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The Fort Collins commuter study: Variability in personal exposure to air pollutants by microenvironment

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Abstract

This study investigated the role of microenvironment on personal exposures to black carbon (BC), fine particulate mass ($PM_{2.5}$), carbon monoxide (CO), and particle number concentration (PNC) among adult residents of Fort Collins, Colorado, USA. Forty-four participants carried a backpack containing personal monitoring instruments for eight nonconsecutive 24-hour periods. Exposures were apportioned into five microenvironments: Home, Work, Transit, Eateries, and Other. Personal exposures exhibited wide heterogeneity that was dominated by within-person variability (both day-to-day and between microenvironment variability). Linear mixed-effects models were used to compare mean personal exposures in each microenvironment, while accounting for possible within-person correlation. Mean personal exposures during Transit and at Eateries tended to be higher than exposures at Home, where participants spent the majority of their time. Compared to Home, mean exposures to BC in Transit were, on average, 129% [95% confidence interval: 101% 162%] higher and exposures to PNC were 180% [101% 289%] higher in Eateries.

KEYWORDS
air pollution, indoor air, microenvironment, personal exposure, traffic-related air pollution, ultrafine particles

1 | INTRODUCTION

Air pollution is a major contributor to the global burden of disease.[1] In the United States, outdoor air pollution is regulated and monitored through a network of fixed sites. Human exposure to air pollution, however, occurs primarily indoors.[2] Air pollution exposures within indoor microenvironments are derived from a combination of indoor sources (eg, cooking, cleaning, movement of people) and infiltration of outdoor (ambient) air pollution indoors.[3-5] The United States Environmental Protection Agency (US EPA) and World Health Organization acknowledge that short- and long-term health effects may be associated with exposure to poor quality indoor air,[6,7] yet indoor air quality monitoring is rare due to the lack of regulations for most indoor microenvironments.

Many studies have provided evidence of an association between exposure to ambient air pollution, which is measured at fixed monitoring sites, and a wide variety of health effects including premature mortality,[8] heart failure,[9] stroke,[10] diabetes mellitus,[11] and many more. However, generally weak correlations between outdoor air pollutant concentrations measured at fixed sites and

personal exposures have been shown on timescales from days to a year[12-16] and little is known about the role of exposures in different microenvironments.

Indoor air pollution concentrations often exceed ambient concentrations due to additional indoor sources and the relatively low levels of air exchange (dilution) indoors.[3,17-20] Many personal exposure studies, however, have been limited to a single microenvironment, due to limitations in monitoring technology.[21] Single-microenvironment studies have evaluated air pollutant concentrations at home,[22-24] at office buildings,[25,26] near roadways,[27,28] or in transit.[29-31] Recent technological advances have enabled a more holistic view of individual exposures as they occur across microenvironments.[19,32-36] Such studies may prove useful for the design of interventions that seek to reduce or mitigate pollutant exposures by targeting specific microenvironments, activities, or pollutants for exposure reduction.[37]

The goals of this study were to evaluate the magnitude of personal exposures to fine particulate matter ($PM_{2.5}$), black carbon (BC), carbon monoxide (CO), and ultrafine particles (quantified by the metric particle number concentration [PNC]), as a function of microenvironment, in a panel of 44 adult participants in Fort Collins, Colorado, USA. We compared personal exposures within and across microenvironments. Variance components analysis was used to examine the between- and within-person variability in microenvironmental exposures.

Practical Implications

- Americans spend 60–90% of their time indoors and within various microenvironments (eg, Home, Work, Eateries), yet data are lacking about how exposure to air pollution varies between such indoor microenvironments and when in transit.
- Personal exposure to multiple air pollutants varied substantially between these microenvironments and from day to day.
- Exposures in Transit and Eateries tended to be among the highest observed and contributed disproportionately to integrated daily exposures compared to the amount of time spent in these microenvironments.

2 | METHODS

2.1 | Personal exposure measurements

The study area, Fort Collins, Colorado is a mid-sized US city with typical ambient $PM_{2.5}$ levels at or below current, health-based regulatory standards established by the US EPA.[38] The city covers approximately 120 km^2 and has approximately 150 000 residents. Personal exposure data were collected as part of the Fort Collins Commuter Study.[39] Briefly, 45 non-smoking participants were recruited to carry backpacks containing lightweight, direct-reading exposure monitors for $PM_{2.5}$ (pDR-1200, Thermo Scientific, Franklin, MA), BC (AE51, AethLabs, San Francisco, CA), and CO (T15n, Langan, Inc, San Francisco, CA). The backpack also contained a global positioning system (GPS) receiver (BT-Q1000XT, QStarz, Taipei, Taiwan) and a temperature, humidity and light sensor (MSR Electronics, GmbH, Switzerland), and an accelerometer used to assess compliance. On a subset of monitoring days, the backpacks also contained an instrument to measure PNC (DiscMini, Testo, Lenzkirch, Germany); the subset was selected based on instrument availability and to maximize within-person replicates. For each sampling day, participants picked up the backpack from the study office in the evening and kept it with them while they completed a full day of their typical activities (including commuting to and from work) and then returned the backpack to the study office on the following morning. Participants were recruited to commute from their home to their workplace on 8 weekdays. On 4 days participants commuted by car (2 days on a high traffic route and 2 days on a lower traffic route) and 4 days participants commuted by bicycle (2 days on a high traffic route and 2 days on a lower traffic route) while carrying the backpack.[39] All sensors recorded data at intervals of 10 seconds or less. Each participant was asked to complete eight sampling days; some participants completed additional sampling days in the case of instrument failure (the range of sampling days per participant was between 1 and 11). Inclusion criteria were the following: 18-65 years old, valid driver's license, current non-smoker, commute at least 1.5 miles (2.4 km) from their home to their workplace, and have no regular exposure to occupational dust and fumes (eg, construction, manufacturing, agriculture). The study was conducted in all seasons; however, participants were not scheduled during official State of Colorado and US Federal holidays. The Colorado State University Institutional Review Board approved all study procedures, and participants provided written informed consent.

