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Atmospheric Particulate Matter and Associated Trace Elements Pollution in Bangladesh: A Comparative Study with Global Megacities

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Abstract Air quality degradation due to high levels of atmospheric particulate matter (PM) of various size fractions and the associated potentially toxic trace elements (PTEs) is a global concern. This article provides a thorough review and analysis of the temporal and spatial distribution of PM and PTEs in Bangladesh, offering a comprehensive assessment with other megacities worldwide based on existing literature. This study provides insights into the sources and transport mechanisms of PM and their link to human health. The level of PM was consistently high in Dhaka (capital of Bangladesh), with occasional higher levels in the surrounding cities. Different functional areas within Bangladesh show varying levels of PM, with total suspended particulates (TSP) being notably prevalent. When compared to megacities worldwide, African and Asian megacities, like India, Pakistan, Nigeria, and Egypt, exhibited higher PM concentrations. The concentration of PM-associated PTEs varies significantly among megacities and PM_{10} tends to have relatively higher concentrations of PTEs compared to other fractions in Bangladesh. Pb in ambient air was found across most megacities, with a temporal increase in Bangladesh. TSP exhibited the highest relative Pb

Highlights
- Review on particulate matter (PM) and associated elements in Bangladeshi air.
- Most Asian megacities are more polluted compared to other global megacities.
- Traffic, industry, and fossil fuel combustion are the major sources of PM pollution.
- PM and associated PTEs are responsible for several diseases in humans.

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content, followed by PM_{10} and $PM_{2.5}$. Temporal factors, geographic locations, meteorological conditions, and anthropogenic activities contribute to the variation in PM and associated PTEs concentration in Bangladesh and global megacities. Ultimately, this study would aid policymakers in assessing the magnitude of PM pollution in Bangladesh compared to other megacities considering regional factors.

Keywords Particulate matter; Potential toxic elements · Global megacities · Human health risks; Source apportionment

1 Introduction

Airborne particulate matter (PM) is a complex mixture of solid and liquid particles suspended in the atmosphere, exhibiting disparate variations in size, shape, density, and chemical composition (Harrison, 2020). Although atmospheric particulates can arise from natural sources, such as volcanoes, bushfires, and re-entrained soil, air quality in both developed and developing countries of the world continues to deteriorate, largely due to increasing urbanization, industrialization, and an upsurge in motorized transport (Gulia et al., 2015; Ramírez et al., 2020). Unlike other pollution vectors, atmospheric particulate matter can be transported over extensive areas, and their resultant impacts can be deleterious to the environment and human health (Ramírez et al., 2020; Wang et al., 2013).

Atmospheric residency time, deposition rate, and particles' ability to penetrate the human respiratory system are predominantly influenced by size. For air quality regulations and management, PM is classified based on aerodynamic diameter (D_a), which is defined as the diameter of a sphere with a density of 1 g cm^{-3}, which settles in still air at the same velocity as the particle in question (DeCarlo et al., 2004; Dockery & Pope, 1994). The United States Environmental Protection Agency (EPA) Air Quality Guidelines categorized PM predominantly in two size fractions: (1) coarse fraction, with an aerodynamic diameter of 10 μm (called PM_{10}), and (2) fine fraction, with an aerodynamic diameter of 2.5 μm (called $PM_{2.5}$) (Kim et al., 2015). Less commonly, total suspended particulates (TSP) are also reported. Ultrafine particles with a diameter of less than 0.1 μm, typically associated with combustion processes, are not regulated or monitored, despite their potentially significant health risk (Baldauf et al., 2016).

The health risks associated with PM arise primarily from deposition within the respiratory system. $PM_{2.5}$ can absorb more toxic substances than coarse PM and can enter the human lung through respiration resulting in various respiratory and cardiovascular diseases (Makkonen et al., 2010). There is also epidemiological evidence that confirms a noteworthy correlation between fine PM and mortality (Lim et al., 2010). Other potential adverse health outcomes from PM exposure also include respiratory and circulatory mortality (Li et al., 2013), mutagenicity and DNA damage (Coronas et al., 2009), preterm-birth risk and adverse birth outcomes (Hooven et al., 2012), increased cancer risk (Díaz-Robles et al., 2013), and inflammatory responses (Silbajoris et al., 2011).

