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Assessment of Personal Exposure to Nitrogen Dioxide in Primary Schoolchildren

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Abstract: This study was designed to assess the level of nitrogen dioxide from several microenvironments including inside the home, outdoors near the home, inside the school, outdoors near the school, and on the road for 42 primary schoolchildren during the month of December 2002 in Seoul, Korea. The average personal, indoor, outdoor NO_2 levels, and indoor/outdoor ratio were 45.08 ppb, 27.89 ppb, 30.96 ppb, and 0.89, respectively. The indoor NO_2 concentrations were significantly associated with the presence of a smoker with a gas stove. The estimated personal NO_2 exposure using time-weighted average equation of 34.64 ± 5.29 ppb was significantly lower than the measured personal exposure of 45.08 ± 5.50 ppb. Our results indicate that indoor NO_2 levels were associated with the presence of a smoker and a gas stove. Moreover, personal NO_2 exposure with a gas stove in the house was significantly higher than those without a gas stove.

Keywords: air pollution, NO_2, time-weighted model, microenvironment, primary schoolchildren

Introduction

For several decades road traffic has been the single largest source of nitrogen oxides (NO_X) in many countries. In Europe NO_X emissions are still of great concern, mainly due to the significant role of NO_X in acid deposition, including adverse effects resulting from the formation of tropospheric ozone. Moreover, there is great concern over the health effects of NO_2 exposure in the population, particularly, in urban areas where traffic emissions accounts for over 80% of ambient NO_2 levels.[1)]

Nitrogen dioxide (NO_2) is formed as a byproduct of high-temperature fossil fuel combustion. The outdoor concentration of NO_2 is closely related to the proximity of the sampled location to a major source of NO_2 such as motor vehicle traffic and fossil fuel power plants. Meteorological factors influencing the transfer and dilution of NO_2 as well as to atmospheric conversion reactions also influence NO_2 levels.[2)] Indoor NO_2 concentration profiles are related to these outdoor levels as well as to the use of unventilated or improperly ventilated indoor combustion appliances (e.g. gas stoves, gas heaters, and unventilated kerosene space heaters) to air infiltration and ventilation, and to chemical reactions and adsorption or absorption of the gas on indoor surfaces.[3-5)]

Indoor sources tend to be the dominant contributors to personal NO_2 exposure due to the large amount of time urban dwellers spend inside their homes.[6,7)] Several studies have shown that individuals spend ≥90% of their time indoors and approximately two-thirds of the day inside their homes.[6,8)] This is particularly true for those segments of the population (e.g. children, the elderly, and the ill) most susceptible to air pollutants.[3)]

When the NO_2 exposure was measured personal exposure levels of 46 children aged 9~11 years in Southampton, England, the personal exposures to NO_2, averaged over seven days, ranged from 6 to 137 ppb with a geometric mean of 19 ppb.[9)]

The presence of a gas range has been identified as one of the major contributing factors to indoor and personal NO_2 exposures.[10)] Significantly higher NO_2 concentrations were measured in homes with gas ranges. Lee *et al.* reported that the mean 2-day indoor NO_2 exposure levels were 13.6 ppb and 9.1 ppb in homes with and without gas ranges, respectively.[5)] The mean indoor/outdoor (I/O) NO_2 ratio of homes with a gas range was 2.27, compared with 1.22 for those homes without gas ranges. In

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addition, the mean I/O NO_2 ratios of the homes with and without an air conditioner were 1.07 and 3.03, respectively.[11] Motor vehicle traffic in urban areas is a major source of pollution which is becoming an increasing concern to public health.[12]

In this study, the NO_2 concentrations were measured inside the home, outdoors near the home, and in the school environment. Additional information on personal NO_2 exposures was acquired using self-administered questionnaires regarding daily activities and household characteristics of each child. These data allowed the assessment of the sources of personal NO_2 exposure from indoor and outdoor microenvironments.

Methods

NO_2 concentrations were taken inside the home, outdoors near the home, inside the school, outdoors near the school, along the road by passive measurement by a filter. Furthermore, each student wore a personal badge with a filter that measured 24-hour NO_2 exposures. NO_2 exposure was measured for 45 subjects (10~12 years of age) from a single school located at 300 meters from a major road in Seoul, Korea. However, personal exposure data from three children were excluded, because one of three children did not place outdoor filters near their home, and the other two did not complete a time-activity diary.

The weather during the study was typically cold and dry, with an outdoor temperature ranging from −6.4 to −3.8°C and a relative humidity ranging from 38.5 to 43.5%. Winter was chosen as the study period because active combustion for heating in winter tends to elevate both indoor and outdoor NO_2 concentrations.