The protocol for personal, spatiotemporal exposure monitoring was similar to previous work,[34,40] and details on the method can be found in Good et al.[39] Collected exposure data and GPS coordinates at 10-second resolution were downloaded into a geographic information system (Igor Pro, version 6.37) and processed with geocoded home and work addresses to define a personal exposure track,[34] from which exposures were extracted and categorized for each participant. The microenvironments were manually assigned based on participant's location, a time-activity diary, and environmental metadata (temperature, light intensity, speed, heart rate, and motion). A custom graphical user interface was built to facilitate the microenvironment assignment. The interface mapped each participant's study day alongside their metadata time series (eg, abrupt changes in these values can help to identify when a participant moves from indoors to outdoors). Microenvironments were assigned using the interface by selecting measurements within areas of the map or measurements on the time series and assigning the corresponding microenvironment using the time-activity diary for reference. For this analysis,

exposure data between 9 PM to 9 PM local time were analyzed across five microenvironments that constituted the majority of location/activities identified: Home, Work, Transit (including any bicycling or driving that was recreational or for transportation), Eateries (including restaurants and coffee shops), and Other. The Other category includes any indoor locations that were not Home, Work, or Eateries and when the participant was outdoors, but not in Transit, such as going for a walk or spending time in their yard. For this analysis, any walking during non-commute (ie, between work and home) times was considered to be recreational and included in the Other category.

Sampling days were excluded for participants with missing GPS data (n = 10 sampling days across seven participants) because we could not determine microenvironments and for one participant where only a single sampling day was completed. Additionally, within a sampling day, if an instrument failure resulted in <22 of 24 hours of data collection, the values from that specific instrument were excluded. The resulting dataset contained 373 days of personal monitoring for 44 participants.

The direct-reading $PM_{2.5}$ measurements were corrected with a gravimetric correction factor (ie, the ratio of the gravimetric concentration over the time-weighted average measurements from the sensor over the same time period) determined from a filter measurement collected downstream of the sensor. The gravimetric limit of detection (LOD) was 31 μg; any filter mass measurements not exceeding the limit of detection were replaced by LOD/√2 in the calculation of the gravimetric mass concentration. The CO sensor concentrations were adjusted for temperature using the manufacturer's recommended calibration. The limits of detection for the PM, BC, and CO sensors were 1 μg m^{-3}, 0.01 μg m^{-3}, and 0.01 ppm, respectively. Any 10-second measures below the LOD were replaced by LOD/√2. We also investigated other imputation methods for values below the LOD without meaningful changes in our results (see Supporting Information for details). None of the PNC measures were below the instrument limit of detection.

The Colorado Department of Public Health and the Environment maintains sites that routinely monitor ambient $PM_{2.5}$ and CO with hourly resolution. Ambient air quality data were downloaded from the US EPA Air Quality System Data Mart from one location that monitored $PM_{2.5}$ and another that monitored CO in Fort Collins, CO. Using the hourly ambient concentration data, 24-hour average ambient concentration from 9 PM to 9 PM on sampling days was calculated to match the personal sampling. The limit of detection for the ambient CO monitor was 0.5 ppm, and all hourly average values below the LOD were replaced by 0.25 ppm. Ambient BC and PNC are not routinely monitored in Fort Collins.

2.2 | Statistical analysis

Descriptive statistics were assessed for exposure measures and for participants age and sex. Mean concentrations, over the time spent in each microenvironment and for the 24-hour period, across all study days were calculated for each participant. Violin plots with overlain boxplots of time-weighted average exposures in each of the microenvironments and for the 24-hour mean were created from all measures (all sampling days across all participants).

Daily integrated exposures were calculated by integrating concentrations over the time spent in the corresponding microenvironment or over the 24-hour sampling duration for each pollutant.[41] Daily mass-time ratio, the proportion of the integrated exposure attributable to each microenvironment divided by the fraction of time spent in that microenvironment,[35] was also created for each participant:

$$\text{mass - time ratio} = \frac{\text{integrated concentration}_{\text{ME/total}}}{\text{time}_{\text{ME/total}}} \quad (1)$$

where ME/total indicates the fraction of integrated concentration or time within a microenvironment over the total integrated concentration or time. The mass-time ratio is a useful metric for examining whether an individual's exposure in a given microenvironment contributes disproportionately to their 24-hour average exposure. A mass-time ratio equal to 1 indicates that the fraction of an individual's integrated daily exposure is equal to the fraction of time spent in that microenvironment (ie, exposure in that microenvironment is indicative of their 24-hour average); values larger than 1 indicate that the fraction of their integrated exposure exceeded the time spent in that microenvironment (ie, exposure in this microenvironment exceeds the expected value based on their 24-hour average); and values less than 1 indicate that the fraction of their integrated exposure was less than the time spent in that microenvironment.

A natural log transformation was applied to all time-weighted average exposures in each microenvironment, the 24-hour average, and the ambient concentration 24-hour average, resulting in exposures that were approximately normally distributed. Pearson's *R* correlation coefficients were calculated to evaluate correlation between pollutants within the same microenvironment and between ambient and personal $PM_{2.5}$ and CO concentrations for the 24-hour sampling day. Statistical analyses were conducted using MATLAB (MathWorks, Inc, Natick, MA) and SAS v9.4 (SAS Institute, Cary, NC).

