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Contrasting intra-urban variability of ultrafine particle number and fine particle mass concentrations in Dhaka, Bangladesh, and Pittsburgh, USA

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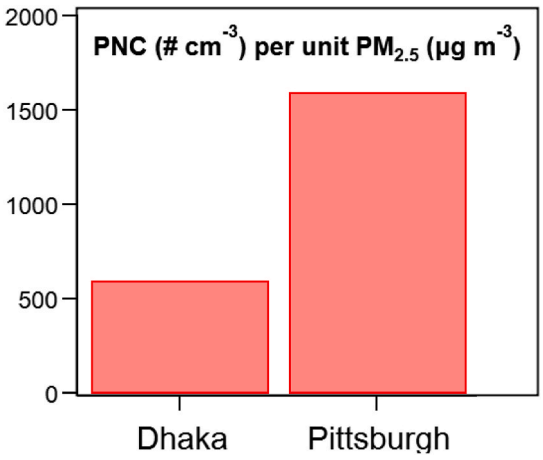
HIGHLIGHTS

- Urban background PNC in Dhaka is three times higher, and $PM_{2.5}$ is twelve times higher compared to Pittsburgh.
- Within-city PNC and $PM_{2.5}$ enhancement increase with source activity, with PNC consistently showing higher local enhancement than $PM_{2.5}$.
- The PNC to $PM_{2.5}$ ratio varies significantly between Pittsburgh and Dhaka, suggesting distinct sources and particle size distributions in these urban areas.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, we investigated the intra-urban variability of fine particle mass ($PM_{2.5}$) and ultrafine particle number concentration (PNC) in Dhaka (Bangladesh), and Pittsburgh (USA), two cities with vastly different pollution sources and levels. We collected measurements of PNC and $PM_{2.5}$ at a wide range of sites spanning a variety of urban land use attributes (35 sites in Dhaka and 30 sites in Pittsburgh). We found that PNC levels exhibited a 3-4-fold variability between sites in each city, ranging from 20,000–100,000 # cm^{-3} in Dhaka and 7,000–28,000 # cm^{-3} in Pittsburgh. $PM_{2.5}$ levels varied within 50% of the urban background level, ranging from 80 to 110 μg m^{-3} in Dhaka and 6–12 μg m^{-3} in Pittsburgh. We observed a moderate level of spatial correlation between PNC and $PM_{2.5}$ measurements in both cities (R^2 0.3 in Dhaka, and 0.4 in Pittsburgh), and consistent within-city spatial patterns in different meteorological seasons. Compared to the Pittsburgh levels, both PNC and $PM_{2.5}$ levels in Dhaka were significantly higher, however, $PM_{2.5}$ was disproportionately higher (10–12 times

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higher) than PNC (3–4 times higher). The slope of PNC versus $PM_{2.5}$ varied significantly between the two cities, suggesting that the sources and size distribution of particles that make up the majority of the $PM_{2.5}$ were different in the two cities. The results found in Dhaka indicate that aerosol particles are influenced by solid fuel combustion, including solid biomass burning, waste burning, and road dust, which have a greater impact on particle mass concentration due to their larger size. Contrary, in Pittsburgh, traffic (gasoline and diesel combustion) particles, relatively smaller in size compared to solid fuel combustion particles, play a dominant role in determining the within-city variability of PNC and $PM_{2.5}$. Our findings suggest that while controlling traffic emissions can contribute to reducing PNC, substantial efforts will be required to mitigate accumulation mode particles from various region-specific sources in order to decrease $PM_{2.5}$ mass concentrations in Dhaka.

1. Introduction

Numerous studies have established a strong link between exposure to fine and ultrafine airborne particles and negative health outcomes (HEI, 2013; Dockery, 2009; Burnett et al., 2018; Pope et al., 2019; Abdillah and Wang, 2023). The distribution of these pollutants varies across cities and regions, with urban areas often reporting higher concentrations (Apte et al., 2015, 2017; Garcia-Marlès et al., 2024). Many south Asian cities, such as Dhaka, Bangladesh, are global hotspots for particulate air pollution (Apte et al., 2015; World Bank, 2023), with fine particulate matter ($PM_{2.5}$; airborne particles with size less than 2.5 μm) levels between 10 and 20 times higher than the World Health Organization guidelines (5 μg m^{-3} for annual average concentrations; 15 μg m^{-3} for a 24-h average).

Besides common sources of $PM_{2.5}$, such as traffic and industrial emissions, South Asian cities like Dhaka have a unique source signature. The use of solid fuels for household cooking and industrial purposes, such as brick manufacturing, as well as activities such as solid waste and agricultural residue burning, construction, and road dust, are reported as important local sources contributing to $PM_{2.5}$ in Dhaka (Begum et al., 2013, 2014; Rahman et al., 2020). Additionally, studies have revealed significant contributions to $PM_{2.5}$ in Dhaka from long-range transported pollution originating within Bangladesh and neighboring countries (Begum et al., 2014; World Bank, 2023).

The $PM_{2.5}$ pollution in Bangladesh is significantly influenced by meteorological factors, with notably higher concentrations (typically, 3–5 times higher) during the dry season (November–March) than during the wet season (April–October) (Afrin et al., 2021). Factors contributing to higher (during the dry season) concentrations may include long-range transported pollution from neighboring countries, especially the Indian Indo-Gangetic Plain (IGP); seasonal sources such as brick kilns and construction activities; and meteorological phenomena like reduced atmospheric mixing (World Bank, 2023). Conversely, heavy precipitation due to the monsoon and a relatively lesser impact from long-range transport, driven by predominant winds from the Bay of Bengal, contribute to relatively lower $PM_{2.5}$ levels during the wet season (Begum et al., 2014; World Bank, 2023).

Health impacts from fine particulate matter ($PM_{2.5}$) have been known for many years. Recently, there is growing awareness of health impacts of ultrafine particles (diameter less than 100 nm) (HEI, 2013; Schraufnagel, 2020). Compared to $PM_{2.5}$, ultrafine particles are smaller, have larger surface area per unit mass, and may have higher toxicity per unit mass basis (HEI, 2013; Heinzerling et al., 2015; Donaldson et al., 2002). When measuring particulate air pollution, particle number concentration (PNC; # cm^{-3}) is typically employed for ultrafine particles; in terms of number concentration, nearly all ambient particles (more than 90–95%) are ultrafine (Kumar et al., 2010, 2014). Conversely, mass concentration (μg m^{-3}) is used for $PM_{2.5}$. The ambient concentration levels of particle number and mass concentrations are influenced by various sources and complex physiochemical processes (Hallquist et al., 2009; Kulmala et al., 2001; Kumar et al., 2010).

