Atmospheric Environment 255 (2021) 118429

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: http://www.elsevier.com/locate/atmosenv

Influence of environmental conditions on the dithiothreitol (DTT)-Based oxidative potential of size-resolved indoor particulate matter of ambient origin

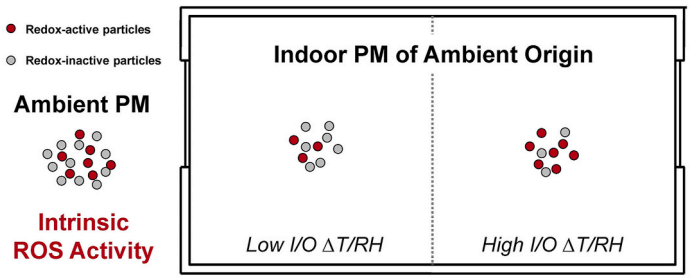
Yicheng Zeng [a], Haoran Yu [b], Haoran Zhao [c], Brent Stephens [a,**], Vishal Verma [b,*]

[a] Department of Civil, Architectural, and Environmental Engineering, Illinois Institute of Technology, Chicago, IL, USA
[b] Department of Civil and Environmental Engineering, University of Illinois at Urbana-Champaign, Urbana, IL, USA
[c] Residential Building Systems Group and Indoor Environment Group, Lawrence Berkeley National Laboratory, Berkeley, CA, USA

HIGHLIGHTS

- Oxidative potential (OP) is an emerging indicator of the toxicity of PM.
- We characterize how the transport of ambient PM from outdoors affects OP indoors.
- The volume-normalized (extrinsic) OP of $PM_{2.5}$ was higher outdoors than indoors.
- The mass-normalized (intrinsic) OP of indoor $PM_{2.5}$ was often greater than outdoors.
- The enhancement in intrinsic OP correlated with environmental conditions.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Indoor air quality
Outdoor air quality
Pollutant transformation
Toxicity
PM2.5
Oxidative potential

ABSTRACT

Much of human exposure to particulate matter (PM) of ambient origin occurs indoors. While the oxidative potential (OP) of PM is an emerging indicator of the intrinsic toxicity of PM, little is known about the OP of indoor PM of ambient origin. Here we characterize the OP of indoor and outdoor size-resolved PM in an unoccupied apartment unit in Chicago, IL, free from indoor emission sources, to explore how the transport of ambient PM from outdoors affects OP indoors. Simultaneous measurements were conducted in both indoor and outdoor locations for at least 5 consecutive days during 11 separate sampling campaigns spanning all seasons. OP was assessed by the dithiothreitol (DTT) assay. Indoor $PM_{2.5}$ concentrations were consistently lower than outdoor $PM_{2.5}$ concentrations, with an average (±standard deviation, SD) indoor/outdoor (I/O) $PM_{2.5}$ mass concentration ratio of 0.36 ± 0.10. The volume-normalized (or extrinsic) OP of the particles was much higher for the outdoor for $PM_{2.5}$ samples across all size-ranges, with an average (±SD) indoor-to-outdoor ratio (I/O) of 0.54 ± 0.57. However, the mass-normalized (or intrinsic) OP of indoor $PM_{2.5}$ samples was greater than concurrent outdoor $PM_{2.5}$ samples during 7 of the sampling periods and had an overall average ± SD I/O of 1.38 ± 0.97. The extent

* Corresponding author. Civil & Environmental Engineering, University of Illinois at Urbana-Champaign, 3230D Newmark Civil Engineering Building, 205 N. Mathews, Urbana, IL, 61801, USA.
** Corresponding author. Department of Civil, Architectural, and Environmental Engineering, Illinois Institute of Technology, Alumni Memorial Hall Room 228E, 3201 South Dearborn Street, Chicago, IL, 60616, USA.
E-mail address: vverma@illinois.edu (V. Verma).
URL: https://www.iit.edu/caee, http://built-envi.com (B. Stephens), http://cee.illinois.edu/faculty/vishalverma, http://publish.illinois.edu/aerosol-research/ (V. Verma).

https://doi.org/10.1016/j.atmosenv.2021.118429
Received 7 January 2021; Received in revised form 5 April 2021; Accepted 16 April 2021
Available online 23 April 2021

of enhancement in intrinsic OP of ambient-infiltrated indoor $PM_{2.5}$ was positively correlated with differences in indoor and outdoor temperature and relative humidity (RH). We hypothesize that changes in the intrinsic OP of ambient PM as it infiltrates indoors may be influenced by transformation processes such as size-resolved penetration, indoor deposition, evaporation of volatile chemical components with lower OP, and/or temperature/RH-dependent partitioning of redox-active substances to indoor-infiltrated PM. However, further investigations, including chemical analyses of the size-segregated indoor and outdoor PM samples and the effect of environmental factors on this chemical composition, will be needed to confirm these hypotheses.

1. Introduction

Epidemiological studies have consistently found associations between elevated concentrations of ambient particulate matter, including fine particulate matter with aerodynamic diameter less than 2.5 μm ($PM_{2.5}$), and adverse health outcomes, including mortality (Bowe et al., 2019; Brook et al., 2010; Di et al., 2017; Pope et al., 2002; Pope and Dockery, 2006; Thurston et al., 2016). However, the heterogeneous and inconsistent nature of these epidemiological associations, including high spatiotemporal variability in the strengths of associations with adverse health outcomes, suggests substantial variability in human exposures and/or the intrinsic toxicities of ambient $PM_{2.5}$ based on composition or other factors (Chen et al., 2012; Dai et al., 2014; Franklin et al, 2007, 2008; Zanobetti and Schwartz, 2009; Zhou et al., 2011). Moreover, much of human exposure to ambient $PM_{2.5}$ likely occurs indoors (Azimi and Stephens, 2018) because people spend nearly 90% of their time inside buildings (Klepeis et al., 2001) and particles of ambient origin can infiltrate and persist in buildings with varying efficiencies (Allen et al., 2012; Chen and Zhao, 2011; Kearney et al., 2014; MacNeill et al, 2012, 2014).

Although the mechanisms of action for PM-related health effects are not completely understood, an increasing body of evidence has demonstrated that PM exposure can induce oxidative stress in the body and, therefore, the oxidative potential (OP) of PM, or the capability of particles to generate reactive oxygen species (ROS) in a biologically-relevant system, may be a useful indicator of the intrinsic toxicity of PM (Bates et al., 2019). Several recent epidemiological studies have shown stronger associations between respiratory and cardiovascular health endpoints and OP of ambient $PM_{2.5}$ than with PM mass concentrations (Abrams et al., 2017; Bates et al., 2015; Delfino et al., 2013; Maikawa et al., 2016; Weichenthal et al., 2016; Yang et al., 2016).

To date, the OP of ambient PM has been characterized in numerous studies using a variety of approaches (Bates et al., 2019). The most commonly used chemical approach for characterizing the OP of PM is the dithiothreitol (DTT) assay. Under the presence of PM, DTT is oxidized by molecular oxygen, leading to the formation of DTT disulfide and superoxide radical ($\cdot O_2^-$). Thus, the DTT assay simulates a similar mechanism for the ROS generation as through the NADPH oxidation occurring in mitochondria (Kumagai et al., 1997). The consumption rate of DTT, also known as the DTT activity, has been found to be correlated with the production rate of H_2O_2 (Tong et al., 2018; Xiong et al., 2017). Both organic compounds [e.g. quinones, humic-like substances] as well as transition metals (e.g., Cu and Mn) have been shown to be active in the DTT assay (Charrier and Anastasio, 2012; Cho et al., 2005; Lin and Yu, 2011; Nicolas et al., 2015; Verma et al., 2012).

