



Mobile Monitoring of Personal NO_x Exposures during Scripted Daily Activities in Chicago, IL

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ABSTRACT

Elevated ambient concentrations of nitrogen oxides (NO_x), including nitric oxide (NO) and nitrogen dioxide (NO₂), are associated with a wide range of adverse human health effects. Most studies have investigated these associations using ambient NO₂/NO_x measurements from fixed-site monitors or modeled ambient NO₂/NO_x concentrations. However, the majority of personal exposures to NO₂/NO_x occur in a variety of different microenvironments in which people spend most of their time. Previous studies have reported widely varying correlations between personal exposures and ambient NO₂/NO_x concentrations over various timescales. To add to the knowledge base of how personal NO/NO₂/NO_x exposures vary spatially, temporally, and within different microenvironments in an urban environment, we conducted roll-around mobile monitoring of NO/NO₂/NO_x with 1-minute resolution during 14 days of scripted activities in and around Chicago, IL. Activities involved time spent in three primary microenvironments: outdoors, indoors inside various building types, and in multiple modes of transportation including walking, personal vehicle, and public transit. Measurement were conducted at a higher time resolution than most prior microenvironmental monitoring studies using a recently developed direct UV absorbance NO/NO₂/NO_x monitor that is designed to minimize interferences that have been observed in some field campaigns using chemiluminescence monitors. The individual microenvironmental categories with the highest median NO_x concentrations included four indoor environments and a variety of public transit environments. The individual transportation microenvironments with the highest median NO_x concentrations were found aboard regional trains, largely driven by high NO from diesel locomotives. Correlations between microenvironmental NO/NO₂/NO_x measurements and simultaneous records from the nearest ambient monitor were extremely low, with coefficients of determination below 0.05 for each NO_x constituent. These data further illustrate the limitations of relying on ambient site regulatory monitors to characterize personal NO/NO₂/NO_x exposures and provide further evidence that personal monitoring is critical for accurately assessing personal exposure to NO_x.

Keywords: Nitrogen oxides; Human exposure; Personal exposure; Mobile samplers; Indoor air pollution.

INTRODUCTION

Elevated ambient concentrations of nitrogen oxides (NO_x), including nitric oxide (NO) and nitrogen dioxide (NO₂), have been associated with a wide range of adverse human health effects including respiratory effects, cardiovascular effects, lung cancer, and mortality (U.S. EPA, 2016). Although most associations with adverse health effects have been made using measurements from fixed-site ambient NO₂/NO_x monitors or modeled ambient NO₂/NO_x concentrations, personal exposures to NO₂/NO_x are more complex, particularly in urban microenvironments that have

a variety of NO₂/NO_x sources. First, because motor vehicle emissions are the single largest contributor to NO₂/NO_x concentrations in ambient air in the U.S. and traffic patterns are highly variable, there are typically high spatial and temporal gradients in ambient NO₂/NO_x concentrations that vary with the distance from central site monitors (Henderson *et al.*, 2007; Novotny *et al.*, 2011; Montagne *et al.*, 2013). Second, people spend most of their time in microenvironments other than outdoors, including inside homes, offices, restaurants, and vehicles (Klepeis *et al.*, 2001), all of which can have varying fractions of ambient NO₂/NO_x that infiltrates and persists (Dimitroulopoulou *et al.*, 2001; Zota *et al.*, 2005; Fabian *et al.*, 2012). Third, there are many indoor sources of NO₂/NO_x in the various microenvironments in which people spend most of their time, including cooking and space-heating using natural gas and other fuels (Yang *et al.*, 2004; Kornartit *et al.*, 2010; Logue *et al.*, 2014). The combination of these effects leads

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to indoor NO_2/NO_x exposures that are often higher than outdoors (Baxter *et al.*, 2007a, b) and personal exposures that are influenced by exposures in a number of different microenvironments (Lee *et al.*, 2000). These issues also complicate our ability to perform accurate personal exposure assessments for ambient $\text{NO}/\text{NO}_2/\text{NO}_x$, particularly on a short-term basis.

Previous studies have reported widely varying correlations between personal or microenvironmental exposures and ambient NO_2/NO_x concentrations, typically increasing with sampling duration. Some of these studies have shown moderate correlations between personal and ambient and/or indoor NO_2/NO_x concentrations for some populations (Sørensen *et al.*, 2005; Meng *et al.*, 2012b), while others have shown almost no correlation (Quackenboss *et al.*, 1986; Kousa *et al.*, 2001; Lai *et al.*, 2004; Meng *et al.*, 2012a). Many previous personal or microenvironmental NO_2/NO_x studies have been limited to long sampling intervals using passive integrated samplers and most have focused on either NO_2 or total NO_x (Esplugues *et al.*, 2010; Borge *et al.*, 2016; Xu *et al.*, 2016), which limits understanding of important spatiotemporal variations in personal exposures to $\text{NO}/\text{NO}_2/\text{NO}_x$ that could affect short-term health effects and elucidate contributions from various sources. Moreover, most field campaigns that have made microenvironmental $\text{NO}/\text{NO}_2/\text{NO}_x$ measurements with higher temporal resolution used chemiluminescence monitors, some of which have been shown to be subject to interference by species common to urban environments including HONO , HNO_3 , and peroxyacyl nitrates (McClenny *et al.*, 2002; Gerboles *et al.*, 2003; Dunlea *et al.*, 2007; Steinbacher *et al.*, 2007; Kebabian *et al.*, 2008).