In urban areas, PNCs are much more variable than mass concentrations, and local sources such as traffic and combustion sources are the dominant drivers of number concentration (Kumar et al., 2014; Saha et al., 2019a). New particle formation is also an important contributor to PNC in certain areas, times of day, and seasons (Kulmala et al., 2012). $PM_{2.5}$ can be primary (i.e., directly emitted, for example from traffic, cooking, biomass burning, or industrial sources (McDuffie et al., 2021)) or secondary (i.e., formed in the atmosphere, for example, from organic compound condensation or gas-to-particle reactions that add to particle mass) (Hallquist et al., 2009; Jimenez et al., 2009; World Bank, 2023).

The size distribution of particles emitted from different sources varies widely, resulting in a different influence on the particle number versus mass concentrations (Saha et al., 2020). For example, particles emitted from natural gas burning, gasoline, and diesel traffic are typically smaller in size than those from biomass burning, cooking and road dust (Asmi et al., 2011; Ban-Weiss et al., 2010; Kaltsonoudis et al., 2017). Traffic, therefore, has a strong influence on PNC on a per unit $PM_{2.5}$ mass basis, while particles from solid fuel burning and road dust are larger in size and have a lesser impact on PNC on a per unit $PM_{2.5}$ mass basis. The sources can vary by region (Apte et al., 2015; Apte and Pant, 2019; McDuffie et al., 2021), and their relative influence on ambient particle number and mass concentrations can vary based on their location and prevalence.

The two goals of this paper are, first, to present a spatially dense measurement dataset of PNC and $PM_{2.5}$ (35 sites within Dhaka, Bangladesh) to quantify within-city variability, in a region with severely lacking air pollution data. Second, we compare against similar measurements from Pittsburgh, USA (30 sites within the city). This allows us to investigate within-city variability in two settings with markedly different pollution sources and levels. The Pittsburgh dataset has been previously described (Saha et al., 2019a).

2. Methods and materials

2.1. Spatially dense intra-city measurements

2.1.1. Measurements in Dhaka

To assess the intra-city spatial variability of PNC and $PM_{2.5}$ in Dhaka, we collected measurements at 35 sites (Fig. 1A). The sites were chosen to capture the variability in a wide range of land-use, and, thus, particle sources. Based on the land-use characteristics surrounding the various sites, sites were categorized into four groups (see Fig. 1A):

- Site type 1: located in areas with relatively low influences from local urban sources. These sites are characterized by low traffic and population densities, and comparatively further distance from major roads/bus routes/business centers/restaurants. In the results and discussion section of this paper, these sites are termed “urban background”, since they are relatively less influenced by local sources.
- Site type 2: situated in residential areas and near local roads with medium traffic and restaurant densities.
- Site type 3: situated in a mixture of residential and commercial areas with relatively higher traffic and restaurant density than pure residential areas.
- Site type 4: is located near arterial roads, highways, or busy commercial areas; characterized by very high traffic density.

We designed a repeated short-term sampling to collect measurements obtained in 10–17 visits to each site, with around 15 min of data collected per visit. These measurements were distributed across seasons (dry and wet) and times-of-day (morning, midday, and afternoon) to capture the variability in sources and their emissions. This short-term sampling approach has also been used in previous research (Blanco et al., 2023; Doubleday et al., 2023; Hankey and Marshall, 2015; Hoek, 2017; Hoek et al., 2011). Table S1 provides detailed information on the sites, the number of visits, and the measured average PNC and $PM_{2.5}$ concentrations in Dhaka.

The PNC measurements were collected using a water-based condensation particle counter (CPC; MAGIC Model 200P (Hering et al., 2014), Aerosol Devices Inc.; lower size cut 5 nm, flow rate 0.3 L/min). An optical low-cost sensor (PurpleAir PA-II-SD, Plantower PMS5003 sensor) was used to measure $PM_{2.5}^{36}$. Data was collected between 2020 and 2022 during daytime, between 7 a.m. and 7 p.m.

For the measurements collected in 2020, we did not gather or include data from the active lockdown period in Dhaka, which lasted only a few weeks in March. Furthermore, a study conducted in Dhaka (Wadud et al., 2021) examined the impact of COVID-19 lockdowns on local air quality and found no measurable changes in $PM_{2.5}$ levels during lockdowns when historical air pollution and meteorological data were considered. That findings is consistent with similar research in the US (Xiang et al., 2020). Since we did not collect or include any PNC and $PM_{2.5}$ data during the active lockdown, we did not expect COVID-19 lockdowns to affect the dataset presented in this paper.

2.1.2. Measurements in Pittsburgh

Continuous measurements of PNC and $PM_{2.5}$ concentrations were collected in Pittsburgh at 30 sites between 2016 and 2017 (Saha et al., 2019a). The Pittsburgh sites also span a range of urban land uses, including urban background, near-local and arterial roads, near-highway, traffic intersections and city center (downtown); we grouped into four categories, similar to those in Dhaka (Fig. 1B).

PNC measurements in Pittsburgh were obtained continuously between 4 and 6 weeks at each site during the winters of 2016 and 2017 using a rotating network of twelve condensation particle counters (MAGIC CPCs, Model 200P). $PM_{2.5}$ concentrations were also collected at these sites using low-cost sensors (either PurpleAir PA-II or MetOne Neighborhood Particulate Monitor, NPM) (Malings et al., 2020), which were monitored for several years (2017–2020) (Rose Eilenberg et al., 2020). While PNC data in Pittsburgh were collected only during winters, $PM_{2.5}$ data were available throughout the year, encompassing both winter and summer. Detail information on the sites and the measurements in Pittsburgh can be found in Table S2.

