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Investigating $PM_{2.5}$ Oxidative Potential and Its Association with Chemical Constituents Measured outside of Urban Residences in Three Metropolitan Cities of India

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Background: Redox-active potent species present in fine particulate matter [PM ≤2.5 μm in aerodynamic diameter ($PM_{2.5}$)] have been suggested as one of the major sources of oxidative stress– and health-related disorders in the urban population.

Objectives: Our objective was to determine $PM_{2.5}$ oxidative potential (OP) in urban residential neighborhoods having different sources of $PM_{2.5}$ (traffic emissions, commercial, and residential activities) in three metropolitan Indian cities.

Methods: We investigated the neighborhood and seasonal variation in $PM_{2.5}$ OP across three metropolitan cities (Delhi, Mumbai, and Bengaluru) in India. Low-cost samplers were used to collect $PM_{2.5}$ outside balconies, ground floors, and first floors of residential buildings for 24 h. We used acellular assays, including dithiothreitol (DTT) and ascorbic acid (AA), to examine the particle toxicity. Bivariate and multiple linear regression analyses were conducted to examine the association of OP with the analyzed PM constituents.

Results: The extrinsic OP^{DTTv} levels, were comparable between the cities, with the highest levels observed in Delhi (mean ± standard deviation: $3.82 \pm 1.56\,\mathrm{nmol/min/m^3}$), exceeding those in Mumbai and Bengaluru by a factor of 1.03 and 1.21, respectively. For intrinsic OP, (OP^{DTTm}), Bengaluru exhibited the maximum toxicity, followed by Mumbai and Delhi. Bengaluru demonstrated significant OP variation compared with both Delhi and Mumbai. OP^{AA} showed comparable trends in both intrinsic and extrinsic variation. Further, on comparing intra-urban variability, OP^{DTTv} was highest in all cities in the high-traffic neighborhoods, ranging from 5.13 to $4.22\,\mathrm{nmol/min/m^3}$. Bengaluru residential neighborhoods were ~4 and 6 times higher in OP^{AAm} compared with Delhi and Mumbai residential neighborhoods, respectively. Among residential neighborhoods, the coefficient of divergence (COD) showed ~1.5 times higher heterogeneity in OP^{AAv} than OP^{DTTv}. Carbonaceous fractions and a few transition elements were strongly correlated ($p < 0.05$) with OP assays. In Mumbai, comparable OP^{DTTv} levels were observed in both seasons, winter and summer, suggesting that toxicity is more likely influenced by the primary-originated traffic aerosols. Water-soluble organic carbon, cobalt (Co), and vanadium (V) were the primary contributors to reactive oxygen species activity.

Discussion: Our study reveals that PM toxicity outside of residential homes in traffic-dominated neighborhoods is significant compared with other neighborhoods across all metropolitan cities. This emphasizes the potential health risks associated with PM originating from traffic sources. https://doi.org/10.1289/JHP1007

Introduction

Rapidly developing cities have been facing significant threats from air pollution and the changing climate[1] that could have detrimental effects on human health and the environment. In low- and middle-income countries, such as India, metropolitan cities have experienced worse air quality than cities in developed countries.[2] Fine particulate matter [PM ≤2.5 μm in aerodynamic diameter ($PM_{2.5}$)] mass concentration in these areas often exceeds the World Health Organization's recommended levels (average 24-h recommended level is $60\,\mu\mathrm{g/m^3}$) for almost all days of the year.[3,4] However, it remains unclear whether $PM_{2.5}$ mass concentration surpassing the recommended levels directly correlates with negative impacts on human health.[5,6] This is because $PM_{2.5}$ mass consists of various chemical components, including carbonaceous species,[7,8] metals,[9,10] ions,[11,12] and others, with their composition varying by place. Not all species present in $PM_{2.5}$ may be associated with toxicity[13,14]; further research is therefore necessary to investigate the toxicity associated with $PM_{2.5}$.

$PM_{2.5}$ is composed of redox-active species, such as organic carbon (OC), water-soluble carbon (WSOC), and metals [e.g., iron (Fe), copper (Cu), zinc (Zn), manganese (Mn), chromium (Cr), nickel (Ni)]. These species can generate reactive oxygen species (ROS), such as hydroxyl, superoxide, and peroxide radicals, as well as others. If the production of ROS exceeds the antioxidants capacity present in the body, it can lead to inflammation and cell death. The measurement of the capability of these particles to generate ROS is known as oxidative potential (OP).[15–17] There are various assays to measure the OP, including acellular and cellular assays. Acellular assays, employing, for example, dithiothreitol (DTT),[5,18,19] ascorbic acid (AA),[9,20–22] epithelial lung lining fluid, are cell-free methods that use chemical agents as surrogates for the antioxidants in the human body. Among all the assays, the agents DTT and AA have been widely used all over the world because they are easy to employ and cost effective, and they are complementary to each other given that they target different molecules.[3–5] Studies have shown that DTT is more sensitive to organics, quinones, and metals,[6–8] whereas AA is more sensitive to transition metals.[9–11]

Numerous studies[13,23–26] have investigated the OP of PM in various locations, including ambient and indoor locations,[23,27] as well as the specific PM sources, using both DTT and AA assays. These studies have shown that in addition to the chemical composition, seasonal variations play a significant role in the OP variability of PM.[28–32] The meteorological differences during seasonal shifts influence the composition and types of sources, leading to diverse chemical compositions and, as a consequence, the generation of ROS varies in different seasons.[33–36] In Asia, a limited number of studies have examined the toxicity of PM through OP at multiple

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spatial scales. For instance, annual OP variations were examined in Gucheng[37] and Beijing[36] in China, and seasonal and spatial variations were examined in Yokohama and Noto in Japan.[38] A recent study in Lahore and Peshawar in Pakistan[29] reported that the OP levels during winters exceeded 5 times those observed in Western countries. Moreover, few studies have been conducted for the Indian subcontinent. For example, the OP of the PM fraction $\leq 10\ \mu m$ in aerodynamic diameter (PM_{10}) was examined at rural (Mt. Abu, Rajasthan)[39] and industrial (Patiala, India)[31] sites, in multiple locations in Ahmedabad,[40] at a specific source study in Bengaluru,[41] and in the urban slums of Mumbai,[23] as well as in marine aerosols in Bhubaneshwar.[42] Recently, a study reported OP levels in New Delhi during post-monsoon and winter through real-time analysis.[35] However, all these studies were carried out in a single city and mostly for one specific season. Moreover, almost all these studies employed the DTT assay in examining the OP of $PM_{2.5}$.

Given that OP serves as an indicator of oxidative stress caused by PM, it is crucial to examine the OP consumption of reducing species such as DTT or AA. Redox-active species present in PM are dependent on factors such as the PM emission source, chemical composition, and meteorology. This type of examination is especially important in areas where most urban dwellers reside. In India, more than two-thirds of the population reside in metropolitan cities,[43,44] which are often associated with high commercialization and dense traffic networks, resulting in poor air quality. Existing OP studies are typically conducted at fixed sites and lack assessments of spatial variations; given that the Indian subcontinent experiences diverse climatic conditions and geographical features, spatial variations in OP are expected.

To the best of our knowledge, no studies have been conducted on Indian-based OPs at residential outdoor areas in distinct neighborhoods for the $PM_{2.5}$ fraction through multiple assays. Given that the PM is composed of metals as well as organics, DTT and AA are the appropriate assays to measure the OP of $PM_{2.5}$. Therefore, this study aimed to understand the OP of $PM_{2.5}$ in three different types of urban residential neighborhoods: traffic, commercial, and purely residential. Further, the relationship between OP and chemical constituents was examined and the effect of the season was assessed using multiple acellular assays.