Fewer studies have investigated the OP of indoor PM of either indoor or outdoor origin, including in European office buildings (Mihucz et al., 2015; Szigeti et al, 2014, 2017) and both rural (Secrest et al., 2016) and urban (Zhan et al., 2018) homes in China. In the only study of which we are aware of the OP of PM in U.S. residences, only the particle-bound ROS (Khurshid et al, 2014, 2016) and the hydroxyl radical ($^{\bullet}OH$) generation rate in the presence of H_2O_2 (Khurshid et al., 2019) have been measured as indicators of OP. These metrics measure the ROS already present on the particle's surface or its ability to generate ROS under specific conditions. Conversely, OP is a generic property of the particle to consume cellular antioxidants directly or indirectly (i.e., by generating ROS in a biological or a surrogate-to-biological system).

Although many studies have investigated size-resolved OP of ambient aerosol (Cho et al., 2005; Godri et al., 2011; Lovett et al., 2018; Saffari et al., 2013), no studies to date have reported size-resolved OP of indoor PM of ambient origin or explored the impacts of varying environmental conditions that may influence the composition (Hodas and Turpin, 2013; Johnson et al., 2017; Avery et al., 2019) and/or size-resolution (Zhao and Stephens, 2017) of indoor PM (and therefore OP) of ambient origin. Although the U.S. Environmental Protection Agency's (EPA's) most recent Integrated Science Assessment on Particulate Matter (US EPA, 2019) acknowledges the importance of indoor exposures to PM of ambient origin, as well as the potential utility of OP as a health-relevant measure of PM, it does not make the explicit connection between ambient-to-indoor transformations of OP, largely because there is limited literature on the subject (Goldstein et al., 2020). Therefore, the goal of this work is to experimentally characterize the OP of size-resolved PM samples collected simultaneously inside and outside of an unoccupied indoor environment, free from indoor emission sources, and to explore the influence of environmental conditions on the OP of indoor PM of ambient origin.

2. Materials and methods

Sampling. Simultaneous indoor and outdoor size-resolved aerosol samples were collected using Sioutas Impactors placed inside an unoccupied apartment unit and on the roof of a mid-rise (9-story) dormitory building on the main campus of Illinois Institute of Technology, Chicago, IL USA (Carman Hall, constructed in 1953). The test apartment unit, which is described in detail in prior publications (Kunkel et al., 2017; Zhao and Stephens, 2016, 2017), is a corner unit on the 3rd floor with two exterior walls and one wall connected to another unoccupied unit. The unit is not air-conditioned and is heated by a radiant floor. The potential entry of particulate matter from the adjacent corridor was minimized by taping the only door to the corridor during measurements (Zhao and Stephens, 2017). The Sioutas Impactor has five stages of aerosol collection: <0.25 μm, 0.25–0.5 μm, 0.5–1 μm, 1–2.5 μm, and >2.5 μm in aerodynamic diameter. Particles in each size range >0.25 μm are collected onto 25-mm PTFE filters; particles smaller than the 0.25 μm cut-point of the last stage are collected onto a 37-mm PTFE after-filter. In both locations, the samplers were connected to an SKC Leland Legacy sampling pump with a flow rate of 9 L/min, verified periodically using TSI Mass Flow Meter Model 4143. Samplers were operated for at least 5 consecutive days during 11 separate sampling campaigns from Spring (2018) to Spring 2019 to capture a wide range in indoor and outdoor environmental conditions and seasonal variations in ambient PM concentrations. This long sampling duration might have caused some loss of highly volatile species but was required to ensure the collection of sufficient particle mass in each sample location for OP analysis on each size range. Three indoor samplers (kept side-by-side) were co-located and operated simultaneously with one outdoor sampler. After each sampling campaign, each filter was enclosed and sealed in aluminum foil and transported to a laboratory where they were stored at −21 °C. Size-resolved indoor and outdoor particle number concentrations (0.3–10 μm) were also measured using a TSI Optical Particle Sizer (OPS) Model 3330. Indoor temperature and relative humidity were measured using an Onset HOBO U12 logging at 1-min

intervals, and outdoor weather data were captured from the closest weather station about 500 m away from the sampling site based on 1-h interval. The indoor and outdoor weather data (i.e., temperature and RH) for each sampling set were computed by averaging all the data of the individual parameters obtained during the corresponding sampling period. The average indoor temperature inside the apartment unit was always higher than that of the outdoor environment, which is typical of this vintage of mid-rise residential building without air-conditioning (Dentz et al., 2014).

Mass concentration determination. Size-resolved mass concentrations were determined by weighing blank and field-collected filters after conditioning in a sealed chamber for at least 48 h at 25 ± 2 °C and 35 ± 5% RH. The samples were weighed 5 times using a microanalytical balance with an accuracy of 0.01 mg (AUW220D, Shimadzu, Japan). Because multiple samplers were co-located in each campaign, the total mass concentration on each size-fraction in each location was calculated as the sum of the mass across all co-located filters divided by the total volume of air sampled summed across multiple samplers.

Oxidative potential analysis. Each size-resolved sample collected from both indoor and outdoor locations was extracted into deionized (DI) water separately. Each filter was immersed into a fixed volume of DI (5 mL for 25 mm size-resolved filters; 15 mL for 37 mm PTFE filters) and was sonicated in an ultrasonic water bath for 30 min. These extracts were filtered through a 0.45 μm PTFE syringe filter to remove the insoluble components and filter fibers. The extracts were diluted to 15 μg PM/mL for the DTT assay. Extracts from the same co-located size fractions were combined and DTT assays were carried out on these combined extracts using an automated DTT instrument based on the protocol described in Fang et al. (2015). Briefly, 3.5 mL of each extracted sample was mixed with 0.5 mL of DTT and 1 mL of phosphate buffer (P-KB, pH = 7.4). At exact time intervals of 3, 15, 27, 39, and 51 min, a small aliquot (100 μL) of the reaction mixture was mixed with 500 μL DTNB, which leads to the formation of a yellow-colored complex TNB. The concentration of this complex was determined by measuring its absorbance at 412 nm with an online miniature spectrophotometer (Ocean Optics, Dunedin, FL, US) coupled to a liquid waveguide capillary cell (LWCC; World Precision Instruments, Sarasota, FL, US). DTT activity (μM/min) was then calculated as the slope of the DTT concentration decay over time. The raw DTT activities of the samples were blank corrected by subtracting the DTT activity of the filter blanks (0.48 ± 0.07 μM/min). The intrinsic mass-normalized OP (DTTm, defined as DTT activity per μg of PM mass, nmol/min/μg) and the extrinsic volume-normalized OP (DTTv, which is a function of both intrinsic OP and the mass concentration, defined as the DTT activity per m^3 of air, nmol/min/m^3) for each sample is calculated by Equations (1) and (2), respectively:

$$\mathrm{DTTm}_{sample} = \frac{\mathrm{OP}_{\mathrm{raw}}^{\mathrm{DTT}}}{\frac{\mathrm{m}_{\mathrm{sample}}}{\mathrm{V}_{\mathrm{DI}}} \times 0.7} \tag{1}$$

$$\mathrm{DTTv}_{sample} = \dot{\mathrm{m}}_{\mathrm{sample}}\mathrm{DTTm}_{sample} \tag{2}$$

where m_{sample} is the total mass of PM on the fraction of the filter used for extraction (μg), V_{DI} is the volume of DI used for extracting the filter, 0.7 is the ratio of the sample extract volume to the total reaction volume in RV, and $\dot{m}_{sample}$ is the mass concentration of the PM in different size bins (μg/m^3).