Linear mixed models were developed to estimate the difference in mean personal exposures by microenvironment. The model is:

$$Y_{ijk} = \mu + b_i + d_{j(i)} + \beta_k + e_{ijk}, \quad \text{(Model 1)}$$

where Y_{ijk} is the log-transformed time-weighted average pollutant concentration (BC, $PM_{2.5}$, CO, or PNC) in a microenvironment *k* for participant *i* and sampling day *j*, μ is the overall mean exposure for that pollutant, and e_{ijk} is the unexplained error. To account for potential within-subject and within-day cluster, we include a participant-specific random intercept b_i and a day-specific random intercept nested within participant $d_{j(i)}$. An unstructured covariance structure was specified. The regression coefficient β_k represents a fixed-effect for microenvironment *k*, where the "Home" microenvironment was

considered the reference and omitted from the model. Due to the transformation of the exposure, results are presented as mean percent difference compared to Home and 95% confidence intervals for the percent difference are presented.

We estimated the relative contribution of within-person and between-person variability in within-microenvironment integrated exposure using a variance components analysis. Since participants do not spend the same amount of time in each microenvironment, the within-microenvironment integrated exposures were used. A variance components analysis was conducted on log-transformed integrated exposure values. A linear mixed model was developed to examine the variance components:

$$Z_{ij} = \mu + b_i + e_{ij}. \quad \text{(Model 2)}$$

In this model, Z_{ij} is the log-transformed integrated pollutant exposure (BC, $PM_{2.5}$ mass, CO, or PNC) in a given microenvironment for participant i and sampling day j; Z_{ij} is modeled as normally distributed with overall mean μ, person-specific intercept b_i assumed to be normally distributed with zero mean and variance σ^2_{btw} (the between-person variability), and error e_{ij} that is assumed to be normally distributed with zero mean and variance σ^2_{with} (the within-person variability). The variance components in each microenvironment and for the 24-hour integrated exposures were modeled separately. The intra-class correlation coefficient (ICC) was calculated as:

$$\text{ICC} = \frac{\sigma^2_{btw}}{\sigma^2_{with} + \sigma^2_{btw}} \quad (2)$$

and takes values between zero and one, where smaller values indicate that the within-person variability is high (relative to between-person variability).[42]

3 | RESULTS

Overall, exposures for 44 participants who completed 373 sampling days were included in the analysis. Demographic information on study participants and sample sizes are listed in Table 1. Participant age varied from 22 to 61 years with a mean age of 37. The sample size for BC and CO is 327 and 339 sampling days per pollutant, respectively. The sample size is lower for $PM_{2.5}$ (287 sampling days) due to missing filter data (n = 10) or malfunctioning sensor (n = 63) and for PNC (123 sampling days) because fewer instruments were available for deployment. Descriptive statistics for personal exposures and time spent in each microenvironment (taken across all participants and replicates) and the 24-hour geometric mean and geometric standard deviation are shown in Table 2. Participants spent approximately 13 hours of their time at Home, 6.6 hours at Work, and a smaller fraction of time in the remaining categories. The majority of participants (90%) spent <10% (2.4 hours) of their time in Transit.

TABLE 1 Participant characteristics and sample sizes for personal monitoring of four pollutants

	N (%) or mean (STD) or median (range)
Participants (N = 44)	
Female	24 (55%)
Male	20 (45%)
Age (years)	37 (12)
Personal sampling days	
Total number of sampling days	373
Median number of observations per participant; range	9 (3-13)

An illustrative example of a single participant's daily exposure track is shown in Figure 1, where the color of the bars on the map indicates the pollutant (BC in black and $PM_{2.5}$ in blue; CO and PNC not shown) and the height of the bar indicates the relative concentration. These concentrations are also shown as a time series (Figure 1; top panel) where color shading represents each microenvironment encountered (Home in red; Work in green; Transit in gray; Eatery in blue; and Other in orange). As was typical, there was high variability in participant exposures on different transit routes and in different microenvironments. Exposures tended to be low overnight at Home and low when at Work. Large increases in BC exposure during Transit (gray shaded portions of the time series) are visible and accompanied by only modest increases in $PM_{2.5}$. The relative increase in BC compared to $PM_{2.5}$ is more apparent on the map, particularly near some intersections, where BC concentrations increased dramatically, while $PM_{2.5}$ concentrations increased only slightly. Conversely, while walking in the natural area (green shaded regions on map; orange shading on the time series), $PM_{2.5}$ exposures were elevated (possibly due to pedestrian and bike traffic on unpaved surfaces) and BC levels were low.

Violin plots with overlain boxplots of the mean concentrations of BC, $PM_{2.5}$, CO, and PNC in each microenvironment and the 24-hour time-weighted average are shown in Figure 2 for all sampling days across all participants. The 24-hour time-weighted average concentrations tended to be less than 3 $\mu g\ m^{-3}$ for BC, 15 $\mu g\ m^{-3}$ for $PM_{2.5}$, 1 ppm for CO, and 2×10^4 # cm^{-3} for PNC. For all pollutants, mean concentrations at Work were the lowest among the five microenvironments. Mean concentrations in Eateries were highly variable, but represented many of the highest observed exposures (along with exposures in Transit). Within-person percent change in mean concentrations compared to the Home microenvironment and 95% confidence intervals are listed in Table 3. Compared to the Home microenvironment, BC mean concentrations were 129% (95% CI: 101%-162%) higher in Transit, 93% (58%-137%) higher in Eateries, and 24% (8%-44%) higher in Other microenvironments. $PM_{2.5}$ mean concentrations were 70% (66%-74%) lower at Work, 14% (1%-25%) lower in Transit, and 39% (29%-47%) lower in Other compared to Home, but not different while at Eateries (−13% to 33%). Mean concentrations of CO were elevated in Transit and lower at Work and Other compared to Home. PNC mean concentrations were elevated in Transit and Eateries and lower at Work compared to Home. On

