#5059

ESTIMATION OF POPULATION EXPOSURE TO NITROGEN DIOXIDE

IRWIN H. BILLICK
Gas Research Institute
8600 West Bryn Mawr Avenue
Chicago, Illinois 60631

INTRODUCTION

The assessment of population risk to exposure of nitrogen dioxide can be estimated by integrating the probability of an adverse health outcome occurring at a given level of exposure with the probability of exposure (Roth 1984). This paper discusses our current ability to estimate the distribution of population exposure to nitrogen dioxide. Another paper in this journal addresses the probability that an adverse health effect will occur at a given level of exposure (Samet 1989).

Exposure to nitrogen dioxide is the result of time varying emissions from a variety of indoor and outdoor sources. For purposes of estimating possible adverse health effect it would be desirable to know the distribution of exposure over both long-term and short-term averaging periods. The short-term (1–3 hour) exposure pattern is characterized by short-intense bursts of nitrogen dioxide above some background level which itself may be varying in time but at a much slower rate. The long-term exposure pattern is an integrated exposure averaged over periods of hours, days and even years. At the present time, little data are available to adequately describe the population distribution or the patterns of short-term NO_2 exposure, but, in recent years a large data base has been generated which provides a great deal of insight into the long-term average NO_2 exposure of different populations. This paper will focus on the state of our knowledge about long-term integrated exposure to NO_2 and the possibility of using these data to estimate population NO_2 exposure in general.

It has been frequently pointed out that individuals spend their time in a variety of different microenvironments with differing levels and patterns of NO_2 and that time-weighted exposure in each must be considered in determining the total exposure (National Research Council 1981). One approach to measuring total exposure is through the use of personal monitors which will reflect the sum of exposures in the different microenvironments (Spengler 1984). In the last few years several large scale personal monitoring studies have been implemented, but, the results from the two largest studies,

1. Address correspondence to: Irwin H. Billick, Gas Research Institute, 8600 West Bryn Mawr Avenue, Chicago, Illinois, 60631.

2. Key words: indoor air quality, microenvironments, nitrogen dioxide, survey data, total exposure.

Toxicology and Industrial Health, 6:2, pp. 325-335

ISSN: 0748-2337

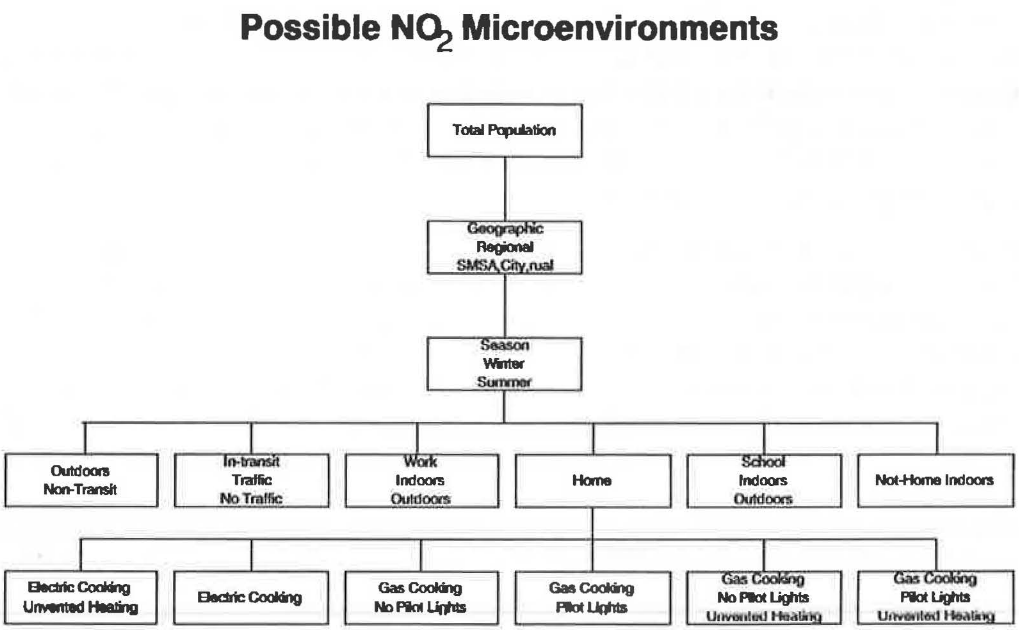
in Boston and Los Angeles, have not been completed (Nitta 1984, Quackenboss 1985, Clausing 1984, Ryan 1987, Schwab 1989). Preliminary examination of the data of these latter two studies indicates that they, along with the earlier studies will provide significant insight into the relative contribution of exposure in specific microenvironments to the total personal exposure (Schwab 1989).