Ambient and indoor air pollution pose significant environmental risk across South Asian countries and the levels of pollution in Bangladesh are among the highest in the region. Fine particulate matter has been found to be responsible for 21% of all yearly deaths in Bangladesh (World Bank, 2018). Though there are few studies on particulate matter and associated potentially toxic elements (PTEs) in Bangladesh, there is a scarcity of comprehensive comparisons among different cities in Bangladesh. Most of the studies in Bangladesh were conducted in the capital city Dhaka and its adjoining areas (Begum & Hopke, 2018; Begum et al., 2004, 2010, 2013). Those studies focused mostly on PM concentration, and/or associated PTEs, and source identification based on size and chemical properties of PM along with some statistical analysis (Begum & Hopke, 2018; Begum et al., 2004, 2010, 2013). However, to our best knowledge, a significant knowledge gap exists with regard to a comprehensive systematic review and data synthesis, especially on airborne PM and associated PTEs when comparing Bangladesh to other global megacities. Therefore, this study aims to:

a. Provide a comprehensive summary of current research on air pollution, with a specific emphasis on PM ($PM_{2.5}$, PM_{10}, TSP, and $PM_{2.5–10}$) and associated PTEs in Bangladesh.

b. Compare the air quality in Bangladesh with that of other global megacities to shed light on relative pollution levels.
c. Highlight the human health risks associated with PM exposure and provide a brief overview of the various models used for source apportionment in air pollution studies.

By addressing these objectives, this study aims to bridge the gap in knowledge regarding air pollution in Bangladesh, while also offering valuable insights into the comparative air quality status between Bangladesh and other major cities worldwide.

2 Methods of Literature Survey and Data Acquisition

This literature survey was conducted according to the methods of Kumar et al. (2019), utilizing various databases such as Web of Science, ScienceDirect, and Google Scholar. Search keywords included "particulate matter pollution in Bangladesh," "ambient particulate matter in Bangladesh," "particulate matter bound trace metals in Bangladesh," "trace elements in the air in Bangladesh," and "trace metals associated with particulate matter in Bangladesh." The same keywords were used for the other global megacities, replacing Bangladesh with the specific megacity name, for example, Dhaka, Delhi, Guangzhou, Beijing, Shanghai, and Kolkata. For Bangladesh, 60 research articles were reviewed, with 42 papers selected for data collection covering years from 1992 to 2021. The papers were selected if they reported PM and/or associated major and trace element concentrations. In the case of the other megacities, 95 research articles were identified as relevant to the context of this study. However, 66 most recent and relevant articles representing 39 megacities worldwide reported both PM and associated major and/or trace element concentration and were used to extract the data of three major PM types ($PM_{2.5}$, PM_{10}, and TSP). The locations of the global megacities as well as study districts in Bangladesh considered in this study are shown in Fig. 1 and a list of articles used for data collection for each study location is provided in the supplementary files. It should be noted that data was also obtained from the referenced work in the aforementioned articles. Data were collected only when it was presented in tabulated form or provided in the text and written in English. A PRISMA flowchart summarizing the data acquisition process is provided in supplementary Fig. S1.

As different units of measurement were reported throughout the collected data, elemental and PM concentrations were converted into μg m^{-3}. The final data set of the reported concentrations of major and trace elements, as well as $PM_{2.5}$, PM_{10}, $PM_{2.5–10}$, and TSP, was collated and includes sampling location type (only for Bangladesh including other cities along with megacity Dhaka), city name, and country (supplementary Tables S1, S2 and S3). The graphs were prepared using the average values for individual locations by averaging spatial (different locations of a city or megacity) and temporal (different years and seasons) values of that location or location type for different parameters. Precisely, for a study location or location type, for example, Dhaka in Bangladesh or residential area, there were several studies carried out at different sites within the location or the region of interest. Dhaka city or residential area data were obtained at different times, thus the data for a specific parameter (e.g., $PM_{2.5}$) was averaged among times within the location. So, for each location and parameter, there were varying numbers of data points resulting in a non-normal distribution of the dataset. Therefore, to check any statistical differences among study locations and parameters, a non-parametric ANOVA, known as the Kruskal-Wallis test was performed. Besides, principal component analysis (PCA) was performed for particulate matter and associated PTEs in Bangladesh.

