The Influence of Human and Environmental Exposure Factors on Personal NO_2 Exposures

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Abstract

The US Environmental Protection Agency's (US EPA) Detroit Exposure and Aerosol Research Study (DEARS) deployed a total of over 2000 nitrogen dioxide NO_2 passive monitors during three years of field data collections. These 24-hr based personal, residential outdoor, and community-based measurements allowed for the investigation of NO_2 spatial, temporal, human and environmental factors. The relationships between personal exposures to NO_2 and the factors that influence the relationship with community-based measurements were of interest. Survey data from 136 participants were integrated with exposure findings to allow for mixed model effect analyses. Ultimately, 50 individual factors were selected for examination. NO_2 analyses revealed that season, exposure to environmental tobacco smoke, and residential gas appliances were strong influencing factors. Only modest associations between community-based measures of nitrogen dioxide and personal exposures impacted by various exposure factors for heating ($r = 0.44$) or non-heating seasons ($r = 0.34$) were observed indicating that use of ambient-based monitoring as a surrogate of personal exposure might result in sizeable exposure misclassification.

Introduction

Allowable community-based concentrations of select air pollutants, including nitrogen dioxide (NO_2) are regulated by the U.S. Environmental Protection Agency (EPA) under the Clean Air Act (CAA, 2004). National Ambient Air Quality Standards (NAAQS) have been established and are required to undergo review on a regular basis. The EPA recently completed an integrated science assessment for the oxides of nitrogen (EPA, 2008). The NAAQS represents community-based air concentration limits believed to provide an adequate margin of safety for those most susceptible to the effects of these pollutants. However, the monitoring for these pollutants is often limited to a small number of measurement locations in a given demographic area (EPA, 2008). Likewise, humans are involved in various time activities and rarely do they spend time either in close proximity to these compliance monitors or even in outdoor settings that might be close approximations to where these monitors are located (Klepis et al 2001; Williams 2000, Williams 2003).

A number of studies have examined the relationship between community-based measures of NO_2 and personal exposures (Alm et al., 1998; Brauer et al., 1989; Sarnat et al., 2001). These studies have focused on convenience populations of specific age groups or some susceptible subpopulation (e.g., children, elderly). These study populations were not recruited with a specific spatial intent in mind as part of their study design. These studies have shown highly variable degrees of agreement between personal and ambient-based measures. Human time activity patterns, environmental factors, residential combustion sources, as well as reactivity of some of these gases themselves in indoor environments, are a number of the parameters that have been reported as influencing these associations.

The EPA's Detroit Exposure and Aerosol Research Study (DEARS) was designed to provide information as to the magnitude and variability of central community site (ambient) measurements as compared to residential and personal-based exposure monitoring for particulate matter (PM) and select air toxics and gases (Williams, 2005; Williams et al., 2009). In doing so, a randomized household recruitment strategy was employed so that participants living in predefined spatial locations would participate. Spatiality of the resulting ambient to personal measurements would be assessed to better define the factors influencing this relationship. A number of exposure or linked epidemiological summaries of DEARS findings have been reported (Baxter et al., 2008; Brook et al., 2010; Rodes et al., 2010).

A separate article being developed will describe multi-pollutant relationship observations of particulate matter (PM), NO_2, and other gases. The current article focuses on the impact of various human and environmental exposure factors on total personal exposures to NO_2. This includes the influence of environmental tobacco smoke (ETS). Models describing these impacts on personal exposures are reported.

Study design

The DEARS study design has been reported in depth (Williams et al., 2009) and is available at the study website (www.epa.gov/dears). A total of 136 non-smoking adults from neighborhoods in Wayne County, Michigan participated in the monitoring over a three year period (2004-2007). We provide a full description of the participant selection process and recruitment/retention and demographics statistics elsewhere (Phillips et al., 2010). Participants had to be ambulatory, able to read or understand English or Spanish and plan on staying in their existing housing through consecutive summer (June-August) and winter (January-March) monitoring periods. They were asked to consent to personal exposure monitoring. More than 70% of those enrolled participated in both seasons allowing seasonal impacts on their exposures to be examined.