An alternative to direct measurement of personal exposure is to measure the NO_2 levels in differing microenvironments and using these observations, along with time-activity data, to estimate the total exposure (Duan 1982). This approach would use empirical modeling, such as regression analysis, to predict NO_2 levels based upon a few readily available predictor variables.

A third and somewhat similar approach would be to use theoretical models, such as the mass balance model for indoor pollutant levels, for estimating the exposure in various microenvironments and simulation modeling to predict the NO_2 levels in a population. This method requires a knowledge of the distribution of the individual parameters, which may be more readily available, for substitution into the model (Traynor 1987, Billick 1988).

Microenvironments of interest for NO_2 can be defined in numerous ways and by a variety of classifications, for example, geographical, indoors, outdoors, transport, places of employment or school, and the residence. Figure 1 is an example of one such alter-

FIGURE 1. Possible nitrogen dioxide exposure microenvironments.



native classification of microenvironments for the study of the personal exposure to NO_2.

Data on the NO_2 concentrations in the different microenvironments may be available from a number of sources. For example, ambient NO_2 levels can be obtained from EPA through its national monitoring network. Unfortunately, in a given geographic area, a city, for example, relatively few fixed site monitors are used and whether or not these can be used to describe the actual distribution of population ambient exposure is open to question. A possible approach to obtaining the geographical distribution of NO_2 which is representative of population distribution is to develop a network of many cheap integrating NO_2 monitors, such as the Palmes tubes, located throughout the area of interest.

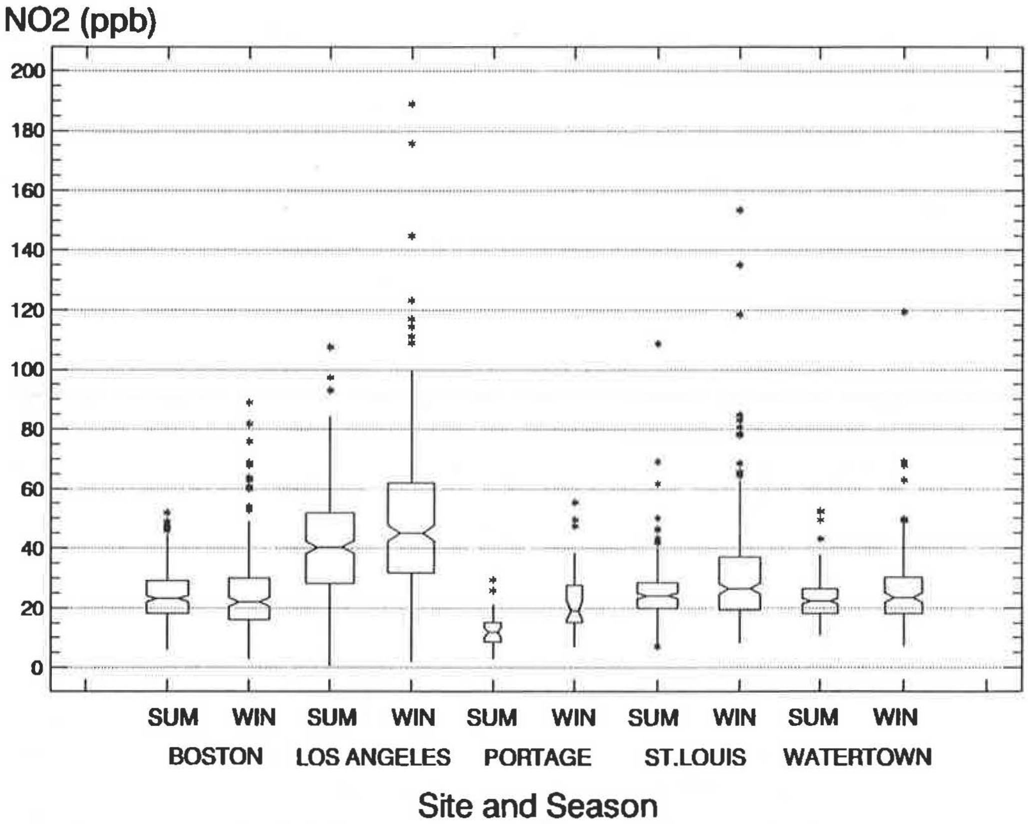
Little information is available about the distribution of NO_2 levels either during transit or employment or school. Much more data are available, however, about the distribution of NO_2 concentrations in the home. As a first approximation, the total NO_2 exposure can be estimated from a time-weighted average of the home NO_2 level and an ambient NO_2 level. Since the time-activity pattern is heavily weighted towards the home, the result may be reasonably acceptable.

