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Sulfur Dioxide and Nitrogen Dioxide Levels Inside and Outside Homes and the Implications on Health Effects Research

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■ This paper presents the results of 1 year's indoor and outdoor monitoring for SO₂ and NO₂ in six communities with widely varying outdoor levels. The representativeness of the monitoring in defining exposure is discussed for each city. In four of the communities, outdoor SO₂ levels are less than 50% of the annual NAAQS, while violations are found in the other two. Annual average indoor levels of SO₂ are never found to exceed the standard. In fact, indoor SO₂ concentrations are 20 to 70% of the outdoor levels. Indoor NO₂ levels, on the other hand, can exceed outdoor levels by a factor of two, depending on the type of cooking appliance used, but do not exceed the standard. The impact of various heating and cooking systems on the indoor concentrations of these gases, as well as the type of appliances used in the house, is evaluated.

A prospective epidemiological study (1) to assess respiratory health effects of particulates and sulfur oxides is now in its fifth year. Over the first 5 years, personal interviews and pulmonary functions have been administered to 9000 adults between ages 25 and 74 and to 11 000 children in grades one through six. The rates of decline of the pulmonary function in adults and the increase in children as they grow are obtained from spirometer tracings of forced vital capacity and forced expiratory volume in 1 s. Adults are reinterviewed at intervals of 3 years. Children are examined on a yearly basis.

In addition to pulmonary function tests, information on symptoms of respiratory disease, smoking, and occupational and residential history is obtained from adults. For the children a questionnaire (completed by parents) asks about living conditions, family composition, heating system and fuel, air conditioning, cooking system, and occupational and smoking habits of the parents.

This study was designed in an attempt to test the adequacy

of the current air pollution standards and to derive new information concerning sulfur oxides and fine particulates. As such, six communities having widely different ambient air pollution levels are being studied: Kingston and Harriman, Tenn.; Portage, Wis.; Steubenville, Ohio; St. Louis, Mo.; Topeka, Kans.; and Watertown, Mass.

It was apparent at the start that air pollution data from a central monitoring site might not provide adequate details of population exposure. A more comprehensive measurement strategy was therefore developed. As part of our assessment of exposure in the six cities, we have devised a three-stage monitoring strategy for each city which includes: central station monitoring, indoor/outdoor monitoring, and personal monitoring. The central station monitor uses standard continuous monitoring technology to collect hourly and 24-h samples every day in each city. The continuously monitored pollutants are SO₂, NO, NO₂, and O₃. Integrated 24-h SO₂, NO₂, mass respirable particulates (MRP), total suspended particulates (TSP), and the sulfate and nitrate fractions of the particulate matter are obtained every other day at the central site. Each community also has 10 or more satellite stations sampling simultaneously the 24-h indoor and outdoor concentrations of SO₂, NO₂, MRP, and respirable sulfate. On a limited basis, personal monitoring for NO₂, MRP, and respirable sulfates will be conducted in each community. The purpose of the personal monitoring is to develop and verify an exposure model. Eventually, exposure values will be assigned to every subject in the health study.

The purpose of this paper is to report the results of the indoor/outdoor SO₂ and NO₂ measurements and to discuss their relevance to exposure assessment in epidemiologic studies of air pollution. The results of the indoor/outdoor MRP and sulfate sampling and the personal monitoring are reported in other papers (2, 3).

It is realistic to expect continued reliance on relatively few

ambient outdoor monitors for regulatory purposes. The relationship between these central site measurements and actual personal pollution exposures of individuals in each of our communities is ongoing research.

The role of indoor pollution may be particularly important, since most people spend so much of their time indoors rather than outdoors. The importance of characterizing indoor air pollution is documented in the results of human activity pattern studies. Chapin (4) and Szalai (5) have reported on extensive surveys on this subject. For comparative purposes their results have been condensed and summarized in Table I. We are interested initially in only the total amount of time spent indoors and outdoors at home, at work, time in transit, and a general category of other activities. Time reported at work in these studies is assumed as indoors. The results are consistent, showing about 22 h per day (92%) spent indoors, and most of that, 16 h, at home. Therefore, the largest determinant of average (and perhaps peak) nonoccupational exposures should be indoor residential concentrations. Air pollution concentrations associated with transportation will have a significant impact on integrated exposure only if the "transportation concentration \times transit time" approaches that for indoor exposure. For specific pollutants, such as lead or CO, transit exposures may be significant. Similarly, considering the relatively small amount of time spent outdoors by the general population, the levels would have to be considerably higher than indoors to override the importance of indoor concentrations. However, outdoor concentrations may be a principal variable in determining indoor concentrations as well as home characteristics that influence ventilation rates, removal processes, and indoor sources.