Methods

Sampling Site

The study area comprised three Indian metropolitan cities: Mumbai, Bengaluru, and Delhi, and a detailed description of each site and selection of homes can be found elsewhere.[45] A brief summary is provided here. Homes in these metropolitan cities were selected to be part of the Climate, Air pollution and Skin Aging in Indian women (CASAI) study. The participants in this study comprised low-to-medium socioeconomic status individuals (1,500 in total; 500 participants from each city). Air pollution monitoring was conducted in each city, and a subset of two to three residences (with consenting participants) participated in the $PM_{2.5}$ sampling. The distribution of the homes and their nearby Central Pollution Control Board station is depicted in Figure S1. Three residential neighborhoods were selected in each city: *a*) an area influenced by high traffic, *b*) a purely residential area, and *c*) a commercial area, defined by industrial emissions within 1 km. These classifications were selected to compare how different neighborhoods' activities affect the concentrations, variations, and OP of $PM_{2.5}$.

$PM_{2.5}$ Monitoring and Meteorological Data

Biweekly $PM_{2.5}$ measurements were conducted outdoors at selected homes during the winter season in the three different cities. The measurements were taken on balconies, ground floors, and first floors, when ground floors were not accessible, in Delhi (9–22 January 2020), Mumbai (25 November–9 December 2019), and Bengaluru (14–28 December 2019). In addition, sampling was repeated during the summer season (24 May–7 June 2019) in Mumbai only owing to the unavailability of samplers. $PM_{2.5}$ samples (in micrograms per meter cubed) were collected on 47-mm diameter polytetrafluoroethylene (PTFE) filters using low-cost samplers (Tactical Air Sampler, AirMetrics) operated at a flow rate of 5 L/min for 24 h. Each of the sites—traffic, commercial, and residential—was sampled daily over a 2-wk period, yielding a total of 42 PTFE samples per city (Delhi, Mumbai, and Bengaluru). The PTFE samples were collected for gravimetric, toxicological, and elemental analysis. Five quartz samples were collected from each site for a total of 15 quartz samples per city. The quartz substrates were used for carbonaceous analysis. Both the quartz ($n = 45$) and the PTFE ($n = 126$) samples were stored at −4°C before undergoing chemical analysis.

The daily meteorological data, including wind speed (WS, in meters per second), relative humidity (RH, in percentage), and ambient temperature (AT, in degrees Celsius), were obtained from the air quality monitoring station (maintained by the Central Pollution Control Board, India) closest to our study sites (Figure S1). For the mixing layer height (MLH; in meters), the data was obtained from the National Oceanic and Atmospheric Administration (NOAA) government website (https://www.ready.noaa.gov/READYamet.php).

OP Analysis

Post-weighed filters were cut and sonicated in ultrapure water (resistivity: 18 mΩ) for 1 h. The resulting suspension was filtered using a nylon syringe [PALL IC Acrodisc (PES), 0.45 μm, 13 mm]. These filtered samples were then subjected to both chemical and OP characterization.

OP analyses were performed on the water-extracted PTFE filters using two different acellular assays: *a*) OP^{DTT} and OP^{AA}. For the OP^{DTT} assay, we used dithiothreitol (DTT; Sigma-Aldrich, D0632), 5,5-dithiol-bis-(2-nitrobenzoic acid) (DTNB; Sigma-Aldrich, D8130) indicator, and 0.1 M potassium phosphate buffer (EMPARTA ACS). For the OP^{AA} assay, we used L-AA (ResearchLabs, 0190D) and phosphate-buffered saline (PBS; Sigma-Aldrich, P4417). The OP^{DTT} protocol was based on the adoption by Anand et al.[23] and Raparthi et al.,[14] and the OP^{AA} protocol was based on the adoption by Massimi et al.[18]

An aliquot of the $PM_{2.5}$ sample was mixed with 3 mL of the DTT solution (0.1 mM) in an amber vial and shaken for 5, 10, 15, 25, 40, and 60 min using a shaker at 37°C. Then, 0.5 mL of the reaction mixture was pipetted out and added to an amber vial containing 50 μL of 10.0 mM DTNB to quench the reaction.[14,23] Next, 200 μL of the bright-colored 5-mercapto-2-nitrobenzoic acid (TNB) complex, the final oxidized product, was dispensed into 96-well plates (nonsterile, flat bottom, TARSON plates), and the absorbance was recorded at 412 nm using a spectrophotometer (Tecan Infinite M NANO$^+$ reader). The whole experiment was conducted in a dark room because it is a light-sensitive OP analysis. For the OP^{AA} assay, each sample was run as a duplicate. We added 0.165 μL of the $PM_{2.5}$ extract with 20 μL of AA (2 mM) and 20 μL of PBS (0.5 mM) in 96-well plates (Hëllma Analytics). The absorbance was recorded at 265 nm every 2 min for 1 h using a spectrophotometer (Tecan Infinite M NANO$^+$ reader).[18,34,46] Each sample was run as a duplicate.

For quality control and assurance of experimental analysis, 9,10-phenathroquinone (9,10-PQN, 1 μM) was used as a positive control and ultrapure water was used as a negative control. The positive control was prepared in dimethyl sulfoxide (DMSO; 99.5% analytical reagent grade). Multiple concentrations were prepared at 0.1, 0.15, 0.20, 0.25, 0.5, 0.1 μm, and the DTT depletion rate was plotted against time as illustrated in Figure S2. The DTT rate obtained from different 9,10-phenathroquinone concentrations lay in the range of (3.54–4.22 nmol/min), which aligns with the reported literature.[47–49]

Chemical Analysis

The detailed chemical (carbonaceous and non-carbonaceous) and optical analyses [$b_{abs\text{-}370}$ (absorbance at 370 nm) and $b_{abs\text{-}880}$] are described in the companion paper.[45] Briefly, the carbonaceous analysis, involving the determination of OC and elemental carbon (EC) fractions, was performed on quartz filters using the Multiwavelength Carbon Analyzer (DRI model 2015) with the IMPROVE A protocol.[50,51] Elemental analysis (total metals) was carried out on PTFE filters using inductively coupled plasma mass spectrometry for transition elements, including Fe, molybdenum (Mo), Zn, lead (Pb), Mn, Cu, titanium (Ti), vanadium (V), cobalt (Co), Ni, selenium (Se), and strontium (Sr). In addition, water-extracted quartz samples were subjected to WSOC through a total organic carbon analyzer (SHIMADZU) and b_{abs} through ultraviolet–visible spectrophotometry.[52] In addition, ionic species [sodium ion (Na^+), ammonium (NH_4^+), potassium ion (K^+), magnesium ion (Mg^{2+}), calcium ion (Ca^{2+}), chloride ion (Cl^-), nitrate ion (NO_3^-), and sulfate ion (SO_4^{2-})] were examined using ion chromatography. Pearson correlation analyses were conducted separately for WSOC, water-soluble $b_{abs\text{-}365}$, $b_{abs\text{-}370}$, and $b_{abs\text{-}880}$ from biweekly measurements on PTFE filters from each city ($n = 42$). However, when conducting correlations with chemical constituents (organics and ions) against OP, the data points were restricted to the 15 measurements from quartz filters.