FIELD SURVEY ANALYSIS

A large data base on the distribution of NO_2 concentrations in homes along with the factors which may impact these levels, such as outdoor NO_2, air-exchange rates, decay rates and presences of NO_2 sources now is available and is undergoing extensive analysis (Drye 1989). The data from these surveys provide the necessary input for estimating population exposures by either of the last two alternatives mentioned above. The two largest studies, Boston (Ryan 1988) and Los Angeles (Wilson 1986), have observations on about 500 homes each in each location during three different seasons. The survey participants were statistically sampled from the larger SMSA population, but, have been over-sampled from homes with gas ranges, which are an indoor source of NO_2. Additional smaller data bases are currently available from Portage, St. Louis, Watertown (Drye 1989) and soon will be available from Tucson and Albuquerque. Survey data are also available for NO_2 levels in special populations such as the users of unvented gas or kerosene space heaters (Koontz 1988, Leaderer 1984). Analysis of these data sets using several relatively available predictors have yielded several regression models which form the basis for estimating the population distribution of NO_2 levels.

The data for gas cooking homes from Boston, Los Angeles, Portage, St. Louis and Watertown have been combined into a single data set of about 1950 observations covering both winter and summer (Drye 1989). The weekly average NO_2, as measured by Palmes tubes, is available for the kitchen, bedroom and outdoors. Figure 2 is a box-whisker plot of the distribution of NO_2 levels in the bedroom for each site and season

FIGURE 2. Distribution of bedroom NO_2 levels by city and season.



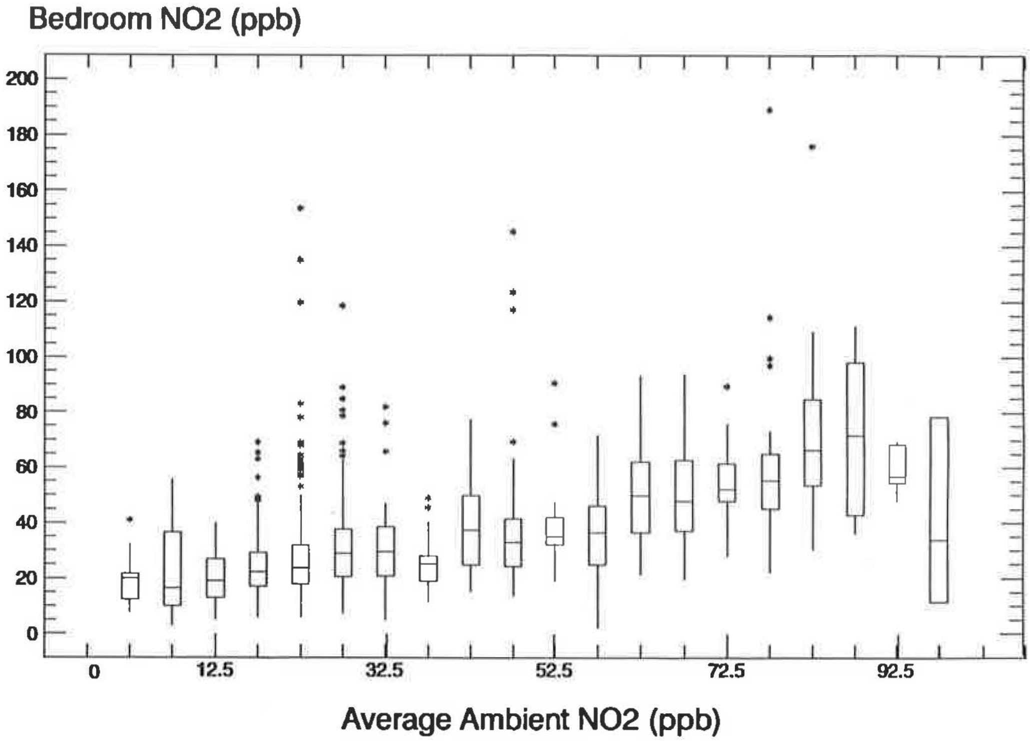
for this data set. The distributions vary between city and between season within a given city. If one categorizes the bedroom NO_2 levels, by ambient NO_2, as shown for the winter season in Figure 3, a distinct dependence of bedroom NO_2 on ambient NO_2 is observed. The difference in distributions between homes with and without pilot lights is apparent in Figure 4, which is also for the winter observations.