Participants were predominately female (77%) with the total cohort representing 51%

African-American, 28% White and 21% other. The median age was 41 years and 67 % were not engaged in work outside the home. All of the participants lived in single-family detached homes. In addition to personal monitoring, a central community monitoring location (ambient) was established at a State of Michigan regulatory air quality platform in Allen Park, MI. We have previously reported on the spatial characteristics of the neighborhoods where the participants lived and their relationship to the ambient monitor (Williams et al., 2009).

Methods

Environmental Monitoring

Ogawa (Pompano Beach, FL) passive dosimeters were deployed for the 24-hr daily collection of NO_2 air mass pollutant concentrations. Specific procedures have been reported (Williams et al., 2009). These devices have been used extensively in other EPA studies and details of their use and validation reported earlier (Williams et al., 2003; Murkerjee et al., 2004, 2009).

NO_2 monitors were deployed each day (Tuesday through Sunday morning) on a consistent 9 am to 9 am schedule (24 ± 2.5 hrs) for a maximum of 5 days per participant per season. Personal monitoring was achieved using a nylon support vest (Williams et al., 2003; Williams et al., 2009) with the monitor affixed in the breathing zone. Ambient monitoring was performed with the monitor affixed to a weather-shielded stand. Method detection limits (MDLs) of 5.1 ppb were established. MDLs represented a three-fold value of the average standard deviation observed in field blank concentrations. More than 99% of all personal and ambient NO_2 measures had environmental concentrations at or above the MDL with replicate precision error of 12.5%.

Estimation of environmental tobacco smoke (ETS) was performed using an active personal $PM_{2.5}$ filter-based monitor collocated with the NO_2 monitor. ETS estimations were performed using procedures defined by Lawless et al. (2004). Even though the DEARS was a non-smoking cohort, we have reported on the significant contribution ETS had on total personal $PM_{2.5}$ exposures (Williams et al., 2009., Brook et al., 2010; Phillips et al., 2010; Rodes et al., 2010). A contribution of $\geq$ 1.5 $\mu g/m^3$ ETS per $PM_{2.5}$ filter sample was deemed to be indicative of significant ETS exposures during any sampling period. The criteria for this mass concentration selection value have previously been reported (Brook et al., 2010; Rodes et al., 2010).

Daily air exchange rates for each home was performed using perfluorocarbon tracer sources and receptor tubes (Williams et al., 2009). The techniques used to perform the monitoring were those reported by Dietz and Cote (1982) with the Brookhaven National Laboratory (Upton, MY) providing all resources and sample analyses.

Survey questionnaires

A major component of the DEARS study design was the collection of detailed time activity pattern data and survey information needed to investigate human and environmental exposure factors. A total of five such data collection instruments were used and are publically available (www.epa.gov/dears). Williams et al. (2009) have explained the use of each instrument. Nearly 50 ordinal or categorical inputs associated with either the environmental monitoring data or the survey instruments were incorporated into the data analysis. These are presented in Supplemental Table S1 and represent potential NO_2 sources the participants might have encountered or factors that might impact such sources contributing to total personal exposures. Variables include residential air exchange, ambient NO_2 mass concentrations, and use of fans or specific gas appliances in the home, among others. Limited exposure factor information pertaining to non-home situations was obtained. This included air quality issues the participants might have encountered while visiting local schools, stores and restaurants among other locations, as well as that involved in commuting.