Estimating the Intra-Urban Variability

To examine the intra-urban variability of OP assays, which included traffic, commercial, and residential neighborhoods, the coefficient of divergence (COD) was employed.[53,54] The COD was calculated using Equation 1:

$$COD_{jk} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{x_{ij}-x_{ik}}{x_{ij}+x_{ik}}\right)^{2}}, \tag{1}$$

where x_{ij} is the individual concentration (ith) measured at a given site during a sampling period, with n representing the number of observations taken at two distinct sites, j and k.[52,54] COD values ranged from 0 to 1, where 0 indicates complete homogeneity, and higher values indicate increasing levels of heterogeneity (values >0.2 are typically indicative of spatial heterogeneity).

Data Analysis

Outliers were defined as data points outside of the range of the mean ±4 standard deviations (SDs) (to include 99.9% of data) and removed from the raw data of the OP assays.[45,55] Student's t-tests and one-way analysis of variance (with post hoc Tukey's test) were run to examine the weekday and weekend OP variation. To investigate the relationship of both OP assays with different chemical species, a Pearson correlation was performed. Statistical significance was evaluated at the 95% confidence level. Stepwise multiple linear regression (MLR) analysis was conducted for the city of Mumbai, combining data from both seasons, to determine the contribution of each component to the OP, as shown in Equation 2:

$$OP^{DTTv} = \sum_{i=1}^{n}\beta_i X_i + \beta_o, \tag{2}$$

where OP^{DTTv} is the volume-normalized DTT activity, n is the number of chemical species, X_i and β_i respectively represent the concentration and coefficient of the ith chemical species, and β_o is the constant term. The model could not be developed for other cities owing to limited data points. Data curation was done using MS Excel, and statistical modeling (bivariate and MLR) was performed using R Studio (version 3.4; RStudio Team) and SPSS (version 22; IBM). Origin software (version 2022; OriginLab) was used for graphics and data visualization. A map of the study sites was created using ArcGIS (version 10.6; ESRI), and the geocodes and corresponding OP^{DTT} values were stored as a comma-separated value file (Excel Table S4) and projected as bubble plots on the world map.

Results and Discussion

Intercity Comparison

All three metropolitan cities showed significant variation in the mass concentration levels across the sites owing to their distinct climatic conditions and geographical locations. Delhi is in the north-central part of India with a temperature range of 5–45°C and a humidity range of 55.4%–98.2%, and Bengaluru has a temperate climate with a temperature in range of 18–36°C, similar to that of Delhi. Mumbai lies on the west side of the country, near the Arabian sea and witnesses less humidity and less fluctuation in temperature ($\sim 27°C$) and humidity ($\sim 57\%$). The descriptive summary of $PM_{2.5}$ and other meteorological variables is presented in Table 1. Mean RH ranged from 67% to 76%, AT ranged from 13°C to 27°C, and MLH ranged from 256 to 453 m during the winter. The average $PM_{2.5}$ concentrations in Delhi were significantly higher (219.33 ± 116.66) compared with those in Mumbai (131.26 ± 71.83) and Bengaluru ($86.24 \pm 80.35\ \mu g/m^3$).

Figure 1 shows the intercity OP variation through two different assays, OP^{AAv} and OP^{DTTv}, expressed in volume-normalized units. OP^{DTTv} levels were found to be comparable among all the cities; highest for Delhi (mean ± SD: 3.82 ± 1.56), followed by Mumbai (3.71 ± 1.61) and Bengaluru (3.15 ± 1.72 nmol/min/m^3). The toxicity of $PM_{2.5}$, through OP^{AAv}, was found to be highest in Delhi (0.25 ± 0.19), followed by Bengaluru (0.21 ± 0.15) and Mumbai (0.17 ± 0.12 nmol/min/m^3). During our biweekly sampling period, no significant variations were observed in OP and $PM_{2.5}$

Table 1. Descriptive summary (nean ± SD) of meteorological parameters ($n = 14$ from each city) and fine particulate matter ($PM_{2.5}$) ($n = 42$ from each city) for all the study sites in winter ($n = 126$) and Mumbai in summer ($n = 14$), 2019–2020.

Meteorological parameters and $PM_{2.5}$	Winter			Summer
	Mumbai	Delhi	Bengaluru	Mumbai
RH (%)	67.45 ± 5.09	61.14 ± 12.13	72.90 ± 5.88	75.4 ± 2.23
WS (m/s)	1.0 ± 0.42	0.81 ± 0.53	0.89 ± 1.11	1.08 ± 0.29
AT (°C)	27.34 ± 1.01	13.82 ± 2.77	24.62 ± 1.68	30.39 ± 0.67
MLH (m)	293.34 ± 54.13	256.52 ± 39.37	442.96 ± 50.60	420.05 ± 73.46
$PM_{2.5}$ ($\mu g/m^3$)	131.26 ± 71.83	219.33 ± 116.66	86.24 ± 80.35	68.38 ± 34.63

Note: The meteorological parameters data were obtained from the Central Pollution Control Board, India. Data collection duration, for winter [Mumbai (25 November–9 December 2019), Delhi (9–22 January 2020), Bengaluru (14–28 December 2019)] and for summer [Mumbai (24 May–7 June 2019)]. AT, ambient temperature; MLH, mixing layer height; $PM_{2.5}$, fine particulate matter (obtained from field measurement); RH, relative humidity; WS, wind speed.

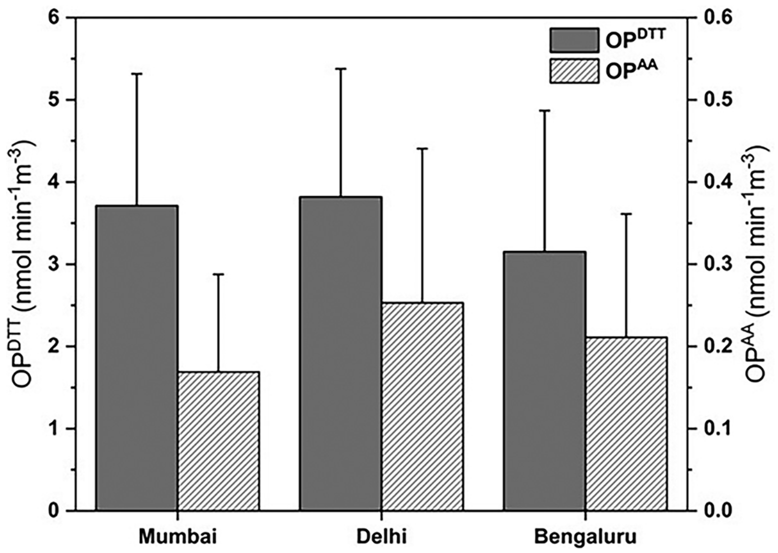

Figure 1. Comparison of volume-normalized dithiothreitol (DTT) and ascorbic acid (AA) oxidative potential assays among three different metropolitan cities: Mumbai, Delhi, and Bengaluru ($n = 42$ from each city). Columns bars and error bars showing mean and standard deviations, respectively. Corresponding numeric data are provided in Excel Table S1. Note: OP^{AA}, ascorbic acid oxidative potential acellular assay; OP^{DTT}, dithiothreitol oxidative potential acellular assay.

values between weekdays and weekends, among neighboring sites, or between cities, as shown in Table 2 (for OP assays) and Table S1 (for $PM_{2.5}$).

On the other hand, intrinsic (mass-normalized) OP activity (OP^{DTTm}) was highest in Bengaluru (0.051 ± 0.035), followed by Mumbai (0.031 ± 0.013) and Delhi (0.022 ± 0.015 nmol/min/μg). Bengaluru showed significant OP variation compared with both Delhi ($p = 0.00001$) and Mumbai ($p = 0.002$). The other assay, OP^{AAm}, showed a trend similar to that of OP^{DTTm} for the three cities, Bengaluru, 0.003 ± 0.002; Mumbai, 0.001 ± 0.0010; and Delhi, 0.001 ± 0.0009 nmol/min/μg (Figure S3).