3. Results and discussion

Fig. 1 shows the resulting time-integrated indoor and outdoor size-resolved PM mass concentrations measured during each sampling campaign throughout the study duration. A full summary of indoor and outdoor sampling results is provided in the supplemental information (SI) (Table S1 to S4). The average indoor and outdoor total PM mass concentrations integrated across all sizes, including >2.5 μm, were 4.0

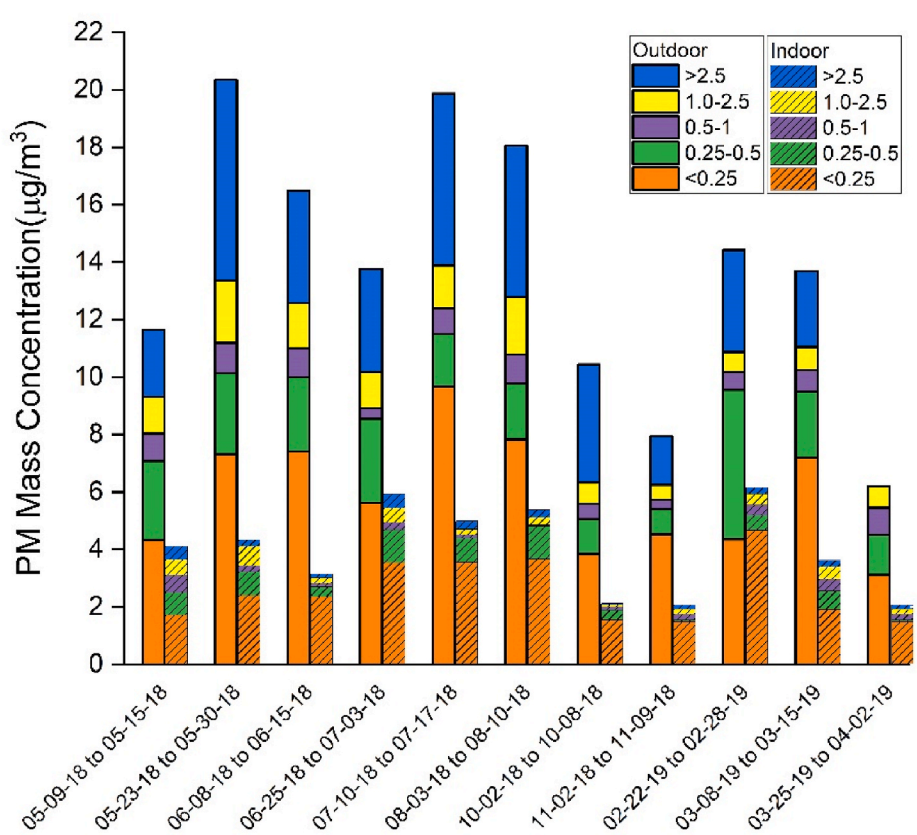


Fig. 1. Time-integrated indoor and outdoor size-resolved PM mass concentrations measured during each sampling campaign.

μg/m^3 and 13.9 μg/m^3, respectively, ranging 1.6–6.2 μg/m^3 and 6.2–20.3 μg/m^3, respectively. Similarly, the average indoor and outdoor $PM_{2.5}$ mass concentrations were 3.7 and 10.3 μg/m^3, respectively, ranging 1.6–5.9 μg/m^3 and 6.2–13.9 μg/m^3, respectively.

Indoor $PM_{2.5}$ concentrations were consistently lower than outdoor $PM_{2.5}$ concentrations, with an average (±standard deviation, SD) indoor/outdoor (I/O) $PM_{2.5}$ mass ratio of 0.36 ± 0.10, ranging from 0.24 to 0.54, which is slightly lower than what has been measured previously in the same unoccupied apartment unit using optical monitors (Zhao and Stephens, 2017). The time series of integrated $PM_{0.3\text{-}2.5}$ mass concentrations for both indoor and outdoor environments as obtained from OPS during all sampling periods are also reported in Figure S1 of the SI. The $PM_{0.3\text{-}2.5}$ mass concentrations in the outdoor samples were significantly higher (2.6–6.4 times) than those from inside the unoccupied apartment unit, which indicated no major indoor sources. Further, we did not observe any other indicators of intermittent indoor PM sources in the OPS data. For example, there were no sudden spikes in the OPS data in which indoor PM concentrations peaked higher than the concurrent outdoor PM concentrations, and we did not observe any sudden large (e.g., at least 5 μg/m^3) increases in indoor PM concentrations without concurrent increases in outdoor concentrations. Finally, indoor $PM_{0.3\text{-}2.5}$ mass concentrations followed the same trends in variation as the outdoor $PM_{0.3\text{-}2.5}$ mass concentrations, which confirmed that there were no obvious indoor sources in the unit or coming from the adjacent apartments or the hallway.

Fig. 2 shows the time-integrated extrinsic (DTTv) and intrinsic OP (DTTm) levels of indoor and outdoor $PM_{2.5}$ (integrated across all size bins <2.5 μm), as well as the I/O ratio of $PM_{2.5}$ DTTv and DTTm, from each approximately weeklong measurement period. Outdoor extrinsic OP (DTTv) of $PM_{2.5}$ samples was consistently higher than indoor DTTv (Fig. 2a), except for one measurement in February 2019, with an average (±SD) indoor-to-outdoor ratio (I/O) of 0.54 ± 0.57. This trend of significantly higher outdoor DTTv than indoors is largely driven by low I/O ratios of $PM_{2.5}$ mass concentrations. Conversely, the mass-normalized (or intrinsic) OP of indoor $PM_{2.5}$ samples was greater than concurrent outdoor $PM_{2.5}$ samples during 7 of the sampling periods and had an overall average ± SD I/O ratio of 1.38 ± 0.97, with significant seasonal variations ($p < 0.05$ from unpaired *t*-test between warmer and colder seasons, defined as an average outdoor temperature higher or lower than 15 °C, respectively) (Fig. 2b). Outdoor $PM_{2.5}$ DTTm was generally higher in warmer months compared to colder months ($p < 0.05$ in unpaired *t*-test), while no significant difference was observed for indoor $PM_{2.5}$ DTTm between warmer and colder seasons ($p = 0.24$). I/O $PM_{2.5}$ DTTm ratios were generally less than 1 during summer months and greater than 1 during fall and winter months. Over half of the $PM_{2.5}$ DTTm I/O ratios were greater than 1, with a maximum of ~4.2.

Distributions of size-resolved DTTv and DTTm I/O ratios are shown in Fig. 3. Most of the I/O ratios for DTTv were below 1 (0.04–0.7), indicating lower indoor DTTv across all particle sizes with only a few exceptions. In contrast, I/O ratios of DTTm showed a large range, varying from 0.1 to 7.3. Interestingly, the ratios were above 1 for more than 60% of the samples (34 out of 55), and appeared to be distributed uniformly throughout all particle sizes. This contrast in I/O ratios for DTTm vs. DTTv indicates a frequent enhancement in intrinsic OP as ambient PM infiltrated and persisted indoors.

To explore potential indoor and outdoor environmental determinants of indoor OP of $PM_{2.5}$ of ambient origin, we plotted I/O ratios of $PM_{2.5}$ OP (both DTTv and DTTm) versus indoor-outdoor differences in temperature (i.e., indoor minus outdoor) and outdoor-indoor differences in RH (i.e., outdoor minus indoor), as shown in Fig. 4. Modest correlations were observed between the I/O ratio of DTTv for $PM_{2.5}$ and indoor-outdoor differences in temperature (Fig. 4a; $R^2 = 0.37$; Spearman rho = 0.48, $p = 0.13$) and outdoor-indoor differences in RH (Fig. 4b; $R^2 = 0.52$; Spearman rho = 0.60, $p = 0.07$). Stronger correlations were observed for the I/O ratio of DTTm for $PM_{2.5}$ and both environmental parameters, including indoor-outdoor differences in temperature (Fig. 4c; $R^2 = 0.47$; Spearman rho = 0.69, $p = 0.018$) and outdoor-indoor differences in RH (Fig. 4d; $R^2 = 0.63$; Spearman rho = 0.80, $p = 0.004$).