2.2. Long-term urban background measurements

In addition to repeated short-term sampling conducted at one of the four site-types mentioned above, long-term continuous measurements of PNC and $PM_{2.5}$ were conducted in each city at an urban background type site. In Dhaka, PNC was collected at the Bangladesh University of Engineering and Technology (BUET) campus with a CPC (MAGIC Model 200P) during thehumid summer (March–April 2020) and dry winter (September–October 2020), about a month in each season. As a long-term $PM_{2.5}$ data set in Dhaka, Beta Attenuation Monitor (BAM) measured continuous hourly $PM_{2.5}$ measurements at the US Embassy Dhaka between 2020 and 2022 were used.

In Pittsburgh, a year-long continuous PNC and $PM_{2.5}$ measurements were obtained at the Carnegie Mellon University campus between September 2016 and August 2017 as the long-term urban background dataset. These measurements were collected as part of the Center for Air, Climate, and Energy Solutions, as described previously (Saha et al., 2018). The PNC were obtained by a Scanning Mobility Particle Sizer (SMPS; TSI DMA 3081, TSI butanol CPC 3772), and $PM_{2.5}$ concentrations were TEOM (Tapered electrode oscillating microbalance) with hourly resolution. Table S3 provides a summary of the instrumentation used in Dhaka and Pittsburgh for this study.

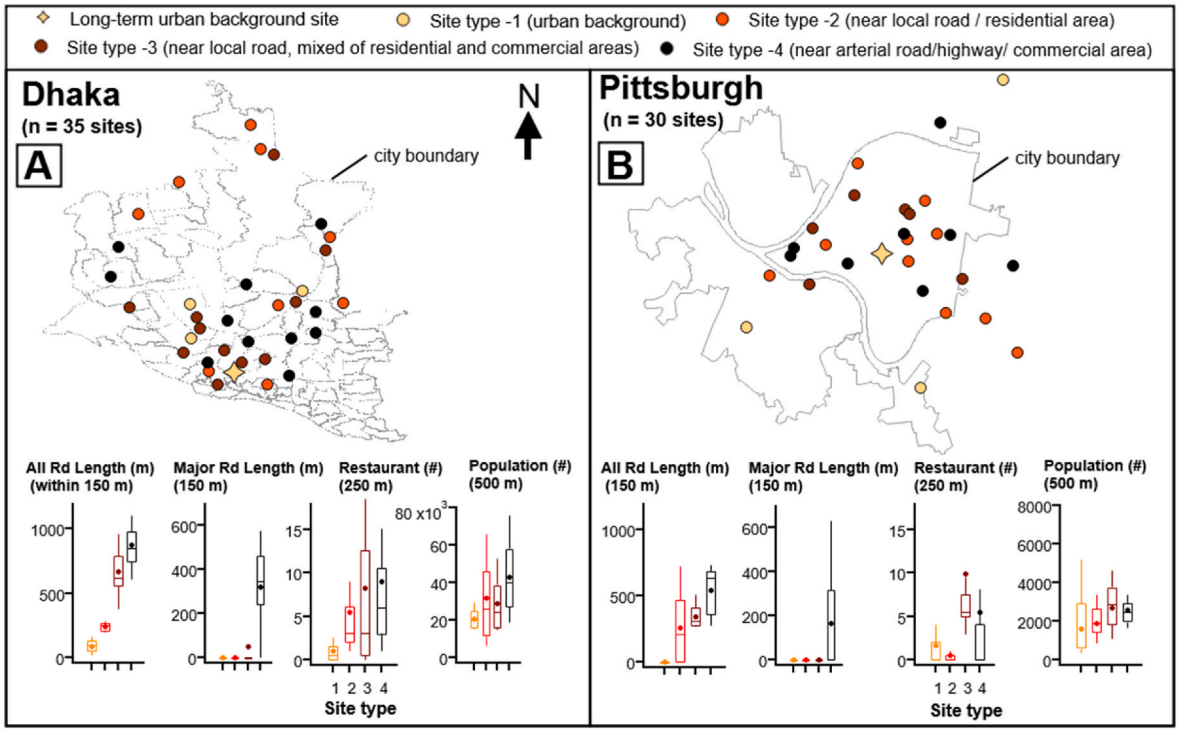

Fig. 1. Sites and surrounding land-uses in (A) Dhaka and (B) Pittsburgh. The symbols on the map show site locations, grouped into four categories (types 1 to 4), plus "long-term urban background sites". Box-and-whisker plots at the bottom illustrate the distribution of land uses for each of the four site types (all road length within 150 m, major road length within 150 m, restaurant count within 250 m and population count within 500 m).

2.3. Data quality assurance

The Purple Air low-cost sensors used in Dhaka to obtain $PM_{2.5}$ concentrations at different sites throughout the city were corrected using beta attenuation monitor (BAM) measurements. Purple Air sensors were co-located at a continuous air monitoring station (CAMS; Darrussalam) in Dhaka in two seasons, obtaining one month of data in each of them. Calibration factors derived from the linear regression indicate slope = 0.94, intercept = 6.3 and $R^2 = 0.90$. These slope and intercept values were used to correct PurpleAir measured $PM_{2.5}$ at different sites throughout the city. $PM_{2.5}$ concentrations measured by PurpleAir and MetOne NPM in Pittsburgh were also corrected using a calibration model developed from co-location experiments against reference monitors. BAM or TEOM were used as reference monitor for calibrating low-cost sensors $PM_{2.5}$ data in Pittsburgh, as described in Malings et al. (2020)

The water-based CPC measurements in Dhaka and Pittsburgh were corrected using calibration factors derived from co-location experiments with a butanol CPC (TSI, model 3772, cut point 10 nm). Since local co-location wasn't possible in Dhaka, Pittsburgh-derived calibration factors were applied for both cities. The same MAGIC CPC unit was used for measuring PNC in the two cities. The intercomparison of the MAGIC water CPC against a TSI butanol CPC showed an approximately 30% undercounting, consistent with findings from other studies (Franklin et al., 2010; Kupc et al., 2013; Mordas et al., 2008). Applying this factor improved the absolute quantification of measured PNC. By using an intercomparison factor based on a butanol CPC with a 10 nm cut point, the resulting PNC can be considered equivalent to a butanol CPC 10 nm PNC. This approach mirrors how we corrected data from PurpleAir $PM_{2.5}$ sensors and compared it with beta attenuation monitor (BAM), referred to as BAM-equivalent $PM_{2.5}$.