Clearly, in an attempt to ascertain air-pollution-related health impairment, it would be illogical to measure ambient air only. This is particularly true for gases and particles for which there may be indoor sources or where the home ventilation rates affect the buildup or dilution of concentrations. Improper or inadequate assignment of exposure would bias results because subjects' exposures would be misclassified.

Previous Work

A number of investigators have indicated that there can be differences between indoor and outdoor levels of SO₂ and NO₂ air pollution. Biersteker et al. (6) noted that SO₂ levels were lower indoors and that the age of the house influenced the level, suggesting that new houses with fresher plaster had lower levels. They also pointed out that faulty flues on heaters could be sources of SO₂ indoors. Anderson (7) compared 24-h samples of sulfur dioxide collected outside and inside a room over a 7.5-month period in Denmark. Inside SO₂ averaged 51% of the outside SO₂ concentrations.

Wade et al. (8) have made continuous measurements in kitchens with gas appliances. Levels of NO₂ up to 1 ppm (1880 $\mu\text{g}/\text{m}^3$) were found. Derouance and Verduyn (9) have pointed out that gas hot-water heaters are a potential source of NO₂ in homes in Europe. Higher levels of NO₂ in dwellings with gas stoves vs. electric stoves were reported by Palmes et al. (10). The mean weekly levels derived from exposing a pair of diffusion tubes in 10 homes with gas cooking and 9 homes with electric cooking were 49 vs. 8.3 $\mu\text{g}/\text{m}^3$. Unfortunately, no outdoor levels were reported.

Aerometric Measurements

In each of the six cities studied, indoor and outdoor air pollution concentrations have been measured in a minimum of 10 homes or public facilities for at least 1 year. Twenty-four-hour integrated samples of sulfur dioxide, nitrogen dioxide, and respirable particulates are collected every sixth day.

The indoor/outdoor sampling network was designed with

Table I. Summary of Time-Use Studies into Indoor and Outdoor Activity Patterns

location	h spent in each location	
	Chapin (1974)	Szalai (1972)
home, indoors	16.03	16.75
work, indoors	4.61	4.03
other, indoors	1.31	1.63
total indoors	21.95	22.41
home, outdoors	0.27	0.23
work, outdoors		
other, outdoors	0.27	0.12
total outdoors	0.54	0.35
transit	1.16	1.25
total	23.65 ^a	24.01

^a Chapin does not explain this difference from 24 h.

two objectives: first, to define the spatial distribution of outdoor air pollution in each of the cities being studied, and second, to define the indoor/outdoor relationships in a variety of homes. No a priori assumptions were made on what home variables were important in determining indoor levels. Rather, homes were selected to represent the range of home characteristics in each community.

The homes were described by eight site characteristics that describe potential sources in the home and ventilation factors. The indoor source variables are: the number of smokers in the building; the estimated number of cigarettes smoked each day inside; the type of heating fuel used (electricity, gas, oil, wood); and the type of cooking fuel used (electricity, gas). The ventilation variables are: the type of heating system (forced hot air, radiative, or convective); the use of storm windows; type of air conditioning used (none, room units, central); and kitchen ventilation (none, recirculation, outside).

At each site, samplers were located in the main activity room of the house (living room, TV room, den). The collecting reagents were located inside to control for temperature. The exposed samples were collected and refrigerated the day following sampling, except for Friday and Saturday test days, when the samples were collected on Monday. The bubbler samples for SO₂ are measured by the West-Gaeke method (11) and NO₂ by the modified sodium arsenite method (12). These samples are analyzed in local cooperating laboratories to prevent loss of sample due to spillage or decay in transit. A detailed description of sampling, analytic procedures, and data validation procedures is available in the "Indoor-Outdoor Quality Assurance and Procedures Manual" (13).