Statistical analyses

Univariate analyses have been described elsewhere (Williams et al., 2009). The impact of ETS as a non-ambient NO_2 source was investigated by determining the distribution of the sample population with concentration values above the ETS screening level. This allowed for ETS-impacted and non-ETS impacted personal monitoring periods to be established. ETS was observed to be a significant factor upon personal winter time NO_2 concentrations ($P < 0.05$). Therefore, personal NO_2 data from this season associated with monitoring days on which specific individuals had ETS exposures above the screening level were excluded from model runs attempting to attribute ambient source contributions to total personal exposures. Of the 50 exposure factors examined, 26 were determined to have sufficient responses (sample size) for testing. This number was further reduced to a total of 13 variables based upon t-tests associated with seasonal association. Mixed models were then employed to account for repeated measures. Backwards elimination of factors was then performed until the smallest number of variables associated with the highest degree of overall accountability in variability in personal exposures was obtained. General linear models were also performed using a reduced set of variables to investigate any improvement in ambient measures predicting personal exposure variability. All statistical analyses were performed using SAS, version 9.1, Cary, NC. Statistical comparisons reported here had to have P values ≤ 0.05 to be significant.

Results

Personal and ambient NO_2 concentrations and home air exchange rates are presented in Table 1. Mean personal winter measurements (35.6 ppb) were 40% higher than the average community-based winter measures (25.5 ppb) and were statistically different. Personal and other exposure factors would therefore appear to be influencing total exposures to this pollutant during colder weather. Residential air exchange rate was an important factor. Mean air exchange rates were vastly different by season (summer = 1.9 ± 1.9; winter 1.1 ± 0.7).

Some of the seasonal differences in total exposures might be explained in part by ETS. Even though the cohort was required to be non-smoking, a small number of the participants were non-compliant and ultimately had to be removed from the study (Phillips et al., 2010). In addition, participants had no ability to stop others from smoking in their presence or prohibited from visiting locations away from the home where smoking was permitted. Table 2 reports the potential impact of ETS on personal measures by season. There was a very strong association ($p < 0.01$) of personal NO_2 exposure concentrations when ETS above the 1.5 $\mu g/m^3$ screening limit occurred. Only measurements performed during the winter seasons revealed this pattern. Mean non-ETS impacted winter levels of personal NO_2 exposures were 26.8 ppb while ETS impacted exposures for this season averaged nearly 50 ppb. Summer personal comparisons averaged approximately 25 ppb regardless of ETS exposures. The higher ETS contributions to total NO_2 exposures in winter are believed to be explained by the seasonal variations in residential air exchange rates reported in Table 1. Few participants employed central air conditioning during the summer and natural (window) ventilation was often observed. Any ETS released indoors in the winter had a far greater capacity to result in environmental concentrations of significance. Activity patterns previously reported (Rodes et al., 2010) indicated participants spent an average 6.1% more time indoors in the winter as compared to the summer.

The impact of various exposure factors are presented in Table 3 and categorized by season. Established ETS-impacted data were removed prior to the analysis. Listed variables represent a reduced version of those described earlier in Table S1. This truncation was the result of determining which variables had a sufficient sampling population to permit a valid examination of effects. It is interesting to note that participants who thought they were exposed to ETS in the winter (but had measured ETS levels below the screening threshold) still had NO_2 exposure concentrations higher ($P= 0.05$) than those who did not respond positively to this question.

The t-tests reported in Table 3 resulted in 13 variables ultimately selected for inclusion into mixed modeling. Participants indicating they employed home air conditioning of any type had on average a 28.1 ppb daily personal exposure during the summer. The use of a gas space heater in the home during the winter was observed to influence daily personal NO_2 exposures by 59.6 ± 46.6 ppb. In fact, a number of gas appliance-related survey questions appeared to potentially represent significant exposure risk estimates during the winter. Having so many survey findings related to a common exposure source (gas combustion products) resulted in some difficulty in ultimately reducing the mixed model inputs to a satisfactory number as described below.