Therefore, to add to the knowledge base of how personal $\text{NO}/\text{NO}_2/\text{NO}_x$ exposures vary spatially, temporally, and within different microenvironments in an urban environment, we conducted roll-around mobile monitoring of $\text{NO}/\text{NO}_2/\text{NO}_x$ with 1-minute resolution during 14 days of scripted activities in and around Chicago, IL. Measurements were made using a new direct UV absorbance $\text{NO}/\text{NO}_2/\text{NO}_x$ monitor that is designed to minimize interferences that have been observed in field campaigns using chemiluminescence monitors. Scripted activities were designed to capture time spent in three primary microenvironments: outdoors, indoors

inside various building types, and in multiple modes of transportation including walking, personal vehicle, and various modes of public transit. Results are intended to more accurately demonstrate the spatiotemporal variability in personal NO_x exposures encountered during typical daily activity in an urban environment and to improve knowledge of how personal $\text{NO}/\text{NO}_2/\text{NO}_x$ exposures correlate with ambient central-site monitors in urban environments.

METHODS

Measurements were made using a 2B Technologies Model 405 $\text{NO}/\text{NO}_2/\text{NO}_x$ direct UV absorbance analyzer installed horizontally inside a roll-around bag connected to a 12V lead-acid car battery for mobile monitoring, similar to that described for mobile measurements of personal ozone concentrations in Johnson *et al.* (2013) (Fig. 1). NO_2 is measured directly by the instrument using absorbance at 405 nm, and NO is measured by alternative sequential conversion to NO_2 with internally generated O_3 . Total NO_x is calculated by adding the resulting NO and NO_2 concentrations. The instrument has a manufacturer reported limit of detection of ~ 1 ppb and an accuracy of 2 ppb or 2% of the reading, whichever is greater. The instrument logged at 1-minute intervals for all measurement periods.

A 1 m length of PTFE (Teflon™) tubing was used for the sample inlet, installed at a height of ~ 0.5 m off the ground, and Tygon™ tubing was used for the analyzer's exhaust port (located on the opposite side of the bag from the sampling inlet). The rechargeable battery allowed for measurements for up to 10 hours on a full charge each day of monitoring. 2B recommends an analyzer operating temperature range between 10 and 50°C . Therefore, the temperature and relative humidity inside the case was monitored using an Onset HOBO U12 recording at 1-minute intervals during all measurements. The minimum temperature was just above 10°C and the maximum temperature was 32°C . Other instrument checks included daily zero checks prior to measurements, with a new offset applied as necessary, as well as weekly NO span calibration using a 2B Technologies Model 408 NO Calibration Source.

Two researchers conducted roll-around measurements



Fig. 1. NO_x analyzer installed in a portable, roll-around case.

during a total of 14 days spanning a period of approximately two months during winter and spring, March 2016 through May 2016. During 10 of the sampling days, measurements were made for approximately 10 consecutive hours following a variety of scripted activities in and around Chicago, IL. A shorter sampling period was used for the remaining four days, which involved only a few hours of sampling near the main campus of Illinois Institute of Technology in Chicago, IL. Scripted activities were designed to capture a wide variety of typical behaviors and microenvironments encountered by residents of Chicago, including travel via multiple modes of transportation (e.g., personal vehicle, city bus, subway, elevated train, regional commuter train, taxi, and walking), residential activities (e.g., inactive periods indoors and cooking activities), work/school activities (e.g., attending class, working in a laboratory, or working in an office building), and dining in restaurants (e.g., both fast food and sit-down). Each activity was scheduled to last at least 10 minutes to ensure adequate data collection in each microenvironment, although many activities were conducted for longer periods of time. A smartphone application (Lat Long) was used to record the latitude and longitude of each measurement location during sampling.

Each day's route is shown graphically in Fig. 2, including a few longer suburban routes, several routes between IIT's main campus and downtown Chicago, and several routes within downtown Chicago.

Time-series data from the roll-around monitor were downloaded every day after sampling. Data processing involved labeling each time-series data point with the specific location, latitude, longitude, and time in which sampling took place, notes on the type of activities that were present in a particular microenvironment, and the corresponding temperature, relative humidity, and ambient NO/NO₂/NO_x concentration data from the analyzer. Each location and activity was then coded with more generalized descriptions and individual locations were grouped into the following three primary and nine secondary microenvironmental (ME) categories listed in Table 1.