Analysis

In presenting the results of the indoor/outdoor SO₂ and NO₂ monitoring, the question of population exposures in a cross-sectional air pollution health study is addressed. Specifically, the initial analysis is directed toward ascertaining whether there are significantly different exposures among the six cities. A standard statistical package computer program (14) was used to compute mean levels by an ordered set of variables. Paired *t* tests for significant differences between indoor and outdoor concentrations and pooled *ts* to test differences among cities were performed. NO₂ levels at each site within a city were analyzed to determine the significance of the observed variability between days, and between homes with gas cooking and those with electric cooking. Prior to performing this analysis of variance, the missing values were filled to conform with a linear model, and the degrees of freedom associated with the error mean square were appropriately adjusted (15, 16).

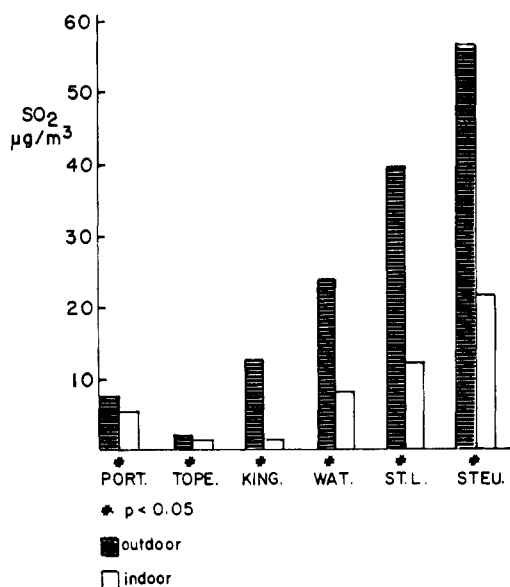


Figure 1. Annual sulfur dioxide concentration averaged across each community's indoor and outdoor network (May 1977–April 1978); # implies $p < 0.05$ significant difference by paired t test

Results

Sulfur Dioxide. The annual sulfur dioxide concentrations averaged across each community's network of indoor/outdoor monitors are displayed in Figure 1. On the basis of ambient concentrations, the six cities appear to have significantly different mean SO₂ levels. The six cities could be divided into three broad exposure groups. Steubenville and St. Louis are the highest, Watertown and Kingston are medium, and Portage and Topeka are the lowest. The mean outdoor SO₂ concentrations were found to be significantly different ($p < 0.01$) by a pooled t test for all cities.

Indoor SO₂ values are dramatically reduced compared to outdoor values. In the cities of modest to high SO₂ concentrations, the reduction is in the range of 50–70%. In Kingston, where all the monitored homes were air conditioned, the reduction of SO₂ was approximately 90%. In Portage and Topeka, where ambient concentrations are the lowest, a 10–30% reduction was found. All these reductions were found to be significant at the 95% confidence level, by a paired t test.

In a comparison of the indoor mean levels between cities, the exposure differences are less dramatic. Only Steubenville remains significantly different from the other cities at the $p < 0.02$ level. The indoor mean levels between Watertown and Portage, Watertown and Topeka, and Watertown and St. Louis are not significantly different.

The range of indoor and outdoor mean exposures for each city is displayed in Figure 2. The composite mean for the 11 sites in Portage, Kingston, Watertown, and Steubenville, the 14 sites in Topeka, and 15 sites in St. Louis is also indicated. The ranges of indoor mean values in St. Louis and Steubenville overlap, even though there is a significant difference in the composite mean concentrations by pooled t test.

Implications of Indoor SO₂ Exposures. From the perspective of an epidemiological study on the health effects of air pollutants, these six cities provide a wide range of outdoor ambient SO₂ exposures. The community mean values range from 52 µg/m³ in Steubenville to 2 µg/m³ in Topeka. The indoor mean levels reveal the same pattern of spread between cities; however, the range is less. Steubenville's indoor mean values are 22 µg/m³, while Kingston and Topeka's levels are 1 µg/m³. The study populations in these communities are experiencing an indoor SO₂ exposure considerably less than



Figure 2. Range of annual mean sulfur dioxide concentrations inside and outside homes for each community

outdoor ambient exposure. Overall, the SO₂ exposure reduction is approximately 50%.