Since the average indoor temperature was always greater than the average outdoor temperature during each sampling period, positive associations between indoor-outdoor temperature differences and DTTv and DTTm I/O ratios mean that as the indoor-outdoor difference increased (i.e., outdoor temperatures decreased), both the extrinsic and intrinsic OP of indoor $PM_{2.5}$ increased relative to any changes in the OP of outdoor $PM_{2.5}$. Combined with similar (and slightly stronger) associations between I/O DTT ratios (both extrinsic DTTv and intrinsic DTTm) and outdoor-indoor RH differences, we may infer that differences in indoor and outdoor environmental driving forces, including both temperature and RH, were associated with the degree of enhancement in both extrinsic and intrinsic OP as ambient PM infiltrated and persisted indoors.

As DTTm is driven largely by PM chemical composition, the stronger correlations observed between environmental parameters and DTTm compared to DTTv imply that both temperature and RH could have some influence in altering the fraction of redox-active substances as ambient PM infiltrates into the indoor environment. Due to higher indoor temperature and lower humidity, the water contained in the aerosol can evaporate, thus shrinking the aerosol after its penetration from outdoor to indoor environment. This shrinking of aerosols could yield into more concentrated redox-active components in the indoor environment, thus leading to a higher I/O ratio of DTTm, as shown in Fig. 4. However, a detailed chemical composition analysis of the PM samples will be required to confirm this hypothesis.

To further explore the influences of indoor and outdoor

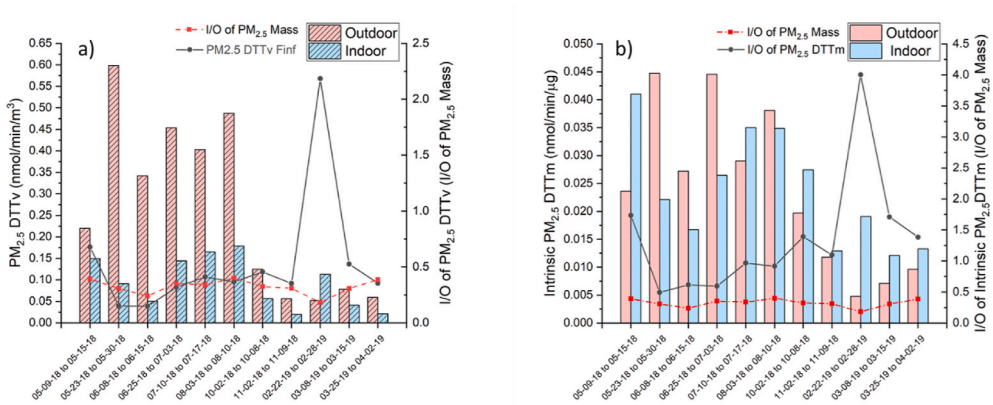


Fig. 2. Time-integrated OP for indoor and outdoor $PM_{2.5}$ samples in the unoccupied apartment unit: (a) extrinsic DTTv levels and I/O ratio of DTTv, and (b) intrinsic DTTm levels and indoor/outdoor (I/O) ratio of DTTm.

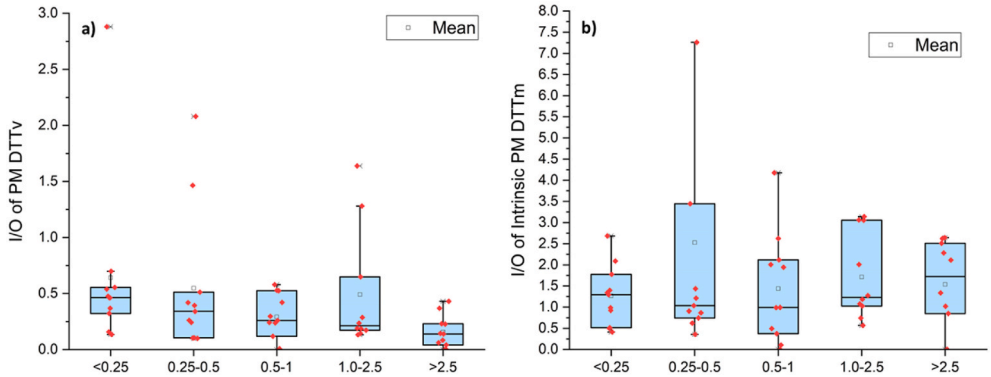


Fig. 3. Size-resolved I/O ratios of (a) DTTv and (b) DTTm levels. Boxes represent 25th and 75th percentiles; whiskers represent upper and lower adjacent values; outside values represent outliers; empty squares represent the mean; and red diamonds represent individual data points. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

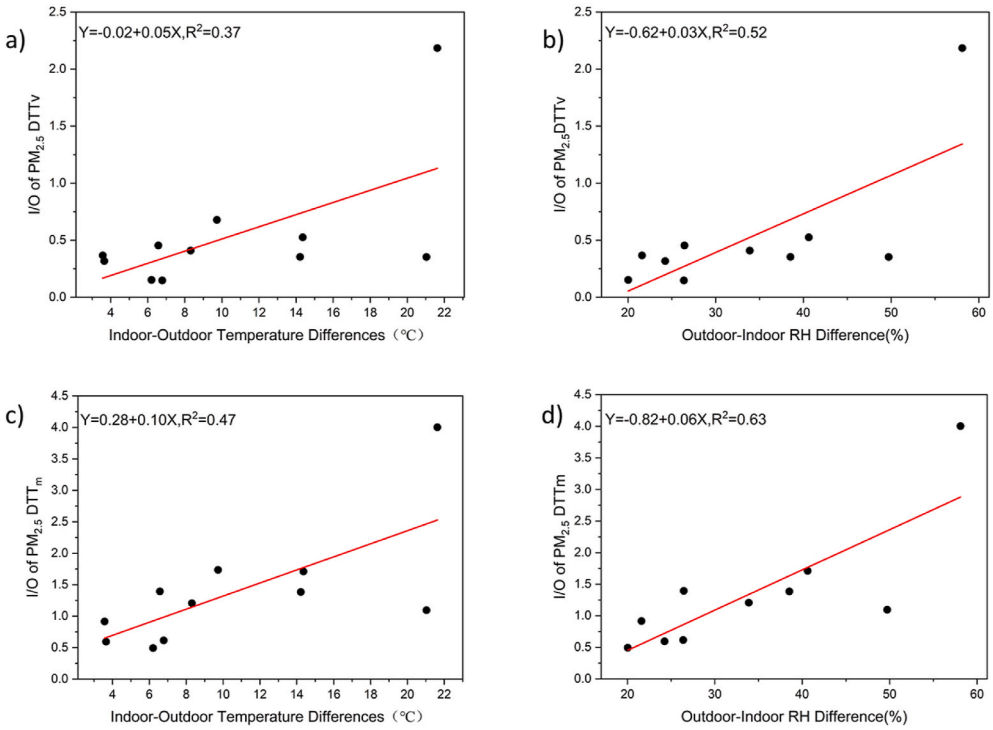


Fig. 4. Time-integrated indoor/outdoor (I/O) ratios of $PM_{2.5}$ OP from the unoccupied apartment unit versus indoor-outdoor differences in environmental conditions: (a) I/O DTTv vs. I–O temperature differences, (b) I/O DTTv vs. O–I relative humidity (RH) differences, (c) I/O DTTm vs. I–O temperature differences, and (d) I/O DTTm vs. O–I RH differences.