Measurements were also conducted immediately outside of one of two local regulatory monitors during the majority of test days, typically for about one hour. This co-location period served to provide a check on the comparability of the roll-around analyzer and the local federal regulatory monitors. Of the five regulatory ambient NO/NO₂/NO_x monitors located within Cook County, two sites were used for comparison:

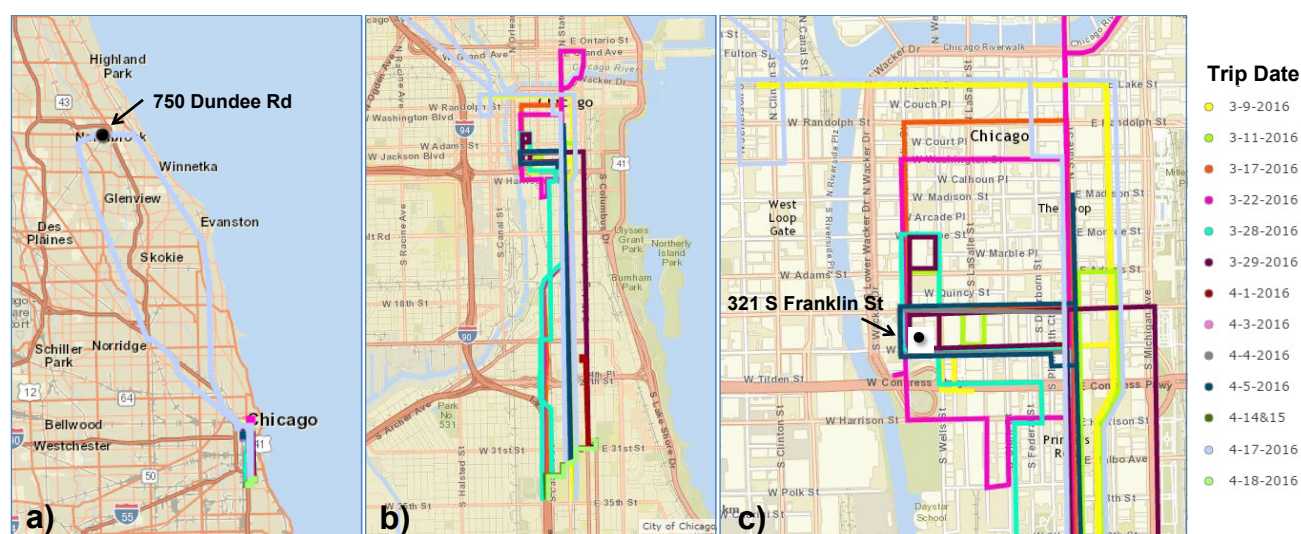


Fig. 2. Map of each sampling day's routes: a) zoomed out to include trips both within downtown Chicago, IL and to surrounding suburban areas; b) zoomed in to include only trips between IIT's main campus and downtown Chicago, IL; and c) zoomed in to include only trips within downtown Chicago, IL. Ambient regulatory monitor locations are marked as 750 Dundee Road and 321 S Franklin Street.

Table 1. List of three primary and nine secondary microenvironment (ME) categories in which measurements were made.

Primary ME	Secondary ME
Indoors	Residential buildings Commercial buildings Retail buildings Educational buildings Restaurants Parking garages (all above-ground)
Transportation	Personal vehicles Public transit (e.g., on a regional or local train or entrance platform)
Outdoors	n/a

an urban site at 321 S Franklin Street in downtown Chicago, IL (visited 11 times), and a suburban site at 750 Dundee Road in the suburb of Northbrook, IL near O'Hare airport (visited once) (IL EPA, 2014). Both locations utilize a Teledyne API Model T200 chemiluminescence NO/NO₂/NO_x analyzer, which is designated by the U.S. EPA as an automated federal reference method (FRM), with a sample inlet height of ~6 m. Hourly-averaged data from these regulatory monitors were kindly provided by personnel at the State of Illinois Environmental Protection Agency. We should note that these data have not yet been assessed for meeting quality assurance thresholds through the EPA's annual data certification process.

RESULTS AND DISCUSSION

After data processing, there were a total of nearly 4000 1-minute average samples, providing approximately 65 hours of useful microenvironmental NO_x concentration data for analysis.

Co-Location Comparisons to Ambient Regulatory Monitoring Stations

Fig. 3 shows the resulting hourly average concentrations measured concurrently with the roll-around NO/NO₂/NO_x analyzer and the two ambient regulatory monitoring stations. Results shown from the roll-around monitor are averages and standard deviations of the 1-minute data summarized over the hour that was spent immediately outside the regulatory monitoring stations. Data from the ambient regulatory monitoring station include either one data point spanning the same hour during which the sampling team was outside the station, or the average and standard deviation across two hourly data points spanning the hours that the team was present outside the station.

