were excluded, since the subject did not place indoor and outdoor samplers. The house characteristic information is shown in Table 1. The mean number of bedrooms in all houses was 3.2. The average number of family members was 2.9. Eighteen houses had gas ranges. None of the houses had pilot lights or kerosene or coal heaters. Even though the monitoring was conducted in June (winter in the southern hemisphere), windows were open in ~89% of the houses at some time during the sampling period. The NO_2 concentrations and the mean I/O NO_2 ratio are shown in Table 2.

The presence of a gas range had the most statistically significant influence on indoor concentrations, as shown in Table 3. The mean indoor concentration in homes with gas ranges was 13.6 ppb, compared with 9.1 ppb for homes without gas ranges. The presence of gas ranges provided a mean I/O ratio of 1.03, compared with a mean I/O ratio of 0.67 for those without gas ranges. When participants lived in homes with a smoker, indoor concentrations were significantly higher. "Single detached house" and "presence of gas water heater" factors increased indoor NO_2 concentrations slightly.

Outdoor NO_2 concentrations were associated significantly with the number of bedrooms, residential area, and house type. Mean outdoor concentration in homes with one or two bedrooms was 18.5 ppb, compared with 13.6 ppb in homes with three or more bedrooms. Outdoor NO_2 levels were higher when the house was in a nonresidential area or a single detached house. The presence of gas ranges and gas water heaters was the most significant factor contributing to personal NO_2 exposure. Average personal exposure in homes with gas ranges was 17.9 ppb, compared with 13.7 ppb for houses without gas ranges. Personal exposures were significantly higher when participants lived in homes with one or two bedrooms.

Fifty-five participants completed an activity diary during the sampling period. The participants spent the majority of their time indoors. The fraction of total indoor time was 88 ± 6%. Participants spent 54 ± 12% of their time in homes. Participants stayed inside the workplace for 29 ± 10% of the time. The fraction of total outdoor time was 4 ± 4%. About one-half of the outdoor time was spent near the workplace. Transportation time accounted for 7.1 ± 3.4%.

Table 1. House characteristics in study dwellings and commuting ($n = 57$).

	Number of Houses with House Characteristic	Number of Houses without House Characteristic
House Type (single detached house)	47	10
Number of Bedrooms (1 or 2)	11	46
Attached Garage	19	38
Presence of Smoker	7	50
Gas Range	18	39
Gas Water Heater	12	45
Window Open	51	6
Commuting with Car	48	9
Commuting Time per Day (<60 min)	34	23

Table 2. NO_2 concentrations (ppb) and I/O ratio.

	Mean	Standard Deviation	Minimum	Maximum
Indoor	10.5	5.6	1.2	31.1
Outdoor	14.5	5.8	2.0	35.0
Workplace	18.2	5.0	10.1	23.3
Personal	15.0	5.2	5.7	31.3
I/O Ratio	0.78	0.55	0.29	3.62

Table 3. House characteristics associated with indoor NO_2 concentrations.

Characteristics	Mean Indoor NO_2 with Characteristic (ppb)	Mean Indoor NO_2 without Characteristic (ppb)	*p*-value
Gas range	13.6 ± 6.2	9.1 ± 4.7	0.004
Presence of smoker	14.9 ± 7.7	9.9 ± 5.0	0.025
Single detached house	13.6 ± 7.4	9.9 ± 5.0	0.054
Gas water heater	13.2 ± 5.1	9.8 ± 5.5	0.059

A simple personal exposure model estimated personal exposure using eq 1. The estimated personal NO_2 exposure was significantly associated with the measured personal exposure, with a Spearman correlation coefficient of 0.58, as shown in Figure 1. However, the estimated personal NO_2 exposure of 11.2 ± 4.0 ppb was significantly lower than the measured personal exposures of 15.2 ± 5.3 ppb (paired t test, $p < 0.001$).