Multi-linear regression results indicate that ambient NO_2 concentrations, season, and range pilot lights are significant predictors of bedroom NO_2 levels. The model used in this analysis is shown in equation 1:

$$C_{bed} = b_0 + b_1{*}C_{out} + b_2{*}SEAS + b_3{*}SEAS{*}C_{out} + b_4{*}PILOTS + b_5{*}SITE + b_6{*}SITE{*}C_{out} + e \quad (1)$$

where C_{bed} and C_{out} are the bedroom and outdoor NO_2 concentration, respectively, SEAS is an indicator variable for season (summer = 0, winter = 1), PILOTS is an indicator variable for the presence of gas range pilot lights (absent = 0, present = 1),

FIGURE 3. Distribution of bedroom NO_2 levels by average ambient NO_2 class.

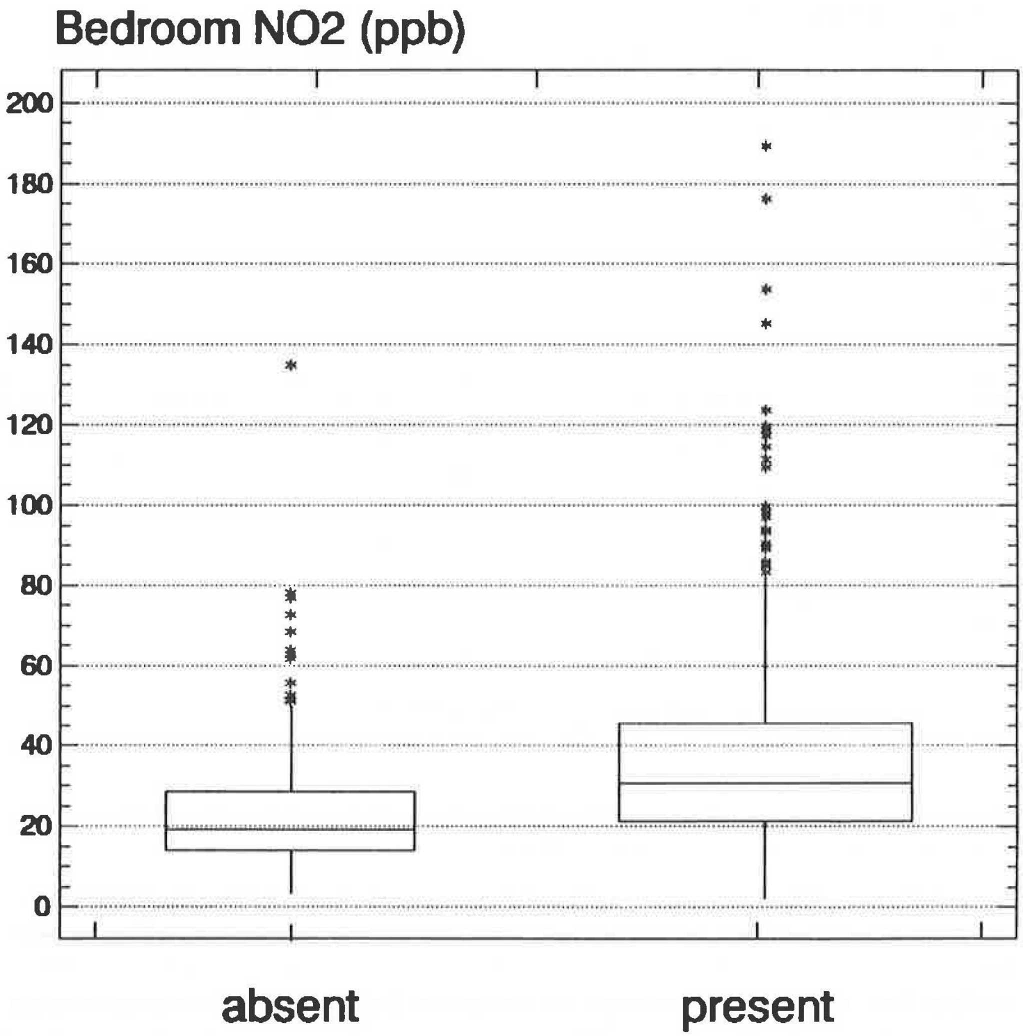


SITE is an indicator variable for survey site (SITE = 0 or 1, depending on the site used in the prediction), and e is the model error term.

Table 1 (Drye 1989) summarizes the regression results. A substantial proportion of the variation in bedroom NO_2 was explained by ambient levels of NO_2 alone ($R^2 = 0.369$). The addition of a sampling location variable further improves the regression model slightly, but, the estimated influence of site on the indoor-outdoor relationship is significantly different from zero in California only. The results indicate that at any given ambient NO_2 level, the predicted bedroom NO_2 levels will be slightly higher in California than in the other sites studied.