Data in Table S2 provide mixed model effect estimates for personal exposure to NO_2 by monitoring season. A number of fixed effects were employed in the analyses such as participant, season and date. Use of a dryer pilot light was a component of the full model but dropped as part of the reduction effort. Two remaining variables reflected significant exposure effects during summer monitoring following stepwise eliminations. Selection of gas as a cooking fuel resulted in an average increase of approximately 9 ppb additional exposure each day. Likewise, use of window air conditioning units appeared to result in an increase of nearly 6 ppb per operating unit. Since such units do not generate NO_2 themselves, this effect must be related to some general air exchange taking place in the home and thus bringing in outdoor sources of NO_2. This result is somewhat contradicted though as residential air exchange in itself was observed to have a modest but positive effect (lowering) of one's personal daily NO_2 exposure (-0.8 ppb/air exchange rate).

Mixed modeling results for the winter seasons resulted in only one significant ($p = 0.0112$) effect following the stepwise elimination process. Both use of a dryer pilot light and kitchen fans were eliminated. Participants using a forced air gas furnace would appear to reap a significant positive benefit relative to one's personal exposure (-16.1 ppb). One would assume that this variable is acting as a surrogate of residential air exchange in some manner by replacing NO_2 tainted indoor air from any number of sources and replacing it with fresher outdoor air, thereby reducing residential indoor source strengths. However, air exchange itself was not determined to be a significant effect during the winter season.

General linear models (GLM) were then applied to the reduced data sets for both seasons In such modeling, we performed naïve analyses (i.e., ignoring potential correlations between repeated measures) using the SAS GLM procedure to generate a traditional coefficient of variation (r^2). Coefficients ($r^2 = 0.19, 0.11$) were established for the summer and winter seasons, respectively. Because a number of questions remained following this round of modeling, additional models (mixed or GLM) were applied to investigate the impact of gas-related survey findings upon personal NO_2 exposures and variance components on the total (non-season categorized data). Select findings from these models are highlighted in Table 4 and defined by model run number. These efforts attempted to constrain the analyses so that data relating to personal exposures from significant home-related NO_2 sources might be removed from the analysis. This would theoretically provide the greatest opportunity to determine the true impact of ambient-based NO_2 concentrations on personal exposures.

The first of these six runs incorporated effects associated with ambient NO_2, air exchange, use of dryer pilot light, operation of home air conditioning, gas as a cooking fuel, heating with a forced gas furnace and use of window AC units. Elimination of the non-significant dryer pilot light variable in the next run (#2) resulted in minimal improvement. Once again, use of a gas furnace had the greatest positive impact upon personal exposures (-19.3 ppb; $p = 0.0031$), while use of gas as a cooking fuel resulted in increased personal exposures (13.6 ppb; $p=0.0357$).

Elimination of the residential gas source variables described immediately above in the next mixed model run provided no improvement relative to ambient NO_2 being a significant predictor of personal exposure ($p = 0.7832$). Model runs # 4 and 5 (GLM) ignored repeated measures using the variables defined earlier in run #2. An r^2 of 0.08 resulted from this exercise in run #4. Model #5 also ignored repeated measures but without the inclusion of heat source variables as potential source variables. Such a run excluded exposure data from which participants had provided a positive response to all residential gas-related source effect questions. Such a dataset would have theoretically increased the potential for an improved association between personal and ambient based measures. No such improvement was observed with an even poorer agreement ($r^2 = 0.012$) between matched personal and ambient measurements obtained. This indicates that even though both of the GLM runs resulted in an extremely poor fit, heat source variables would appear to account for most of the variability in personal NO_2 exposures.

Discussion

DEARS personal and community-based (ambient) daily NO_2 measures were successfully measured across all spatial settings and seasons using a passive monitor with a relatively short time integral (24 hr). Data were consistent with other recent human exposure studies in regard to the wide range in concentration and spatial variability observed in other personal and ambient-based settings in the U.S. and Europe for individuals not overly exposed to automotive emissions (Spengler et al., 1994; Alm et al., 1998; Harrison et al., 2002; Nerriere et al., 2005; Brown et al., 2009).