The difference between the measured level and the estimated level was used to determine concentrations at other nonmeasured microenvironments. When the multiple regression was conducted using indoor other than home and workplace, outside workplace, outside other than workplace and home, and transportation, only the regression coefficient for transportation was statistically significant. The results for Brisbane are shown in Table 4. Time spent outside workplace and other than home and workplace were only 1.9% and 1.1%, respectively. Therefore, outside workplace and outside other than home and workplace were combined into one microenvironment in eq 2.

When personal exposures in the multinational database were estimated by eq 1, the estimated personal NO_2 exposure of 22.5 ± 15.7 ppb was significantly lower than the measured personal exposures of 28.8 ± 18.1 ppb.

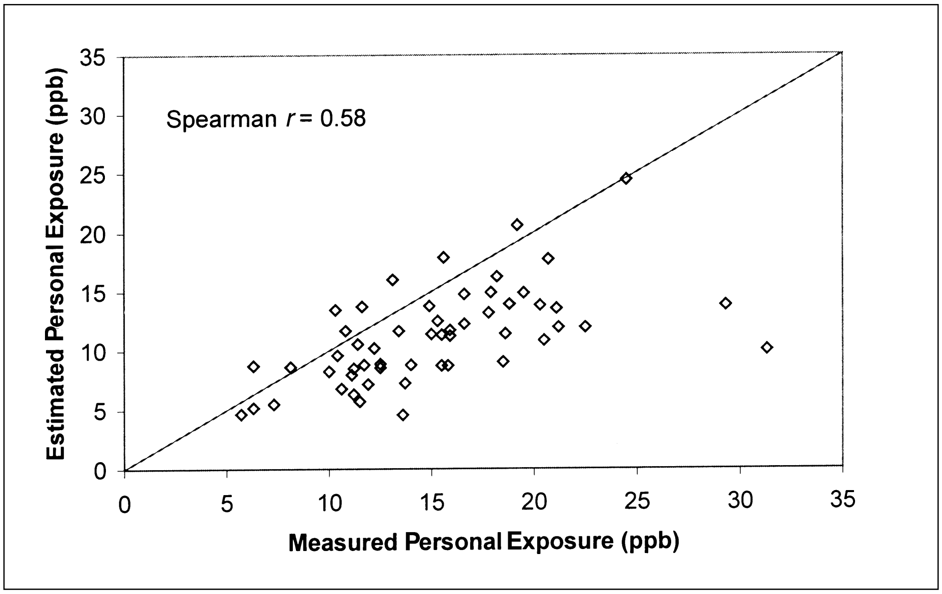

Figure 1. Association between measured personal NO_2 exposure and estimated personal exposure by microenvironmental model.

in the seven regions is shown in Figure 2.

DISCUSSION

Average indoor and outdoor NO_2 levels were 10.5 and 14.5 ppb, respectively. The NO_2 measurements in this study were relatively low compared with the levels in the multinational study with 18 cities. These low levels were observed in only four cities (i.e., Kuopio, Finland; Erfurt, Germany; Kjeller, Norway; and Geneva, Switzerland). These levels were significantly lower than levels reported in Perth, western Australia.[4] However, the indoor levels were slightly higher than the levels observed in Latrobe Valley, Victoria.[5] The indoor levels in Latrobe Valley were reported as annual average.

The average ratio of indoor to outdoor levels was 0.78. Previous studies in the United States reported that I/O NO_2 ratios were higher in winter than in summer. The annual indoor level is estimated to be ~60% of the outdoor level in homes without gas ranges.[9] Considering 18 out of 57 houses in this study had gas ranges, the I/O ratio is slightly lower than expected, possibly due to the fact that 89% of participants opened windows during the sampling period.