While linear regression models allow us to predict the mean NO_2 levels for a population of homes, they do not allow us to estimate directly the distribution of indoor levels for a given set of predictors. An estimate of the distribution of NO_2 from indoor sources for a group of homes can be made based on an assusmption about the shape of the distribution curve, and a predicted or known mean and variance. As an alternative to this approach, a model for predicting the distribution of bedroom levels, for the same independent variables, based on logit analysis has been used. The logit regression analysis uses the model shown in equation 2:

FIGURE 4. Distribution of bedroom NO_2 levels by presence of pilot light.



$$P_j = \exp(b_{0j} + b_{1j}{*}C_{out-m}) \quad / \quad (1 + \exp(b_{0j} + b_{1j}{*}C_{out-m})) \qquad (2)$$

where P_j is the proportion of a population of bedrooms with NO_2 concentrations greater than a threshold level, T_j and an ambient NO_2 level of C_{out-m}.

For NO_2, logit regression analysis was performed separately on summer and winter data, for homes which use gas ranges (Drye 1989). The data was categorized by 5 ppb ranges of ambient NO_2 to and the mid-point of the range was set equal to C_{out-m}. Separate regressions were performed for values of T_j between 10 and 80 ppb. The influence of other predictor variables (such as the presence or absence of range pilots) on model estimates has not yet been investigated; hence, the applicability of the results to other

TABLE 1
Summary of Linear Regression Results (Drye 1989)
(standard error of coefficients in parenthesis)

Model: $C_{bed} = b_0 + b_1{*}c_{out} + b_2{*}SEAS + b_3{*}SEAS{*}C_{out} + b_4{*}PILOTS + b_5{*}SITE + b_6{*}SITE{*}C_{out} + e$

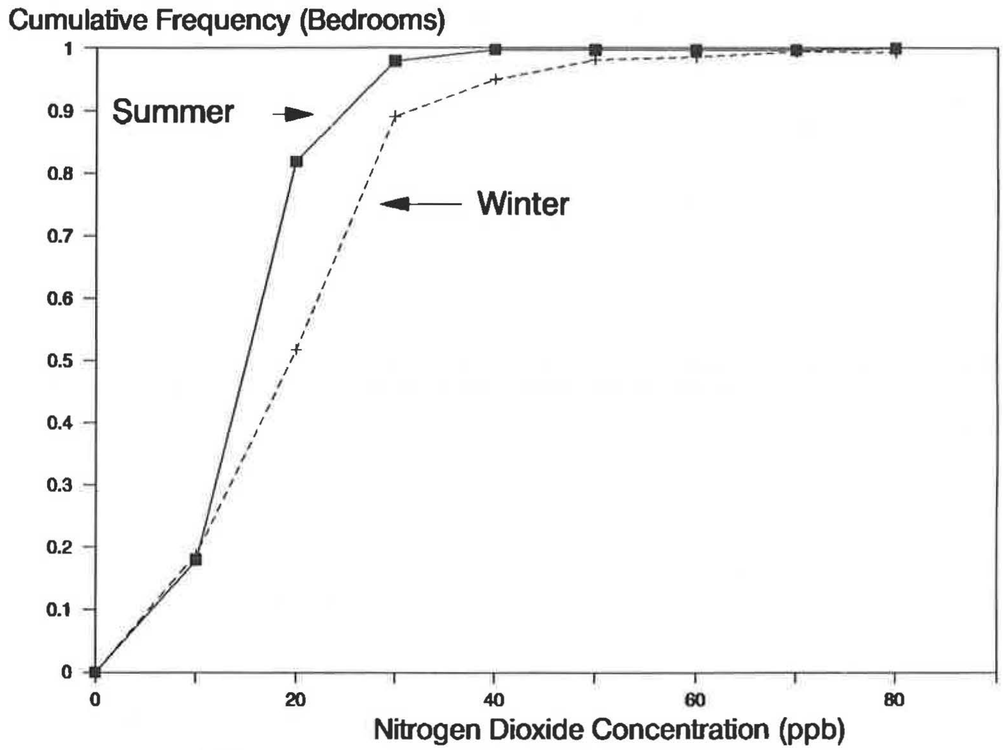
Variable Added	N	R-sq	b_0	b_1	b_2	b_3	b_4	SITE	b_5	b_6	Part F Pr > F
C_{out}	1948	0.369	12.42	0.61							0.0001
			(0.66)	(0.02)							
SEAS	1948	0.371	11.79	0.61	1.6						0.0223
			(0.72)	(0.02)	(0.70)						
C_{out}*SEAS	1948	0.376	8.87	0.71	6.23	−0.15					0.0001
			(1.02)	(0.03)	(1.36)	(0.04)					
PILOTS	1924	0.0401	3.79	0.67	5.97	−0.14	7.68				0.001
			(1.16)	(0.03)	(1.35)	(0.04)	(0.83)				
SITE	1924	0.405	3.97	0.62	6.12	−0.14	7.54	Boston	−0.04*		0.0130
			(1.34)	(0.04)	(1.35)	(0.04)	(0.83)		(0.83)		
								L.A.	3.85		
									(1.07)		
								Portage	1.08*		
									(2.00)		
								St.Lou	1.92*		
									(1.08)		
C_{out}*SITE	1924	0.406	9.85	0.35*	6.23	−0.15	7.53	Boston	−4.42*	0.20*	0.3887
			(4.41)	(0.20)	(1.37)	(0.04)	(0.83)		(4.81)	(0.22)	
								L.A.	−3.22	0.30*	
									(4.68)	(0.20)	
								Portage	−5.91*	(0.43)*	
									(6.21)	(0.61)	
								St.Lou	−0.07*	(0.10)*	
									(5.98)	(0.26)	