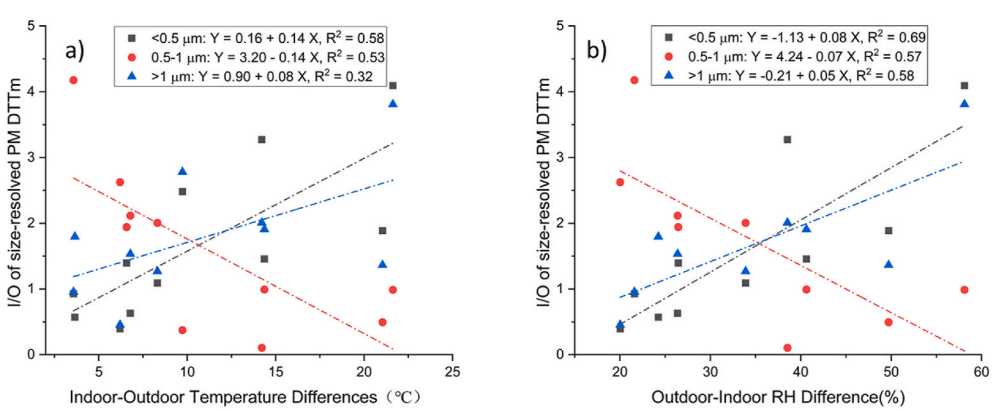


Fig. 5. The correlations of indoor/outdoor (I/O) ratios of intrinsic DTTm of size-resolved PM (i.e., <0.5 μm, 0.5–1 μm, and >1 μm) versus indoor and outdoor differences in environmental conditions: (a) I/O DTTm vs. Indoor-Outdoor (I–O) temperature differences and (b) I/O DTTm vs. Outdoor-Indoor (O–I) RH differences. The black, red and blue dash lines denote the lines of simple linear regression for $PM_{0.5}$, $PM_{0.5\text{-}1}$ and supermicron particles, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

environmental conditions on the intrinsic OP (DTTm) of PM in different size ranges, we conducted a simple linear regression analysis on size-resolved I/O ratios of DTTm for the PM samples from all size bins (including the largest >2.5 μm bin) versus indoor-outdoor temperature differences and outdoor-indoor RH differences (Fig. 5). Here, we recombined two size bins of <0.25 μm and 0.25–0.5 μm into the bin of <0.5 μm ($PM_{0.5}$), and another two size bins of 1.0–2.5 μm and >2.5 μm into the bin of >1 μm (i.e., supermicron particles), while keeping the particles in the moderate size range (i.e., 0.5–1 μm, $PM_{0.5\text{-}1}$) as is. Moderate positive correlations were observed between I/O DTTm ratios and indoor-outdoor differences in both temperature and RH for particles <0.5 μm (Temperature: $R^2 = 0.58$; Spearman rho = 0.827, p = 0.0017; RH: $R^2 = 0.69$; Spearman rho = 0.915, p = 0.0002) and for particles >1 μm (Temperature: $R^2 = 0.32$; Spearman rho = 0.624, p = 0.05; RH: $R^2 = 0.58$; Spearman rho = 0.717, p = 0.03). Conversely, a negative correlation was observed between I/O DTTm ratios and indoor-outdoor differences in both temperature and RH for 0.5–1 μm particles (Temperature: $R^2 = 0.53$; Spearman rho = −0.77, p = 0.009; RH: $R^2 = 0.57$, Spearman rho = −0.85, p = 0.004). These results suggest that intrinsic OP enhancement increased with increasing indoor-outdoor differences in temperature and RH only for the smallest and largest particles in the measured size range, while intrinsic OP decreased for particles in the middle of the size range (0.5–1 μm).

This phenomenon might be attributed to size-dependent penetration of the particles, which could alter the chemical composition of the PM within these different size bins as well as the total PM. In our previous study at the same indoor location (Zhao and Stephens, 2017), we observed a decreasing infiltration factor with increasing particle size for particles greater than 0.3 μm, which could explain higher fraction of smaller particles indoors in our study (Fig. 1). Several other studies have also reported a greater fraction of water-soluble redox-active species [like WSOC (Cabada et al., 2004; Velali et al., 2016) or transition metals (Saffari et al., 2013)] and thus higher intrinsic OP for smaller-sized particles (Cabada et al., 2004; Cho et al., 2005; Lovett et al., 2018; Saffari et al., 2013; Velali et al., 2016). Rivas et al. (2015) also reported higher infiltration factors for redox-active transition metals (e.g. Cd, V, Zn) compared to redox-inactive inorganic ions (e.g. NO_3^- and NH_4^+), further supporting a higher I/O ratio of PM DTTm for smaller particles (<0.5 μm, as shown in Fig. 3). However, this hypothesis is applicable to only smaller particles (<0.5 μm), and we do not have a clear explanation for other particle sizes (i.e., those >0.5 μm).

4. Conclusion

Our study provides the very first comparison of indoor vs. outdoor levels of a biologically relevant property (i.e., OP) of ambient PM as it infiltrates indoors. However, there were several limitations in our work. First, we limited our sampling only to a single unoccupied environment with a relatively small sample size (N = 11). The OP of the particles in an occupied environment can be substantially altered by other factors, such as occupant movement and activities, including behaviors to modify both natural (i.e., opening the windows) and mechanical (turning on/off HVAC system) ventilation, and most importantly, the presence of indoor sources. Second, the long sampling duration, which was dictated by the requirement of sufficient mass for OP analysis, can cause some negative artifacts (e.g., loss of highly volatile species), thus marginally biasing the OP towards stable (or less volatile) PM species. Third, since the DTT assay is a purely chemical method, it is limited in capturing all the possible mechanisms involved in the generation of ROS. Our previous study (Xiong et al., 2017) found that the consumption rate of DTT could not capture the redox-activity of Fe, which was active in forming hydroxyl radical (•OH) in DTT via Fenton reaction (Held et al., 1996). Similar to the DTT assay, other OP endpoints such as the consumption rate of ascorbic acid and glutathione also show different sensitivities towards transition metals (Ayres et al., 2008; Künzli et al., 2006; Mudway et al., 2005) and organic species (Calas et al., 2018) in PM. Therefore, a better assessment of OP can be obtained by employing different OP endpoints in addition to the DTT assay. Finally, due to limited mass available on the filters, we could not conduct any chemical analyses of the collected samples. Despite these limitations, our results indicate that the shifts in the OP of ambient PM as it infiltrates indoors, enhanced by the magnitude of differences in indoor and outdoor environmental (temperature and RH) conditions, may be occurring due to transformation processes that influence chemical composition differently across different PM sizes, such as size-dependent penetration through the envelope (Chen and Zhao, 2011; Liu and Nazaroff, 2001; Zhao and Stephens, 2017), indoor deposition (He et al., 2005; Wallace et al., 2013), and/or phase changes(Hodas and Turpin, 2013; Johnson et al., 2017; Avery et al., 2019). For example, we suspect this could be due to the evaporation of volatile chemical components with lower OP as ambient PM infiltrates indoors (albeit only under some indoor-outdoor environmental conditions), as such evaporation may be enhanced on smaller particle size (<0.5 μm) that count for ~80% of the total mass of indoor $PM_{2.5}$, leading to an overall increase on DTTm level of total $PM_{2.5}$, or perhaps due to the temperature and/or RH dependent partitioning of redox-active substances to indoor-infiltrated PM. Broadly, these associations highlight the importance of indoor/outdoor PM chemical composition changes over PM mass concentrations changes for OP. Future work should strive to further elucidate the role of chemical and physical drivers, such as the presence of occupants and indoor sources, building characteristics, and meteorological conditions, in altering the OP of ambient PM as it infiltrates indoors, as well as the resulting net OP of the particles in the indoor environment. This in turn can help in better characterizing the exposure of $PM_{2.5}$ to the occupants and relating this exposure to the observed health endpoints.