Additionally, the supplementary information includes a curated list of research articles, systematically categorized based on different study locations worldwide. Depending on the nature of the samples, the number of replications, the methodology employed in elemental extraction, and the instruments used in quantifying different elements and PM, a significant variability may exist in the final data set of PM and associated elemental concentrations. Furthermore, temporal and seasonal differences, variations in geographic locations, and meteorological conditions among the study sites made the data comparison more complicated. As a result, we acknowledge potential biases in the dataset and advise that the data should be viewed cautiously.

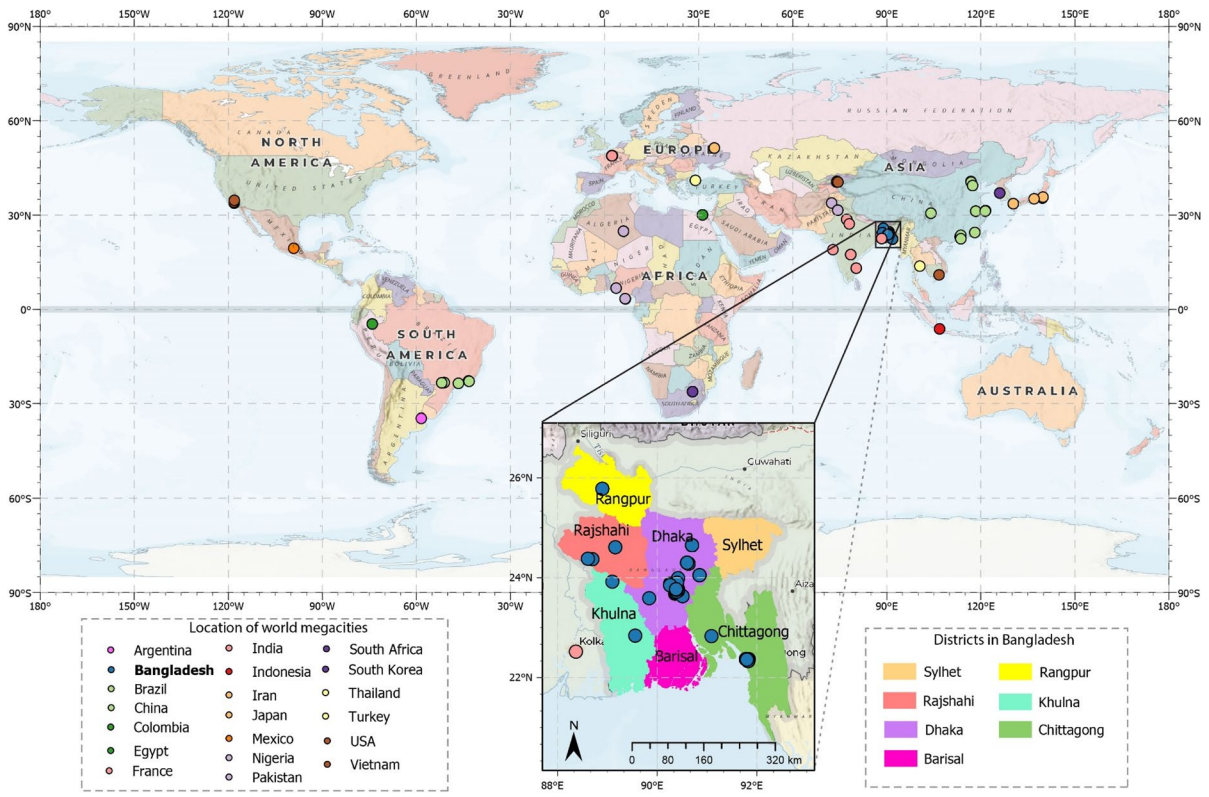

Fig. 1 Locations of air particle sampling stations in world megacities and in Bangladesh (inset). Small circles with different colors represent the locations

