THE ASSESSMENT OF HUMAN EXPOSURE TO NITROGEN DIOXIDE

EVALUATION DE L'EXPOSITION HUMAINE AU DIOXIDE DE NITROGÈNE

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ABSTRACT

The assessment of human exposure to air pollutants has become a focus of recent research. This interest stems from the need to identify exposed populations and levels of such exposure from both indoor and outdoor sources in order to assess the overall risk associated with these pollutants. One such pollutant, nitrogen dioxide (NO_2), is of particular interest to the natural gas industry and consumer in that it is produced in the presence of high temperature combustion such as that found in the flames of gas ranges and ovens. Exposure to this pollutant in high concentrations is known to produce adverse health effects, but the effects of exposure to low levels of ambient NO_2 have not been completely investigated. Additionally, prior to 1984, no systematic exposure assessment for this pollutant in residential environments had been undertaken. In 1983, the Gas Research Institute contracted Harvard School of Public Health researchers to design and implement a complete exposure assessment study for this pollutant. In this work, we report some of the major findings of this exposure assessment.

RÉSUMÉ

L'évaluation de l'exposition humaine aux polluants de l'air est devenu un centre d'intérêt pour la recherche actuelle. Cet intérêt émane du besoin d'identifier les populations exposées ainsi que les niveaux d'exposition aux sources de polluants intérieures et extérieures afin d'évaluer le risque total por la santé associé avec ces polluants. L'un de ces polluants, le dioxide de nitrogène (NO_2), présente un intérêt particulier pour l'industri du gas naturel et ses consommateurs parce que le gas est utilisé en présence d'une température de combustion élevée dans les flammes et les fours. Les effets négatifs sur la santé de l'exposition à de hautes concentrations de dioxide de nitrogène sont connus, mais, les effets de l'exposition aux faibles concetrations du NO_2 ambiont n'ont pas encore été entièrement recherchés. En outre, avant 1984, aucune évaluation systématique de l'exposition au NO_2 dans l'habitat n'avait été entreprise. En 1983, l'Institute Americain de Recherche sur le Gas (Gas Research Institute) établit un contrat avec les chercheurs de l'Ecole de Santé Publique de l'Université de Harvard (Harvard School of Public Health) afin qu'ils mettent en ouevre et ménant à bien une étude complète pour évaluer l'exposition humaine due au dioxide de nitrogène. Le travail présenté ici relate les principaux résultats obtenus lors cette étude.

INTRODUCTION

The assessment of human exposure to air pollutants has become a focus of recent research. This interest stems from the need to identify exposed populations and levels of such exposure from both indoor and outdoor sources in order to assess the overall risk associated with these pollutants. Nitrogen dioxide (NO_2) exposure risk assessment has been at the forefront of such research. The National Ambient Air Quality Standard (NAAQS) in the United States for this pollutant (53 ppb as an annual average) presupposes that: one year is the proper averaging time for NO_2-related health effects; and, ambient air is the major contributor to exposure. While exposures to extremely high concentrations of NO_2 can cause death due to pulmonary edema (1), the effects of exposure to low levels of ambient NO_2 have not been completely investigated. Also, the impact or repeated exposures to short-term (several minutes) peaks exceeding the 53 ppb by as much as an order of magnitude are, likewise, in need of adequate investigation. Finally, as most individuals spend the majority of their time indoors, exposures experienced in such environments often dominate total exposure.

Indoor environments, especially residential environments, are important in the overall picture of exposure assessment. Because of the large proportion of time people spend indoors, the indoor contribution to total exposures to pollutants can be substantial. Nitrogen dioxide may be present in higher concentrations in indoor environments than outdoors if unvented combustion appliances are present and are used. Therefore, a detailed characterization of the home environment is an important starting point for a complete exposure assessment for NO_2.

In a detailed exposure assessment study, personal exposures experienced by individuals are the main prerequisite. But an understanding of the factors that influence such exposures may allow prediction of personal exposures for locations or cohorts not actually monitored. Toward this end, it is reasonable to carry out a controlled monitoring program designed not only to assess the exposure of the study population but also to assess the factors most strongly influencing personal exposures with an eye toward developing models of personal exposure which use as input variables factors which are more easily, and cheaply, measured.