In this study, the time activity diaries were simplified so that locations and time were preprinted on the pages, which the children would mark to indicate their location. The form was divided into 30-min increments and the locations included inside home, outside at or near the home, inside the school, outside or near school, or on road. The children were also asked to note if they were exposed to passive cigarette smoke, other types of smoke or irritating smells. This type of diary was easier for the children to complete and made information collection more efficient. An additional questionnaire noted the children' household characteristics such as the presence of smokers, a gas stove, and air cleaners.

All NO_2 concentrations were measured using passive filter badges without the use of pumps or other equipment (Tokyo Roshi Kaisha, Ltd.). The filter badge was small ($5 \times 4 \times 1$ cm^3) and light (16 g) and had a high sensitivity (detection limit 66 ppb × hr). The filter badges absorbed NO_2 on a cellulose fiber filter coated with a triethanolamine solution.[13] The overall mass transfer coefficient of the sampler was 0.10 cm/s.[14] Quality assurance was performed by the use of blanks from each site to account for the differences in the sealing quality and lag time before each analysis. Once the sampling was completed, the NO_2 badges were analyzed using the procedure described in the Yanagisawa study using a Shimadzu Model UV-1601 spectrophotometer.[13] The overall detection limit of the sampler calculated from the sum of the average blank value and 3 times the standard deviation of the blanks (sample size = 8) was 1.97 ppb over the 24-hours sampling period.

Each child wore a filter badge on his or her chest during all waking hours, and placed it on a table or dresser at night or while showering. Filters were placed outside of each child's home, at least one meter off the ground or balcony, where they could not be affected by rain or snow. Filters were also placed inside each child's house, at least 3 m from any unventilated combustion source and window. All personal, indoor, and outdoor samplers were exposed for 24 hours. Filters placed in the school were exposed for eight hours during class hours in each child's classroom and playground in front of the main school building. Furthermore filters placed on the road were exposed for 24 consecutive hours during the same sampling day. Finally, the number of filters placed in the school and on the road was sixteen and six, respectively.

Personal exposure can be approximated as the time-weighted average of the microenvironmental concentrations. Although not all the microenvironments were measured in this study, the personal NO_2 exposure was estimated using the levels inside the home, outdoors near the home, inside the school, outdoors near the school, and near the road according to the following equation.

$$C_i = (CIH_iTIH_i + COH_iTOH_i + CIS_iTIS_i + COS_iTOS_i + CTTT_i) / T$$

Where C_i is the estimated time-weighted average personal NO_2 exposure for children i. CIH_i is the measured average NO_2 concentrations inside the home for children i. COH_i is the measured average NO_2 concentrations outdoors near the home for children i. CIS_i is the measured average NO_2 concentrations inside the school for children i. COS_i is the measured average NO_2 concentrations outdoors near the school for children i. CT is the measured average NO_2 concentrations in the road. TIH_i is number of hours spent inside the home for children i. TOH_i is the number of hours spent outside the home for children i. TIS_i is the number of hours spent inside the school for children i. TOS_i is the number of hours spent outside the school for children i. TT_i is the number of hours spent on the road for children i, and T is the total exposure time.

Results

A total of 43 children were recruited from a single elementary school located at 300 meters from a heavily trafficked road in Gangseo-Gu, Seoul, Korea (Fig. 1).

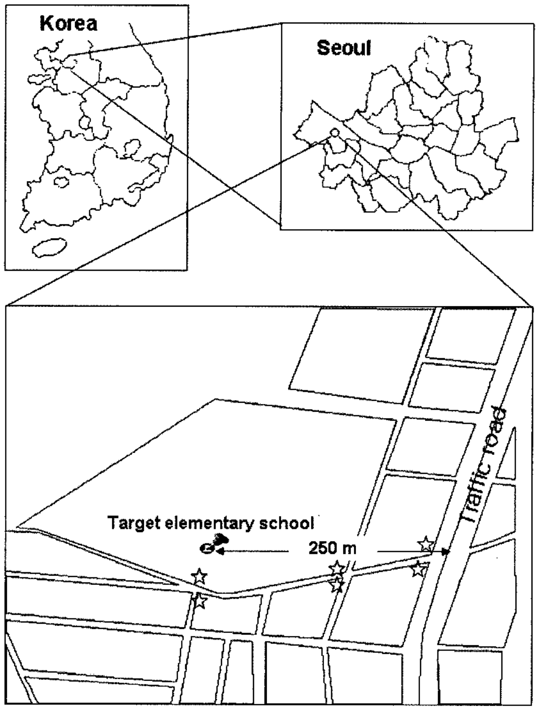

Fig. 1. The map of the sampling locations measured transportation NO_2 exposure and target elementary school in this study. The star markers are sampling sites for transportation from traffic road.