3 Health Risks of Airborne Particulate Matter

Particulate matter can seriously impact human health and has been linked to a wide range of diseases, some of which can be fatal (Fig. 2). It has been estimated that life expectancy is reduced by 8.6 months on average due to PM exposure (Krewski, 2009). In urban air, PM_{10} is a major pollutant responsible for millions of premature deaths each year (WHO, 2018). Additionally, $PM_{2.5}$ is responsible for ~ 4.58 million premature deaths worldwide annually (Stanaway et al., 2018).

Inhalation and dermal contact are the main routes of exposure to PM. The negative health impacts of airborne particulates arise from their ability to penetrate into the lungs, including the bronchioles and alveoli, during breathing (Phan et al., 2020). Extensive experimental and epidemiological studies have revealed that not all particles are equally toxic and that they individually pose different health risks (Kim et al., 2015). The size of particles plays a crucial role in determining toxicity to human health. PM with an aerodynamic diameter between 2.5 and 10 μm is primarily deposited in the tracheal and bronchial region. These particles are either expelled through coughing and sneezing or transported to the gastrointestinal tract via mucociliary processes (Elmes & Gasparon, 2017). $PM_{2.5}$, however, can penetrate the bronchioles and alveoli to enter the human circulatory system and translocate to extrapulmonary organs such as the liver, spleen, heart, and brain (Darquenne, 2012; Hussain et al., 2011). Consequently, $PM_{2.5}$ has been linked to several diseases, including cardiovascular disease and stroke (Burnett et al., 2014; Du et al., 2016), respiratory disorders, pulmonary diseases (Cowie et al., 2010; Guo et al., 2014), and even cognitive problems (Weuve, 2012). Fine PM has a higher capacity to absorb toxic substances compared to coarse PM (Du et al., 2021; Wu et al., 2018). Moreover, fine PM can stay suspended in the atmosphere for

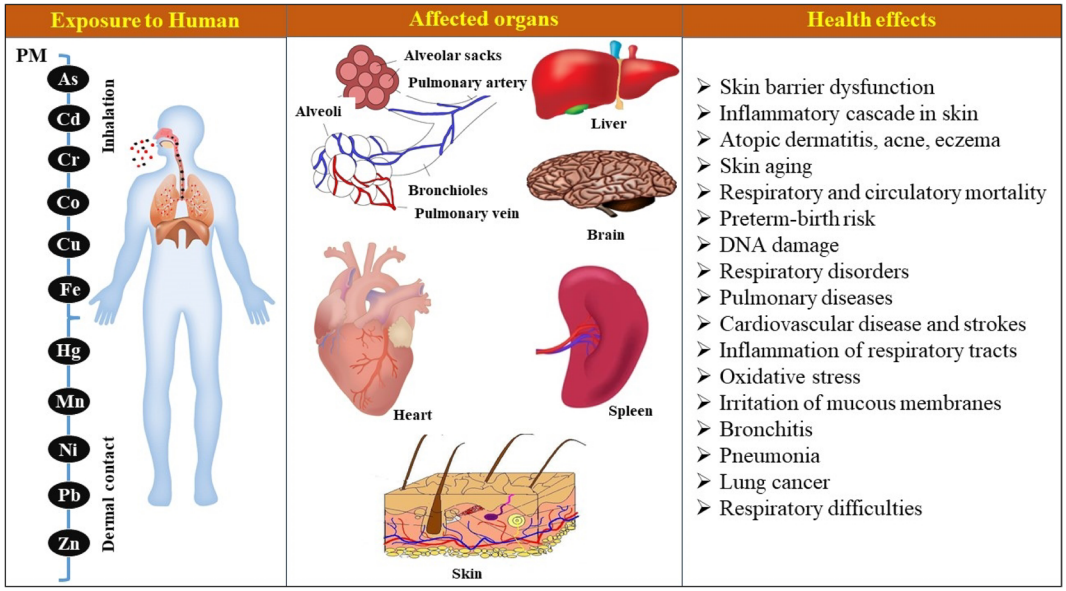

Fig. 2 An schematic representation of exposure routes, possible affected organs, and human health risks from exposure to PM and associated PTEs