Previous studies have shown that an average concentration derived from a series of targeted short-term sampling can provide a value similar to the long-term average concentration (Apte et al., 2017; Blanco et al., 2023; Doubleday et al., 2023; Li et al., 2019; Saha et al., 2019b). That finding is consistent with the central-limit theory, which says that, if randomly sampling from a distribution, the mean value of the sample will tend to approach the mean of the population However, to obtain a stable representative average, 10–15 visits are required over a range of temporal conditions (as was done in Dhaka). To examine the representativeness of the spatially distributed measurements through short-term visits concerning the long-term, mean concentrations were compared to the long-term continuous $PM_{2.5}$ measurements from the US Embassy in Dhaka. The mean concentration from short-term visits agreed within 10% of the long-term continuous mean (Fig. S2).

We compared the long-term continuous PNC data collected at the BUET campus with data from short-term visits to the same location (Fig. S3). Mean PNC levels from long-term continuous and short-term (daytime only) sampling exhibited agreement within 20%, and within 5% when compared against long-term daytime-only mean PNC (Fig. S3). These findings suggest that mean concentrations from targeted repeated short-term visits can represent long-term average exposure concentrations at these locations. However, short-term sampling may not capture all extreme scenarios detected by long-term continuous sampling.

A limitation of our short-term sampling in Dhaka is that it was restricted to daytime only (between 7 a.m. and 7 p.m.). Concentrations of pollutants vary at different times of the day. Our analysis of long-term continuous and short-term PNC data collected at the urban background location in Dhaka indicates that the mean PNC concentration from daytime short-term sampling is about 12% higher compared to the mean estimated from long-term continuous sampling covering day and night. That finding is consistent with recent evidence from Seattle, USA, suggesting that nighttime sampling may be important for correctly determining long-term averages (Blanco et al., 2023; Doubleday et al., 2023); further investigation of that aspect for Bangladesh may be useful in follow-on research. $PM_{2.5}$ concentrations also show variability over different times of the day, with slightly higher concentrations during nighttime and morning, and lower concentrations in midday. However, since all the concentrations were collected during the daytime and covered different parts of the day (morning, midday, and afternoon), using the average concentrations from each site for site-to-site comparison does not introduce a systematic bias for spatial variation, which is the main focus of this paper.

2.4. Quantification of spatial variability

We used quality-assured datasets to quantify the spatial variability of PNC and $PM_{2.5}$ in Dhaka and Pittsburgh and compared it. Specifically, we employed four approaches: (i) comparing measured average concentrations at each site by grouping them according to their categories (site type 1 to 4), (ii) quantifying spatial heterogeneity between pairs of sites using the coefficient of divergence (COD), (iii) examining local increases (concentrations above urban background level) of PNC and $PM_{2.5}$ concentrations according to their categories (site type 1 to 4), and (iv) examining the spatial correlation between PNC and $PM_{2.5}$ across the range of sites in each city.

The coefficient of divergence (COD) has been widely employed in previous studies to measure spatial variation between pairs of sites (Krudysz et al., 2009; Moore et al., 2009; Wilson et al., 2005). The COD value for a pair of sites ranges from 0 to 1: 0 indicates identical concentrations at both sites; a value nearing 1 suggests significantly different concentrations. In this paper, if the COD exceeds 0.2, pollutant concentrations at the two sites are deemed spatially heterogeneous (Wilson et al., 2005).

3. Results and discussion

3.1. Urban background concentrations

Fig. 2 compares the urban background levels of PNC and $PM_{2.5}$ in Dhaka and Pittsburgh. Fig. S4 displays the measured time series data in urban background sites. The urban background air pollution levels in Dhaka are, on average relative to Pittsburgh, approximately three times higher for PNC and approximately twelve times higher for $PM_{2.5}$. This indicates that the relative level of $PM_{2.5}$ is disproportionately higher in Dhaka.

Ambient PNC and $PM_{2.5}$ levels are influenced by various sources and processes. Relative to PNC, the disproportionately higher levels of $PM_{2.5}$ in Dhaka suggest a relatively greater abundance of sources and processes contributing to particles that are accumulation mode particles or larger. PNC levels are primarily influenced by nucleation and Aitken mode particles (particles less than 100 nm), with a significant contribution from traffic sources (Brown et al., 2000; Morawska et al., 2008). On the other hand, $PM_{2.5}$ mass concentrations usually consist mainly of accumulation mode particles (particles in the size range 0.5–2 μm in diameter), with an important contribution from secondary particle mass resulting from gas-particle conversion of precursor gases (Hallquist et al., 2009), as well as primary particles from traffic, wood smoke, cooking, and others.

A recent modeling-based analysis (World Bank, 2023) in Dhaka supports a significant contribution from secondary $PM_{2.5}$ and primary $PM_{2.5}$ resulting from solid fuel combustion. According to a chemical transport modeling in that study, approximately 45–50% of $PM_{2.5}$ in Dhaka originates from secondary sources, with the second-largest contribution (25–30%) coming from primary $PM_{2.5}$ emitted by the residential sector, mainly due to wood usage for cooking. Furthermore, that study reported that primary $PM_{2.5}$ from agricultural residue burning, municipal waste, livestock fertilizer, small industries such as brick kilns, and mobile sources are significant contributors, each accounting for 2–7% of the overall $PM_{2.5}$ levels.

Fig. 2C and D shows the average diurnal profiles of PNC and $PM_{2.5}$ in Dhaka and Pittsburgh. Although the absolute magnitudes of PNC and