TABLE 2 Mean and quartiles of time-weighted average exposure for BC (μg m^{-3}), $PM_{2.5}$ (μg m^{-3}), CO (ppm), and PNC (particles/cm^{-3}) by microenvironment, among sampling days for which each microenvironment was visited and for the 24-h average

	N	Geometric mean	GSD	Min	25th	50th	75th	Max
Home								
Hours per day	373	13.0	1.4	0.5	12.7	13.8	14.6	24.0
BC (μg m^{-3})	326	0.34	2.25	0.02	0.19	0.35	0.60	3.21
$PM_{2.5}$ (μg m^{-3})	287	8.0	2.3	1.0	4.6	7.3	13.1	78.3
CO (ppm)	338	0.38	2.32	0.01	0.24	0.41	0.63	3.98
PNC (# cm^{-3} × 10^4)	123	0.81	2.18	0.20	0.42	0.77	1.39	9.62
Work								
Hours per day	357	6.6	1.6	0.1	6.2	7.5	8.4	12.5
BC (μg m^{-3})	313	0.28	2.48	0.01	0.17	0.29	0.50	16.40
$PM_{2.5}$ (μg m^{-3})	275	2.4	2.5	0.7	1.1	2.0	4.4	68.6
CO (ppm)	324	0.16	2.86	0.01	0.10	0.18	0.30	15.17
PNC (# cm^{-3} × 10^4)	119	0.27	2.47	0.01	0.15	0.27	0.49	2.56
Transit								
Hours per day	373	1.2	1.7	0.2	0.9	1.3	1.7	4.1
BC (μg m^{-3})	326	1.02	2.01	0.05	0.65	1.04	1.59	15.54
$PM_{2.5}$ (μg m^{-3})	287	6.8	2.2	0.7	4.3	6.9	10.8	95.0
CO (ppm)	338	0.58	2.18	0.01	0.38	0.58	0.85	10.52
PNC (# cm^{-3} × 10^4)	122	1.07	2.02	0.18	0.66	1.02	1.50	22.75
Eatery								
Hours per day	108	0.7	3.1	0.04	0.3	0.8	1.6	7.1
BC (μg m^{-3})	96	0.71	2.42	0.06	0.38	0.63	1.15	9.29
$PM_{2.5}$ (μg m^{-3})	90	8.4	3.5	0.7	4.5	8.4	19.9	130.3
CO (ppm)	95	0.41	2.81	0.01	0.24	0.41	0.79	2.94
PNC (# cm^{-3} × 10^4)	36	2.26	4.26	0.13	1.03	1.64	5.35	53.45
Other								
Hours per day	285	1.5	3.1	0.04	0.8	1.8	3.0	17.0
BC (μg m^{-3})	249	0.46	2.37	0.03	0.28	0.46	0.77	6.03
$PM_{2.5}$ (μg m^{-3})	219	4.9	2.9	0.7	2.5	5.5	8.4	245.0
CO (ppm)	259	0.30	3.14	0.01	0.19	0.32	0.61	5.77
PNC (# cm^{-3} × 10^4)	86	0.70	2.76	0.06	0.36	0.63	1.30	19.38
24 h								
BC (μg m^{-3})	327	0.43	1.94	0.03	0.29	0.42	0.65	3.54
$PM_{2.5}$ (μg m^{-3})	287	7.4	1.9	1.7	4.9	6.6	10.2	51.3
CO (ppm)	339	0.37	2.03	0.01	0.26	0.40	0.56	4.77
PNC (# cm^{-3} × 10^4)	123	0.79	1.79	0.20	0.50	0.76	1.19	6.69

average, PNC mean concentrations were 180% larger (101%-289%) in Eateries compared to the Home microenvironment (the largest within-person difference detected). The average contributions of these microenvironments to mean daily $PM_{2.5}$ exposures are shown by participant in Figure 3. The large day-to-day variability that existed for many participants is exemplified for participant 42 in the inset, where the contribution of the different microenvironments is shown for each of the eight sampling days. The 24-hour average exposures for each sampling day, by participant, are shown in the right panel. The variation in daily $PM_{2.5}$ mean concentrations was large both within and between participants. However, the within-person variability of integrated exposure tended to exceed the between-person variability. Intra-class correlation coefficients were between 0 and 0.49 for all pollutants and microenvironments (Table S1). ICC values were smallest for PNC for the 24-hour average. Large within- and between-participant variability was also observed for the other pollutants (see Figures S1-S3). Participants with the highest mean 24-hour $PM_{2.5}$ exposures did not necessarily have the highest exposures to the other pollutants. Pearson's *R* correlation coefficients were calculated for the 24-hour mean