long periods of time and be transported extensively via atmospheric circulation (Harrison, 2020).

The inflammatory response caused by PM is not solely dependent on the physical properties of the particle (e.g., size, mass, and particle number), but also on the chemical properties of the individual particle, particle's solubility in the biological fluid, the health status of the exposed population, and the content of PTEs in various size fractions of PM (Harrison, 2020). The PTEs have been observed to accumulate in different fractions of PM and can account for ~ 2–8% of the total mass of $PM_{2.5}$ (Fan et al., 2021). Despite their small mass fraction, PTEs still pose a threat to human health by increasing the risk of cancer and respiratory diseases (Fan et al., 2021). The non-biodegradable and bio-accumulative nature of PTEs cause exceedance of their threshold/safe limits that can consequently harm human health. For example, Pb affects the hematopoietic and nervous systems leading to cognitive function impairment and growth issues (Charkiewicz & Backstrand, 2020). Inhalation of Cr irritates mucous membranes and causes bronchitis, pneumonia, and lung cancer (Briffa et al., 2020). Reactive oxygen species (ROS) arising from atmospheric Fe, Mn, Cu, Co, Ti, and As can cause inflammation of respiratory tracts and generate oxidative stress (Xu et al., 2020).

4 Transportation Mechanisms of PM

The resuspension and transportation of PM in the atmosphere are influenced by a complex interplay of environmental and meteorological factors. Suspension of the particles into the atmosphere occurs when the lift force of airflow exceeds the adhesive forces of the particles, either among themselves or with the attached surface (Ali et al., 2019). Once in the atmosphere, the size and chemical composition of individual particles change in response to gas-to-particle conversion processes. The growth and aging of primary and newly formed particulates are facilitated by the presence of organic matter, sulfate, nitrate, and ammonium (Guo et al., 2014; Huang et al., 2014). Following these processes, PM interacts with solar radiation and other meteorological parameters and is either deposited locally by dry or wet deposition or transported by air masses to areas remote from their origin. Suspended dust particles can remain in the atmosphere for extended periods and can travel

thousands of kilometers, which is typical for particles less than 20 μm (Ali et al., 2019).

In regions like South Asia, particularly in Bangladesh, PM related air pollution becomes severe during winter (December–February) due to low temperatures, humidity and rainfall, and winds carrying aerosols from India and Arabia over large areas of the Arabian Sea, and northern Indian Ocean (Begum et al., 2013). Studies of back trajectories have found that the air masses travel through Iran, Afghanistan, Pakistan, and India to Bangladesh before curving down to Sri Lanka (Begum et al., 2013; Hopke et al., 2008).

Wind speed and direction are crucial factors influencing the concentrations of PM and various elements in the air during transportation. Strong winds disperse the pollutants, while weak winds result in their accumulation, leading to higher concentrations in the air (Hsu & Cheng, 2019). During winter and spring, the wind direction in Dhaka city (capital of Bangladesh) is mainly from the north/north-west quadrants and during pre-monsoon and monsoon, it is from the south/south-east quadrants (supplementary Fig. S2) (Salam & Salam, 2011). Consequently, Asian dust particles transported over long distances by westerly winds in the late winter and spring seasons substantially enhance the ambient aerosol concentrations in downwind countries like Bangladesh. Along with windblown dust, significant pollutant loads accompany these dust-rich atmospheric systems derived from the Indian sub-continent, primarily due to increased development in industries, biomass burning, and household coal combustion (Begum et al., 2013; Rahman et al., 2020; Schleicher et al., 2011). As a result, the air quality in winter, with its minimal rainfall, becomes extremely poor. Both local and regional sources contribute to degrading winter air quality in Bangladesh (Begum et al., 2013). Conversely, the air pollution problem is insignificant during the wet (pre-monsoon and monsoon seasons) season when the wind derives mainly from the south and south-east.