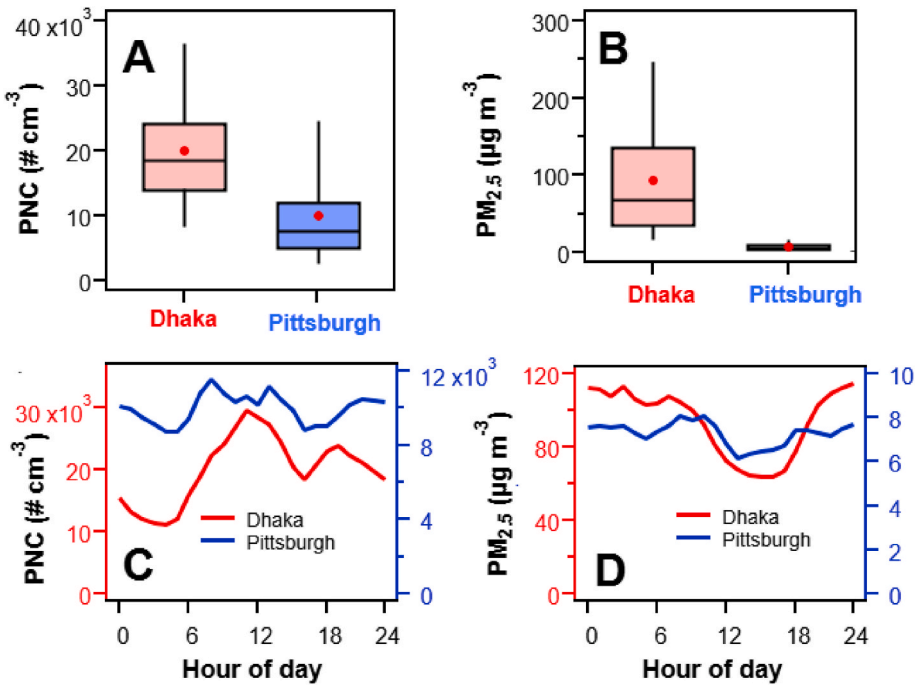

Fig. 2. Comparison of urban background particle number concentration (PNC) and $PM_{2.5}$ levels in Dhaka and Pittsburgh. Box-and-whisker plots of long-term of (A) PNC and (B) $PM_{2.5}$ measurements. Boxes indicate interquartile range, whiskers represent 5^{th}-95^{th} percentile range, horizontal lines within the boxes indicate the median, and circles represent the mean. Average diurnal profiles of measured (C) PNC and (D) $PM_{2.5}$ concentrations are shown. The left axis (red) represents Dhaka concentrations, and the right axis (blue) Pittsburgh concentrations.

$PM_{2.5}$ levels are significantly higher in Dhaka, their diurnal variation shows somewhat similar trends in both cities. PNC levels are lower during nighttime and higher during daytime, with local PNC levels in urban areas mostly dominated by traffic, resulting in peaks observed during morning and evening traffic rush hours in both cities.

Atmospheric new particle formation (nucleation) is another phenomenon that causes peaks in PNC levels, particularly at midday (Kulmala et al., 2012). Elevated midday PNC levels are observed in both cities. Prior research has found that in Pittsburgh, nucleation events occur on average 30% of days in a year (Saha et al., 2018). PNC time series data at the urban background location in Dhaka indicated frequent midday peaks (between 11 a.m. and 3 p.m.), occurring on 41% of days in March–April (12 days out of 29) and 26% of days in September–October (7 days out of 27). While new particle formation was previously thought to be a pristine phenomenon, substantial new particle formation and elevated PNC levels are reported in many polluted cities like Beijing, China, and others (Wang et al., 2017; Wu et al., 2007; Xiao et al., 2021). Fig. S5 provides an example of a midday PNC peak from a possible nucleation versus a non-nucleation day. Previous research has reported that these elevated PNC peaks at midday lasting 1–3 h and contributed between 5 and 10% to the long-term average urban background concentration (Kulmala et al., 2012; Saha et al., 2018).

In addition, $PM_{2.5}$ levels in both cities are highest during nighttime and morning, and lowest at midday, indicating a strong influence from meteorological conditions and diurnal variation of the atmospheric mixing layer (Fig. 2C and D). The atmospheric boundary layer is typically higher during midday and lower at night, resulting in lower $PM_{2.5}$ concentrations during midday and higher concentrations during the night, as this pollution is more regionally distributed. Midday $PM_{2.5}$ levels are 30% lower-than-average in Dhaka and 10% lower-than-average in Pittsburgh. Local sources have a greater influence on urban PNC levels, leading to elevated levels during high source activity periods such as traffic rush hours and midday local nucleation (Brines et al., 2015; Kalkavouras et al., 2021; Kalkavouras et al., 2021, 2021; Kerminen et al., 2018; Saha et al., 2018). In contrast, $PM_{2.5}$ levels are more influenced by regional sources and processes, with atmospheric mixing being an influential factor in the observed diurnal variation.

3.2. *Intra-city spatial variability of PNC and $PM_{2.5}$*

The PNC and $PM_{2.5}$ data measured at multiple sites in both Dhaka (35 sites) and Pittsburgh (30 sites) are summarized in Fig. 3. The measured PNC levels in Dhaka ranged from 20,000 to 100,000 # cm^{-3}, while in Pittsburgh, they ranged from 7,000 to 28,000 # cm^{-3}. The within-city $PM_{2.5}$ levels in Dhaka ranged from 80 to 110 µg m^{-3}, whereas in Pittsburgh, they varied from 6 to 12 µg m^{-3}. Similar to the urban background observation, spatially distributed measurements across the cities also indicate that the relative levels of $PM_{2.5}$ in Dhaka were disproportionately higher.

Despite significant differences in the absolute concentration levels of PNC and $PM_{2.5}$ between Dhaka and Pittsburgh, the within-city variability of these pollutants follows a similar spatial trend in both cities. In terms of spatial patterns, as expected, PNC levels are lower-than-average at urban background sites, and higher-than-average in areas with higher relevant source-activity. Across the selected sites, temporal-mean PNC levels vary by a factor of 3–4 relative to the urban background level, while $PM_{2.5}$ levels show a spatial variability of 10–50% of the urban background level.

In Dhaka, at the urban background sites (site type-1), PNC levels range from 20,000 to 30,000 # cm^{-3}; in Pittsburgh, the same range is from 7,000 to 8,000 # cm^{-3}. PNC near local roads and residential areas (site type-2) are about 50% higher than the urban background levels in both cities, while site type-3 exhibits PNC levels about 1.5–2 times higher than the urban background levels. Near arterial roads, highways, and commercial areas (site type-4), the PNC levels are about 3–4 times higher than the mean urban background level. In Dhaka, PNC levels in these areas ranged from 65,000 to 100,000 # cm^{-3}, while in Pittsburgh, concentrations in high source-activity areas (site type-4) ranged from 15,000 to 28,000 # cm^{-3}.