Intra-City Comparison

Figure 2A–C depicts the intra-city comparison for all the three distinct residential neighborhoods of Mumbai, Delhi, and Bengaluru. OP^{DTTv} at high-traffic neighborhoods in each city were reported for Mumbai as 5.13 ± 2.06, for Delhi as 4.27 ± 2.05, and for Bengaluru as 4.22 ± 1.95 nmol/min/m^3. Across all the cities, commercial and residential neighborhoods had ~1.6-fold lower OP^{DTT} activity than high-traffic neighborhoods. In Mumbai, the OP^{DTTv} for the commercial neighborhood was 2.99 ± 1.38 nmol/min/m^3 ($p = 0.02$), and for the residential neighborhood, it was 3.02 ± 1.38 nmol/min/m^3 ($p = 0.03$). These values differed significantly from the DTT activity observed in the Mumbai high-traffic neighborhood. In Bengaluru, both the commercial and residential neighborhoods were comparable with each other, 2.63 ± 2.17 and 2.61 ± 1.03 nmol/min/m^3. However, in Delhi, the residential neighborhood had a slightly higher DTT activity (3.97 ± 1.38 nmol/min/m^3) than the commercial neighborhood (3.21 ± 1.25 nmol/min/m^3).

The OP^{AAv} assay, in contrast to the OP^{DTTv} assay, showed incongruence for traffic-influenced neighborhoods in Mumbai, Delhi, and Bengaluru, with mean values of 0.16 ± 0.09, 0.31 ± 0.27, and $0.10 \pm 0.0.07$ nmol/min/m^3, respectively. In commercial neighborhoods, mean OP^{AAv} values were as follows for Mumbai (0.22 ± 0.20), for Delhi (0.20 ± 0.15), and for Bengaluru (0.29 ± 0.25 nmol/min/m^3). On the contrary, the residential neighborhood of Mumbai, at 0.13 ± 0.07 nmol/min/m^3, showed significant variation compared with the residential neighborhoods of Delhi [0.25 ± 0.15 nmol/min/m^3 ($p = 0.03$)] and Bengaluru [0.32 ± 0.20 nmol/min/m^3 ($p = 0.001$)]. Variability in OP^{AAv} was observed within cities, such as in Bengaluru between commercial and high-traffic neighborhoods ($p = 0.02$), as well as in Mumbai between residential and commercial neighborhoods ($p = 0.04$).

In the case of intrinsic OP (Figure S4) for all the three cities, OP^{DTTm} for the high-traffic neighborhood was found to be the slightly higher in Bengaluru (0.074 ± 0.052) than in Mumbai (0.046 ± 0.016), but significantly higher than in Delhi [0.024 ± 0.021 nmol/min/μg ($p = 0.0002$)]. In addition, among the two neighborhoods, the Bengaluru high-traffic neighborhood was significantly higher than the residential ($p = 0.02$) and commercial ($p = 0.01$) neighborhoods. In the commercial neighborhoods, OP levels were as follows for Bengaluru, 0.040 ± 0.034, for Mumbai, 0.026 ± 0.014, and for Delhi, 0.020 ± 0.010 nmol/min/μg. In the residential neighborhoods, OP levels were as follows for Bengaluru,

Table 2. Descriptive summary (mean ± SD) of weekdays ($N = 10$) and weekends ($N = 4$) for all the residential study locations (inter and intra-city) for winter season dithiothreitol (DTT) assay and ascorbic acid (AA) assay measuring oxidative potential (OP, in nanomoles per minute per meter cubed) of $PM_{2.5}$, 2019–2020.

		High traffic		Commercial		Residential		Overall	
Cities/site	Time period	OP^{DTT}	OP^{AA}	OP^{DTT}	OP^{AA}	OP^{DTT}	OP^{AA}	OP^{DTT}	OP^{AA}
Mumbai	Weekday	5.42 ± 2.34	0.17 ± 0.14	2.65 ± 1.28	0.24 ± 0.22	2.67 ± 1.37	0.12 ± 0.07	3.58 ± 1.66	0.18 ± 0.13
	Weekend	4.30 ± 1.29	0.14 ± 0.07	3.89 ± 1.58	0.13 ± 0.13	3.98 ± 1.22	0.12 ± 0.05	4.06 ± 1.36	0.13 ± 0.08
Delhi	Weekday	5.42 ± 2.34	0.17 ± 0.10	2.65 ± 1.28	0.24 ± 0.22	4.10 ± 1.39	0.26 ± 0.16	4.06 ± 1.67	0.22 ± 0.16
	Weekend	4.33 ± 1.59	0.23 ± 0.11	3.91 ± 0.61	0.27 ± 0.23	3.56 ± 1.63	0.20 ± 0.10	3.93 ± 1.27	0.23 ± 0.15
Bengaluru	Weekday	3.99 ± 2.08	0.15 ± 0.03	2.49 ± 2.42	0.26 ± 0.27	2.45 ± 107	0.27 ± 0.20	2.98 ± 1.86	0.23 ± 0.22
	Weekend	4.80 ± 1.97	0.09 ± 0.07	2.99 ± 2	0.36 ± 0.19	3.02 ± 1.08	0.44 ± 0.18	3.60 ± 1.68	0.29 ± 0.14

Note: $PM_{2.5}$, fine particulate matter; SD, standard deviation.

