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Indoor NO_2 Sampling in a Large University Campus in Benin City, Southern Nigeria, Using Palmes Diffusion Tubes

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Abstract. Monitoring of NO_2 in different indoor environments (without cooking and with cooking using different fuels) was done. Palmes diffusion tubes were used for the monitoring. The sampling duration was two weeks. The highest NO_2 concentration of 38.61 ppb (73.74 μg/m^3) was monitored in the room where the cooking was done with a gas burner. This was followed by the room with firewood cooking, where the concentration was 36.75 ppb (70.19 μg/m^3) and the least concentration of 24.05 ppb (46.80 μg/m^3) was noted in the room, where kerosene stove was used for cooking. It is of significance to observe that the WHO annual average guideline value of 40 μg/m^3 was exceeded in all the rooms where cooking was done. Levels obtained in this study, therefore, suggest a need for precautionary mitigation. However, the outdoor concentration of NO_2 was almost the same as that obtained indoors in the rooms without cooking. This suggests high penetration indoors of outdoor NO_2. A background level of 3.40 ppb (6.49 μg/m^3) was established for the environment in Ugbowo, Benin City, Nigeria.

Keywords: indoor No_2, outdoor No_2, cooking fuels, nitrogen dioxide, air pollution, Palmes diffusion tubes

Introduction

Although overwhelming evidence exists on the anthropogenic source of air pollutants in Nigeria, yet only scanty information and data are available on this subject. Informations on the tropospheric levels of the solid and liquid droplets have been provided (Baumbach *et al.,* 1995; Adejumo *et al.,* 1994; Ogunsola *et al.,* 1993). Ambient concentrations of some gaseous pollutants have also been reported recently (Ukpebor and Ahonkhai, 2000). Most people spend more time indoors than outdoors, thus making indoor spaces important microenvironments when addressing risks from air pollution. A recent report indicates that a person is perhaps 1000 times more likely to inhale a chemical molecule if it is emitted indoors than outdoors (Nazaroff and Singer, 2004). Furthermore, estimates from the World Health Organization indicate that indoor air pollution from the use of solid fuels, accounts for 1.6 million deaths globally per year (WHO, 2002).Though it has been estimated that about 1.9 million people die annually as a result of exposure to high concentrations of suspended particulate matter and other pollutants in the indoor air environment in the developing countries (WHO, 2000), information and resources to control the indoor air quality are often lacking (Ferrari *et al.,* 1995). The only reported indoor measurement till date in Nigeria is the 1999 study on the indoor levels of NO_2 in an operating room using a nitrous oxide as an anaesthetic (Ukpebor and Imarengiaye, 2002). Prohibitive cost of air sampling equipment, erratic electric power supply, lack of trained personnel and unreliability of some of the available passive samplers are some of the reasons advanced for the lack of data on indoor air quality status. NO_2 has been primarily selected for air monitoring because of the recent confirmation of the reliability and sensitivity of Palmes diffusion tubes for NO_2 (Ukpebor *et al.,* 2004; Hansen *et al.,* 2001; Hangartner, 1999). Secondly, NO_2 is an important indicator of air pollution because the concentration of NO_2 is well correlated with the concentration of carbon monoxide, particulates, polycyclic aromatic hydrocarbons (Lewis *et al.,* 1995), and soot (Bower *et al.,* 1991).

The indoor NO_2 is generated from a series of sources, such as tobacco smoke or from cooking with biomass fuelled stoves. While cooking, unvented gas and kerosene heaters may increase the indoor NO_2 levels (Bardana, 2001). Elevated levels of NO_2 have been measured routinely in kitchens during conventional gas cooking (Spengler and Cohen, 1985). NO_2 may cause irritation of nose and eyes and may induce lower respiratory system symptoms through penetration of the conducting airways (Alberts, 1994). In healthy subjects, any exposure to NO_2 at levels found indoors may cause airway inflammation, affect blood cells, and augment susceptibility of airway epithelial cells to injury from respiratory viruses (Frampton *et al.,* 2002). High indoor levels of NO_2 have been associated with an increased prevalence of respiratory symptoms in healthy children (Garret *et al.,* 1998) and adults (Simoni *et al.,* 2002), or asthmatic subjects (Ng *et al.,* 2001).

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The work reported here is a pilot study designed to assess the NO_2 levels in different indoor environments within the Ugbowo Campus, University of Benin, Nigeria. It is hoped that this report will stimulate further indoor air pollution studies. Furthermore, it is anticipated that the data generated would help in policy development and prioritization of management actions.

Materials and Methods

Study areas. Ugbowo Campus, University of Benin, Nigeria, is populated with about 40,000 inhabitants, approx 30,000 of which are students and the rest include staff and their families. About 70% of the student population resides on the campus. Different indoor environments were carefully selected on the campus in this pilot study, which included a room with a kerosene stove used exclusively for cooking, a room with a gas burner, a kitchen with firewood as the means of cooking, and a room with no cooking to serve as the control. Two outdoor measuring sites were also selected, a remote site about three km from the residential area with no motorized traffic density and another site with very low traffic density. The former remote site was to act as a control and also to provide the background NO_2 concentration in the environment. The latter outdoor site was to facilitate the calculation of outdoor/indoor NO_2 ratio.