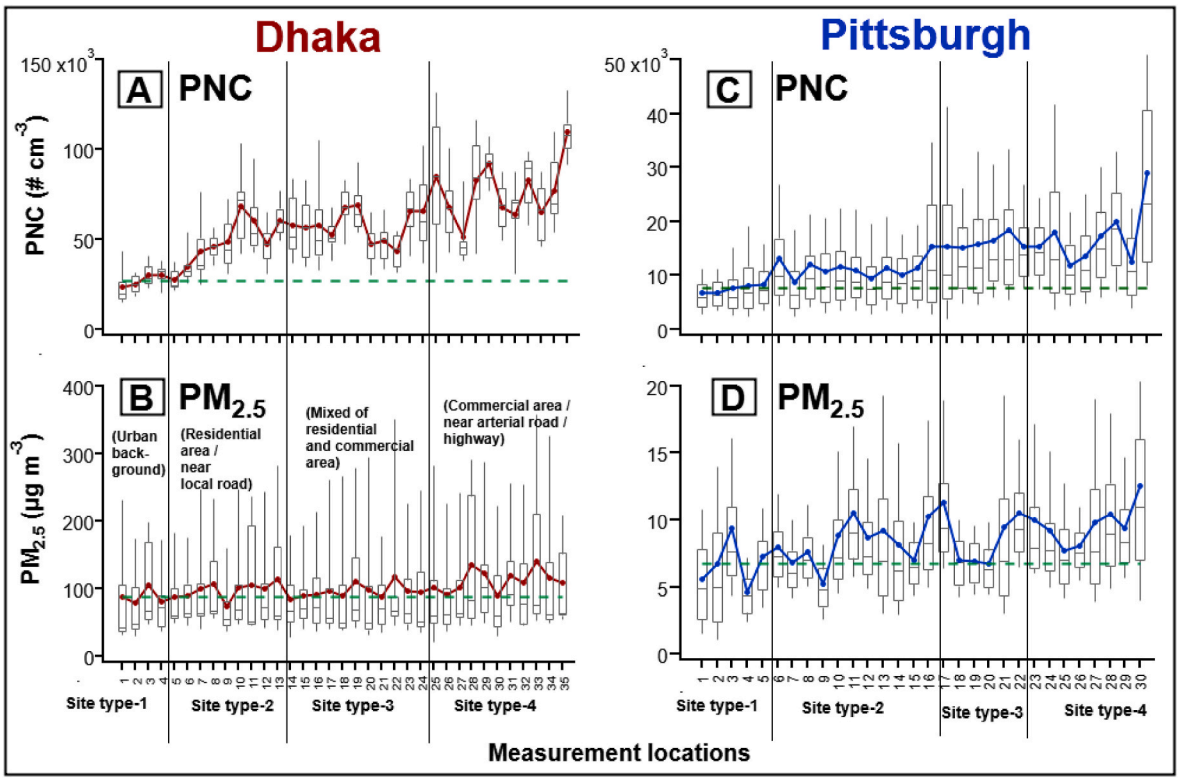

Fig. 3. Within-city spatial variability of PNC and $PM_{2.5}$ in Dhaka (A–B) and Pittsburgh (C–D). The sites are grouped by site type, as defined in Fig. 1. The box-and-whisker plot shows the range of measured concentrations at each site, where the box represents the interquartile range, the whiskers show the 5^{th}-95^{th} percentile range, the horizontal line within the box is the median, and the dot represents the mean. A line connecting the mean concentration at each site is provided for visual guidance. The horizontal dashed green line in each panel represents the average concentrations measured across all urban background sites (site type 1) in each city.

On the other hand, $PM_{2.5}$ levels exhibit less spatial variability than PNC in both cities, with levels in residential and mixed areas about 10–20% higher than the urban background level, and in sites with higher traffic and busy commercial areas between 30 and 50% higher.

The coefficient of divergence (COD) was used to assess the spatial heterogeneity of PNC and $PM_{2.5}$ in Dhaka and Pittsburgh. A COD value greater than 0.2 typically indicates substantial differences in concentrations measured at a pair of sites. Fig. S6 shows the estimated COD values for each site pair for both cities. The COD values for PNC exhibit higher variability than those for $PM_{2.5}$ in both cities, ranging from 0.02 to 0.6 in Dhaka and from 0.02 to 0.55 in Pittsburgh. The $PM_{2.5}$ COD values ranged from 0.01 to 0.25 in Dhaka and from 0.02 to 0.3 in Pittsburgh. In the case of specific site types, COD values are smaller, indicating similar measured concentrations. For the PNC, the highest COD value is observed between the urban background (site type-1) and sites in commercial areas and near major roads (site type-4).

3.3. Seasonal variability of PNC and $PM_{2.5}$

We conducted a seasonal analysis of within-city spatial variability in PNC and $PM_{2.5}$ levels (Figs. S7 and S8). Our findings indicated that, while concentration magnitudes varied across seasons, the within-city spatial patterns were similar for each season.

In Dhaka, $PM_{2.5}$ concentrations were found to be about a factor of three higher in the dry season (November–March) compared to the wet season (April–October). However, the within-city spatial variability of $PM_{2.5}$ remained consistent for each season, ranging from 10 to 50% of urban background levels (Fig. S7). This result is consistent with large seasonal variation in $PM_{2.5}$ levels in Dhaka being largely driven by regional and meteorological factors whereas the within-city spatial variation being attributable to local sources. The within-city spatial variability of PNC followed a similar pattern, with concentrations varying within a factor of 2–4 across different sites in both seasons, and slightly higher levels (between 5 and 20%) observed during the dry season.

In Pittsburgh, $PM_{2.5}$ data were available for both summer and winter suggestions and analysis indicate that within-city spatial variability of $PM_{2.5}$ remained consistent across seasons (Fig. S8). However, Pittsburgh PNC data were only collected during the winter season, hence seasonal analysis was not feasible. Long-term PNC data from the urban background site in Pittsburgh revealed that PNC levels remained within 20% of the annual mean across all seasons.

3.4. Influence of local sources on intra-city variability of PNC and $PM_{2.5}$

Fig. 4 presents the average PNC and $PM_{2.5}$ across different site types, separating between urban background levels and those likely from highly localized emissions (labeled as “local enhancement”). The urban background level represents the mean concentrations measured across site type 1. Concentrations from highly localized emissions are characterized as the differences between measured concentrations in other site types and the urban background level. In Dhaka, PNC local enhancement ranges from 20,000 to 50,000 # cm^{-3} and $PM_{2.5}$ varies between 10 and 20 µg m^{-3}. In Pittsburgh, PNC local enhancement ranges from 3,000–10,000 # cm^{-3}, and $PM_{2.5}$ ranges from 1.5 to 3 µg m^{-3}.