Examining the range of indoor mean values for individual homes in each city reveals the overlap of exposures between cities. In Steubenville and St. Louis, the range of mean concentrations implies that a portion of the population in each community has the same SO₂ exposures, despite significantly different mean outdoor concentrations. Similarly, depending on location and activity patterns of populations in the other four cities (Watertown, Kingston, Topeka, and Portage), exposures of subpopulations in the cities will be equal to those in other cities with significantly different ambient levels.

A cross-sectional study attempting to associate pulmonary functions and/or prevalence of respiratory illness would be biased if SO₂ exposures were determined only by outdoor ambient measurements. By virtue of reduced indoor SO₂ levels and the overlapping range of indoor exposures, the subjects could be misclassified. In spite of the great reduction of SO₂ levels indoors, we are encouraged that populations in Watertown, Kingston, Portage, and Topeka are never exposed to levels as high as those found inside homes in St. Louis and Steubenville. Individual exposures will depend on indoor/outdoor activity patterns. Seasonal variability in both air pollution concentrations and amount of time spent outdoors will be important factors in defining SO₂ exposures in these communities. Time-weighted exposure models are being developed as part of this epidemiological study.

Some epidemiological studies have indicated health effects at outdoor SO₂ levels of 115 (17) and 120 µg/m³ (18). These studies indicated increased morbidity in the presence of 160 µg/m³ smoke and increased severity of respiratory disease with 100 µg/m³ smoke, respectively. In these British studies, atmospheric particulate burden was inferred from smoke shade. A precise conversion of smoke shade to TSP concentrations is not possible because of dependency on particle composition. These studies were for annual averaged concentrations, and actual population exposure to indoor SO₂ may have been 50% lower. As Waller (19) recently clarified, increased respiratory symptoms were discernible in London bronchitic patients at 24-h averages around 500 µg/m³ SO₂ with 250 µg/m³ smoke (20). One assumes these patients were indoors where actual SO₂ levels may have been 20–50% lower, or 400–200 µg/m³.

Reduced indoor SO₂ values underscore the need for more precise determination of exposures. Inadequate definition of exposure in previous epidemiological studies may have led to incorrect interpretation of results. True health effects response may have been masked by the biasing influence of indoor exposures. Health effects associated with ambient SO₂ concentrations may be overestimating the actual levels where effects are occurring. This presumes that SO₂ is the active

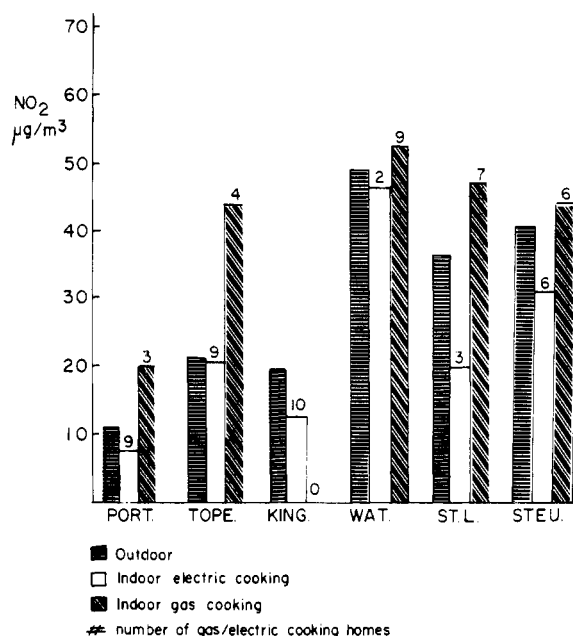


Figure 3. Annual nitrogen dioxide concentration outside and inside electric and gas cooking homes, averaged across each community's indoor and outdoor network (May 1977–April 1978)

component. On the other hand, SO₂ may be acting as a surrogate for some other more reactive compound whose indoor/outdoor relationship may be entirely different from SO₂.