NO_2 monitoring. NO_2 measurement was carried out by using Palmes diffusion tubes. The diffusion tubes used in this study consisted of a small acrylic tubing, 8.20 cm long with a cross-sectional area of 0.82 cm^2, having two stainless steel mesh as support for adsorbing material at its one end. Triethanolamine was used as the adsorbent for NO_2. The sensitivity and utmost accuracy of this particular tube length and the selectivity and specificity of triethanolamine for NO_2 have been documented (Ukpebor *et al.*, 2004; Gold, 1977). In preparing the tubes, uniformity was maximized, for example, the same drying time for each of the steel grids, and the use of a freshly prepared triethanolamine/acetone mixture. The required steel grids were cleaned with acetone and dried properly. A mixture of two parts of acetone and one part of triethanolamine was prepared and stirred thoroughly. The grids were dipped in the mixture and dried for 20 min. Dipping in solution prior to assembly was found to give significantly more precise NO_2 concentrations than by the pipetting method (Hamilton and Heal, 2004). After drying, two of the steel grids were placed at one end of the tube and the tube was capped. The prepared tubes were stored in a refrigerator and finally exposed at the different monitoring sites. Two tubes from each of the prepared sets were retained in storage as 'blanks' for later analysis along with the exposed tubes. A two-week sampling period was observed to allow a reasonable quantity of nitrogen dioxide to be adsorbed.

After exposing the passive samplers, the amount of NO_2 adsorbed was determined colourimetrically as nitrite with Saltzmann reagent (Palmes *et al.*,1976). A visible spectrophotometer (Spectronic, 21D) at zero extinction, previously calibrated with known concentrations of nitrite (NO_2^-), was used to determine the absorbance of both blanks and the air samples at 540 nm, using the reagent as referred. Absorbance readings from the unexposed 'blank' diffusion tubes were averaged for each preparation set and the value subtracted from the readings for the exposed tubes. The atmospheric concentration of NO_2 obtained during the measuring period was calculated as described by Palmes *et al.* (1976), using Fick's first law, the dimensions of the tube, and the diffusion coefficient of NO_2 in the air with the following equation:

$$C = \frac{22.4 \times 10^3 \times \frac{T}{273} \times F \times Z \times E}{t \times D \times A}$$

where:
C = NO_2 conc (ppb)
T = temperature (K)
F = calibration factor
Z = length of the diffusion tube (cm)
E = extinction minus extinction of the blank
t = exposure time (sec)
D = diffusion coefficient of the compound in air (cm^2/sec)
A = cross–sectional area of the tube (cm^2)

note: 1 ppb NO_2 = 1.91 μg NO_2/m^3

Results and Discussion

The time-weighted average concentrations of NO_2 obtained from the different indoors and the two outdoor environments during June and July are presented in Table 1. NO_2 levels measured from three of the four indoor environments seem high. However, these values can best be assessed by a comparison with the approved National and International Standards. The Federal Environmental Protection Agency (FEPA, 1991) allows daily average limit for NO_2, which is presently 75-113 μg/m^3. The US National Ambient Air Quality Standard (USEPA, 1990) for NO_2 is presently 100 μg/m^3 annual average, while the World Health Organization annual average guideline for exposure to NO_2 is 40 μg/m^3, and the 1 h guideline is of 200 μg/m^3 (WHO, 2000). It is significant to indicate that the outdoor air had a different pollutant composition than that found in the indoor air. Not all of these compositions have been taken into account in developing the air quality guide-

Table 1. Measured indoors and outdoors NO_2 concentrations during the two months of study

Sampling site	NO_2 conc ppb (μg/m^3)		
	Analysis during June	Analysis during July	Mean values
Room with kerosene stove	25.98 (49.62)	23.02 (43.97)	25.05 (46.80)
Room with gas burner	40.83 (77.98)	36.38 (69.49)	38.61 (73.74)
Room with firewood	37.86 (72.31)	35.64 (68.07)	36.75 (70.19)
Room with no cooking	11.88 (22.69)	8.91 (17.02)	10.40 (19.86)
Vice-chancellor's lodge	11.92 (22.77)	8.17 (17.60)	10.05 (19.19)
Control site	3. 82 (7.30)	2.97 (5.67)	3. 40 (6.49)

lines given above and, therefore, may not be applicable under all circumstances. Despite this limitation, the data obtained would be discussed in line with the set standards.