Correlations between the roll-around analyzer and the regulatory monitors differed for NO₂ and NO_x, as might be expected due to several possible interferences noted in the NO_x FRM (ASTM, 2005), but surprisingly also for NO. Coefficients of determination (R^2 values) for NO, NO₂, and NO_x were 0.29, 0.56, and 0.69, respectively. Moreover,

NO_x measurements with the roll-around analyzer were slightly lower than the regulatory monitor (slope = 0.82), much lower for NO₂ (slope = 0.51), and higher for NO (slope = 1.44). Discrepancies between the monitors are likely due to a combination of differences in measurement methods (e.g., chemiluminescence vs. UV), inlet sample heights (e.g., ~0.5 m vs. ~6 m), and sampling intervals (e.g., 1-minute vs. 1-hour) and sampling timeframes that did not exactly overlap that may have captured different temporal phenomena such as highly varying traffic sources.

Detailed Microenvironmental Comparisons

Fig. 4 shows an example of time-series NO and NO₂ data collected on March 22, 2016 from both the roll-around monitor (at 1-minute average intervals) and the nearest fixed-site ambient regulatory monitor (at 1-hour average intervals, measured at the 321 S Franklin Street monitor downtown). The roll-around data reflect measurements made in several different microenvironments, with the highest NO₂ peaks occurring in indoor microenvironments (including retail and educational settings), the highest combined NO/NO₂ peaks occurring in transportation microenvironments (chiefly public transit), and the highest NO peaks occurring in outdoor microenvironments. None of these peak values were reflected in the hourly average concentrations measured at the fixed-site monitor.

Fig. 5 shows distributions of 1-minute average NO, NO₂, and NO_x concentrations measured in each of the 9 individual (i.e., secondary) microenvironmental categories listed in Table 1. These same distributional data are also summarized in Table 2 by the number of observations (i.e., 1-minute interval data points in each category) and summary statistics (i.e., mean, standard deviation, and 10th, 50th, and 90th percentiles).

The individual microenvironmental categories with the five highest median NO_x concentrations included four indoor environments and a variety of public transit environments. The median 1-minute average NO_x concentration was highest in the residential microenvironments, which included measurements made in a bedroom and a kitchen inside an apartment unit while occupants were cooking on a natural

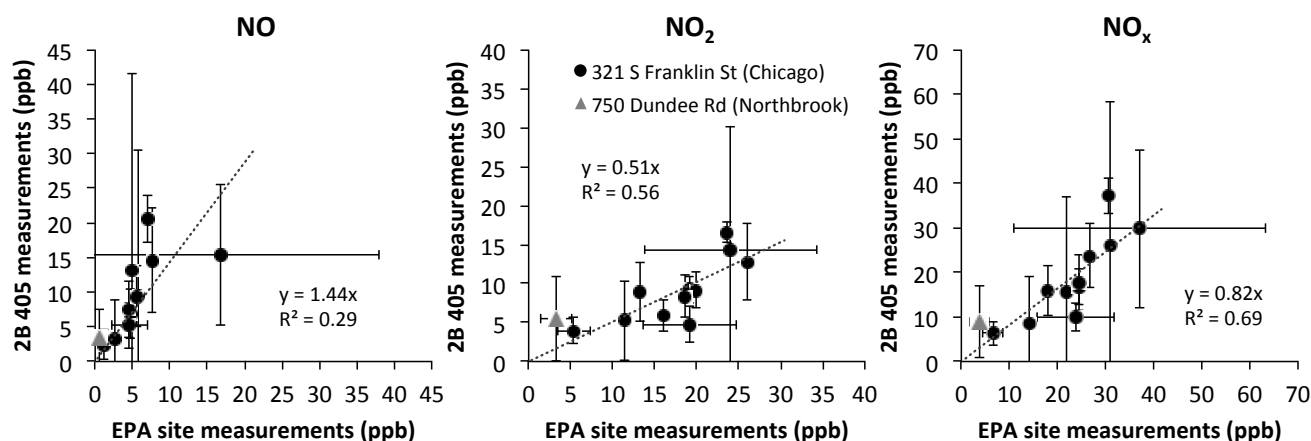


Fig. 3. Hourly average concentrations resulting from co-location measurements alongside two ambient regulatory monitors in Cook County.

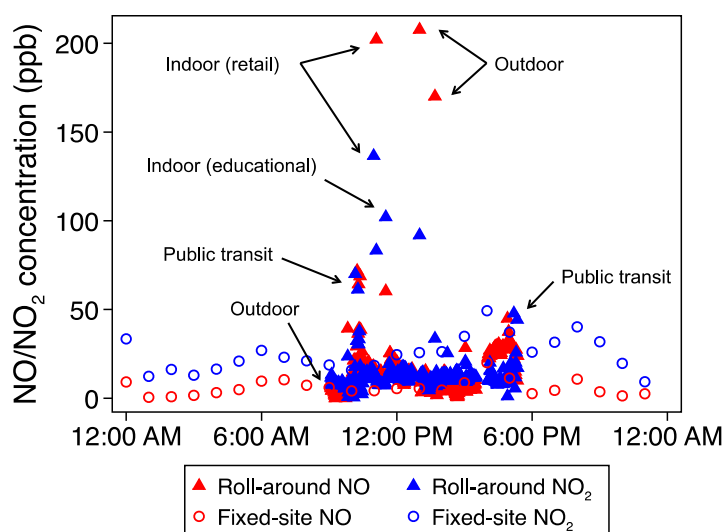


Fig. 4. Example time-series NO_x data collected on March 22, 2016 from (a) the roll-around monitor (sampling interval = 1 minute) and (b) the nearest fixed-site regulatory monitor located at 321 S Franklin Street in Chicago (sampling interval = 1 hour). Microenvironmental categories are marked for several periods of roll-around sampling.