From the point of view of risk management, the understanding of population exposures to any pollutant is critical. Variability among seemingly-similar sub-populations is especially noteworthy. Additionally, conflicting results from epidemiologic investigations of health effects due to nitrogen dioxide exposure (2),(3) have signaled the need for a detailed investigation of exposures. It is believed that the potential for misclassification of individuals in such studies, owing to the use of surrogates for exposure, may have been part of the cause of equivocal results (4). Another potential difficulty in assessing risk stems from the difference between exposure and dose (5): dose to the target organ is responsible for adverse health effects. Damage to the lung is the major mechanism of damage for NO_2. Variability in uptake due to activity and work patterns further clouds the understanding of NO_2-related health effects. Increased understanding of inter-individual variability in breathing rate and response to given concentration of this pollutant is needed.

Cost considerations often preclude the use of state-of-the-art technology for large scale monitoring. For example, in the case of NO_2 exposure assessment, the use of continuous instrumentation, while giving real-time measurements of pollutant concentration, would be prohibitively expensive. Inexpensive time-integrating devices, on the other hand, may provide insufficient time resolution to assess fully the impact of various microenvironments. To overcome this obstacle, it is incumbent upon exposure assessment researchers to develop a study design which compromises between cost-efficient methods and methods with greater time resolution. This can be accomplished by selecting certain individuals and environments for more detailed analysis. Parallels can then be drawn between such monitoring and the longer-term, but less expensive, monitoring.

PROJECT DESCRIPTIONS

The Harvard-GRI Nitrogen Dioxide Exposure Assessment Study was designed to be a multiphased, population-based study making use of modern survey design techniques applied to air pollution and time/activity monitoring. Care in selection of participants in the study would results in the ability to extrapolate to larger populations. Phases included detailed characterization of residences followed by personal monitoring of individuals within the residences. Additionally, a detailed assessment of the short-term exposures associated with cooking events was included to appraise the magnitude of such exposures and their impact on total exposure. A brief description of each component follows in the next few paragraphs.

Boston Residential Characterization Study

A year-long survey of NO_2 concentrations and housing factors, the Boston Residential Characterization Study, was conducted in over 500 dwelling units (DUs) Boston using a stratified, clustered approach to sampling (6),(7). The presence of gas range was used as the stratification variable at the census tract level. A two-stage clustering was performed which ensured that each dwelling unit had a equal probability of selection. Dwelling units ranging from single-family, rural homes to multistory apartment units were monitored for nitrogen dioxide concentration, air exchange rate and water vapor concentrations. Monitoring occurred in three separate rounds encompassing the winter-spring season, the summer season and the fall season of 1985. Multiple locations were also monitored: kitchen, bedroom, living room and outdoors. Information was gathered via questionnaire on characteristics of the home and personal activities believed to influence exposure to nitrogen dioxide.

Personal Monitoring

To quantify the contribution of indoor sources, ambient air and the effect of varied activity patterns on NO_2 exposures, a multi-phased personal monitoring study was conducted in Boston, Massachusetts, and Los Angeles, California. The personal exposure studies were carried out to determine: a) population distributions of NO_2 exposures; and, b) the factors influencing these exposures. Nitrogen dioxide exposures were obtained for approximately 300 individuals in Boston and 650 individuals on Los Angeles utilizing an integrating, diffusion badge. Participants wore one personal sampler while indoors and a different sampler when outdoors. Outdoor measurements were also taken. The study was designed to evaluate the effect on personal exposures to nitrogen dioxide of the very different ambient concentrations of this pollutant found in these locations. The designs of these studies were quite different and merit brief description here. More detailed descriptions can be found elsewhere (8),(9).

Boston Personal Monitoring. Participants for the Boston Personal Monitoring Study were selected from among residents on the DUs monitored in the Boston Residential Characterization Study. During the summer sampling period, initial screening of residents was done with a simple questionnaire outlining their activities which might influence exposure to NO_2 such as cooking and commuting habits. Based on the results of this survey and monitoring in the homes, individuals were categorized as either high or low with respect to their potential exposure from activities and similarly with respect to their within-home concentration. Targets within each of the four categories created by cross-tabulating these results were selected randomly for participation in the personal monitoring study. Other members of the same household were solicited for participation and also classified.

Monitoring took place over two 48-hr periods; once in the winter, once in the summer in 1986. Individuals wore a single personal monitor while in their home and a separate monitor while out of their home. Additionally, a fixed-location monitor of the same type was placed in the bedroom of the participant. A detailed time-activity diary was kept by each participant. A total of approximately 300 individuals completed each season of monitoring. Of these, approximately 80% participated in both seasons.