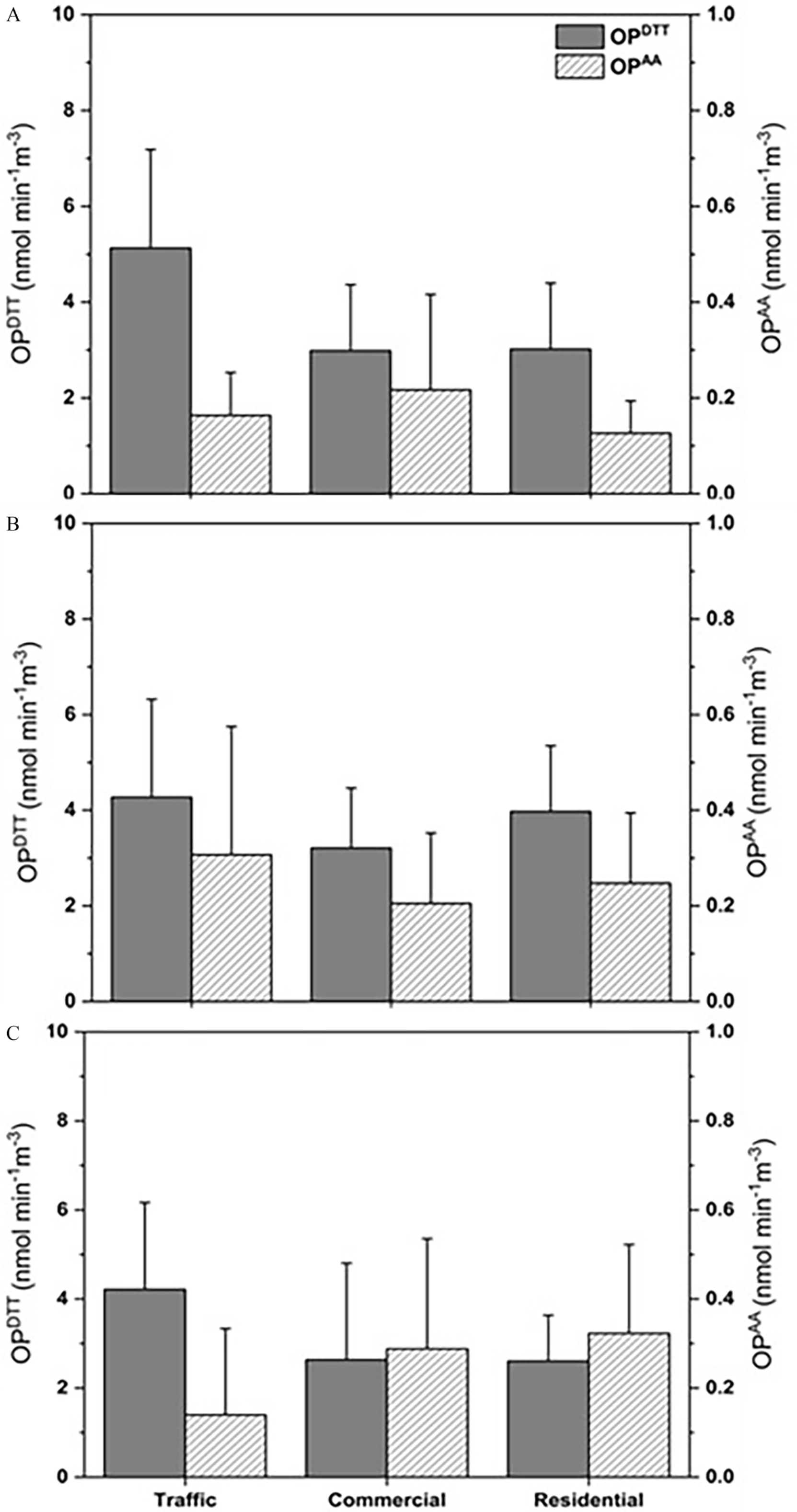

Figure 2. Comparison of volume-normalized dithiothreitol (DTT) and ascorbic acid (AA) oxidative potential assays (high traffic, commercial, and residential) of three metropolitan cities: (A) Mumbai, (B) Delhi, and (C) Bengaluru ($n = 14$ from each site). Columns bars and error bars showing mean and standard deviations, respectively. Corresponding numeric data are provided in Excel Table S2. Note: OP^{AA}, ascorbic acid oxidative potential acellular assay; OP^{DTT}, dithiothreitol oxidative potential acellular assay.

Table 3. Coefficient of divergence (COD) for the dithiothreitol (DTT) assay and ascorbic acid (AA) assay measuring oxidative potential (OP) of $PM_{2.5}$ collected at three locations in Mumbai, Delhi, and Bengaluru, India, 2019–2020 ($N = 42$, each city, 14 from each neighborhood).

		COD	
Metropolitan cities	Neighborhoods[a]	DTT	AA
Mumbai	Traf-Comm	0.40	0.43
	Comm-Resi	0.29	0.46
	Traf-Resi	0.36	0.38
Delhi	Traf-Comm	0.33	0.45
	Comm-Resi	0.34	0.51
	Traf-Resi	0.17	0.41
Bengaluru	Traf-Comm	0.42	0.58
	Comm-Resi	0.25	0.48
	Traf-Resi	0.30	0.57

Note: Comm-Resi, commercial-residential; $PM_{2.5}$, fine particulate matter; Traf-Comm, traffic-commercial; Traf-Resi, traffic-residential.

[a] Neighborhoods are defined as Traf-Comm (examining spatial heterogeneity within the city at two different sites influenced by traffic and industrial emissions), Comm-Resi (examining spatial heterogeneity within the city at two different sites influenced by pure residential emissions and industrial emissions), and Traf-Resi (examining spatial heterogeneity within the city at two different sites influenced by traffic and pure residential emissions).

0.039 ± 0.021, for Mumbai, 0.021 ± 0.010, and for Delhi, 0.023 ± 0.015 nmol/min/µg.

The OP^{AAm} assay levels were the highest in high-traffic neighborhoods in Mumbai, followed by in Delhi and then Bengaluru, with average values of 0.002 ± 0.001, 0.0013 ± 0.0012, and 0.0012 ± 0.0.0009 nmol/min/µg, respectively. In commercial neighborhoods, each city was found to exhibit similar OP^{AAm} levels (Mumbai, 0.002 ± 0.001; Delhi, 0.001 ± 0.0007; and Bengaluru, 0.004 ± 0.003 nmol/min/µg). In residential neighborhoods, the values were 0.0009 ± 0.0005, 0.0011 ± 0.0006, and 0.0049 ± 0.0046 nmol/min/µg for Mumbai, Delhi, and Bengaluru, respectively. The neighborhoods of Bengaluru showed distinct OP activity compared with the other cities, as well as within the city neighborhoods. The high-traffic neighborhood of Bengaluru was significantly different from both Bengaluru commercial ($p = 0.00006$) and residential neighborhoods ($p = 0.01$). OP^{AAm} compared for the commercial neighborhoods of Delhi and Bengaluru ($p = 0.0001$) and Mumbai and Bengaluru ($p = 0.002$) were found to be statistically different. It was observed that the OP^{AAm} for the Bengaluru residential neighborhood was significantly higher: ∼4 and 6 times the OP^{AAm} of Delhi ($p = 0.01$) and Mumbai ($p = 0.008$) residential neighborhoods.

Table 3 shows the city-wise COD values. A substantial degree of variability was observed in the OP measurements, encompassing both assays. In Mumbai, the most pronounced intra-city variation in OP^{DTT} was identified in traffic-commercial areas (COD = 0.4), followed by traffic-residential areas (0.36) and commercial-residential areas (0.29). These findings indicate that the sources of redox-active species such as primary and secondary organic aerosols exhibit diversity within the city, suggesting a source-driven nature of OP.[38,39] Likewise, in Delhi (traffic-commercial: 0.33, traffic-residential: 0.34, commercial-residential: 0.17) and Bengaluru (traffic-commercial: 0.42, traffic-residential: 0.25, commercial-residential: 0.30), similar trends were observed.

Furthermore, OP^{AA} displayed ∼1.5 times greater spatial heterogeneity compared with OP^{DTT} across all cities. The higher COD value associated with OP^{AA} implies that the ROS active species, exhibiting a higher affinity toward the AA assay, exhibited notable variation across different locations compared with the DTT assay.

Association with Chemical Constituents

Table 4. Association of volume-normalized dithiothreitol (DTT) and ascorbic acid (AA) oxidative potential assays with chemical constituents ($n = 42$ for $PM_{2.5}$, $b_{abs\text{-}370}$, $b_{abs\text{-}880}$, AAE, WSOC) and ($n = 15$ for OC-EC and their fractions, water-soluble $b_{abs\text{-}365}$, metals, and ions) of $PM_{2.5}$, collected during the winter season in Mumbai, Delhi, and Bengaluru, India, 2019–2020.