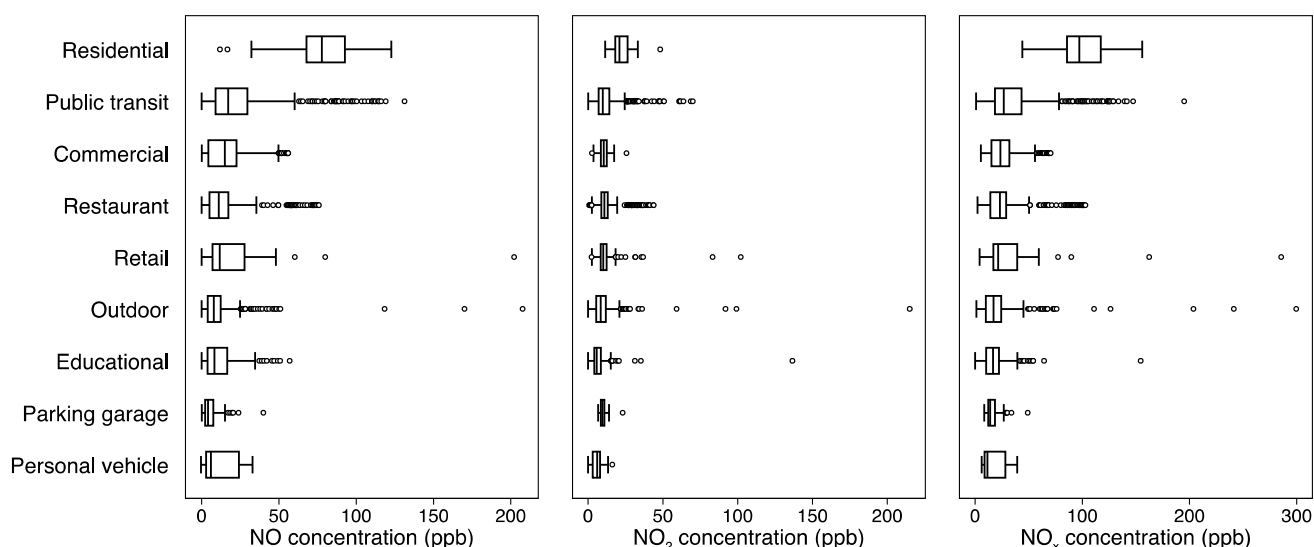


Fig. 5. Distributions of 1-min average NO , NO_2 , and NO_x concentrations measured in 9 different types of microenvironments.

gas stove (median NO_x = 97 ppb, mostly NO). The next highest median microenvironmental NO_x concentrations were those in the public transit category (median NO_x = 27 ppb, mostly NO), which included a combination of measurements in regional train cars, local elevated train cars, local underground subway train cars, outdoor elevated train platforms, and underground subway and regional rail stations. Outdoor measurements had the fourth lowest median NO_x value of 17 ppb. However, peak 1-minute average concentrations of $\text{NO}/\text{NO}_2/\text{NO}_x$ were all highest outdoors and in one of the retail environments visited, with single readings reaching as high as 200 ppb for NO and NO_2 and as high as 300 ppb for NO_x .

Results (i.e., p -values) from non-parametric statistical comparisons of NO , NO_2 , and NO_x concentrations measured in each of the 9 individual microenvironments

made using two-sample Wilcoxon rank-sum (i.e., Mann-Whitney) tests are also shown in Table 3. The majority of comparisons revealed statistically significant differences in NO , NO_2 , and NO_x concentrations between the individual microenvironments. The microenvironmental comparisons that did not yield statistically significant differences in at least one measure of $\text{NO}/\text{NO}_2/\text{NO}_x$ were (i) commercial buildings, restaurants, and retail stores, and (ii) outdoors, educational buildings, parking garages, and personal vehicles.

Summary of Microenvironmental Comparisons

Fig. 6 shows distributions of the same 1-minute average NO , NO_2 , and NO_x data grouped by the three primary microenvironmental categories: transportation, indoor, and outdoor. Median NO_x concentrations were highest in the transportation microenvironments (median = 26 ppb),

Table 2. Summary statistics of 1-min average NO, NO₂, and NO_x concentrations measured in 9 different types of microenvironments.