Personal NO_2 exposure distribution for the DEARS would indicate the 50th percentile being well below the current national ambient standard. With rare exception, individuals experiencing elevated daily exposures to NO_2 (in one instance 474 ppb), had identifiable sources potentially linked to indoor gas combustion appliances via questionnaire data.

ETS exposures had a significant impact on NO_2 exposures during winter measurements. Increased NO_2 exposures in populations of smokers or those exposed to ETS have been reported elsewhere (Alm et al., 1998; Harrison et al., 2002). This complication compounds the difficulty in trying to accurately assess the relationship of ambient NO_2 on personal exposures in the general population. It would appear that the presence of ETS needs to be carefully screened for in human panel studies for accurate spatial relationships to be investigated.

Mixed modeling revealed that pinpointing specific exposure factors (beyond ETS exposures) would be problematic in this population and sometimes contradictory. While use of gas as a cooking fuel was significant for the summer seasons relative to an increase in personal exposures, use of a forced air gas furnace actually had a positive (lowering effect) upon participants during the winter. Refined GLM and mixed models attempting to consolidate or exclude a number of these factors to simplify the analyses yielded little additional insight. At best, participants having a higher extent of exposure to various gas combustion-related factors had more of their overall personal exposure variability explained. A number of other studies have also attempted to determine the factors influencing personal exposures to NO_2. Spengler et al. (1994) reported from a Los Angeles basin study that persons living in homes with or without gas as a heating fuel, gas water heaters or gas clothes dryers were statistically equivalent. Homes with gas appliances have been linked to potentially higher personal exposures in the Boston area (Brown et al., 2009). Results from the large Genotox ER study reported that personal exposures were higher in individuals using a gas stove but that inclusion of this factor did not significantly improve the overall understanding of personal versus ambient relationship (Nerriere et al., 2005).

Piechocki-Minguy et al. (2005) reported that a major component of personal NO_2 exposure variability could be explained using factors related to mobile emissions (transportation survey questions). A specific question related to travel by auto in the DEARS did not yield data supporting such a finding.

Considering that the DEARS was designed specifically to help address issues related to how ambient sources impact personal exposures and that more than a thousand source-based questionnaires were collected in support of this objective, it is surprising that a better understanding in personal variability could not be obtained. Residential gas-related factors did help explain a minor component of the overall variability. Ambient-based measures as a surrogate for true personal exposures were determined to be an extremely poor predictor regardless of season. Even when exposure data potentially impacted by ETS and residential gas appliance use were removed from the analysis, the resulting models showed little overall improvement. Other studies have been more successful in associating ambient-based measures with personal exposures, especially those that have incorporated time activity diary input into the models (Kousa et al., 2001; Talar et al., 2009). It is apparent that studies even more focused than the DEARS need to be designed and performed concerning this environmental pollutant.

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Table 1. Summer and Winter Seasons Pollutant Descriptive Statistics.

Variable	N	Mean	STD	Min	25^{th} PCT	50 PCT	75 PCT	95^{th} PCT	Max
Summer									
Ambient NO_2	759	23.7	11.6	8.0	17.0	22.0	27.0	40.0	100.0
Personal NO_2	508	25.5	20.3	3.5	17.0	23.0	29.9	45.0	298.7
Air exchange	492	1.9	1.9	0.1	0.5	1.3	2.5	5.3	17.0
Winter									
Ambient NO_2	653	23.9	8.6	3.5	19.0	24.0	28.0	41.0	48.0
Personal NO_2	401	35.6	50.6	3.5	17.0	24.0	36.0	83.0	474.0
Air exchange	424	1.1	0.7	0.2	0.6	0.9	1.4	2.7	3.7

Table 2. Effect of Environmental Tobacco Smoke (ETS) on Personal NO_2 Exposures

	Variable	**ETS (<1.5)**	**ETS ≥ 1.5**	**P value**
Summer	NO_2	25.2 ±18.0	25.9 ± 25.7	0.75
Winter	NO_2	26.8 ± 20.4	49.3 ± 74.9	<0.01

NO_2 exposure concentration values in units of ppb. ETS concentrations of ≥ 1.5 µg/ used to challenge personal gas exposures for effect.