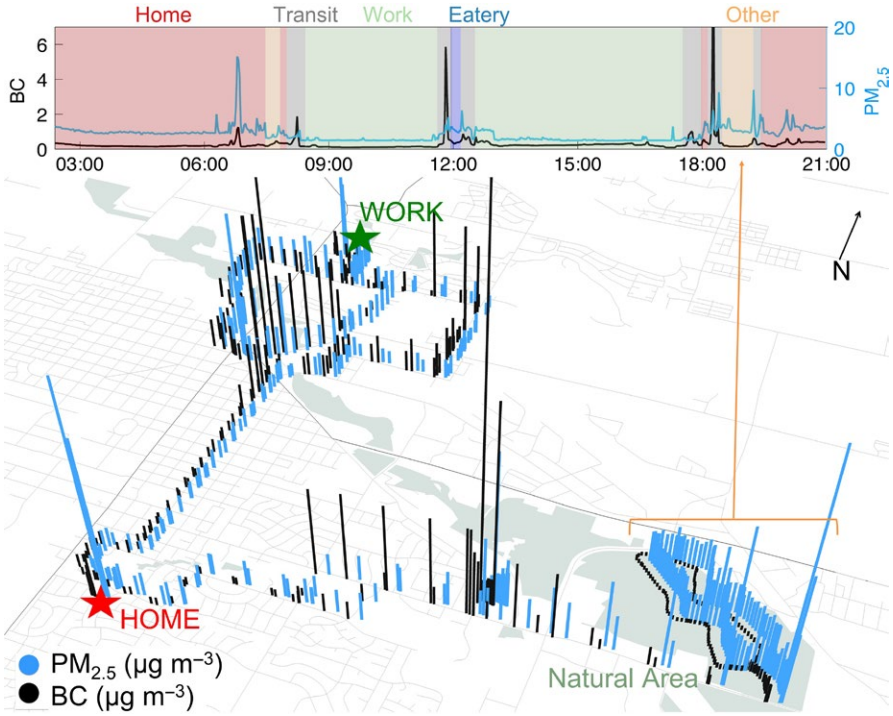

FIGURE 1 GPS trace for one sampling day for a single participant. Bar color denotes the pollutant (BC in black or $PM_{2.5}$ in blue), and bar height denotes the relative concentration. The red star denotes the participant's home location, and the green star denotes the participant's work location. The concentrations of BC and $PM_{2.5}$ over time are shown in the top panel. The shading indicates the microenvironment in which the participant was located during that time period (Home in red; Work in green; Transit in gray; Eatery in blue; and Other in orange). Natural areas are denoted in green on the map

and microenvironment means between each pair of pollutants (see Table S2; results were similar for Spearman's correlation coefficients). Correlations tended to be highest between $PM_{2.5}$ and BC and weakest between CO and PNC. The highest correlation observed was between $PM_{2.5}$ and BC at Eateries and weakest between CO and PNC at Eateries. Correlations at Home varied between 0.06 and 0.50.

The mass-time ratio represents the proportion of integrated exposure attributable to each microenvironment divided by the fraction of time spent in that microenvironment.[35] Violin plots, with overlain boxplots, of the mass-time ratio in each microenvironment are shown in Figure 4. At Home, mass-time ratios tended to be somewhat less than 1 for BC and somewhat greater than 1 for $PM_{2.5}$, CO, and PNC. Mass-time ratios at Work tended to be less than 1 for all pollutants. Although Transit typically accounted for less than 7% of participant's time per day (75th percentile of time), this microenvironment contributed disproportionately to BC, CO, and PNC integrated exposure. Up to 71% of some individual's daily BC-integrated exposure occurred in Transit (mass-time ratio up to 11). Eateries also tended to contribute a disproportionate fraction to integrated exposures for all four pollutants. Mass-time ratios for the other microenvironments tended to center around 1, with higher variability compared to Home and Work.

We examined the association between ambient (outdoor) and personal exposure for both $PM_{2.5}$ and CO (both log-transformed) on a daily basis (24-hour averages) and found that the correlation (Pearson's *R*) was 0.30 and 0.40, respectively. Figure 5 shows the relationship between ambient concentrations and personal exposures (without log transformation). During the days of our study (9/12/2012 to 2/4/2014), daily average ambient measures of $PM_{2.5}$ and CO were 7.1 $\mu g\ m^{-3}$ and 0.34 ppm, respectively. Personal exposures showed a wider range of concentrations for both pollutants than was observed in the ambient measurements.

4 | DISCUSSION

Few studies have examined personal exposure to multiple pollutants as a function of microenvironment due to challenges associated with monitoring; historically, personal sampling has been time-integrated across 8- or 24-hour durations (eg, filter-based analysis of $PM_{2.5}$ mass). Relatively little is known about the variability of personal exposure to air pollution within and between microenvironments. This study, with a median of nine 24-hour measurements per person, is one of the largest studies of personal, multi-pollutant exposure by microenvironment to date. The proportion of time spent by participants in each microenvironment was roughly consistent with national averages for working adults.[43] In comparison with the National Human Activity Pattern Survey (NHAPS), participants in this study typically (mean values reported) spent somewhat less time per day at Home (13.0 hours compared to 16.6 hours), a similar amount of time at Work as was reported for working adults (6.6 hours compared to 6.5 hours), a similar amount of time in Transit (1.2 hours compared to 1.6 hours), and less time in Eateries compared to time in bars/restaurants reported by those who visited that microenvironment (0.7 hours compared to 1.8 hours).[2]

Variance components analysis (Table S1) suggests that within-person variability dominates integrated exposure in a given microenvironment (ie, all ICC values were between 0 and 0.49); this variability is also evident in Figure 3 (inset) where the magnitude and relative contribution (to daily integrated exposure) of each microenvironmental exposure can change considerably from one monitoring