Location type	N	Mean (SD)	10 th percentile	50 th percentile	90 th percentile
NO_x (ppb)					
Residential	76	99 (26)	60	97	133
Public transit	832	35 (26)	12	27	68
Commercial	241	28 (18)	10	24	63
Restaurant	637	27 (22)	8	23	50
Retail	678	28 (18)	11	22	48
Outdoor	707	20 (20)	6	17	33
Educational	431	18 (12)	7	17	32
Parking garage	62	16 (7)	10	14	27
Personal vehicle	178	17 (10)	7	12	33
NO (ppb)					
Residential	76	77 (23)	43	78	102
Public transit	832	23 (22)	4	17	47
Commercial	241	17 (16)	2	15	48
Restaurant	637	15 (15)	2	11	26
Retail	678	17 (14)	3	12	34
Outdoor	707	10 (13)	2	8	20
Educational	431	11 (9)	1	8	21
Parking garage	62	6 (7)	2	4	15
Personal vehicle	178	11 (11)	1	6	26
NO₂ (ppb)					
Residential	76	23 (6)	16	21	32
Public transit	832	12 (9)	4	10	21
Commercial	241	11 (3)	7	11	14
Restaurant	637	12 (8)	6	11	24
Retail	678	11 (6)	7	10	15
Outdoor	707	10 (11)	4	8	16
Educational	431	7 (7)	3	6	12
Parking garage	62	10 (2)	8	10	12
Personal vehicle	178	6 (3)	2	6	9

followed by the indoor environments (median = 21 ppb), and lowest in the outdoor environments (median = 17 ppb). Similar patterns were also observed for NO, as NO drove most of the variability in total NO_x. Differences in NO and NO_x between each microenvironment were all highly statistically significant ($p < 0.0001$ according to a two-sample Wilcoxon rank-sum, i.e., Mann-Whitney, test). NO₂ distributions were more similar across each of the three microenvironmental categories, although differences in NO₂ between indoor and outdoor and indoor and transportation microenvironments were highly statistically significant ($p < 0.0001$ for both). Differences between outdoor and transportation microenvironments were not as highly statistically significant ($p = 0.006$). Peak 1-min values of NO/NO₂/NO_x were quite similar across all microenvironmental categories, suggesting that NO, NO₂, and NO_x concentrations in excess of 100, 50, and 150 ppb, respectively, can all be encountered at times in each type of microenvironment depending on the nearby source characteristics.

Looking more closely into the indoor and transportation microenvironments, Figs. 7 and 8 show distributions of 1-minute average NO, NO₂, and NO_x concentrations measured in six specific categories of indoor microenvironments and

eight specific categories of transportation microenvironments, respectively. Residential indoor microenvironments had the highest median NO_x concentrations, while all other indoor microenvironments were similar to each other. Interestingly, the lowest indoor NO_x concentrations were observed in the above ground, open air parking garages that were visited. The individual transportation microenvironments with the highest median NO_x concentrations were surprisingly found aboard regional trains, largely driven by high NO from diesel locomotives. The transportation microenvironments with the next highest median NO_x concentrations were underground train stations (e.g., subway and/or regional rail). Personal vehicles and outdoor train platforms had the lowest median NO_x concentrations in this sample.

Correlations between Microenvironmental Measurements and Ambient Regulatory Monitors

Finally, Fig. 9 shows correlations between hourly average records of NO, NO₂, and NO_x taken from the 321 S Franklin Street ambient regulatory monitor in downtown Chicago and the simultaneous microenvironmental measurements made during those same time periods (regardless of location). This ambient monitor was chosen for comparison because it is the nearest monitor for the vast

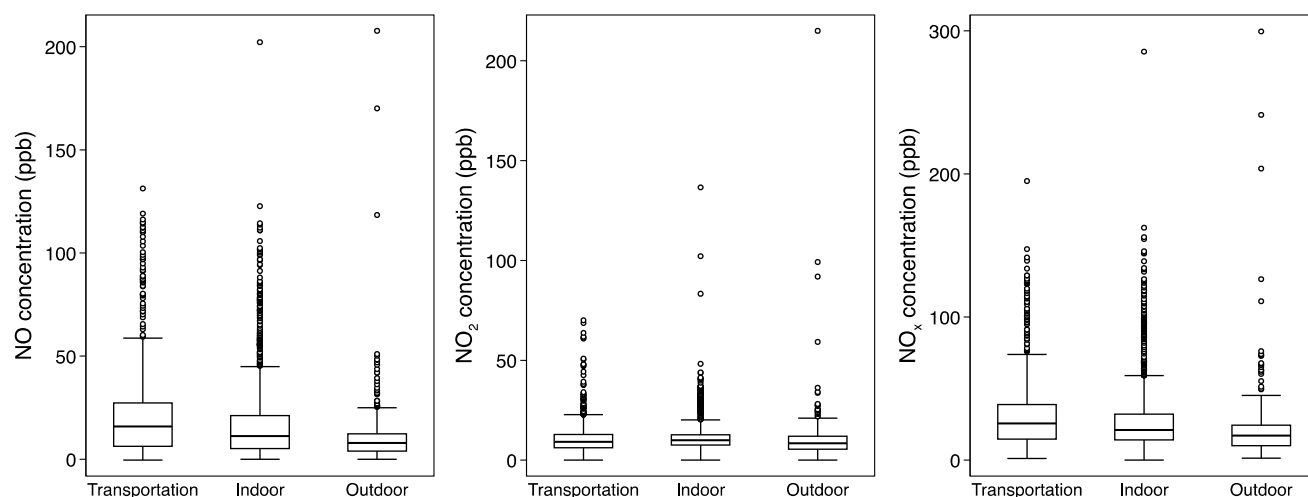


Fig. 6. Distributions of 1-min average NO, NO₂, and NO_x concentrations measured in 3 main categories of microenvironments (n = 1010 for transportation, n = 2125 for indoor, and n = 707 for outdoor).