* not significant at alpha = 0.05

populations assumes that the population of interest and that represented by the analysis sample are comparable with respect to the predictor variables included in the model.

From the values of b_{0j} it is possible to construct an apparent distribution of bedroom NO_2 concentrations of a population living in an area where the outdoor level of NO_2 is zero which can be interpreted as the distribution of NO_2 produced by the indoor sources alone. Figure 5 illustrates the distribution of indoor source contribution for the winter data in homes with gas ranges. Using the values for the coefficients in equation 2 and population-weighted values for C_{out-m} an estimated distribution of NO_2 levels for other populations with homes with gas ranges can be produced. Figure 6 shows a comparison between the predicted winter bedroom NO_2 distribution using this approach and the actual observations for Watertown, Mass. Possible improvement between the predicted

FIGURE 5. Calculated distribution of indoor NO_2 source contribution.

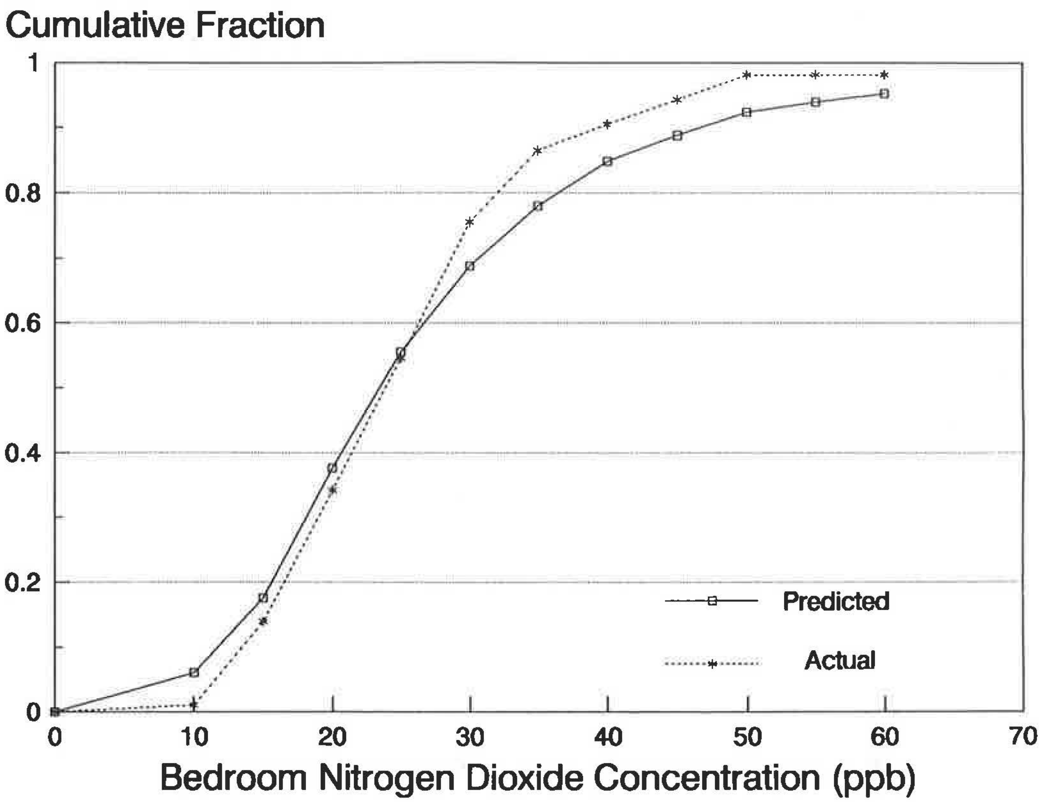


and observed curves might be obtained by employing additional predictors such as the presence or absence of pilot lights.