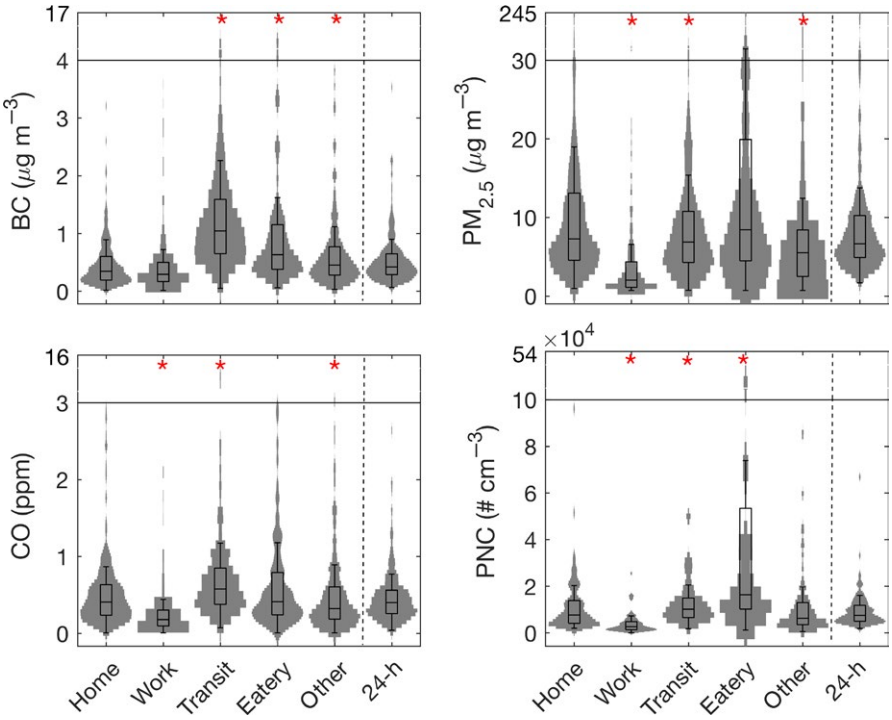

FIGURE 2 Violin plots overlain with boxplots of time-weighted average concentration in each microenvironment and over 24-h for BC, $PM_{2.5}$, CO, and PNC for all participants. The top and bottom line of each box shows the 25th and 75th percentile of exposures, and the middle line shows the median. Whiskers on the boxplots show 95% coverage of the data; the axis is broken along the line to show outliers. Asterisks indicate that exposures in a given microenvironment were significantly different than in the Home microenvironment, when accounting for within-subject autocorrelation

TABLE 3 Mean percent difference [95% confidence intervals for mean percent difference] in personal exposure in each microenvironment compared to the Home microenvironment

	N	Home	Work	Transit	Eatery	Other
BC	1310	Ref.	−1 [−13, 14]	129 [101, 162]	93 [58, 137]	24 [8, 44]
$PM_{2.5}$	1158	Ref.	−70 [−74, −66]	−14 [−25, −1]	8 [−13, 33]	−39 [−47, −29]
CO	1354	Ref.	−57 [−62, −52]	51 [34, 70]	8 [−11, 30]	−22 [−32, −11]
PNC	468	Ref.	−67 [−74, −59]	32 [6, 64]	180 [101, 289]	−11 [−30, 13]

day to the next. Large within-person variability was also observed for the daily measures, as shown in the right-hand side of Figure 3, where an individual's daily personal mean concentration often spans over a factor of five across repeated measurement days. Our results demonstrate lognormal variations in personal exposure between subjects, within subjects, and across the various microenvironments encountered. These results have implications for exposure modeling, because capturing such variability may prove challenging using traditional modeling approaches to capture short-term exposures (eg, temporal land-use regression). These results also reinforce the need for repeated measures when seeking to describe an individual's personal exposure to air pollution; a single, 24-hour measurement can vary by as much as an order of magnitude from one day to another.

We are unaware of any other studies that have considered variance components across different microenvironments. Recent studies have examined the variance components for daily averaged $PM_{2.5,}$ microbial agents, and volatile organic carbon.[44-47] In a study in Gothenburg, Sweden,[45] daily $PM_{2.5}$ concentrations measured among 29 subjects (<2 samples per participant, total n = 43) and variance was partitioned within and between participants and resulted in an ICC value equal to 0.62. ICC values tended to be less than 0.5 and approached zero for some elements, in $PM_{2.5}$ trace analysis.[45] ICC values (with variance partitioned within and between participants) less than 0.5 were also observed for 24-hour $PM_{2.5}$ exposures among adults in Hong Kong[47] and for exposure to microbial agents in buildings.[44] Rappaport and Kupper used a nested design with person nested within city (no person is in both cities) to examine 24-hour VOC exposures in the United States and found that typically half or more of the variance was within-person.[46] A large proportion of the 24-hour average $PM_{2.5}$ variability was also within-person among elderly non-smoking participants in Helsinki, Finland (ICC = 0.34) and Amsterdam, Netherlands (ICC = 0.11).[48] High microenvironmental variability was also observed in a study of ultrafine particulate exposures among adult participants in Copenhagen,

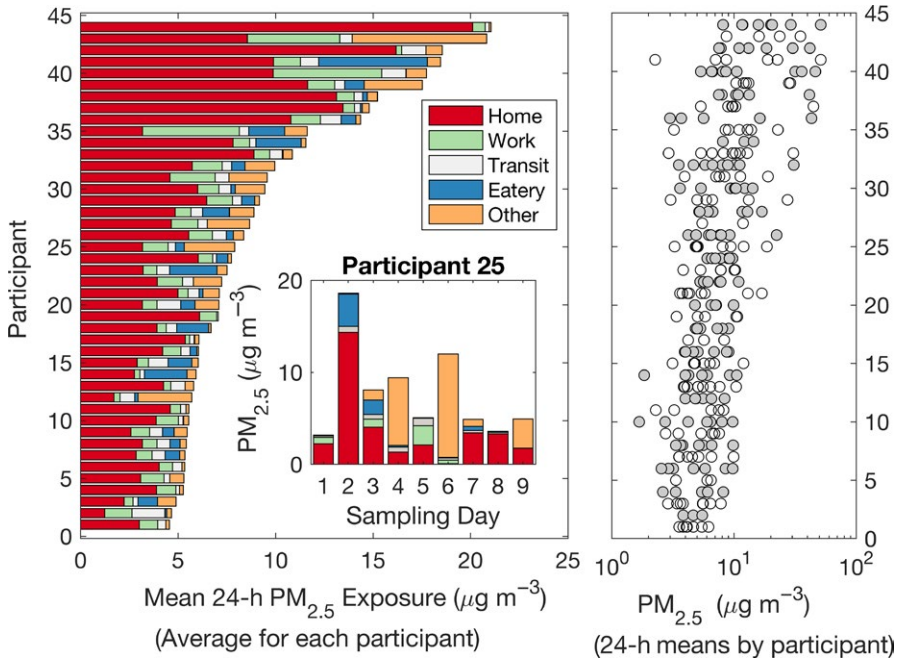

FIGURE 3 Left: the contribution of the five microenvironments to mean exposures of $PM_{2.5}$ mass (average for each participant; μg m^{-3}). Mean exposures to $PM_{2.5}$ by microenvironment for all eight sampling days for a single participant are shown in the inset. Right: 24-h average $PM_{2.5}$ exposures for all sampling days by participant. Participants are ordered from lowest to highest in terms of integrated $PM_{2.5}$ exposure