5 Source Apportionment Methods for PM and PTEs in Air

Identifying the source of atmospheric particulates and evaluating the relative contribution of the identified sources are an essential aspect of air pollution research and vital for implementing mitigation techniques and managing air quality (Hopke et al., 2020). Several analytical methods and computer-based predictive models have been developed for source apportionment, each with its own strengths and limitations. However, uncertainties related to particulate dispersion and atmospheric processes currently limit the attainment of absolute certainty in source apportionment.

In Bangladesh, several methods have been widely employed for source apportionment studies. These include the enrichment factor (EF) (Akther et al., 2019; Boman et al., 2005; Salam et al., 2003), correlation analysis (Boman et al., 2005; Rahman et al., 2019a; Salam et al., 2003), principal component analysis (PCA) (Begum et al., 2006; Rahman et al., 2019a), reconstructed chemical mass balance (RCM) (Begum et al., 2006; Rahman et al., 2019a), and positive matrix factorization (PMF) (Begum & Hopke, 2018; Begum et al., 2004, 2010, 2013). A shift from factor analysis followed by chemical mass balance to PCA and PMF can be observed by analyzing the historical perspective of source apportionment techniques (Hopke, 2016).

The enrichment factor method involves normalizing the concentration of elements in ambient air mass to pre-historic background values of reference elements like Al, Fe, Mn, Ti, Sc, Li, and Cs (Amin et al., 2009; Karbassi et al., 2008; Salati & Moore, 2010). This is a simple, but functional, technique for discriminating pollution sources, either natural or anthropogenic. In general, an arbitrary average EF value less than 10 indicates a significant crustal origin for an element in the PM, whereas an EF value greater than 10 is considered a non-crustal source (Kumar et al., 2021). In Bangladesh, about 10% of studies used the EF for source identification (supplementary Fig. S3). However, this technique has limitations as it can only indicate the nature of the source (natural or anthropogenic) without specifically identifying individual sources or their contributions to contaminant origins. Furthermore, the lack of local background data often necessitates the use of pre-historic background values from different geographical locations, leading to potential misinterpretation of data due to regional geological and mineralogical variations.

Another simple method of source identification used in PM pollution research is the reconstructed mass calculation (RCM), in which pseudo sources are identified and calculated using the basic convergences

of their constituent components as outlined in Eqs. 1–7 (Malm et al., 1994):

$$\begin{aligned}\text{RCM} = [\text{Soil}] + [\text{OMH}] + [\text{BC}] + [\text{Smoke}] \\ +[\text{Sulfate}] + [\text{Seasalt}] + [\text{Cu}] + [\text{Zn}]\end{aligned} \quad (1)$$

where BC is the black carbon and OMH is the organic matter containing hydrogen, where the pseudo sources are characterized as follows, with the brackets representing the concentration of each element (Rahman et al., 2019a):

$$\begin{aligned}[\text{Soil}] = 2.20 \times [\text{Al}] + 2.49 \times [\text{Si}] + 1.63 \\ \times[\text{Ca}] + 2.42 \times [\text{Fe}] + 1.94 \times [\text{Ti}]\end{aligned} \quad (2)$$

$$[\text{BC}] = [\text{Soot}] \quad (3)$$

$$[\text{Smoke}] = [\text{K}] - 0.6 \times [\text{Fe}] \quad (4)$$

$$[\text{Seasalt}] = 2.54 \times [\text{Na}] = [\text{Na}] + [\text{Cl}] \quad (5)$$