	Mumbai		Delhi		Bengaluru	
Variables	DTT	AA	DTT	AA	DTT	AA
$PM_{2.5}$	0.34	−0.14	0.10	−0.16	−0.48	0.38
$b_{abs\text{-}370}$	0.14	0.04	0.05	0.16	0.26	0.06
$b_{abs\text{-}880}$	0.10	−0.07	0.01	0.17	0.30	0.04
AAE	0.15	0.39	−0.03	−0.11	−0.07	0.37
Water-Soluble $b_{abs\text{-}365}$	0.12	0.21	−0.12	0.69	0.12	−0.03
OC	−0.25	0.65	−0.05	−0.19	0.50	−0.38
EC	−0.09	0.62	0.01	−0.31	0.59	−0.33
OC1	0.52	−0.08	−0.11	−0.27	0.70	−0.39
OC2	0.61	−0.20	−0.17	−0.24	0.75	−0.52
OC3	0.67	−0.25	0.06	−0.10	0.42	−0.34
OC4	0.55	−0.27	0.1	−0.20	0.44	−0.34
EC1	0.66	−0.05	−0.14	−0.20	0.69	−0.40
EC2	0.42	−0.13	−0.31	0.01	−0.61	0.42
EC3	−0.03	0.01	0.28	−0.21	0.13	0.09
TC	−0.37	0.33	0.03	−0.06	0.72	−0.44
OC/TC	−0.20	0.28	0.58	−0.22	0.10	−0.07
WSOC	0.13	−0.19	0.24	0.05	0.08	0.08
WSOC/OC	0.30	−0.07	−0.25	−0.03	−0.73	0.59
Ammonium	0.34	0.41	0.34	0.18	−0.34	0.26
Nitrate	−0.22	0.38	−0.03	−0.48	−0.24	0.35
Sulfate	−0.28	0.41	0.28	−0.09	−0.19	0.13
Fe	0.19	0.34	−0.17	−0.19	−0.31	0.38
Mo	0.26	0.26	−0.05	0.03	0.09	−0.16
Zn	0.41	0.51	−0.14	−0.10	0.13	−0.15
Pb	−0.24	0.23	−0.36	−0.07	−0.61	0.46
Mn	0.14	0.81	−0.45	0.27	−0.09	0.02
Cu	0.38	0.05	−0.23	0.40	0.29	−0.17
Ti	−0.19	0.37	−0.21	0.65	−0.52	0.41
V	−0.10	0.45	−0.46	−0.05	−0.38	0.29
Co	−0.32	0.38	−0.20	0.49	−0.68	0.65
Ni	−0.22	0.40	−0.26	0.50	−0.63	0.52
Se	−0.12	0.41	−0.23	0.71	−0.56	0.38
Sr	−0.07	0.32	−0.18	0.56	−0.25	−0.05
Total metals	−0.21	0.56	−0.43	0.37	−0.05	0.09

Note: The Pearson correlation statistic (r) has been reported in the table for the association between different PM constituents with OP. The significant association was checked at $p < 0.05$. The p-value is given in the bracket for the statistically significant association. Association with OP^{DTT}, Mumbai: OC1 ($p = 0.03$), OC2 ($p = 0.01$), OC3 ($p = 0.01$), OC4 ($p = 0.04$), EC1 ($p = 0.006$); Bengaluru: EC ($p = 0.03$), OC1 ($p = 0.002$), OC2 ($p = 0.002$), EC ($p = 0.02$), TC ($p = 0.002$). Association with OP^{AA}, Mumbai: AAE ($p = 0.04$), OC ($p = 0.01$), EC ($p = 0.01$), Mn ($p = 0.001$), Total metals ($p = 0.03$); Delhi: $b_{abs\text{-}370}$ ($p = 0.002$) Ti ($p = 0.002$), Se ($p = 0.003$), Sr ($p = 0.03$); Bengaluru: AAE ($p = 0.04$), Co ($p = 0.02$), Ni ($p = 0.03$). AAE, absorption angstrom exponent; $b_{abs\text{-}365}$, absorption of organics at 365 nm; $b_{abs\text{-}370}$, absorption of organics at 370 nm, commonly known as brown carbon; $b_{abs\text{-}880}$, absorption of organics at 880 nm, commonly known as black carbon; Co, cobalt; Cu, copper; EC, elemental carbon; Fe, iron; Mn, manganese; Mo, molybdenum; Ni, nickel; OC, organic carbon; OP, oxidative potential; OP^{AA}, ascorbic acid oxidative potential acellular assay; OP^{DTT}, dithiothreitol oxidative potential acellular assay; Pb, lead; $PM_{2.5}$, fine particulate matter; Se, selenium; Sr, strontium; TC, total carbon; Ti, titanium; total metals (Fe, Mo, Zn, Pb, Mn, Cu, Ti, V, Co, Ni, Se, Sr); V, vanadium; WSOC, water-soluble organic carbon; Zn, zinc.

Both assays showed unique associations with the chemical species for different cities, as shown in Table 4 (extrinsic OP). In the case of OP^{DTTv}, ROS activity was strongly correlated with the species originating from combustible sources (metals from the exhaust and non-exhaust emissions and other primary emissions) in Mumbai and Bengaluru. Similarly, in Mumbai, positive correlation was found with OC-EC fractions except for EC2 [OC1 ($r = 0.52$, $p = 0.02$), OC2 ($r = 0.61$, $p = 0.01$), OC3 ($r = 0.67$, $p = 0.01$), OC4 ($r = 0.55$, $p = 0.04$), and EC1 ($r = 0.66$, $p = 0.006$)]. In Bengaluru, carbonaceous species, such as EC ($r = 0.59$, $p = 0.032$), and fractions, such as OC1 ($r = 0.70$, $p = 0.002$), OC2 ($r = 0.75$, $p = 0.002$), EC1 ($r = 0.69$, $p = 0.026$), and total carbon (TC; $r = 0.72$, $p = 0.002$) were found to be significantly correlated with OP^{DTTv}.

The OP^{AAv}, OC ($r = 0.65$, $p = 0.012$), EC ($r = 0.62$, $p = 0.01$), absorption angstrom exponent (AAE; $r = 0.39$, $p = 0.04$), Mn

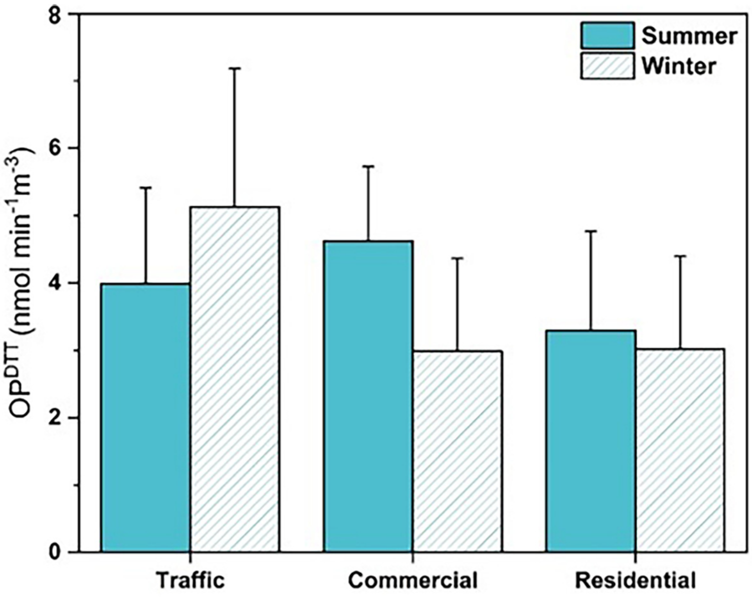

Figure 3. Seasonal variation of volume-normalized dithiothreitol (OP^{DTT}) at three different sites (high traffic, commercial, and residential) in Mumbai ($n = 14$ from each site, both seasons). Columns bars and error bars showing mean and standard deviations, respectively. Corresponding numeric data are provided in Excel Table S3. Note: DTT, dithiothreitol; OP, oxidative potential; OP^{DTT}, dithiothreitol oxidative potential acellular assay.