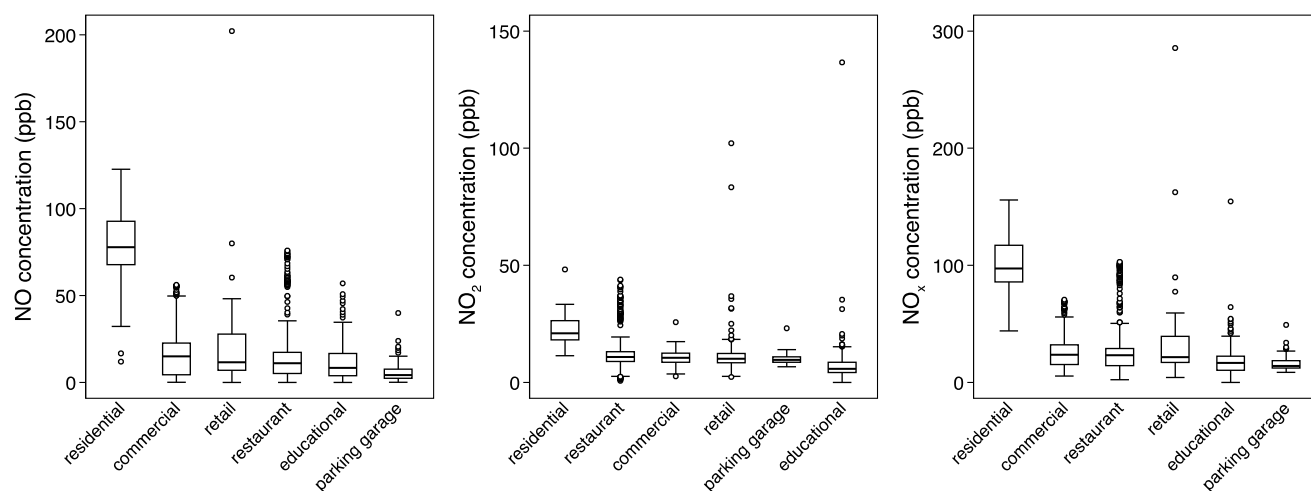


Fig. 7. Distributions of 1-min average NO, NO₂, and NO_x concentrations measured in 6 specific categories of indoor microenvironments.

majority of the microenvironmental measurements that were made (Fig. 2). Because the regulatory monitor only provides a single hourly average value, roll-around microenvironmental NO_x data were matched to the EPA data on an hourly basis via matching time stamps, and an average and standard deviation were obtained using as many 1-minute interval data points as were available in each hour. Comparisons between the roll-around and simultaneous ambient monitor data in Fig. 9 were limited to those hours that had at least 30 data points (i.e., 30 minutes of 1-minute interval data recorded during the same hourly timestamp as that recorded for the EPA monitor). This provided a total of 72 simultaneously recorded hourly NO_x concentrations for comparison.

Correlations between microenvironmental NO/NO₂/NO_x measurements and simultaneous records from the nearest ambient monitor were extremely low, with R^2 values below 0.05 for all comparisons. This is consistent with observations from several prior studies that have reported essentially no

correlation between personal NO₂/NO_x exposures and simultaneous NO₂/NO_x measurements from local ambient monitoring stations or nearby outdoor measurements (Quackenboss *et al.*, 1986; Kousa *et al.*, 2001; Lai *et al.*, 2004; Meng *et al.*, 2012a). These data further illustrate the limitations of relying on ambient site regulatory monitors to characterize personal NO/NO₂/NO_x exposures and provide further evidence that personal monitoring is critical for accurately assessing personal exposure.

Limitations

There are several limitations to this study, as well as limitations to applicability of the measurement methods used here. First, the roll-around monitoring system we used is portable, yet bulky enough that it is not easily carried from one place to another in some areas of typical urban environments (e.g., up and down stairs). Second, measurements are limited to only a short time frame of about two weeks worth of data collection and limited only