In order to obtain a distribution of the bedroom NO_2 exposure for the entire population in a given geographic location, it would be necessary to perform similar analyses on homes without gas ranges based on the distribution of ambient NO_2 levels for that population. In addition, the analysis must include the population which may have unusual sources of NO_2 emission, such as unvented fossil fueled space heaters or who may use the gas range or oven as a source of heat.

The model for predicting the NO_2 levels in homes with unvented gas space heaters is different from that with only gas ranges. The regression analyses of the survey data from homes in Texas which use unvented space heaters as their primary source of heat (Koontz 1988) show that the significant predictor for indoor NO_2 is the difference between indoor and outdoor temperatures. Since the latter data are readily available, the calculation of the residential indoor NO_2 distribution for this special population is straight forward (Hemphill 1987).

FIGURE 6. Predicted and observed distribution of bedroom NO_2 levels for Watertown, Mass.



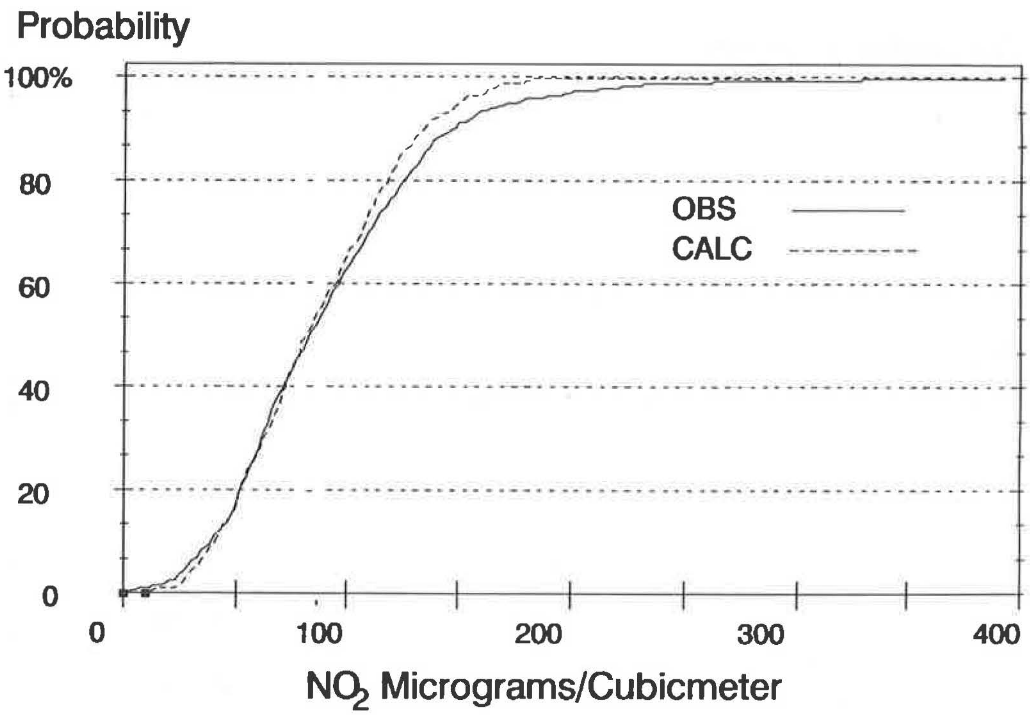
SIMULATION MODELING

Simulation modeling of homes with gas ranges and space heaters also has been carried out using the well known mass balance model

$$f(C_{bed}) = \frac{f(a)*f(C_{out}) + f(S)/f(V)}{f(a) + f(k)} \quad (3)$$

where f(x) is the distribution factor for a given variable. C_{bed} and C_{out} are the bedroom and outdoor concentrations of NO_2, a, the infiltration rate, k, the reactive decay rate, S, the source emission rate, and V, the dwelling unit volume. The distributions for the input terms on the right side of equation 3 are obtained from survey or other published data. Figure 7 is a comparison between the predicted distributions of NO_2 levels using this model and the observed distribution for the Los Angeles area (Billick 1988). The difference between the NO_2 distribution levels using simulation modeling and the observed are small for most of the curve but tend to increase in the high tail. One reason for the possible difference is that the simulation has not included all possible significant sources of NO_2 emissions, such as the use of the gas range and oven for space heating.

FIGURE 7. Observed and calculated distribution of bedroom NO_2 levels for Los Angeles.