($r = 0.81$, $p = 0.001$), and total ROS metals ($r = 0.56$, $p = 0.031$) were found to be significantly correlated for Mumbai. In Delhi, transition metals, Ti ($r = 0.65$, $p = 0.002$), Se ($r = 0.71$, $p = 0.003$), and Sr ($r = 0.56$, $p = 0.03$) were found to be correlated, along with water-soluble $b_{abs\text{-}370}$ ($r = 0.69$, $p = 0.002$). OP^{AA} showed statistically significant positive correlation with AAE ($r = 0.37$, $p = 0.042$), Co ($r = 0.65$, $p = 0.023$), and Ni ($r = 0.52$, $p = 0.033$) in Bengaluru.

Mass-normalized OP (intrinsic OP; Table S2) exhibited a positive association with a few chemical constituents for all three cities. Interestingly, OP^{DTTm} showed a strong correlation with OC/TC ($r = 0.55$, $p = 0.046$) and Ni ($r = 0.90$, $p = 0.001$). On the other hand, Bengaluru showed a significant correlation with EC ($r = 0.56$, $p = 0.029$), OC1 ($r = 0.64$, $p = 0.017$), OC2 ($r = 0.72$, $p = 0.003$), EC1 ($r = 0.67$, $p = 0.006$), TC ($r = 0.72$, $p = 0.002$), and Co ($r = 0.58$, $p = 0.042$).

Seasonal Variation of OP^{DTT} in Mumbai

$PM_{2.5}$ concentrations during winter (131.32 μg/m^3) were higher than during summer (68.08 μg/m^3) by a factor of 2, showing statistically significant ($p = 0.001$) seasonal variation in Mumbai. During summer, the OP^{DTT} value was comparable to that of winter (3.97 ± 0.66 vs. 3.69 ± 1.16 nmol/min/m^3, respectively). However, mass-normalized OP was 2-fold higher in summer (0.06 ± 0.05 nmol/min/ μg) compared with winter (0.03 ± 0.02 nmol/min/μg) ($p = 10^{-6}$).

Figure 3 and Figure S5 respectively illustrate the volume and mass-normalized OP^{DTT} variation in different neighborhoods (intra-OP variability) of Mumbai. All of the neighborhoods showed comparable variation in extrinsic OP for both of the seasons: traffic neighborhood (summer, 3.83 ± 1.47; winter, 5.13 ± 2.06 nmol/min/m^3); commercial neighborhood (summer, 4.41 ± 1.34; winter, 2.99 ± 1.38 nmol/min/m^3); and residential neighborhood (summer, 3.18 ± 1.45; winter, 3.02 ± 1.38 nmol/min/m^3). However, intrinsic OP showed a significant difference among the neighborhoods; high-traffic neighborhood (summer, 0.07 ± 0.03; winter, 0.05 ± 0.02 nmol/min/μg), commercial neighborhood (summer, 0.06 ± 0.03; winter, 0.03 ± 0.01 nmol/ min/μg), and residential neighborhood (summer, 0.06 ± 0.05; winter, 0.02 ± 0.01 nmol/min/μg). Seasonal variation of OP^{DTT} assays within Mumbai and association with different chemical species was observed during the winter and summer as shown in Table S3. In carbonaceous species, the OC1 fraction ($r = 0.85$, $p = 0.01$) and transition element, V ($r = 0.57$, $p = 0.03$), were significantly associated with OP during summer.

Multiple linear regression. The obtained regression model for Mumbai is presented in Equation 3 with an adjusted R^2 value of 0.47 ($p = 0.03$). From the MLR results, we found that the transition metals Co and V are significant predictors of volume-normalized DTT activity. In addition to these, WSOC, which is most likely predominantly associated with secondary organic aerosols, also contributed to the DTT activity.

$$OP^{DTTv} = 0.15(\text{WSOC}) + 4.51(\text{Co}) + 0.9(\text{V}) - 0.94. \tag{3}$$

Discussion

The intercity and intra-city OP variation indicate that volume-normalized levels from both assays are comparable across the three cities, and the high-traffic neighborhood is responsible for the maximum PM-induced toxicity in the residential outdoor homes. In terms of mass-normalized results, Bengaluru was significantly higher than the other two cities, and each of its neighborhoods (high-traffic, commercial, and residential) significantly differed from each other.

Several OP^{DTT} studies have been conducted across the globe and a comparative assessment among all available PM OP^{DTT} studies are provided in Figure 4. It should be noted that the present study used a different aliquot volume, optimized for our samples and protocols, redefining those previously used in studies from the United States, Europe, and other Asian countries. In addition, a few PM_{10} Indian studies, such as in Mount Abu[39] (in 2018; 1.23 ± 0.68 nmol/min/m^3) and Patiala[31] (in 2018; 3.81 ± 1.4 nmol/min/m^3), showed a range to the findings similar to that of the present study. At an international level, most European countries[18,23,26,28,61] that have reported OP levels greatly

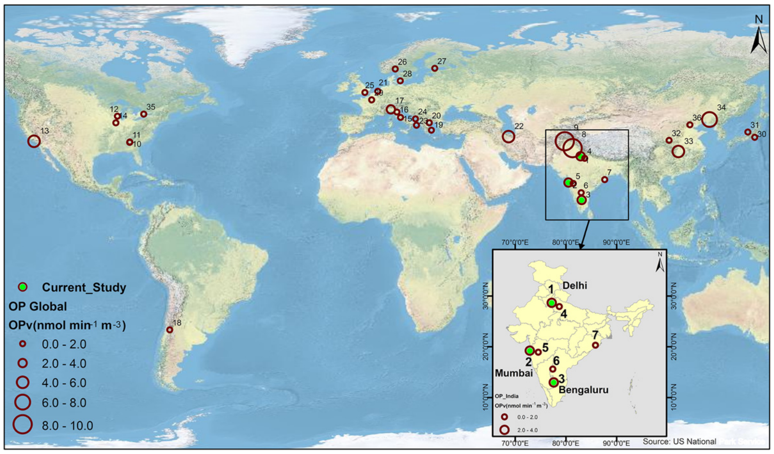

Figure 4. Comparison of different volume-normalized OP^{DTT} values across the globe. 1: Delhi (present study); 2: Mumbai (present study); 3: Bengaluru (present study); 4: Delhi[35]; 5: Mumbai[23]; 6: Bengaluru[41]; 7: Bhubaneshwar[56]; 8: Lahore[29]; 9: Peshwar[29]; 10: Atlanta[48]; 11: Atlanta[10]; 12: Urbana-Champaign[57]; 13: Los Angeles[58]; 14: Illinois[59]; 15: Terni[18]; 16: Bologna[60]; 17: Milan[61]; 18: Santiago[62]; 19: Athens[28]; 20: Thessaloniki[63]; 21: Rotterdam[26]; 22: Tehran[64]; 23: Salento's peninsula[65]; 24: Salento's peninsula[65]; 25: London[66]; 26: Oslo[66]; 27: Helsinki[66]; 28: Copenhagen[66]; 29: Paris[66]; 30: Yokohama[67]; 31: Noto[38]; 32: Xi'an[68]; 33: Jinzhou[69]; 34: Liaoning[69]; 35: Toronto[70]; 36: Beijing.[5] Corresponding numeric data are provided in Excel Table S4. Note: OP, oxidative potential; OP^{DTT}, dithiothreitol oxidative potential acellular assay; OPv, volume normalized oxidative potential.

comply to the range of current studies, suggesting common sources for the ROS generation. However, in studies conducted in the United States,[58,71–74] OP^{DTT} values lay in the range of $0–6\,\mathrm{nmol/min/m^3}$, whereas other Asian studies[13,37,75,76] (excluding India) showed OP values of $0.03–14\,\mathrm{nmol/min/m^3}$. Such studies show the tremendous spatial OP variation across the globe, influenced by climatic factors, as well as by specific sources.