Los Angeles Personal Monitoring. The Los Angeles personal monitoring study made use of previously-collected data on home characteristics obtained in a study funded by the Southern California Gas Company (10). This study afforded a characterization of the home types in Southern California and the impact of these characteristics on NO_2 concentration in homes. Participants were solicited by telephone via a random-digit dialing technique, also designed to obtain a statistically-representative sample.

Modeling work (11),(12) suggested that an adequate representation the population for the personal monitoring could be obtained by a simple random sample of 600 individuals with 200 monitored each season. Individuals were monitored only once during the year commencing in May 1987 and completed in May 1988. Because of this fact, an additional study was implemented to assess the between season variability of such measurements. Fifty individuals were monitored repeatedly; a total of

eight times throughout the year. These individuals, as well as acting as surrogates for the larger population, were asked to monitor their activities much more closely and to change monitors upon entering certain microenvironments. To distinguish between the larger group monitored only once and the smaller, repeatedly-monitored individuals, we assign the former group the name Main Study participants and the smaller group the Microenvironmental Study participants.

Short-term Exposure Studies

To examine exposures that occur over short durations, a study was conducted in which volunteers wore small, light-weight, portable, continuous NO_2 monitors (13). Volunteers were either the researcher himself or participants from other studies solicited by telephone. Air sampling was proximate to the nose throughout a cooking event. Data were stored as 5-second averages. Fixed-location, area monitors were operated in the kitchen and, in many cases, another room of the DU. Approximately 80 cooking events were monitored. Additionally, a smaller number of commuting samples were taken with the researcher simulated actual commuting routes and modes.

MEASUREMENT TECHNIQUES

Air pollutant sampling was performed in two different modes: integrated, passive sampling; and, continuous monitoring. Palmes tubes (14),(15), inexpensive, time-integrating devices, were used for large-scale survey sampling while the continuous instruments were used to assess peak exposures in a small number on individuals in selected environments. In a separate study in a research-house setting, relationships were established between continuous monitoring and the longer-term monitoring.

The analytical methods for sample analysis were adequate to ensure high quality data. Quality assurance plans were developed early in the study to ensure adequate control of preparation, instrumentation and sample chain-of-custody. The passive techniques used have an uncertainty of $\pm$ 15% which is acceptable for the investigation of exposure effects.

The quality assurance program was extensive in this study. In the Boston Residential Characterization Study, blank and replicate samplers were assigned to 10% of the field packages, each corresponding to monitoring in one residence. Random visits by senior staff to homes set up by field technicians revealed no procedural discrepancies. Furthermore, telephone sampling of a random 5% of the sample requesting cross-referencing information to collected questionnaire data revealed few inconsistencies. Extensive computer cross-checking was performed to minimize coding and keypunch errors. In the laboratory, standard operating procedures were developed. Systems audits are performed regularly at Harvard to ensure all such protocols are followed. Detailed control charts for instruments, calibration curves and other standards are kept on a daily basis. Round robin and inter-laboratory comparisons of the techniques are done on a routine basis several times per year.

Microenvironmental studies were performed using a real-time electrochemical analyzer modified for stability and portability. It uses a battery-operated pump which draws approximately 1 L/min of air. The response is linear over the range of 20 ppbv to 2.0 ppmv with a response time to 66% of final value in less than 5 s. Modifications were made to the electrochemical cell to improve time and temperature stability. The sampling manifold was designed with personal sampling in mind. The whole apparatus was fit into a small backpack to offer the least limitation in movement of the subject. Portable chemiluminescent analyzers were used as fixed-site monitors in this study as well.

RESULTS

Boston Residential Characterization Study

Figure 1 displays graphically the cumulative probability for indoor NO_2 in Boston residences. The data for the three seasons monitored has been averaged to estimate an annual average concentration. Note that only DUs for which data in all three seasons were available has been included. The results presented account for oversampling of the gas population and include only data for homes monitored in all three seasons. These data suggest that approximately 10% of the DUs in Boston which are equipped with gas ranges violate the NAAQS annual standard for NO_2. Results showed that the contribution of ambient NO_2 to indoor air varied with season, housing structure and geographical location, with effective penetration of outdoor NO_2 ranging from 0.5 to 0.8 of the outdoor level. The presence of gas ranges added, on average, 15 ppb to the indoor concentration of NO_2, whereas continuously-burning pilot lights contributed an additional 10 ppb. The relationship of residential concentrations and presence of continuously-burning pilot lights is borne out across the entire distribution as indicated in Figure 3. For each percentile, the pilot-light contribution remains constant. Additional analyses suggested that the variability in indoor concentrations is caused mainly by differences in individual houses and individual activities, rather than strong geographical differences as differences in ambient concentrations across the metropolitan area were not great. This was evidenced by a relatively small design effect, about 1.5 to 2.0, associated with geographical clustering of homes. Such a finding suggests that characteristics of the home itself as well as the characterization of individuals' activities within the home should produce a useful predictive model of exposure.