Our analysis reveals that, on average, within-city local enhancement of PNC and $PM_{2.5}$ increases with source activity, with site type 4 exhibiting the highest enhancement compared to site types 2 and 3 (see Fig. 4). However, fractional local enhancement of PNC is consistently higher than $PM_{2.5}$, when compared to the urban background levels.

In Dhaka, high-source activity areas (site type 4) exhibit PNC local enhancement that is approximately 200% higher than the urban background levels; in Pittsburgh, it is 150% higher. For $PM_{2.5}$, enhancement levels are 25% higher than background in Dhaka and 40% higher in

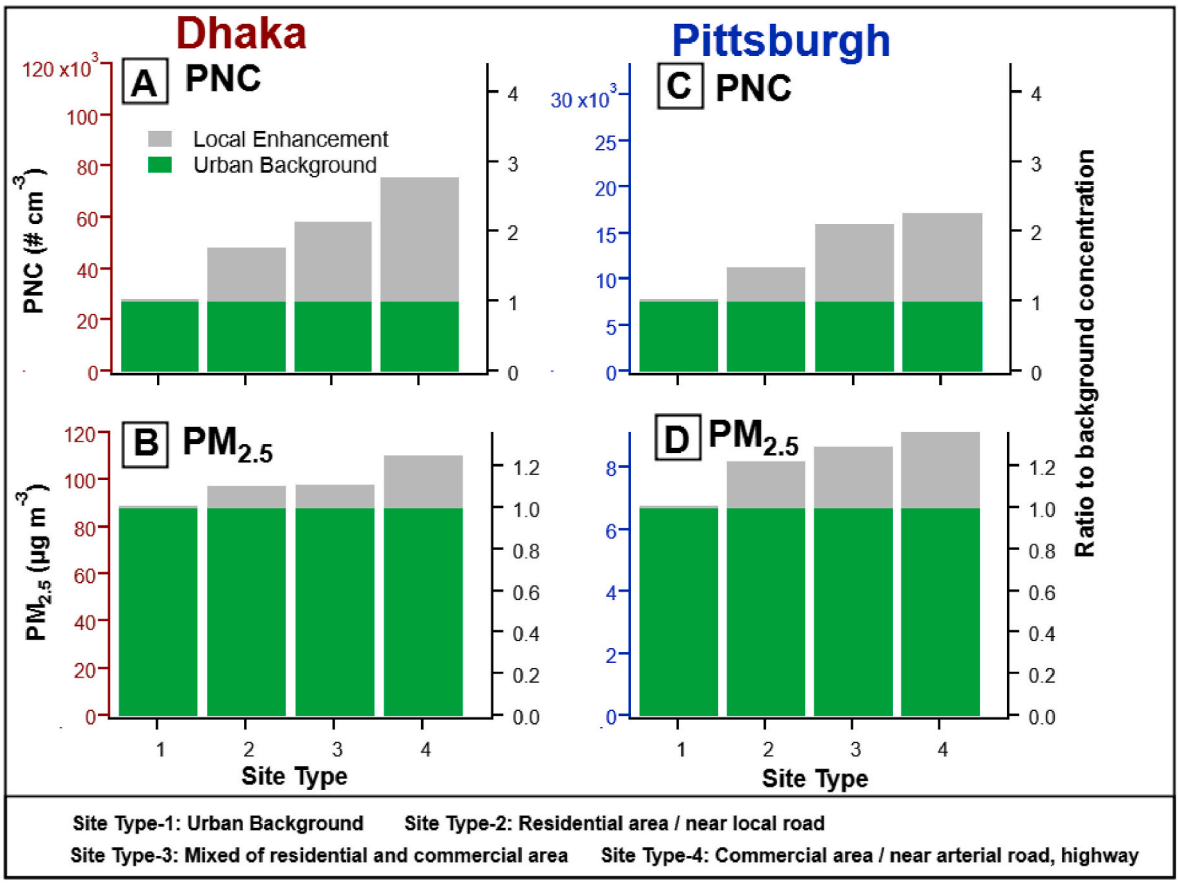

Fig. 4. Within-city local enhancement of PNC and $PM_{2.5}$ in different site types in (A–B) Dhaka and (C–D) Pittsburgh. Each bar displays the average concentrations for each site type, further categorizing the concentrations into two groups: urban background levels (green) and those likely from local emissions (gray; "local enhancement").

Pittsburgh at high-source activity areas.

Residential areas and local road sites (site type 2) exhibit a local PNC enhancement that is 78% higher than background in Dhaka and 50% higher in Pittsburgh, while $PM_{2.5}$ enhancement is 10% in Dhaka and 20% in Pittsburgh. Mixed areas (site type 3) exhibit a local PNC enhancement that is 115% higher than background in Dhaka and 110% higher in Pittsburgh, while $PM_{2.5}$ enhancement is 11% in Dhaka and 29% in Pittsburgh.