The presence of gas ranges increased personal exposure by 4 ppb in this study, significantly lower than the increase of 15 ppb reported by Levy et al.[3] for homes with a gas range. Unlike the multinational study, no house in this study had a pilot light. Considering the absence of a pilot light, the increase was comparable to the study in the United States; personal exposure in homes with gas ranges without a pilot light was 6 ppb higher than in those with electric ranges.[2]

The impact of indoor NO_2 on personal exposure can be affected by activity patterns. The participants (all office workers) spent 88% of their time indoors and an average of 7.1% of their time on transportation. While the recruitment of office workers allowed for direct comparison with the multinational study, it is acknowledged that office workers may spend the least amount of time outdoors, and consequently, the impact of residential NO_2 on personal exposure in this study and the multinational study may not be applicable to different population groups.

Personal exposure was estimated by eq 1. The Spearman correlation coefficient between the measured

Using the multinational study database, NO_2 concentrations in other nonmeasured microenvironments were estimated by eq 2. The results in eight regions are shown in Table 5. Regression coefficients for transportation were significant for all countries or areas, except China. It is significant that 19 subjects among the 43 participants in China did not commute during the sampling period. Regression coefficients for other microenvironments were not consistently significant.

NO_2 exposure during transportation was not measured in this study. The estimated NO_2 levels in transportation were compared with residential outdoor levels. The NO_2 level in transportation was ~3 times higher than the residential outdoor level in Korea and Japan. In western Europe, North America, and Brisbane, the ratio of NO_2 in transportation to residential outdoor level was ~2. The ratio was ~1 in Scandinavian cities and the United Kingdom. The relationship between average residential outdoor levels and the estimated NO_2 levels in transportation

Table 4. Estimated NO_2 concentrations and fraction of time in four microenvironments in Brisbane.

	Regression Coefficient ± SE (ppb)	Fraction of Time (%)	Sig
Indoor other than Home and Workplace	−14.4 ± 10.4	4.9 ± 2.6	0.176
Outside Workplace	−8.9 ± 24.8	1.9 ± 2.6	0.720
Outside other than Home and Workplace	36.0 ± 24.9	1.1 ± 2.3	0.151
Transportation	32.0 ± 13.2	7.1 ± 3.4	0.019

Table 5. Estimated NO_2 concentrations in microenvironments from multinational measurements.

Country	Regression Coefficient ± SE for Other Indoors (ppb)	Regression Coefficient ± SE for Outdoors other than Home (ppb)	Regression Coefficient ± SE for Transportation (ppb)	Sample Number
North America (Boston and Ottawa)	96.0 ± 27.1[a]	13.5 ± 9.8	60.8 ± 16.2[a]	48
Western Europe (Berlin, Erfurt, and Geneva)	20.3 ± 21.5	16.9 ± 8.7	43.8 ± 15.1[a]	89
UK (London and Wartford)	27.0 ± 16.8	35.8 ± 8.6[a]	35.3 ± 11.9[a]	57
Scandinavian (Kjeller and Kuopio)	39.0 ± 9.6[a]	33.2 ± 6.4[a]	16.1 ± 6.4[b]	56
Korea	66.9 ± 31.6[b]	28.5 ± 32.5	134.9 ± 48.0[a]	66
Japan	33.3 ± 44.5	48.1 ± 23.3[b]	71.9 ± 20.0[a]	88
Brisbane	32.4 ± 16.3	5.4 ± 9.8	36.2 ± 12.2[a]	55
China[c]	34.0 ± 16.4[b]	42.1 ± 30.5	18.6 ± 37.0	43
Total	28.3 ± 7.1[a]	49.6 ± 9.3[a]	59.3 ± 9.2[a]	502

[a]$p < 0.01$; [b]$p < 0.05$; [c]19 of 43 subjects did not spend any time on transportation.

personal exposure and the estimated personal exposure was less than the coefficient of 0.81 reported by Levy et al.[3] It should be noted that the denominator of the model in this study was different from the denominator in the study by Levy et al.[3] When personal exposure was calculated with a total of the three microenvironments (residential indoor, residential outdoor, and workplace indoor), as in the multinational study, the estimated personal exposure in Brisbane was 13.2 ± 4.4 ppb. The estimated personal exposures showed better correlation. When personal exposure was estimated by the equation with total sampling time, the model provided personal exposure of 11.2 ppb.