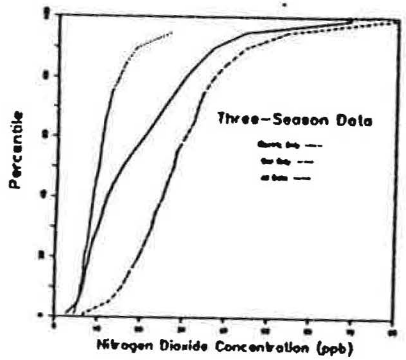

FIGURE 1. CUMULATIVE PROBABILITY DISTRIBUTIONS FOR NITROGEN DIOXIDE CONCENTRATIONS IN BOSTON RESIDENCES.

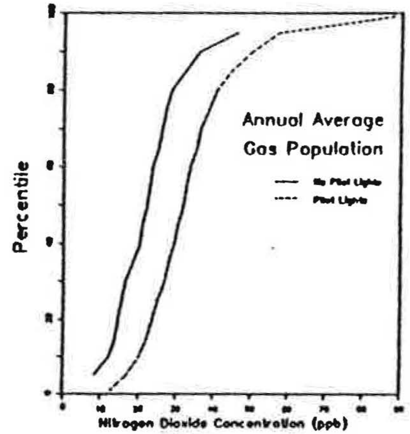

FIGURE 2. CUMULATIVE PROBABILITY DISTRIBUTIONS FOR NITROGEN DIOXIDE CONCENTR
TIONS FOR PILOT LIGHT AND NON-PILOT LIGHT HOMES

Personal Monitoring

Results from the Boston and Los Angeles Personal Monitoring Studies show both similarities a
differences between the two locations. In Boston, the ambient NO_2 concentrations are approximatel
to 30 ppb. Personal exposures are generally lower than indoor concentrations, but, nevertheless, re
the influence of indoor sources. Furthermore, categorization of individuals based upon home conce
tion and cooking and commuting results in highly significant exposure prediction in the winter seaso
In Los Angeles, ambient NO_2 levels range from 30 to 70 ppb. Similar analyses suggest less predicti
power. One may speculate that the higher, and concomitantly more variable, ambient concentratio
coupled with a more mobile population results in less easily categorized exposures.

Boston Personal Monitoring. Results for the Boston Personal Monitoring Study are presented in Ta
1 and 2. Insufficient exposure time experienced in the Winter season required that a modification o
protocol be made for Summer season. The two out of home badges were "combined" such that on
single badge was worn out of home in the Summer season.

As part of the analysis, analysis of variance was performed to assess the validity of the class
tion scheme used in the selection process. For the Winter season (Table 1), one sees a significant
ration of in-home exposures is afforded by the classification scheme. This is not surprising as an
essential component of the classification scheme was the previously-measured in home concentr
Little predictive power is evident for the non-home badges suggesting that information gathered o
muting habits offers little insight into NO_2 exposures. This observation must be tempered by the f
that many badges received exposure indistinguishable from zero owing to the short exposure dur
This resulted in modification of the protocol in the summer period to ensure adequate sample dur

Table 1 Boston Personal Monitoring Study. Winter Badges by Recruitment Classification Scheme

Badge/Class	Mean	St. Dev	Cases	F	Prob.	Eta^2
Day 1, Inhome				16.46	0.000	0.16
High Potential High Indoor	29.2	18.2	62			
Low Potential High Indoor	23.5	16.7	54			
High Potential Low Indoor	17.7	9.2	45			
Low Potential Low Indoor	13.0	9.0	72			
Day 2, Inhome				12.22	0.000	0.14
High Potential High Indoor	28.5	18.1	60			
Low Potential High Indoor	25.0	12.3	48			
High Potential Low Indoor	17.33	11.7	43			
Low Potential Low Indoor	14.80	15.2	72			
Day 1, Nonhome				1.71	0.165	0.02
High Potential High Indoor	29.2	22.0	56			
Low Potential High Indoor	23.3	13.0	51			
High Potential Low Indoor	33.3	32.0	43			
Low Potential Low Indoor	25.3	24.7	65			
Day 2, Nonhome				0.91	0.440	0.01
High Potential High Indoor	29.4	24.5	55			
Low Potential High Indoor	25.7	17.2	46			
High Potential Low Indoor	33.8	22.5	38			
Low Potential Low Indoor	28.0	25.1	67			