The OP^{DTT} and OP^{AA} assays capture different natures of aerosols; the former is sensitive to both metals, as well as organics, whereas the latter is better at detecting metals. To the best of our knowledge, only one Indian[77] study has reported using the OP^{AA} technique ($0.22\,\mathrm{nmol/min/m^3}$). In comparison, some international studies reported the following OP^{AA} values: Italy,[21] urban site—$0.52 \pm 0.09\,\mathrm{nmol/min/m^3}$, rural site—$0.27 \pm 0.06\,\mathrm{nmol/min/m^3}$; Central Mediterranean[65]—$0.09 \pm 0.01\,\mathrm{nmol/min/m^3}$; and France[34]—with relatively a higher range, $5.0\,\mathrm{nmol/min/m^3}$, but showing an OP range within that of our present study. These findings suggest that the geographical distribution of pollutants may influence the nature of ROS in $PM_{2.5}$ across different regions.

Several aerosol studies have examined redox-active PM through the DTT assay at various locations influenced by different sources. For example, in an urban background site in Pakistan,[29] reported OP^{DTT} values were $8.90 \pm 3.80\,\mathrm{nmol/min/m^3}$ during the winter season. Source-specific studies have indicated values such as $1.40–1.70\,\mathrm{nmol/min/m^3}$ at a high-traffic site in the Netherlands,[26] $1.23 \pm 0.68\,\mathrm{nmol/min/m^3}$ at a rural site in India,[31] and $0.66–1.03\,\mathrm{nmol/min/m^3}$ at a site impacted by traffic and trash burning in India.[41] A recent study on intense paddy residue burning in a semi-urban location of India reported the highest OP^{DTT}, $14 \pm 5\,\mathrm{nmol/min/m^3}$.[78] Moreover, at an urban background site in the United States,[79] OP^{DTT} values ranged from 0.10 to $1.50\,\mathrm{nmol/min/m^3}$. Our study observed an ~2.5-fold lower OP range compared with these referenced studies, which is likely attributed to specific emission sources at urban residential outdoors. However, the highest OP^{DTT} activity was reported in the urban outdoors near high-traffic neighborhoods in all three cities, which was probably due to the dominance of vehicle exhaust and non-exhaust sources. The OP^{DTT} activity between residential and commercial neighborhoods showed similar levels. However, in Mumbai, there was a significant difference in OP^{DTT} activity between commercial-traffic ($p = 0.027$) and residential-traffic ($p = 0.03$) neighborhoods. The mass-normalized OP^{DTT} and OP^{AA} assay results in this study show good agreement with European studies,[23,29,31,40] indicating that similar ROS-induced PM species are responsible for OP activity. This suggests the ROS present per unit of PM mass in Mumbai is higher than the ROS levels in the other two cities.

According to existing studies,[8,34,56,80–84] elemental species show a stronger affinity toward OP^{AA}. Primary aerosols, consisting of more elemental fractions originating from exhaust and non-exhaust sources, contributed to increased traffic-generated ROS, with domestic household emissions also contributing to OP^{AA} levels. Our study shows that OP^{DTT} has more association with carbonaceous fractions[31,33,85] and few traffic related trace elements.[10,81,86] Several US–European studies[8,46,87] have also reported a list of transition elements (Cu, Fe, Mn, Co, Ni) that participate in ROS generation and that are present in the redox-active oxidation state. The correlation with optical properties suggests that these species are providing a platform to ROS generation (acting as a proxy to it) as is also suggested by existing studies.[85,88–90] It is well known that OP^{AA} positively correlates to redox-active total metals (Cu, Fe, Mn).[24,71,85] Several studies are incongruent with our findings; in those studies winter levels are reported as 20%–40% higher than summer levels.[43,78] However, our study shows weak correlation with the transition metals, except for V ($p < 0.05$), similar to the study conducted in Noto, Japan,[20] because the elements are most likely present in the non–redox-active state.[38,67,91]

In Bengaluru, a positive correlation was found with organics, indicating the local wood burning and other combustible sources and traffic aerosols emissions.[38,41] In Mumbai, transition metals showed a strong association with OP^{DTT} originating from the primary emission sources. Non-exhaust emission sources, such as Cd, associated with lubricating oil emissions, were correlated with DTT activity.[23,92–94] A strong correlation between OC/TC, reveals that emissions from biogenic (burning events) and anthropogenic sources (natural gas home appliances), atmospheric transport, and transformation are contributing to the ROS.[74,95,96] composition.

Seasonal variation in Mumbai clearly revealed significant variation in PM concentration; however, the OP variation was not found to be different. Commercial and residential neighborhoods showed higher OP during summer, suggesting primary, as well as aged, aerosols contributing to the OP levels. However, no statistically significant difference was observed in Mumbai between the two seasons, suggesting that toxicity is more likely influenced by the primary-originated ROS-induced aerosols and limited secondary aerosols. This implies that the chemical composition that influenced the ROS-induced toxicity is predominant. In addition, the seasonal spatial heterogeneity of OP^{DTT} was calculated through the COD was found to be ~ 1.6 times greater during winter than summer across the aforementioned neighborhoods ($p < 0.05$). Mass-normalized OP levels also show a similar range of OP^{DTT} level across all the neighborhoods during winter. However, in summer the trend is higher in high-traffic, followed by residential, then commercial neighborhoods, indicating the lower mass concentration during summer increases the respective intrinsic (mass-normalized) OP levels, complying with the study in Fresno, California,[90] and Greece,[7] suggesting that the higher intrinsic OP in summer is due to higher ROS in per unit PM mass, mainly coming from the fresh and transformed organic aerosol (increased photochemical activity during summer). The association with few carbonaceous species, the OC fraction (OC1), and the transition element, V, suggests that long-range transport organic aerosols, consisting of more soluble secondary species and volatile organic species, are contributing to OP activity.[33,97]

MLR analysis predicted that Co and V are major species contributing to DTT activity in residential outdoor areas of Mumbai. Co and V likely originate from commercial (industrial) emissions, specifically from catalyst synthesis in the metal industry.[47,98,99] WSOC is a major precursor of biogenic emissions[29,100,101] and secondary aerosols produced from combustible sources.[29] These predictor species (WSOC, Co, and V) are associated with OP^{DTT}, indicating ROS that also originates from small-scale commercial firms.

Limitations

The limitations of this study are that the OP assays are peculiar to the nature of aerosols and that cell-free assays can only act as a surrogate of the living cells and cannot explain the remaining unexamined aerosols. The seasonal variation was examined only in Mumbai, which gives some insights to the OP variation due to meteorological parameters; however, this needs to be further validated with the other cities where extreme weather fluctuations are observed, such as Delhi.

Conclusions

To the best of our knowledge, this is the first study in India examining the OP in different types of urban residential neighborhoods in Delhi, Mumbai, and Bengaluru: three different agroclimatic zones of the country, using multiple acellular OP assays. PM concentrations in all the three cities were different, but the corresponding OP levels did not align in the same proportion, suggesting the toxicity is derived through chemical composition and meteorological variables rather mass concentration. The study also highlights that low-levels of PM found in some parts of the country suggest a lack of PM toxicity.

Although the study was conducted in three metropolitan cities, the results can be expanded to all tier 1 and tier 2 cities (average population >100,000), which have significantly high levels of PM that likely put the population at higher toxicity risk of air pollution. Most of the time continuous emphasis is on mass-based PM, but to understand the associated risk, mass-based PM needs to be supplemented by the OP metric or chemical composition-based metric. These significant findings shed light on the potential toxicity of PM in residential neighborhoods, advocating for effective measures to mitigate its impact on public health in densely populated urban areas.

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