Table 1 shows the fraction of time the 42 children spent in each microenvironment. The 42 children completed a time-activity diary during the sampling period. The children spent the majority of their time indoors. The fraction of the total indoor time was 88.32 ± 7.02%. The children spent 69.27% of their time in their homes and 19.05% of their time in the school. The fraction of total outdoor time was 5.58 ± 4.80%. Approximately two-thirds of outdoor time was spent near the home. Traffic road time accounted for 6.10 ± 6.46%. The fraction of time spent in each microenvironment was similar to those in Australia and America.[5,15] Several studies have shown that individuals in industrialized countries spend approximately 90% of their time indoors, with approximately two-thirds of the day spent inside the home in both summer and winter.[6-8]

A descriptive summary of the NO_2 concentrations

Table 1. Percent of time the 42 children spent in each microenvironment

	Indoor		Outdoor		Traffic road
	Home	School	Near home	Near school	
Mean hours (Mean ± SD)	16.63 ± 1.75	4.57 ± 0.74	0.84 ± 0.98	0.46 ± 0.70	1.46 ± 1.55
%	69.27	19.05	3.65	1.93	6.10
Total %	88.32 ± 7.02		5.58 ± 4.80		6.10 ± 6.46

Note) SD is standard deviation.

Table 2. Descriptive summary of the NO_2 concentrations (ppb) and the I/O* ratio at home

	No.	Mean ± SD	Median	Min	Max	90th percentile	95th Percentile
Personal	42	45.08 ± 5.50	45.58	33.42	56.70	51.91	53.82
Indoor	42	27.89 ± 7.38	25.79	15.72	47.12	38.14	41.38
Outdoor	42	30.96 ± 5.75	32.12	13.29	39.56	36.63	38.29
I/O Ratio	42	0.93 ± 0.28	0.88	0.50	1.65	1.33	1.48

Note) SD is standard deviation; Min is minimum; Max is maximum.
I/O is indoor NO_2 concentrations/outdoor NO_2 concentrations.

Table 3. Household characteristics associated with the indoor and indoor/outdoor nitrogen dioxide concentrations at home

Variable		Indoor		Indoor/Outdoor	
		No.	Mean ± SD	No.	Mean ± SD
Presence of smoker					
Yes		21	29.67 ± 7.52	21	0.90 ± 0.25
No		21	26.11 ± 6.96	21	0.87 ± 0.30
Presence of gas stove					
Yes		17	30.37 ± 7.94	17	0.99 ± 0.30
No		25	26.21 ± 6.62	25	0.89 ± 0.26
Presence of gas stove with smoker					
Gas stove	Smoker				
Yes	Yes	10	32.98 ± 8.38	10	1.04 ± 0.26
Yes	No	7	26.63 ± 5.90	7	0.92 ± 0.36
No	Yes	11	26.66 ± 5.39	11	0.95 ± 0.25
No	No	14	25.85 ± 7.63*	14	0.84 ± 0.27
Total		42	27.89 ± 7.38	42	0.93 ± 0.28

Note) SD is standard deviation, *Statistically significant difference (p-value < 0.05) between home with both gas stove & smoker and home without gas stove & smoker.

and the mean indoor/outdoor ratio at home are shown in Table 2. The average personal, indoor, outdoor NO_2 levels, and indoor/outdoor ratio were 45.08 ppb, 27.89 ppb, 30.96 ppb, and 0.89, respectively. All NO_2 levels were higher than the levels observed in Australia, America, Canada, Finland, Germany, Norway, Philippines, and Switzerland, and similar to those in observed China.[5,10,11] However, the indoor levels were lower than those observed in India, Japan, Mexico, and Poland.[10] The average ratio of the indoor to outdoor levels was 0.93. However, the I/O NO_2 ratios are higher in Australia.[5]

The household characteristics associated with the indoor levels and indoor/outdoor ratios for NO_2 concentrations at home are shown in Table 3. The indoor NO_2 concentrations and I/O ratios were

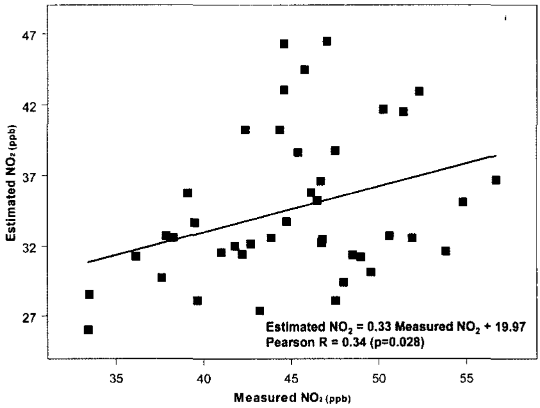

Fig. 2. The relationship between the directly measured personal NO_2 exposure level and the estimated personal NO_2 exposure level according to the microenvironmental model.