DISCUSSION

Several modeling approaches exist which offer promise for estimating the population exposure to nitrogen dioxide in different microenvironments. These models require input which may be readily available data from such sources as the EPA National Sampling Network, Census or DOE residential surveys, weather bureau data and indoor air quality or energy field surveys. When combined with time-activity data it should be possible to calculate a reasonably reliable estimate of the distribution of NO_2 levels in a wide variety of locations and exposure conditions and should be adequate for national exposure estimates needed for policy making or standard setting.

REFERENCES

BILLICK, I.H., BAKER, P.E. and COLOME, S.D. (1988). Simulation modeling of indoor nitrogen dioxide concentrations. Paper presented at the APCA Specialty Conference on Combustion Products, Niagara Falls, NY.

CLAUSING, P., MAK, J.P., SPENGLER, J.D. and LETZ, R. (1984). Personal NO_2 exposure of high school students. Proc. The Third Int. Conf. on Indoor Air Quality and Climate.

DRYE, E.E., OZKAYNAK, H., BURBANK, B., BILLICK, I.H., SPENGLER, J.D., RYAN, P.B., BAKER, P.E. and COLOME, S.D. (1989). Development of models for predicting the distribution of indoor nitrogen dioxide concentrations. JAPCA, to be published.

DUAN, N. (1982). Models for human exposure to air pollution. Environ. Int. **8**:305.

HEMPHILL, C.P., RYAN, P.B., BILLICK, I.H., NAGDA, N., KOONTZ, M.D. and FORTMAN, R.C. (1987). Estimation of nitrogen dioxide concentration. Proc. The Fourth Int. Conf. on Indoor Air Quality and Climate **1**:420.

KOONTZ, M.D., FORTMAN, R.C. and NAGDA, N.L. (1988). A topical report on a filed monitoring study of homes with unvented gas space heaters. Gas Research Institute Report GRI-87/0044, Chicago, IL.

LEADERER, B.P., ZAGRANISKI, R.T., HOLFORD, T.R., BERWICK, M. and STOLWIJK, J.A.J. (1984). Multivariate model for predicting NO_2 levels in residences based upon source and source use. Proc. The Third Int. Conf. on Indoor Air Quality and Climate **4**:43.

NITTA, H. and MAEDA, K. (1982). Personal exposure to nitrogen dioxide. Environ. Int. **8**:243.

NRC (1981). Indoor Pollutants. Committee on Indoor Pollutants, National Research Council, National Academy Press, Washington DC.

QUACKENBOSS, J.J., SPENGLER, J.D., KANAREK, M.S., LETZ, R. and DUFFY, C.P. (1985). Personal exposure to nitrogen dioxide: relationship to indoor/outdoor air quality and activity patterns. Environ. Sci. Technol. **20**:775.

ROTH, H.D., BILLICK, I.H. and JOHNSON, D.O. (1984). Comparative risk assessment of unvented gas appliances. Proc. AGA Oper. Sec. 484.

RYAN, P.B., SPENGLER, J.D., TREITMAN, R.D., SOCZEK, M.L., YANAGISAWA, Y., WALLACE, L.A. and BILLICK, I.H. (1987). Nitrogen dioxide exposure assessment in greater Boston: Evaluation of personal monitoring. Proc. The Fourth Int. Conf. on Indoor Air Quality and Climate **1**:493.

RYAN, P.B., SOCZEK, M.L., TREITMAN, R.D., SPENGLER, J.D. and BILLICK, I.H. (1988). The Boston residential NO_2 characterization study II. Atm. Environ. **22**:1010.

SAMET, J.M. and UTELL, M.J. (1989). The risk of nitrogen dioxide: What have we learned from epidemiological and clinical studies? (reference to this journal).

SCHWAB, M., SPENGLER, J.D., RYAN, P.B., COLOME, S.D., WILSON, A.L., BECKER, E. and BILLICK, I.H. (1989). Describing activity patterns for use in exposure assessment: Data from the Los Angeles personal monitoring study. Paper at the APCA 82nd annual meeting. Anaheim, Ca.

SPENGLER, J.D. and SOCZEK, M.L. (1984). Evidence for improved ambient air quality and the need for personal exposure research. Environ. Sci. Technol. **18**:268A.

TRAYNOR, G.W., ACETI, J.C., APTE, M.G., SMITH, B.V., GREEN, L.L., SMITH-REISER, A., NOVAK, K.M. and MOSES, D.O. (1987). Macromodel for assessing indoor exposures to combustion-generated pollutants. Proc. The Fourth Int. Conf. on Indoor Air Quality and Climate **1**:273.

WILSON, A.L., COLOME, S.D., BAKER, P.E. and BECKER, E.W. (1986). Residential indoor air quality characterization study of nitrogen dioxide: Phase I Final Report. Southern California Gas Company, Los Angeles.