CRediT authorship contribution statement

Yicheng Zeng: Sampling, data collection, analysis, Writing – review & editing, Writing – original draft, Writing – review & editing. **Haoran Yu:** Sampling, data collection, analysis, Writing – review & editing. **Haoran Zhao:** Sampling, data collection, analysis, Writing – review & editing. **Brent Stephens:** Conceptualization of study design, Methodology, Writing – review & editing. **Vishal Verma:** Conceptualization of study design, Methodology, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported in part by an ASHRAE New Investigator Award to Brent Stephens and by the Armour College of Engineering at Illinois Tech. The OP analysis on the filters was supported from Vishal Verma's startup fund from UIUC.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2021.118429.

References

Abrams, J.Y., Weber, R.J., Klein, M., Samat, S.E., Chang, H.H., Strickland, M.J., Verma, V., Fang, T., Bates, J.T., Mulholland, J.A., Russell, A.G., Tolbert, P.E., 2017. Associations between ambient fine particulate oxidative potential and cardiorespiratory emergency department visits. Environ. Health Perspect. 125, 107008. https://doi.org/10.1289/EHP1545.

Allen, R.W., Adar, S.D., Avol, E., Cohen, M., Curl, C.L., Larson, T., Liu, L.-J.S., Sheppard, L., Kaufman, J.D., 2012. Modeling the residential infiltration of outdoor