Table 4. NO_2 levels according to the distance from the road

	< 100 meters (N = 2)	101-200 meters (N = 2)	201-300 meters (N = 2)
NO_2 concentration (ppb)	38.77 ± 4.16	34.64 ± 0.47	31.29 ± 2.58

not significantly associated with the presence of smokers or the presence of a gas stove. However, the indoor NO_2 concentrations were significantly associated with the presence of a smoker with a gas stove.

A simple personal exposure model estimated personal exposure using a time-weighted average equation. This study found that the estimated personal NO_2 exposure was associated with the directly measured personal exposure, with a Pearson correlation coefficient of 0.34, as shown in Figure 2. However, the estimated personal NO_2 exposure of 34.64 ± 5.29 ppb was significantly lower than the measured personal exposure of 45.08 ± 5.50 ppb (paired t test, $p = 0.000$).

Table 4 shows the average NO_2 levels on the road, which was classified by the distance from the main traffic road to the school. This table shows a decrease in NO_2 concentration in relation to the distance from the main road, categorized in three classes. The highest level was observed at a distance ≤ 100 m. Above 100 m, NO_2 levels gradually decreased and were noticeably lower at a distance of 201 m to 300 m.

Discussion

Our data showed a weak correlation between indirectly estimated personal exposure and directly measured personal exposure of 42 children in Seoul, Korea. Furthermore, our findings suggest that indoor NO_2 concentration was significantly associated with the presence of a smoker with a gas stove, but not the presence of smoker or gas stove alone. School-age children were chosen since they are particularly sensitive to air pollution. Limitations of this study include insufficient sample size and little variation of microenvironments.

In this study, the mean area of the homes was 27.67 m^2 and the average number of family members was 4.2 persons. Seventeen houses had a gas stove but only three houses had an air cleaner. None of the houses had pilot lights, nor kerosene and coal heaters. Because the monitoring was conducted in December, the windows were closed in approximately 90% of the houses during the sampling period. Houses with smokers and gas stoves had higher indoor NO_2 concentrations and houses with an air conditioner had lower indoor NO_2 concentrations. Mean personal NO_2 exposure in houses with and without a gas stove during the study was 46.58 ± 4.13 ppb and 44.07 ± 6.14 ppb, respectively. These results are lower than the 4 ppb increase between with and without gas stove reported by Lee *et al.* and 14.3 ppb by Levy et al. for houses with a gas stove.[5,9] However, in this study, personal NO_2 exposure levels significantly correlated with the presence of smokers with gas stoves in the home (Table 3).

One of the major contributors to indoor NO_2 exposure, and thus one of the most important contributors to personal exposure, is the presence of a gas stove. Large variations in the frequency of gas stove use and in the quantity of time spent inside the home, as well as in the quantity of time spent in other microenvironments linked to combustion process, are likely to result in variations in personal NO_2 exposures. Therefore, different countries around the world with varying climates, cultures, and socioeconomic distributions would be expected to exhibit large differences in both the indoor and outdoor NO_2 concentrations, and in the relative contributions of the sources of personal exposure. In terms of the outdoor pollutants, densely populated and highly industrialized cities should have a larger quantity of NO_2 emissions from traffic, energy production, and industry, although this might be attenuated by advanced pollution control technology and policy.

Personal, indoor, outdoor, and indoor/outdoor ratio levels of NO_2, and the correlation coefficient between personal *vs* indoor and outdoor level using passive NO_2 samplers in the same region for the three different adult studies and in this study of

Table 5. Comparison of data in adults in other studies and data in children in this study for nitrogen dioxide level using passive NO_2 sampler in Seoul, Korea