3.5. Spatial correlation between within-city variation of PNC and $PM_{2.5}$

Fig. 5 displays a scatter plot that depicts the mean PNC and $PM_{2.5}$

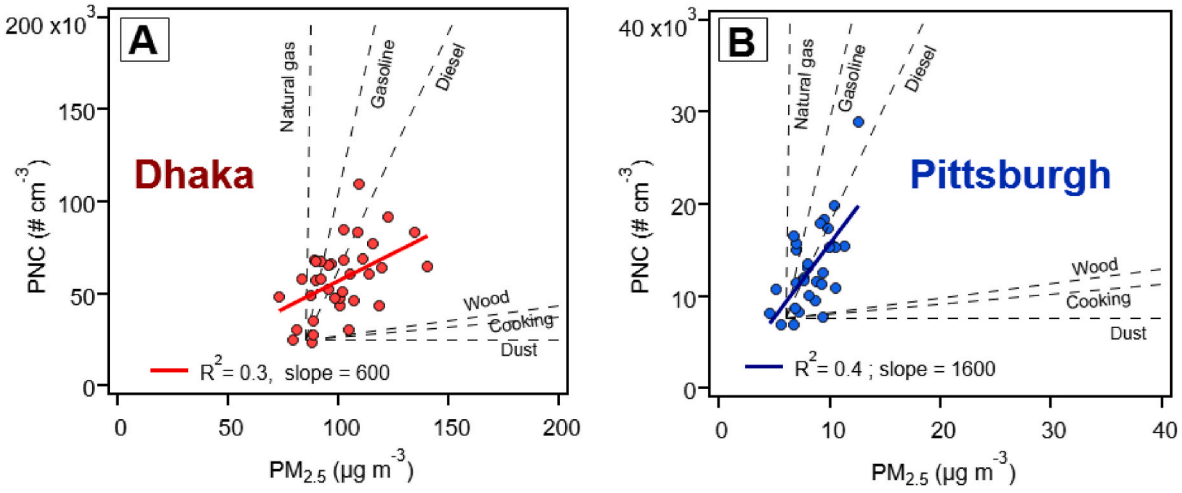

Fig. 5. Scatter plot showing the relationship between within-city spatial variation of PNC and $PM_{2.5}$. Each circle represents the average concentration measured at each site (n = 35 in Dhaka and 30 in Pittsburgh). The solid line represents the linear regression fit of PNC and $PM_{2.5}$ concentrations. The dashed black lines represent the PNC and $PM_{2.5}$ relationship for particles emitted from different sources, shown from literature (Asmi et al., 2011; Ban-Weiss et al., 2010; Hennigan et al., 2012; Kaltsonoudis et al., 2017; Saha et al., 2020), with the origin of source lines set at mean urban background PNC and $PM_{2.5}$ levels in each city.

concentrations measured at various sites, as well as their relationship, considering particles from different sources, such as natural gas, diesel and gasoline, wood smoke, cooking, and dust particles. The lines for PNC versus $PM_{2.5}$ for different sources are based on literature (Asmi et al., 2011; Ban-Weiss et al., 2010; Hennigan et al., 2012; Kaltsonoudis et al., 2017; Saha et al., 2020). Particles emitted from natural gas, gasoline, and diesel combustion are smaller in size, resulting in higher PNC per unit $PM_{2.5}$ mass, while those from wood smoke, cooking, and dust particles are larger (lower PNC per unit $PM_{2.5}$ mass).

The scatter plots of PNC and $PM_{2.5}$ provide valuable insights into the sources that may affect the observed relationship PNC and $PM_{2.5}$ in each city. The slope of PNC versus $PM_{2.5}$ is 1600 # cm^{-3} PNC per μg m^{-3} $PM_{2.5}$ for Pittsburgh and 600 # cm^{-3} PNC per μg m^{-3} $PM_{2.5}$ for Dhaka. This finding indicates that the sources and size distribution of particles that make up the majority of the $PM_{2.5}$ mass are likely different in the two cities.

In Dhaka, the average PNC to $PM_{2.5}$ slope is closer to the source line for wood smoke and dust, which have lower PNC per unit $PM_{2.5}$. Previous studies have identified that biomass burning, waste burning, and dust particles are important contributors to $PM_{2.5}$ in Dhaka city (Apte and Pant, 2019; Begum et al., 2010, 2013; World Bank, 2023). Since particles emitted from these sources are larger in size, they contribute significantly to $PM_{2.5}$ mass concentration. In Pittsburgh, the average PNC to $PM_{2.5}$ slope is closer to the source line for diesel and gasoline, indicating traffic particles likely have significant contributions to the within-city variability of PNC and $PM_{2.5}$ concentration in Pittsburgh. The different particle sources between the two cities located in different continents likely explains the observed disproportionate relationship between PNC to $PM_{2.5}$ ratios.

We found a moderate level of spatial correlation between within-city spatial variation in measured PNC and $PM_{2.5}$ in each city, with a linear regression R^2 of 0.3 for Dhaka and 0.4 for the Pittsburgh. However, we also observed variability in the spatial relationship between PNC and $PM_{2.5}$ (see Fig. 5), indicating the complexity of the sources and processes that contribute to their spatial distribution within each city. The observed relationship is consistent with measurements from other sites in the literature (Cattani et al., 2017; Eeftens et al., 2015; Saha et al., 2020; Wolf et al., 2017).

4. Conclusions

In this study, we present a comprehensive assessment of spatial variability in fine and ultrafine particle concentrations within Dhaka city, Bangladesh, utilizing a spatially dense measurement dataset of PNC and $PM_{2.5}$ collected at 35 sites. We compared the Dhaka dataset with similar measurements collected from Pittsburgh, USA, comprising 30 sites, facilitating an investigation into within-city variability across settings with distinct pollution sources and levels.

The comparison of PNC and $PM_{2.5}$ levels between Dhaka and Pittsburgh yields valuable insights into the sources and processes influencing ambient particle concentrations. In Dhaka, $PM_{2.5}$ levels are primarily driven by solid fuel combustion, including solid biomass burning, waste burning, and soil dust, which significantly contribute to particle mass due to their larger size. Conversely, in Pittsburgh, the higher PNC level per unit $PM_{2.5}$ mass suggests a notable influence of traffic particles (from diesel and gasoline combustion), which are relatively smaller in size compared to particles from solid fuel combustion and road dust. Our findings underscore the importance of controlling traffic emissions to mitigate PNC while emphasizing the necessity of substantial efforts to address accumulation mode particles originating from various region-specific sources, such as solid fuel combustion for cooking, brick manufacturing, waste burning, road dust, and other sources, to reduce $PM_{2.5}$ mass concentrations in Dhaka, Bangladesh.

CRediT authorship contribution statement

Provat K. Saha: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Writing – original draft, Writing – review & editing. **Tanbhir M. Shovon:** Data curation, Formal analysis, Writing – review & editing. **Sheikh M. Rahman:** Conceptualization, Investigation, Methodology, Writing – review & editing. **Julian D. Marshall:** Conceptualization, Investigation, Methodology, Resources, Writing – review & editing. **Allen L. Robinson:** Conceptualization, Funding acquisition, Investigation, Methodology, Writing – review & editing. **Albert A. Presto:** Conceptualization, Investigation, Methodology, Resources, Supervision, Writing – review & editing.

Declaration of competing interest

All authors declare they have no actual or potential competing financial interest.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2024.120497.

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