The mean background NO_2 concentration obtained in this study was 3.40 ppb (6.49 μg/m^3). This level was obtained in the control site, a very remote location devoid of any of the known anthropogenic source group of NO_2. This, therefore, indicates a natural unpoluted source and hence the expected minimum in the monitoring environment. The highest NO_2 range of 36.38 ppb (69.49 μg/m^3) to 40.83 ppb (77.98 μg/m^3) measured in this study was recorded in the indoor site where exclusively gas burner was used for cooking. The mean NO_2 concentration in this room was found to be a factor of 11 times higher than the background concentration and a factor of 4 time higher than what was measured at the outdoor control environment selected for the study (near the Vice Chancellor's lodge). The second highest mean NO_2 concentration of 36.75 ppb (70.19 μg/m^3) was obtained in the room where firewood was used as the means of cooking. This room gave a calculated outdoors/indoors NO_2 ratio of 0.27, and a factor of 10 times higher than the background NO_2 level (Table 2). The least concentration range of 23.02-24.05 ppb NO_2 was measured in the room with kerosene stove. The room with no cooking recorded a mean NO_2 concentration of 19.86 μg/m^3. This is a factor of 3 times higher than the background NO_2 concentration. The calculated outdoors/indoors NO_2 ratio for this room was found to be 0.97 (Table 2). This reveals no significant difference between the outdoor control site concentration and the concentration measured in this room. The outdoors site selected near the Vice-chancellor's lodge gave NO_2 level of 10.05 ppb (19.19 μg/m^3). This value is a factor of 3 times higher than the background concentration. The difference of (13.37 μg/m^3) between the background level (6.49 μg/m^3) and the measured NO_2 concentration (19.86 μg/m^3) in the room with no cooking may be due to penetration indoors of the outdoors NO_2. Indoor concentrations of air pollutants are influenced by outdoors levels, indoor sources, the rate of exchange between indoor and outdoor air, and the characteristics and furnishings of the buildings (WHO, 2000). Indoor values of 20% to 80% of the outdoor concentrations have been reported (Yocum, 1982).

Table 2. Calculated outdoors/indoors NO_2 ratio

Sampling site	Outdoors/indoors ratio
Room with kerosene stove	0.42
Room with gas burner	0.26
Room with firewood	0.27
Room with no cooking	0.97

The differences in the NO_2 levels obtained in the different indoor environments could be a function of the mode of combustion, the type of fuel used for cooking and the quantity of fuel combusted. However, since the samplers gave only average values for the exposure period, no attempt was made to record the pattern or extent of fuel usage. In the developed countries, indoor levels of NO_2, for example, are affected by gas heaters and cooking ranges (used in 20-80% of houses in some countries). In five European countries, the average NO_2 concentrations (over 2-7 days) for dwellings with gas equipment were in the range of 20-40 μg/m^3 in living rooms and 40-70 μg/m^3 in kitchens, and 10-20 μg/m^3 in dwellings without gas equipment (WHO, 2000). In addition, short-term measurements reveal NO_2 concentrations that may be five-folds higher than those averaged over several days. Peak values of up to 3800 μg/m^3 for 1 min have been measured in the Netherlands in kitchens with unvented gas cooking ranges (Seifert, 1993; ECA, 1989).

Relatively few studies have been conducted to determine the health effects of indoor exposures to air pollutants in the developing countries. Enough data have become available in recent years, however, to obtain some preliminary information on the type and very approximate magnitude of effects (Chen *et al.,* 1990). NO_2 levels of about 940 μg/m^3 (0.5 ppm) increase

susceptibility to bacterial and viral infections. Epidemiological studies evaluating the effects of NO_2 exposures in homes with gas cooking appliances have been conducted. In general, epidemiological studies of adults and infants (less than 2-year olds) show no significant effect of the use of gas cooking appliances on respiratory illnesses, nor do the few available studies of infants and adults show any association between pulmonary function changes and gas stove use. However, children 5-12 years old are estimated to have a 20% increased risk for respiratory symptoms and disease for each increase of 28 $\mu g/m^3$ NO_2 (2-week average), where the weekly average concentrations are in the range of 15-128 $\mu g/m^3$ or possibly higher (WHO, 2000).

Conclusions

With growing public concerns about the indoor air quality, action has been taken in many developed countries to characterize the levels of indoor air pollutants. This present pilot study is the first attempt at initiating a large-scale indoor air pollution characterization process in Nigeria. The following can be inferred from this study. The NO_2 levels in the entire indoor/outdoor environments monitored, complied with the threshold values set nationally. However, it is of great significance that the WHO annual average guideline value of 40 $\mu g/m^3$ was exceeded in all the rooms where cooking was done. Levels obtained in this study, therefore, suggest a need for precautionary mitigation. Cooking with a gas burner indoors generated the highest NO_2 concentration of 77.98 $\mu g/m^3$. The room where cooking was done with a kerosene stove gave the lowest NO_2 level of 49.62 $\mu g/m^3$. There was no significant difference between the outdoor NO_2 concentration and the concentration in the room where no cooking was done, suggesting high penetration indoors of the outdoor NO_2.The background NO_2 concentration for Ugbowo environment was found to be 3.40 ppb (6.49 $\mu g/m^3$).

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