Table 3. Impact of Select Factors on Personal NO_2 Exposures

	Yes	No	P value
Summer			
Air conditioning	28.1±26.9	23.4±9.2	0.08
Candles burned	25.8±9.2	25.1±20.3	0.86
Travel by auto	23.4±26.9	29.8±30.4	0.03
Central AC in home	28.1±15.8	23.4±8.7	<0.01
Doors opened	23.5±10.2	27.2±32.9	0.17
ETS exposure believed to have occurred	20.3±9.9	26.1±26.1	0.22
Cooking fuel type	20.5±33.1	26.1±20.4	0.07
Furnace pilot light	23.1±21.8	27.3±23.3	0.09
Use of gas fireplace	10.4±3.4	23.4±23.7	0.04
Use of gas appliance	24.3±19.6	28.3±37.5	0.24
Window AC used	26.5±21.3	19.9±9.9	<0.01
Windows opened	24.6±26.9	26.7±21.6	0.47
Kitchen exhaust fan operated	25.2±28.5	24.9±20.3	0.84
Winter			
Central AC in home	20.5±14.0	28.3±19.9	<0.01
ETS exposure believed to have occurred	34.3±20.7	26.5±21.0	0.05
Cooking fuel type	29.3±26.9	17.8±6.9	<0.01
Furnace pilot light	25.5±13.4	31.1±29.9	0.03
Use of gas fireplace	10.5±1.73	28.3±21.6	<0.01
Use of gas appliance	29.2±23.6	22.1±9.3	<0.01
Use of gas space heater	59.6±46.6	25.3±16.2	<0.01
Kitchen exhaust fan operated	21.7±9.5	32.4±26.5	<0.01

Values represent ppb concentrations and their standard deviations. T-test P values at the 0.05 level indicate significant concentration differences between the yes/no response groups.

Table 4. Final Mixed and General Linear Models

Model	Effect	Estimate	Std Error	DF	t value	Pr >| t |
1	Intercept	50.3852	9.1650	105	5.50	<0.0001
	Ambient NO_2	-0.06399	0.1052	419	-0.61	0.5432
	Home air exchange	-2.4808	0.8951	419	-2.77	0.0058
	Dryer pilot light	-3.2426	4.5342	419	-0.72	0.4749
	Air conditioning	-8.3337	3.3837	418	-2.46	0.0142
	Cooking fuel type	13.8680	7.2333	418	1.92	0.0559
	Forced air gas furnace	-23.1700	7.3684	418	-3.14	0.0018
	Use of window AC	-6.7595	4.6282	418	-1.46	0.1449
	Full mixed model CS=988.90; ICC = 0.67; r^2= 0.12					
2	Intercept	41.3105	7.7830	121	5.31	<0.0001
	Ambient NO_2	-0.04436	0.09752	500	-0.45	0.6494
	Home air exchange	-2.6173	0.8242	500	-3.18	0.0016
	Air Conditioning	-7.6253	2.9093	500	-2.62	0.0090
	Cooking fuel type	13.6172	6.4659	500	2.11	0.0357
	Forced air gas furnace	-19.3218	6.5128	500	-2.97	0.0032
	Reduced mixed model CS= 860.23; ICC = 0.65; r^2=0.08					
3	Intercept	24.0974	5.2364	5	4.60	<0.0058
	Ambient NO_2	-0.03960	0.1422	22	-0.28	0.7832
	Home air exchange	-2.506	1.5921	22	-1.58	0.1276
	Air Conditioning	1.8196	3.3711	22	0.54	0.5948
	Reduced mixed model w/o heating source variable CS= 71.80; r^2= 0.13					
4	GLM (ignoring repeated measures) r^2= 0.0815					
5	GLM (ignoring repeated measures w/o heat source variable) r^2= 0.0129					