Although eq 1 does not include all possible microenvironments, it accounts for 85% of the exposure period. However, the underestimation by the model was greater than the missing sampling time. The underestimation indicates that there are other environmental factors significantly contributing to personal exposure. Multiple regression using other nonmeasured microenvironments showed that personal exposure could be significantly affected by the NO_2 level during transportation.

The importance of transportation on the personal exposure model was confirmed in separate regressions of eight regions from the multinational data. Regression coefficients for transportation were significant in all regions, except China. The results in China may be due to the large number of noncommuters (19 noncommuters among 43 subjects). This is another piece of indirect evidence for this methodology. The difference from the personal exposure model may be caused by various errors. The passive sampler can produce a measurement error of less than 20%.[8,9] The activity diary was determined with a resolution of 1/2-hr intervals. Considering the measurement errors, the finding can conclude that exposure during transportation is significant for the working population that commutes.

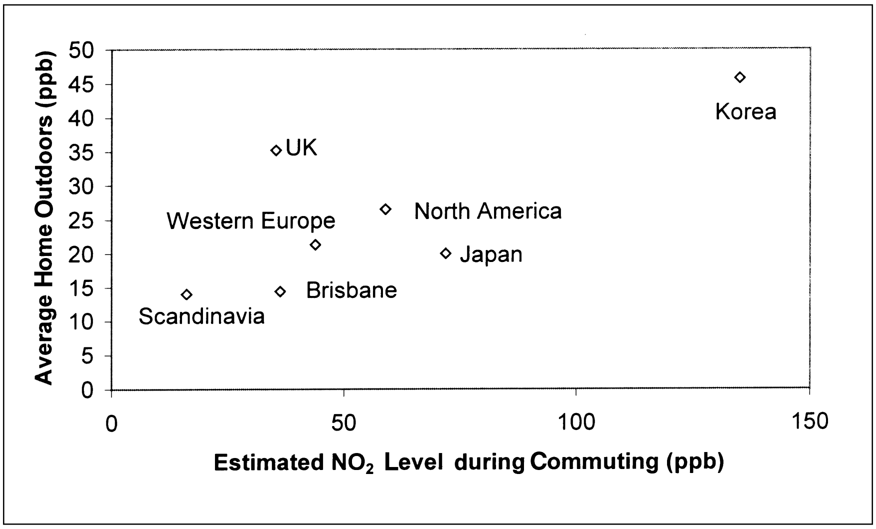

Figure 2. Relationship between estimated NO_2 level during transportation and average outdoor level.

The methodology using eq 2 demonstrated consistent results in the large database from the multinational study. However, the results of the analysis could not be independently verified due to lack of separate measurement of NO_2 exposure during transportation. Most previous studies measured exposure to traffic-related air pollutants with control of routes and transport type.[10,11] Personal exposure to the air pollutant, often higher than concentrations at nearby stationary monitors, was strongly associated with transport mode, route, and city. The higher concentration during transportation is better shown from continuous CO measurements.[12] The estimated NO_2 concentrations in transportation were equal to or higher than the average residential outdoor level in all seven regions. Further measurement of exposure during transportation is needed to verify the impact of transportation on total personal exposure.

CONCLUSIONS

This study demonstrated that the presence of gas ranges was the predominant factor affecting indoor concentrations and personal exposures to NO_2 in Brisbane, Australia. Personal exposure by a microenvironmental model was closely correlated with the measured personal exposure. However, the estimated personal exposure was significantly lower than the measured personal exposure. Although the difference may be contributed by measurement error and other nonmeasured microenvironments, regression analysis with nonmeasured microenvironments strongly suggested that transportation is a major contributor to the difference. The importance of transportation in the prediction of personal exposure was confirmed using the database of the multinational study. The findings conclude that exposure during transportation needs to be considered for better personal exposure models.

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