Table 2 Boston Personal Monitoring Study. Summer Badges by Revised (Proximate) Classification Scheme

Badge/Class	Mean	St. Dev	Cases	F	Prob.	Eta^2
Day 1, Inhome				0.71	0.547	0.01
High Potential High Indoor	24.7	8.6	26			
Low Potential High Indoor	22.5	9.3	17			
High Potential Low Indoor	19.4	25.6	59			
Low Potential Low Indoor	17.7	26.1	98			
Day 2, Inhome				2.59	0.054	0.04
High Potential High Indoor	24.9	11.1	26			
Low Potential High Indoor	23.2	14.4	16			
High Potential Low Indoor	15.2	10.9	60			
Low Potential Low Indoor	16.7	21.4	100			
Nonhome				7.97	0.000	0.11
High Potential High Indoor	26.4	11.4	26			
Low Potential High Indoor	40.7	39.0	16			
High Potential Low Indoor	21.0	13.2	59			
Low Potential Low Indoor	19.2	13.8	91			

During the Summer season, a different result is found. For Day 1, little predictive power is evident for the in-home exposures. The Day 2 results suggest a marginally significant predictive trend. This contradictory result is mitigated by the presence of outliers in the Day 1 data: one individual with low expected indoor concentrations demonstrate anomalously high exposures for Day 1. The same individual demonstrated exposures more in line with those expected on Day 2. It is interesting to note the Nonhome results during the Summer. Those who were expected to have higher indoor concentrations also experienced elevated personal exposures when out of the home. We speculate that the fact that such results were not found in the Winter season may be due to indoor concentrations acting as a surrogate for outdoor in the Summer. That is, individuals with higher indoor concentrations in the Summer season may be getting a significant penetration of outdoor NO_2. Higher indoor concentrations would then be indicative of higher outdoor concentrations which, in turn would be manifested in higher out-of-home exposures if the individual spent most of his out-of-home time in the same geographic area.

Los Angeles Personal Monitoring. Results for two components of the Los Angeles Personal Monitoring Study are presented in Figures 3 and 4. Figure 3 presents a histogram for exposures experienced by Main Study participants throughout the year. The data have the characteristic right-skewness common to exposure and air pollution data. The summary statistics suggest slightly higher exposures are experienced in Southern California with respect to Boston.

```
Count  Midpoint
  18      4 ******
  71     11 ********************
 130     18 ***********************************
 128     25 **********************************
 106     32 *****************************
  76     39 *********************
  35     46 ***********
  20     53 ******
   9     60 ***
   2     67 **
   1     74 *
   0     81 *
   0     88 *
   0     95 *
   0    102 *
   0    109 *
   1    116 *
          L...+...I...+...I...+...I...+...I...+
          0      40      80     120   140
                   Frequency
```

Mean	27.363	Median	26.000	Mode	17.800
Std Dev	12.915	Skewness	1.026	Kurtosis	3.412
Minimum	3.500	Maximum	116.400	N = 597	

FIGURE 3. LOS ANGELES PERSONAL MONITORING STUDY. HISTOGRAM AND DESCRIPTIVE STATISTICS OF DAY 1 PERSONAL BADGE.

Figure 4 presents data from the Microenvironmental component of the investigation. These data are quite similar to the Main Study data. Summary statistics indicate similar means and medians for the two populations. The larger standard deviation may be due to sampling error as the Main Study sample is substantially larger than the Microenvironmental Study group. Detailed analysis of the data reveals small differences in the population sampled. The differences are not believed to be significant in terms of population based exposures to nitrogen dioxide. Further analysis of the data will afford evaluation of the assumptions intrinsic to the design as well as an evaluation of the variability within individuals across seasons.


```
Count  Midpoint
  62      3 ******************
 149     18 ****************************************
  79     33 **********************
  26     48 *********
  12     63 ****
   6     78 ***
   1     93 *
   3    108 **
   1    123 *
   0    138 *
   0    153 *
   0    168 *
   1    183 *
   0    198 *
   0    213 *
   0    228 *
   1    243 *
          L...+...I...+...I...+...I...+...I...+
          0      40      80     120   140
                   Frequency
```

Mean	26.404	Median	21.700	Mode	21.800
Std Dev	23.219	Skewness	4.165	Kurtosis	29.444
Minimum	2.500	Maximum	244.400	Cases = 343	

FIGURE 4. LOS ANGELES PERSONAL MONITORING MICROENVIRONMENTAL STUDY. HISTOGRAM AND DESCRIPTIVE STATISTICS OF AT-HOME PERSONAL BADGE.