Nitrogen Dioxide. The relationship between indoor/outdoor NO₂ levels is quite different from that for SO₂. The NO₂ level can be higher or lower inside, depending on the indoor NO₂ sources and ventilation. Figure 3 displays the annual NO₂ concentrations outside and inside homes with electric and gas cooking averaged across sites in each community. In every community the overall indoor to outdoor NO₂ ratio is less than one for homes with electric stoves and greater than one for homes with gas stoves. There are very few homes with gas cooking in Kingston, and none were included in this initial survey.

Comparing cities on the basis of ambient NO₂ levels, there appear to be two groups. Ambient NO₂ levels in Portage, Topeka, and Kingston are significantly different ($p < 0.001$) from the levels in Steubenville, St. Louis, and Watertown. This reflects the differences in density of vehicular traffic and combustion sources.

When comparing the indoor mean levels, a different pattern emerges. Now indoor NO₂ in Steubenville is not significantly different from indoor values in Topeka. The level of significant difference between Watertown and Topeka has been reduced from $p < 0.001$ (outdoors) to $p = 0.0025$ when mean indoor NO₂ levels are compared. The similarity of the mean indoor NO₂ levels between cities and the overlap in the range of individual site mean values (see Figure 4) will make it difficult to characterize population exposures across these cities on the basis of ambient or even an aggregate indoor measurement. Clearly there are people living in the relatively clean outdoor air of Topeka that have the same or even higher indoor exposure to NO₂ as people living in the relatively polluted air of Watertown, Steubenville, or St. Louis.

Analysis of variance indicated that in each city, for homes with both gas and electric cooking, there was a significant difference among sites. For both gas and electric cooking, the variability indoors was greater than the outdoor variability. There were, however, two exceptions: in homes with gas cooking in Portage, the variability was small, both indoors and

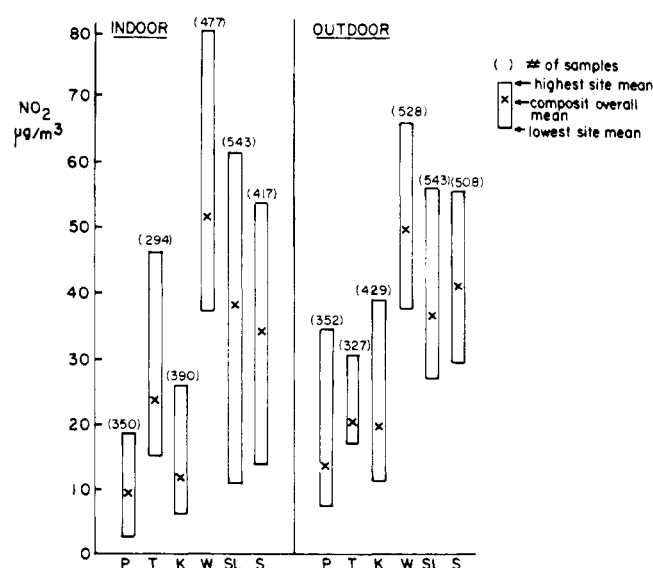


Figure 4. Range of annual mean nitrogen dioxide concentrations inside and outside homes for each community

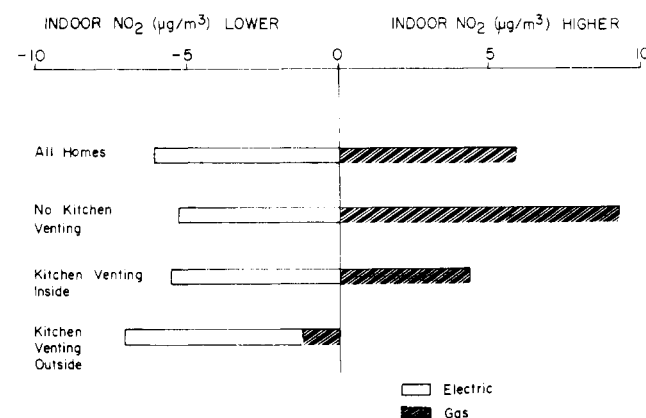


Figure 5. Mean indoor/outdoor difference in nitrogen dioxide concentrations by cooking fuel and kitchen ventilation, averaged across all indoor/outdoor sites (May 1977–April 1978)

outdoors, and in homes with electric cooking in Watertown, the variability was greater outdoors than indoors. The difference in Watertown is related to the location of the homes with respect to the major traffic routes.