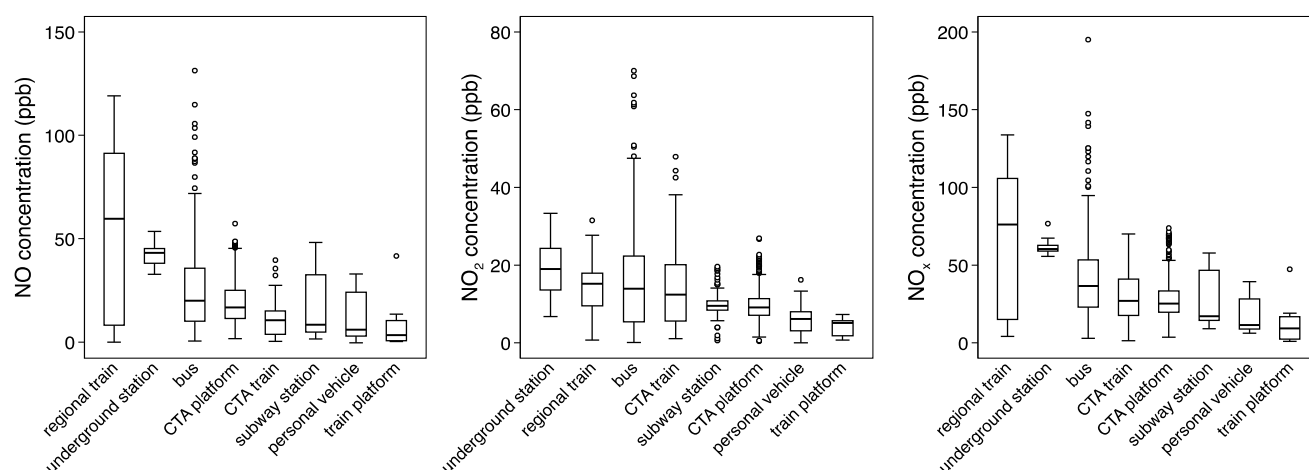


Fig. 8. Distributions of 1-min average NO, NO₂, and NO_x concentrations measured in 8 specific categories of transportation microenvironments.

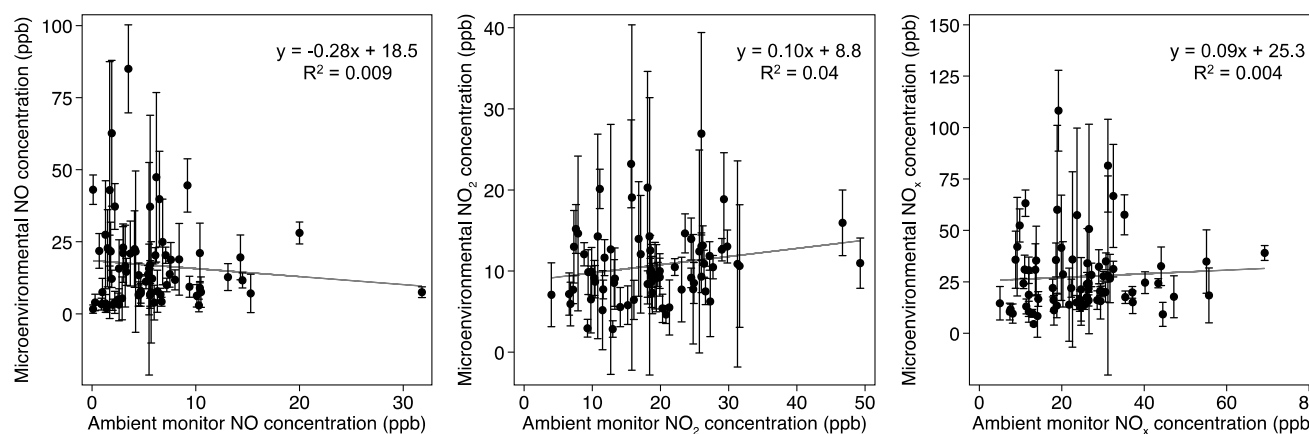


Fig. 9. Correlations between hourly average (\pm SD) microenvironmental NO, NO₂, and NO_x measurements (minimum of 30 1-minute interval data points) and concurrent hourly average concentrations taken from the EPA ambient regulatory monitoring station at 321 S Franklin Street, downtown Chicago, IL ($n = 72$).

to the specific locations in and around Chicago, IL. These data may not be representative for other urban environments. Third, the comparison between the roll-around monitor co-located near the fixed-site regulatory monitor cannot be taken as a direct side-by-side comparison because of differences in sampling inlet heights. Last, each microenvironment was sampled for a relatively short period of time to capture a wide variety of activities, so they may not be representative of longer-term exposures.

CONCLUSIONS

In this work, roll-around mobile monitoring of NO/NO₂/NO_x was conducted with 1-minute resolution during 14 days of scripted activities in and around Chicago, IL. Results demonstrated that residential exposures and exposures in certain types of transit (e.g., regional train and city bus) are likely to drive NO/NO₂/NO_x exposures during typical daily activities in and around Chicago, IL. Correlations between microenvironmental NO/NO₂/NO_x measurements and simultaneous records from the nearest ambient

monitor were extremely low, which further illustrates the limitations of relying on ambient site regulatory monitors to characterize personal NO/NO₂/NO_x exposures and provide further evidence to a growing body of literature that personal monitoring is critical for accurately assessing personal exposure to NO_x.

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DISCLAIMER

Reference to any companies or specific commercial products does not constitute endorsement.

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