PM2.5 in the multi-ethnic study of atherosclerosis and air pollution (MESA air). Environ. Health Perspect. 120, 824–830. https://doi.org/10.1289/ehp.1104447.
Avery, A.M., Waring, M.S., DeCarlo, P.F., 2019. Seasonal variation in aerosol composition and concentration upon transport from the outdoor to indoor environment. Environ. Sci.: Processes Impacts 21, 528–547. https://doi.org/10.1039/C8EM00471D.
Ayres, J.G., Borm, P., Cassee, F.R., Castranova, V., Donaldson, K., Ghio, A., Harrison, R. M., Hider, R., Kelly, F., Kooter, I.M., Marano, F., Maynard, R.L., Mudway, I., Nel, A., Sioutas, C., Smith, S., Baeza-Squiban, A., Cho, A., Duggan, S., Froines, J., 2008. Evaluating the toxicity of airborne particulate matter and nanoparticles by measuring oxidative stress potential—a workshop report and consensus statement. Inhal. Toxicol. 20, 75–99. https://doi.org/10.1080/08958370701665517.
Azimi, P., Stephens, B., 2018. A framework for estimating the US mortality burden of fine particulate matter exposure attributable to indoor and outdoor microenvironments. J. Expo. Sci. Environ. Epidemiol. https://doi.org/10.1038/s41370-018-0103-4.
Bates, J.T., Fang, T., Verma, V., Zeng, L., Weber, R.J., Tolbert, P.E., Abrams, J.Y., Sarnat, S.E., Klein, M., Mulholland, J.A., Russell, A.G., 2019. Review of acellular assays of ambient particulate matter oxidative potential: methods and relationships with composition, sources, and health effects. Environ. Sci. Technol. 53, 4003–4019. https://doi.org/10.1021/acs.est.8b03430.
Bates, J.T., Weber, R.J., Abrams, J., Verma, V., Fang, T., Klein, M., Strickland, M.J., Sarnat, S.E., Chang, H.H., Mulholland, J.A., Tolbert, P.E., Russell, A.G., 2015. Reactive oxygen species generation linked to sources of atmospheric particulate matter and cardiorespiratory effects. Environ. Sci. Technol. 49, 13605–13612. https://doi.org/10.1021/acs.est.5b02967.
Bowe, B., Xie, Y., Yan, Y., Al-Aly, Z., 2019. Burden of cause-specific mortality associated with PM $_{2.5}$ air pollution in the United States. JAMA Netw Open 2, e1915834. https://doi.org/10.1001/jamanetworkopen.2019.15834.
Brook, R.D., Rajagopalan, S., Pope, C.A., Brook, J.R., Bhatnagar, A., Diez-Roux, A.V., Holguin, F., Hong, Y., Luepker, R.V., Mittleman, M.A., Peters, A., Siscovick, D., Smith, S.C., Whitsel, L., Kaufman, J.D., 2010. Particulate matter air pollution and cardiovascular disease. Circulation 121, 2331–2378. https://doi.org/10.1161/CIR.0b013e3181dbece1.
Cabada, J.C., Rees, S., Takahama, S., Khlystov, A., Pandis, S.N., Davidson, C.I., Robinson, A.L., 2004. Mass size distributions and size resolved chemical composition of fine particulate matter at the Pittsburgh supersite. Atmos. Environ. 38, 3127–3141. https://doi.org/10.1016/j.atmosenv.2004.03.004.
Calas, A., Uzu, G., Kelly, F.J., Houdier, S., Martins, J.M.F., Thomas, F., Molton, F., Charron, A., Dunster, C., Oliete, A., Jacob, V., Besombes, J.-L., Chevrier, F., Jaffrezo, J.-L., 2018. Comparison between five acellular oxidative potential measurement assays performed with detailed chemistry on PM$_{10}$ samples from the city of Chamonix (France). Atmos. Chem. Phys. 18, 7863–7875. https://doi.org/10.5194/acp-18-7863-2018.
Charrier, J.G., Anastasio, C., 2012. On dithiothreitol (DTT) as a measure of oxidative potential for ambient particles: evidence for the importance of soluble transition metals. Atmos. Chem. Phys. 12, 11317–11350. https://doi.org/10.5194/acpd-12-11317-2012.
Chen, C., Zhao, B., 2011. Review of relationship between indoor and outdoor particles: I/O ratio, infiltration factor and penetration factor. Atmos. Environ. 45, 275–288. https://doi.org/10.1016/j.atmosenv.2010.09.048.
Chen, C., Zhao, B., Weschler, C.J., 2012. Indoor exposure to "outdoor PM10. Epidemiology 23, 870–878. https://doi.org/10.1097/EDE.0b013e31826b800e.
Cho, A.K., Sioutas, C., Miguel, A.H., Kumagai, Y., Schmitz, D.A., Singh, M., Eiguren-Fernandez, A., Froines, J.R., 2005. Redox activity of airborne particulate matter at different sites in the Los Angeles Basin. Environ. Res. 99, 40–47. https://doi.org/10.1016/j.envres.2005.01.003.
Dai, L., Zanobetti, A., Koutrakis, P., Schwartz, J.D., 2014. Associations of fine particulate matter species with mortality in the United States: a multicity time-series analysis. Environ. Health Perspect. 122, 837–842. https://doi.org/10.1289/ehp.1307568.
Delfino, R.J., Staimer, N., Tjoa, T., Gillen, D.L., Schauer, J.J., Shafer, M.M., 2013. Airway inflammation and oxidative potential of air pollutant particles in a pediatric asthma panel. J. Expo. Sci. Environ. Epidemiol. 23, 466–473. https://doi.org/10.1038/jes.2013.25.
Dentz, J., Varshney, K., Henderson, H., 2014. Overheating in multifamily residential buildings in New York. Energy Efficiency 7, 401–415. https://doi.org/10.1007/s12053-013-9228-4.
Di, Q., Wang, Yan, Zanobetti, A., Wang, Yun, Koutrakis, P., Choirat, C., Dominici, F., Schwartz, J.D., 2017. Air pollution and mortality in the medicare population. N. Engl. J. Med. 376, 2513–2522. https://doi.org/10.1056/NEJMoa1702747.
Fang, T., Verma, V., Guo, H., King, L.E., Edgerton, E.S., Weber, R.J., 2015. A semi-automated system for quantifying the oxidative potential of ambient particles in aqueous extracts using the dithiothreitol (DTT) assay: results from the Southeastern Center for Air Pollution and Epidemiology (SCAPE). Atmos. Meas. Tech. 8, 471–482. https://doi.org/10.5194/amt-8-471-2015.
Franklin, M., Koutrakis, P., Schwartz, P., 2008. The role of particle composition on the association between PM2.5 and mortality. Epidemiology 19, 680–689.
Franklin, M., Zeka, A., Schwartz, J., 2007. Association between PM2.5 and all-cause and specific-cause mortality in 27 US communities. J. Expo. Sci. Environ. Epidemiol. 17, 279–287. https://doi.org/10.1038/sj.jes.7500530.
Godri, K.J., Harrison, R.M., Evans, T., Baker, T., Dunster, C., Mudway, I.S., Kelly, F.J., 2011. Increased oxidative burden associated with traffic component of ambient particulate matter at roadside and urban background schools sites in london. PloS One 6, e21961. https://doi.org/10.1371/journal.pone.0021961.
Goldstein, A.H., Nazaroff, W.W., Weschler, C.J., Williams, J., 2020. How do indoor environments affect air pollution exposure? Environ. Sci. Technol. acs.est., 0c05727 https://doi.org/10.1021/acs.est.0c05727.
He, C., Morawska, L., Gilbert, D., 2005. Particle deposition rates in residential houses. Atmos. Environ. 39, 3891–3899. https://doi.org/10.1016/j.atmosenv.2005.03.016.
Held, K.D., Sylvester, F.C., Hopcia, K.L., Biaglow, J.E., 1996. Role of Fenton chemistry in thiol-induced toxicity and apoptosis. Radiat. Res. 145, 542. https://doi.org/10.2307/3579272.
Hodas, N., Turpin, B.J., 2013. Shifts in the gas-particle partitioning of ambient organics with transport into the indoor environment. Aerosol. Sci. Technol. 131207050823004 https://doi.org/10.1080/02786826.2013.871500.
Johnson, A.M., Waring, M.S., DeCarlo, P.F., 2017. Real-time transformation of outdoor aerosol components upon transport indoors measured with aerosol mass spectrometry. Indoor Air 27, 230–240. https://doi.org/10.1111/ina.12299.
Kearney, J., Wallace, L., MacNeill, M., Héroux, M.-E., Kindzierski, W., Wheeler, A., 2014. Residential infiltration of fine and ultrafine particles in Edmonton. Atmos. Environ. 94, 793–805. https://doi.org/10.1016/j.atmosenv.2014.05.020.
Khurshid, S.S., Emmerich, S., Persily, A., 2019. Oxidative potential of particles at a research house: influencing factors and comparison with outdoor particles. Build. Environ. 106275 https://doi.org/10.1016/j.buildenv.2019.106275.
Khurshid, S.S., Siegel, J.A., Kinney, K.A., 2016. Particulate reactive oxygen species on total suspended particles - measurements in residences in Austin, Texas. Indoor Air 26, 953–963. https://doi.org/10.1111/ina.12269.
Khurshid, S.S., Siegel, J.A., Kinney, K.A., 2014. Indoor particulate reactive oxygen species concentrations. Environ. Res. 132, 46–53. https://doi.org/10.1016/j.envres.2014.03.026.
Klepeis, N.E., Nelson, W.C., Ott, W.R., Robinson, J.P., Tsang, A.M., Switzer, P., Behar, J. V., Hern, S.C., Engelmann, W.H., 2001. The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants. J. Expo. Anal. Environ. Epidemiol. 11, 231–252. https://doi.org/10.1038/sj.jea.7500165.
Kumagai, Y., Arimoto, T., Shinyashiki, M., Shimojo, N., Nakai, Y., Yoshikawa, T., Sagai, M., 1997. Generation of reactive oxygen species during interaction of diesel exhaust particle components with NADPH-cytochrome p450 reductase and involvement of the bioactivation in the DNA damage. Free Radic. Biol. Med. 22, 479–487. https://doi.org/10.1016/S0891-5849(96)00341-3.
Kunkel, S.A., Azimi, P., Zhao, H., Stark, B.C., Stephens, B., 2017. Quantifying the size-resolved dynamics of indoor bioaerosol transport and control. Indoor Air 27, 977–987. https://doi.org/10.1111/ina.12374.
Künzli, N., Mudway, I.S., Götschi, T., Shi, T., Kelly, F.J., Cook, S., Burney, P., Forsberg, B., Gauderman, J.W., Hazenkamp, M.E., Heinrich, J., Jarvis, D., Norbäck, D., Payo-Losa, F., Poli, A., Sunyer, J., Borm, P.J.A., 2006. Comparison of oxidative properties, light absorbance, and total and elemental mass concentration of ambient PM $_{2.5}$ collected at 20 European sites. Environ. Health Perspect. 114, 684–690. https://doi.org/10.1289/ehp.8584.
Lin, P., Yu, J.Z., 2011. Generation of reactive oxygen species mediated by humic-like substances in atmospheric aerosols. Environ. Sci. Technol. 45, 10362–10368. https://doi.org/10.1021/es2028229.
Liu, D., Nazaroff, W.W., 2001. Modeling pollutant penetration across building envelopes. Atmos. Environ. 35, 4451–4462. https://doi.org/10.1016/S1352-2310(01)00218-7.
Lovett, C., Sowlat, M.H., Saliba, N.A., Shihadeh, A.L., Sioutas, C., 2018. Oxidative potential of ambient particulate matter in Beirut during Saharan and Arabian dust events. Atmos. Environ. 188, 34–42. https://doi.org/10.1016/j.atmosenv.2018.06.016.
MacNeill, M., Kearney, J., Wallace, L., Gibson, M., Héroux, M.E., Kuchta, J., Guernsey, J. R., Wheeler, A.J., 2014. Quantifying the contribution of ambient and indoor-generated fine particles to indoor air in residential environments. Indoor Air 24, 362–375. https://doi.org/10.1111/ina.12084.
MacNeill, M., Wallace, L., Kearney, J., Allen, R.W., Van Ryswyk, K., Judek, S., Xu, X., Wheeler, A., 2012. Factors influencing variability in the infiltration of PM2.5 mass and its components. Atmos. Environ. 61, 518–532. https://doi.org/10.1016/j.atmosenv.2012.07.005.
Maikawa, C.L., Weichenthal, S., Wheeler, A.J., Dobbin, N.A., Smargiassi, A., Evans, G., Liu, L., Goldberg, M.S., Pollitt, K.J.G., 2016. Particulate oxidative burden as a predictor of exhaled nitric oxide in children with asthma. Environ. Health Perspect. 124, 1616–1622. https://doi.org/10.1289/EHP175.
Mihucz, V.G., Szigeti, T., Dunster, C., Giannoni, M., de Kluizenaar, Y., Cattaneo, A., Mandin, C., Bartzis, J.G., Lucarelli, F., Kelly, F.J., Záray, G., 2015. An integrated approach for the chemical characterization and oxidative potential assessment of indoor PM2.5. Microchem. J. 119, 22–29. https://doi.org/10.1016/j.microc.2014.10.006.
Mudway, I.S., Duggan, S.T., Venkataraman, C., Habib, G., Kelly, F.J., Grigg, J., 2005. Combustion of dried animal dung as biofuel results in the generation of highly redox active fine particulates. Part. Fibre Toxicol. 2, 6. https://doi.org/10.1186/1743-8977-2-6.
Nicolas, J., Jaafar, M., Sepetdjian, E., Saad, W., Sioutas, C., Shihadeh, A., Saliba, N.A., 2015. Redox activity and chemical interactions of metal oxide nano- and micro-particles with dithiothreitol (DTT). Environ. Sci.: Processes Impacts 17, 1952–1958. https://doi.org/10.1039/C5EM00352K.
Pope, C.A., Burnett, R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K., Thurston, G.D., 2002. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. J. Am. Med. Assoc. 287, 1132–1141.
Pope, C.A., Dockery, D.W., 2006. Health effects of fine particulate air pollution: lines that connect. J. Air Waste Manag. Assoc. 56, 709–742.
Rivas, I., Viana, M., Moreno, T., Bouso, L., Pandolfi, M., Alvarez-Pedrerol, M., Forns, J., Alastuey, A., Sunyer, J., Querol, X., 2015. Outdoor infiltration and indoor contribution of UFP and BC, OC, secondary inorganic ions and metals in PM2.5 in schools. Atmos. Environ. 106, 129–138. https://doi.org/10.1016/j.atmosenv.2015.01.055.