The most striking aspect of these analyses is the difference between homes with gas and electric cooking. When we look at the outdoor values, the variance is of the same order of magnitude as the variance between homes with the same cooking fuel. By contrast, the indoor values show very large differences between homes using gas and electricity in every city. The indoor difference between homes using gas and electricity is greater than the difference between homes with the same fuel, and, except for Steubenville, ranges from 3 to 7 times larger for gas.

Another factor influencing indoor NO₂ is kitchen ventilation. For each home in the sample, we determined whether there was kitchen ventilation and, if so, whether it vented to the outside or not. No attempt was made to ascertain the frequency of use. To illustrate the importance of this variable, the differences between the means of all indoor/outdoor pairs were calculated and associated by cooking and kitchen venting code to obtain the overall effect on the aggregated data. Figure 5 displays these results. A positive value means indoor levels are higher. There are no differences among the electric cooking venting values. However, all the gas-venting mean concentrations are different from each other ($p < 0.05$). All values,

both for gas and electric cooking, are statistically different from zero ($p < 0.05$).

Implications of Indoor NO₂ Exposure. The respiratory effects from cooking with gas or electricity have been investigated on the assumption that gas homes may produce potentially harmful amounts of nitrogen oxides. Melia et al. (21) reported that children in homes with gas cooking had more instances of respiratory disease than children in homes with electric cooking. They noted that girls showed a greater difference than boys and suggested that this may reflect their spending more time in the kitchen; however, they point out that this is speculative. No indoor levels of pollutants were reported.

Keller et al. (22), on the other hand, were not able to show any difference in respiratory illness rates among members of households using gas or electricity. This may reflect the relatively small number of subjects involved.

Upon examining this evidence, it becomes clear that people cooking with gas without kitchen ventilation, or with only recirculating vents, will have an NO₂ exposure greater than ambient. People with electric stoves or gas stoves vented outside will have exposures lower than outdoor ambient and substantially lower than people with unvented gas stoves. Epidemiological studies on the health effects of NO₂ and other air pollutants must take these two variables into account. Further, cities with significantly different mean ambient outdoor NO₂ concentrations could have comparable population exposures because of cooking and kitchen ventilation variables.

Other investigators (10, 23) have taken an initial look at NO₂ levels inside homes with gas cooking. Palmes indicated with limited monitoring that kitchen NO₂ concentrations averaged over a week are approximately twice the values measured in living rooms in homes with gas cooking. Our continuous NO₂ measurements in a residence with gas stoves demonstrate that levels exceeding 500 $\mu\text{g}/\text{m}^3$ and even 1000 $\mu\text{g}/\text{m}^3$ can occur during cooking. These high levels can last from minutes to hours. Similar results were reported by Cote et al. (23).

This has interesting implications for our measurements. All our home samplers are in the activity room of the house, which is usually the living room. Kitchen annual mean values in our study may exceed 100 $\mu\text{g}/\text{m}^3$ if we extrapolate from other studies. Further, the short-term hourly kitchen NO₂ levels during cooking may be 5 to 10 times higher than measured mean values.

Conclusion

These observations indicate that actual population exposures to NO₂ and SO₂ may not be adequately estimated by ambient outdoor measurements. This has serious implications for previous epidemiologic studies. Air Quality Standards based on these studies may be inadequate to protect the health of the entire population with an appropriate margin of safety.

For nitrogen dioxide, increased indoor levels associated with gas stoves may mask any health effect attributed to outdoor nitrogen dioxide levels. Removing this bias from previous epidemiologic studies might increase the strength of association between ambient NO₂ levels and observed health effects, or even lower the levels where health effects are significant.

For sulfur dioxide, indoor levels are lower than outdoors. The health effects attributed to outdoor SO₂ concentrations may actually occur at lower exposures. To the extent that the indoor/outdoor relationship has remained the same, the current SO₂ NAAQS are appropriate. However, if housing characteristics and/or activity patterns have changed sub-

stantially over the years, then the present standard may not be appropriate.

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