Short-term Exposure Studies

Summary results for the Short-term Exposure Studies are presented in Table 3. These data represent average concentrations and exposures experienced during a series of one-hour cooking events. The whole-house average is a simple mean of the kitchen and bedroom samplers. It is meant to represent the exposure experienced by an individual other than the cook during the cooking event. Inspection of these data allows several conclusions to be drawn. In general, the fixed-location monitor in the kitchen slightly over-predicts the cook's exposure to NO_2. This is true whether the summer or winter season is being monitored or whether the range or the oven is being used. Exposures and concentrations in each location are substantially lower during the summer season when compared with winter. Note the similarity between the mean exposure experienced by the cook and the whole-house average concentration.

The data presented in Figure 5 show the differences in exposure patterns experienced by the cook. Although the cook received a mean exposure of only 81 ppbv for this case, the exposure was actually punctuated by short periods of much higher exposure. Indeed, a maximum, short-term exposure of approximately 600 ppbv was experienced. Observation of individuals during the cooking process has suggested that the cook's short-term exposure profile is strongly influenced by activities which take him or her in and out of the direct plume of the gas appliance. Short bursts of very high concentrations can be experienced.

Table 3. Mean values for continuous nitrogen dioxide samples taken in various locations in residences during one-hour cooking events. Data are grouped by season and appliance use. Average values are weighted according to use patterns, time and location. All values are in ppbv.

	Cook	Kitchen	Other Room	Whole-House Average
Season				
Winter	139.9	145.4	130.9	138.7
Summer	76.5	80.1	69.8	75.3
Seasonal Average	108.0	114.1	100.3	107.4
Gas Appliance Used				
Range	135.8	139.8	123.2	132.8
Oven	67.9	76.9	69.2	69.9
Gas Appliance Average	108.0	114.1	100.3	107.4

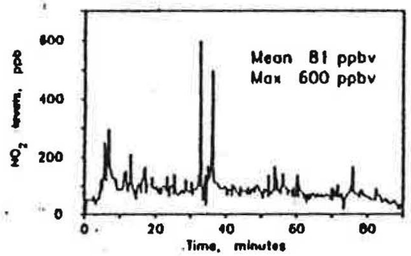

FIGURE 5. CONTINUOUS TRACE OF COOK'S EXPOSURE TO NITROGEN DIOXIDE.

The very short-term average (5 sec - 1 min) maximum personal exposures were uncorrelated with the 30-min averaged exposures. However, correlation was apparent between longer averaging times (e.g. 5-min) and the 30-min results. Thus we conclude that very-short term averaged peak exposures are influenced by the personal interaction with the source. Longer-term averages are influenced by source use, mixing volume and conditions of the room. These results have improved the understanding of the relationship between fixed-site monitors and personal exposures for short-duration activities influencing NO_2 exposure.

SUMMARY

The Harvard-GRI Nitrogen Dioxide Exposure Assessment Study was designed to perform a detailed assessment of personal exposures. The investigations were intended, in part, to investigate the contribution of all sources, both indoors and outdoors, to exposures actually experienced by individuals. Previous studies had indicated that indoor concentrations could be higher than outdoor, at times

exceeding the NAAQS for NO_2. Evaluation of this relationship on a population-based sample was a central theme of these studies. A second objective was to investigate the possibility that human exposures could be modeled from activity patterns and microenvironmental concentration measurements. Of interest as well was the distribution of short-term peaks in concentration and their relationship to the mean concentration.

Essentially all objectives were met in the study. A detailed characterization of residences in the Boston, Massachusetts area was carried out. Characteristics influencing indoor NO_2 concentrations were found. Most notable among these were the presence of a gas range and the presence of continuously-burning pilot lights. Categorization of individuals with respect to their likely exposure to NO_2 was also effected through the use of data collected from the Home Characterization Study and a short personal characteristics questionnaire. Finally, the influence of higher ambient NO_2 concentrations was investigated in the California study. The influence of ambient levels was found to be stronger in the California study in a statistical sense than in Boston. Ongoing work suggests that the influence of gas appliances on human exposure to NO_2 is additive: the exposure experienced indoors is associated with both appliance and on the ambient concentration. The relative contribution from indoor and outdoor sources is site and use specific.

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