Reference	Subject	Study period	Major results
Yang *et al.*[16]	30 office workers	October 1999	Personal : 47.90 ppb Indoor : 43.20 ppb Outdoor : 52.20 ppb I/O : 0.83
Son *et al.*[17]	70 subway workers	Feb. to Mar. 1999	Subway office : 27.87 ppb Platform : 35.76 ppb Outdoor : 52.60 ppb I/O : 0.49 Personal vs indoor R : 0.65 Personal vs outdoor R : 0.67
Yang *et al.*[16]	122 ward officer	October 1999	Personal : 40.40 ppb Indoor : 40.40 ppb Outdoor : 49.90 ppb I/O : 0.81 Personal vs indoor R : 0.70 Personal vs outdoor R : 0.68
This study	42 elementary students	December 2002	Personal : 45.08 ppb Indoor : 27.89 ppb Outdoor : 30.96 ppb I/O : 0.88 Personal vs indoor R : 0.28 Personal vs outdoor R : 0.04

children are shown in Table 5. Personal NO_2 levels and indoor/outdoor ratio (45.08 ppb and 0.88, respectively) in this study of children data were similar to office workers (47.90 ppb and 0.83, respectively) and ward officers (40.40 ppb and 0.81, respectively). Furthermore, the correlation coefficients of personal *vs* indoor levels and personal *vs* outdoor levels for our study were lower than occupational groups as well as non-occupational groups. These results might be explained by the heterogeneity of the sampling group, differing sampling time, and the differing microenvironment of each study.

The relationship between the indoor, outdoor, and personal concentrations of NO_2 was determined. Personal NO_2 concentrations correlated slightly with indoor NO_2 concentrations. However, outdoor NO_2 concentrations did not correlate with personal and indoor NO_2 concentrations. These results contrast with those reported in the study by Lee *et al.*.[11] Since the majority of children in this study spent greater time inside their homes, the personal NO_2 exposure showed a stronger correlation with the indoor NO_2 concentrations ($r^2 = 0.28$) than with the outdoor NO_2 concentrations ($r^2 = 0.04$). This stronger correlation between personal and indoor NO_2 levels could also be seen in other studies.[4-6,10,11] This might be explained by the heterogeneity of the sampling group (children, elderly, adult, worker, and student etc.) of each study, varying types of indoor combustion sources, and differences in other demographic characteristics.

The average NO_2 levels on the road correlate with the distance traveled to the traffic road, which implies that the ambient air NO_2 exposure was affected by a traffic emission source. In addition, the measured NO_2 levels on the road were compared with the residential outdoor levels. The NO_2 level on the road was approximately 10~20% higher than the residential outdoor levels. In Western Europe, North America, and Brisbane Australia, the ratio of NO_2 concentration of traffic emissions to residential outdoor levels was < 2.[10] The ratio was less than 1 in Scandinavian cities and the

United Kingdom. This suggests that NO_2 levels in traffic emissions correlated with the status of the ambient air pollution in each country. A study by Cyrys *et al.* reported indoor and outdoor concentrations of NO_2 for Erfurt in East Germany and Hamburg in West Germany. The authors observed that the presence of gas cooking stoves, smoking and traffic exhaust emissions accounted for a large proportion of indoor NO_2 levels.[15] The findings are similar to those in our study.

The results of this study suggest that while the majority of an individual's NO_2 exposure was attributed to the location in which they spent the majority of the time, there were significant contributors to NO_2 exposure from locations where they spent the least time. While some of the locations and activities tested here are limited due to the small sample size and various measurement errors, this analysis may be useful for predicting the locations and activities that may lead to a higher NO_2 exposure according to the activity level of a given study population. Future analyses should examine a more active and diverse set of participants. The predicted NO_2 profiles for the various microenvironments and study populations may ultimately be useful for developing a more sophisticated NO_2 exposure model.

Conclusions

This study measured the NO_2 concentrations inside the home, outdoors near home, and the school environment. In addition, personal NO_2 exposure levels were measured using a passive sampler and information on household characteristics and daily activities gathered using questionnaires. The indoor and outdoor NO_2 concentrations were measured and compared with the simultaneously measured 24-hour personal exposures of 42 primary schoolchildren during the month of December 2002 in Seoul, Korea.

The household characteristics and daily activities were used to determine the contribution of these factors on personal NO_2 exposure. The indoor NO_2 level was weakly associated with the presence of a gas stove; however the personal NO_2 exposure levels in houses with a gas stove was significantly higher than those without a gas stove. Personal NO_2 exposure levels, which were due, in large part, to the use of a gas stove, was 2.5 ppb higher in houses with a gas stove than those without.

This study reinforced the importance of indoor sources of NO_2 and activities in the various microenvironments that contribute to personal NO_2 exposure. Personal NO_2 exposure correlated more with indoor NO_2 ($r = 0.28$) than the outdoor NO_2 concentrations ($r = 0.04$). The time-weighted average of personal exposure was estimated in a model using NO_2 levels inside the home, outdoors near the home, inside the school, outdoors near the school, and on the traffic road. The estimated personal NO_2 exposure is associated with the measured personal exposure as indicated by a Pearson correlation coefficient of 0.34. The measured NO_2 levels did not exceed the current ambient air quality standard of Korea.

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