Saffari, A., Daher, N., Shafer, M.M., Schauer, J.J., Sioutas, C., 2013. Seasonal and spatial variation in reactive oxygen species activity of quasi-ultrafine particles (PM0.25) in the Los Angeles metropolitan area and its association with chemical composition. Atmos. Environ. 79, 566–575. https://doi.org/10.1016/j.atmosenv.2013.07.058.

Secrest, M.H., Schauer, J.J., Carter, E.M., Lai, A.M., Wang, Y., Shan, M., Yang, X., Zhang, Y., Baumgartner, J., 2016. The oxidative potential of PM2.5 exposures from indoor and outdoor sources in rural China. Sci. Total Environ. 571, 1477–1489. https://doi.org/10.1016/j.scitotenv.2016.06.231.

Szigeti, T., Dunster, C., Cattaneo, A., Spinazzè, A., Mandin, C., Le Ponner, E., de Oliveira Fernandes, E., Ventura, G., Saraga, D.E., Sakellaris, I.A., de Kluizenaar, Y., Cornelissen, E., Bartzis, J.G., Kelly, F.J., 2017. Spatial and temporal variation of particulate matter characteristics within office buildings — the OFFICAIR study. Sci. Total Environ. https://doi.org/10.1016/j.scitotenv.2017.01.013.

Szigeti, T., Kertész, Z., Dunster, C., Kelly, F.J., Záray, G., Mihucz, V.G., 2014. Exposure to PM2.5 in modern office buildings through elemental characterization and oxidative potential. Atmos. Environ. 94, 44–52. https://doi.org/10.1016/j.atmosenv.2014.05.014.

Thurston, G.D., Burnett, R.T., Turner, M.C., Shi, Y., Krewski, D., Lall, R., Ito, K., Jerrett, M., Gapstur, S.M., Diver, W.R., Pope, C.A., 2016. Ischemic heart disease mortality and long-term exposure to source-related components of U.S. Fine particle air pollution. Environ. Health Perspect. 124, 785–794. https://doi.org/10.1289/ehp.1509777.

Tong, H., Lakey, P.S.J., Arangio, A.M., Socorro, J., Shen, F., Lucas, K., Brune, W.H., Pöschl, U., Shiraiwa, M., 2018. Reactive oxygen species formed by secondary organic aerosols in water and surrogate lung fluid. Environ. Sci. Technol. acs.est.8b03695. https://doi.org/10.1021/acs.est.8b03695.

Us EPA, 2019. Integrated Science Assessment for Particulate Matter. Center for Public Health and Environmental Assessment, Office of Research and Development, Research Triangle Park, NC. No. EPA/600/R-19/188.

Velali, E., Papachristou, E., Pantazaki, A., Choli-Papadopoulou, T., Planou, S., Kouras, A., Manoli, E., Besis, A., Voutsa, D., Samara, C., 2016. Redox activity and in vitro bioactivity of the water-soluble fraction of urban particulate matter in relation to particle size and chemical composition. Environ. Pollut. 208, 774–786. https://doi.org/10.1016/j.envpol.2015.10.058.

Verma, V., Rico-Martinez, R., Kotra, N., King, L., Liu, J., Snell, T.W., Weber, R.J., 2012. Contribution of water-soluble and insoluble components and their hydrophobic/hydrophilic subfractions to the reactive oxygen species-generating potential of fine ambient aerosols. Environ. Sci. Technol. 46, 11384–11392. https://doi.org/10.1021/es302484r.

Wallace, L., Kindzierski, W., Kearney, J., MacNeill, M., Héroux, M.-È., Wheeler, A.J., 2013. Fine and ultrafine particle decay rates in multiple homes. Environ. Sci. Technol. 47, 12929–12937. https://doi.org/10.1021/es402580t.

Weichenthal, S., Lavigne, E., Evans, G., Pollitt, K., Burnett, R.T., 2016. Ambient PM2.5 and risk of emergency room visits for myocardial infarction: impact of regional PM2.5 oxidative potential: a case-crossover study. Environ. Health 15, 46. https://doi.org/10.1186/s12940-016-0129-9.

Xiong, Q., Yu, H., Wang, R., Wei, J., Verma, V., 2017. Rethinking dithiothreitol-based particulate matter oxidative potential: measuring dithiothreitol consumption versus reactive oxygen species generation. Environ. Sci. Technol. 51, 6507–6514. https://doi.org/10.1021/acs.est.7b01272.

Yang, A., Janssen, N.A.H., Brunekreef, B., Cassee, F.R., Hoek, G., Gehring, U., 2016. Children's respiratory health and oxidative potential of $PM_{2.5}$: the PIAMA birth cohort study. Occup. Environ. Med. 73, 154–160. https://doi.org/10.1136/oemed-2015-103175.

Zanobetti, A., Schwartz, J., 2009. The effect of fine and coarse particulate air pollution on mortality: a national analysis. Environ. Health Perspect. 117, 898–903. https://doi.org/10.1289/ehp.0800108.

Zhan, Y., Johnson, K., Norris, C., Shafer, M.M., Bergin, M.H., Zhang, Y., Zhang, J., Schauer, J.J., 2018. The influence of air cleaners on indoor particulate matter components and oxidative potential in residential households in Beijing. Sci. Total Environ. 626, 507–518. https://doi.org/10.1016/j.scitotenv.2018.01.024.

Zhao, H., Stephens, B., 2017. Using portable particle sizing instrumentation to rapidly measure the penetration of fine and ultrafine particles in unoccupied residences. Indoor Air 27, 218–229. https://doi.org/10.1111/ina.12295.

Zhao, H., Stephens, B., 2016. A method to measure the ozone penetration factor in residences under infiltration conditions: application in a multifamily apartment unit. Indoor Air 26, 571–581. https://doi.org/10.1111/ina.12228.

Zhou, J., Ito, K., Lall, R., Lippmann, M., Thurston, G., 2011. Time-series analysis of mortality effects of fine particulate matter components in detroit and seattle. Environ. Health Perspect. 119, 461–466. https://doi.org/10.1289/ehp.1002613.