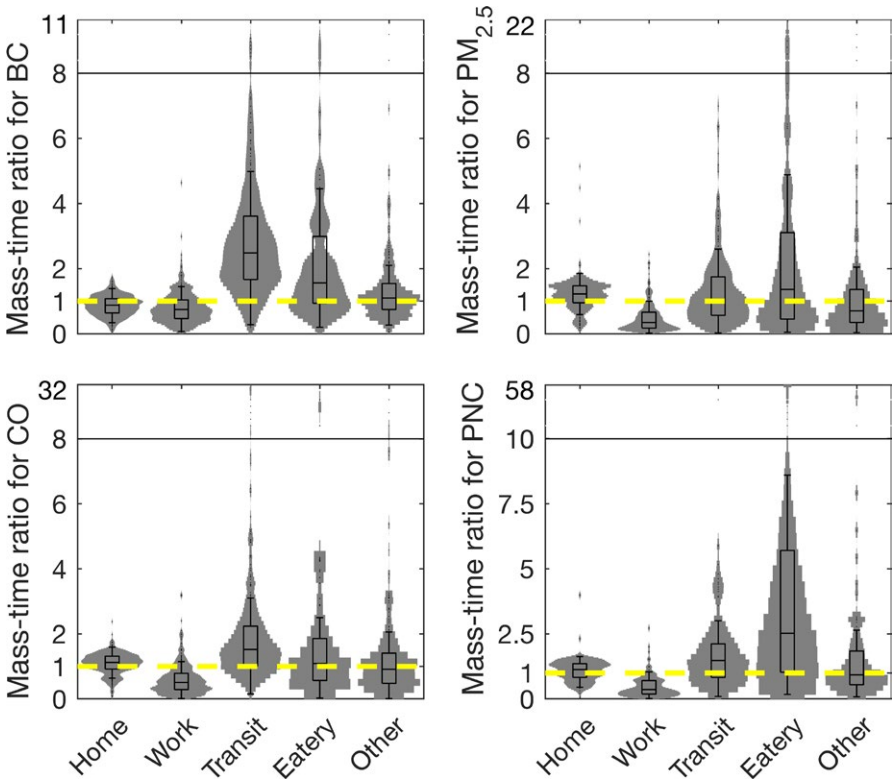

FIGURE 4 Violin plots overlain with boxplots of mass-time exposure ratios in each microenvironment for BC, $PM_{2.5}$, CO, and PNC. The top and bottom line of each box shows the 25th and 75th percentile of exposures, and the middle line shows the median. Whiskers on the boxplots show 95% coverage of the data; the axis is broken along the line to show outliers. The dashed line indicates a value of 1, meaning that the fraction of time in a microenvironment equals the fraction of daily integrated exposure in that microenvironment

Denmark.[41] The low values of ICC observed for Transit may suggest that even with eight replicates, between-day variability may still impact the ability to evaluate exposure-health relationships.[49] Even when our dataset was restricted to sampling days for which participants commuted to work by car, ICC values were somewhat smaller for BC, $PM_{2.5}$, and CO than observed for the full dataset including bicycle commuting, but somewhat larger for PNC. ICC values in Transit and Eateries were less than 0.35; however, the times spent in Transit and Eatery microenvironments were generally small for our participants.

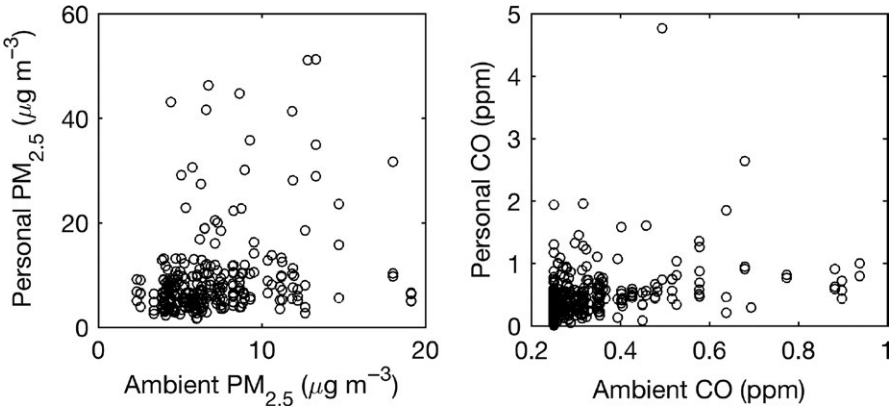

FIGURE 5 Scatter plot of mean 24-h personal exposures and mean 24-h ambient concentrations for $PM_{2.5}$ and CO

Exposure levels were generally low compared to the United States National Ambient Air Quality Standards and WHO guidelines for indoor air for $PM_{2.5}$ and CO. Mean microenvironment exposure levels were below the 24-hour WHO guideline of 25 μg m^{-3} for $PM_{2.5}$ 91% of the time at Home, 98% of the time at Work, 97% of the time in Transit, and 94% of the time for the 24-hour average. Only one Work average exceeded the 8-hr WHO guideline for CO of 8.7 ppm and the 24-hour guideline of 6.1 ppm was not exceeded for any of the 24-hour averages. Air quality regulations for BC and PNC do not exist in the United States, and the WHO has not established guidelines for these pollutants. Ambient air quality in Colorado is generally below regulatory limits, except for higher levels of ozone during summer months that occasionally exceed the 24-hour ozone standard.[38] During the days of our study (9/12/2012 to 2/4/2014), daily average ambient measures of $PM_{2.5}$ and CO were 7.1 μg m^{-3} and 0.34 ppm, respectively. Ambient BC and PNC are not routinely monitored in Fort Collins.

For many participants, the majority of their time is spent at Home, and thus, their integrated exposures tend to be dominated by exposures in the Home (see Table 2 and Figure 3). Compared to integrated exposures, mass-time ratios can be helpful for determining which microenvironments contribute disproportionately to personal exposures to identify potential ways to reduce exposure. Large mass-time ratios for the Transit and Eatery microenvironments were expected due to the close proximity of participants to traffic and cooking sources, respectively. Larger mass-time ratios in Transit compared to Home were observed for BC and CO, but not for $PM_{2.5}$ because BC and CO are considered stronger markers for traffic-related air pollution. These microenvironments with high mass-time ratios could be targets for future interventions studies seeking to reduce exposures. Rea et al[50] found that the fractional contribution of Transit to integrated $PM_{2.5}$ exposure was higher than the fraction of time spent in that microenvironment among elderly adults in Baltimore, Maryland and Fresno, California (ie, mass-time ratio greater than one), but in this study, mass-time ratios centered near one for $PM_{2.5}$ in Transit. Branis and Kolomaznikova[35] reported personal $PM_{2.5}$ exposures for a single participant in the Czech Republic over 239 days and calculated mass-time ratios. The highest concentrations were observed in restaurants and bars (where smoking was allowed) with an average mass-time ratio greater than 20. Typical mass-time ratios in the Eatery category were smaller for our study, likely due to indoor smoking bans in the United States, but values as high as 23 for $PM_{2.5}$ and as high as 57 for PNC were observed. Mass-time ratios at Home were larger in this study than observed by Branis and Kolomaznikova (mean value in this study was 1.2 for $PM_{2.5}$, compared to 0.62), but smaller than those observed by Levy Zamora et al[36] in a population of mostly non-working, women in Texas, USA. The difference in the Home mass-time ratios may reflect the fact that participants in this study tended to work, but in relatively in clean environments with mass-time ratios generally <1 at Work. Additionally, few of our participants reported any exposure to secondhand smoke.

There are a number of strengths to the study design. Each participant was monitored for three pollutants ($PM_{2.5}$ mass, BC, and CO) and a subset of participants were additionally monitored for PNC exposure, on approximately 8 days resulting in one of the largest datasets of personal, spatiotemporal exposure for multiple pollutants. In this study, participants wore chest-mounted accelerometers and the backpack contained an accelerometer and a GPS unit, which allowed us to assess participant compliance with wearing the backpack during waking hours while participants were moving. Compliance was assessed by matching a threshold level of movement (estimated to represent a spatial change) by the participant (determined from a chest-mounted accelerometer) to the backpack accelerometer in 2-minute windows. On average, 66% (standard deviation 22%) of participant movements were matched by backpack movement during each 24-hour measurement period. Compliance was highest in the Transit microenvironment (96%) and lowest in Home (53%) microenvironment. Participants were asked to carry the backpack containing monitoring instruments with them as they moved between rooms, but were not asked to carry the backpack while moving around a room. The potential impacts of participant non-compliance on our results are unclear.

Some limitations of the study include the representativeness of our study population and study design. Participants were non-smokers who lived within the city of Fort Collins and lived at least 1.5 miles from their workplace and were asked to begin their commute during peak commute hours (between 0700 and 0900 hour in the morning and 1630 and 1800 hour in the evening). Thus, our results may not be representative of individuals who telecommute, are

unemployed, or retired. Participant commutes were scripted (mode and route), splitting days between driving and bicycling and driving on higher and lower trafficked routes. Our previous work showed that participants experienced higher exposures to $PM_{2.5}$, BC, and PNC while cycling compared to driving and that they may reduce their exposures to BC and CO by using an alternate commuting route.[39] This may have contributed to the within-person variability demonstrated in this microenvironment (see Table S1). Participants may have changed their behavior under this study design, because they were required to carry a backpack, relative to their typical time-activity patterns. Furthermore, only weekdays were sampled. The impacts on time spent in each microenvironment and on ICC are unclear. Participants were recruited from a wide range of occupations; however, none of our participants were employed in health care or food preparation, and those with regular exposure to occupational dust or fumes (eg, construction, manufacturing, agriculture) were not eligible for inclusion in the study. Additionally, this study may not be reflective of working adults as a whole, as the variability in exposures between urban, suburban, and rural environments is not well established. Furthermore, there is the potential for measurement error in the personal exposure measurements.

5 | CONCLUSIONS

This study is among the largest to quantify time-resolved exposures to $PM_{2.5}$, BC, CO, and PNC and to apportion those exposures into five common microenvironments. Despite representing a small proportion of participant's time, mean exposures to PNC, BC, and CO in Transit were 32%, 129%, and 51% higher than Home exposures, respectively. Exposures, particularly to PNC, were higher in Eateries than other microenvironments and were highly variable. The relative importance of some microenvironments, like Transit and Eateries, to integrated exposures was apparent, with mass-time ratios as high as 20 observed in Transit and as high as 57 in Eateries. Variance component analyses showed that within-person, day-to-day variability dominated the total variance in the dataset and exemplifies the importance of repeated measurements in epidemiologic studies of chronic personal exposure to air pollution.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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