$$[\text{Sulfate}] = 4.125 \times [\text{S}] \quad (6)$$

$$[\text{OMH}] = 11 \times ([\text{H}] - 0.25 \times [\text{S}]) \quad (7)$$

Although this is a straightforward method, the above calculations can be speculative, and the robustness of the outcomes is questionable. More importantly, different authors use different factors to infer the value of the same variable. Consequently, it is a comparatively less commonly used method for source identification, representing approximately 6% of the studies in Bangladesh. In the chemical mass balance model, specific trace metals, organic compounds, and molecular markers can be used to identify exact source types, although knowledge of the emission sources, meteorological conditions, and ambient concentrations is required. The results, however, can be deceptive because of different physicochemical fractionation processes for the different elements, thus, their relative abundances may change significantly during transportation (Bollhöfer & Rosman, 2001).

In Bangladesh, about 4.1% of studies utilized principal component analysis (PCA) to identify possible sources of PM and associated elements in the air. The PCA is a qualitative source identification method wherein each principal component assigns pollution sources based on marker elements. PCA assigns variance based on the difference from the mean to a measured data value and cannot provide a quantitative allocation for each source (Hopke et al., 2020). For quantitative source apportionment, the absolute principal component scores (APCS) of variables are obtained from PCA by analyzing normalized elemental concentrations, which is collectively called the PCA-APCS method. In addition, multiple linear regression (MLR) of the mass concentration data for these APCS can be used to estimate the coefficients that convert the APCS into pollutant source mass contributions from each source for each sample. Therefore, it is also known as the PCA-APCS-MLR method (Harrison et al., 1996; Khan et al., 2010; Rahman et al., 2021a; Thurston & Spengler, 1985). A major drawback of this method is that the analytical uncertainty cannot be taken into account in the PCA-APCS-MLR method. In addition, the uncertainty of the model outcomes due to the heterogeneity of the particle size fractions cannot be estimated. Although it is a widely used method globally, very few studies (about 2%) in Bangladesh considered using this method for source identification.

Recent studies have shown that the PMF method can avoid many uncertainties associated with PCA (Zhou et al., 2004). This is because analytical errors can be integrated with measured values to reduce the uncertainty of source profiling. The size distribution of particles emitted from a source is not perfectly constant, and such variation also needs to accommodate in the PMF model to reduce the uncertainty. To do that, a least-squares method is run separately for number and volume size distributions to estimate uncertainty (Zhou et al., 2004). As a result, this technique provides a more robust quantitative estimation of individual source contributions to pollution levels. Interestingly, in Bangladesh, most studies have used the PMF method (about 20.4%) for source apportionment of PM and PTEs. In contrast, EF along with factor analysis has been extensively used in the studies of other megacities incorporated in this review.

Instead of relying on receptor models, a significant number of studies conducted in Bangladesh have utilized simpler methods such as elemental tracing (approximately 18.4%) and PM size differentiation (approximately 21%) for source identification. For example, the presence of Al, Mg, Na, P, sulfur (S), and Si in PM was considered a soil source. The presence of Mg, Na, Pb, and Si has been

reported as traffic and road dust emissions, and BC and S as motor vehicle emissions (Begum et al., 2005). In another study, the presence of K, Pb, S, and Si was considered a traffic emission source of PM (Begum et al., 2010). Regarding PM size differentiation, the source of air pollution has been hypothesized to be fossil fuel combustion when the concentration of $PM_{2.5}$ (fine particles) exceeds that of PM_{10} (coarse particles). This assumption stems from the belief that coarse PM (PM_{10}) originates from mechanical processes, while fine PM ($PM_{2.5}$) is emitted during combustion processes (Nayeem et al., 2020). However, it is important to note that these methods of source identification are often arbitrary and subjective, relying heavily on individual knowledge about emission sources of PM and associated PTEs. Such approaches lack a systematic and comprehensive framework, leading to inconsistencies and uncertainties in the interpretation of results. To improve the reliability and accuracy of source identification, there is a need to adopt more rigorous and scientifically validated approaches that account for the complex interplay of various factors influencing PM composition and sources. Integrating advanced analytical techniques, multivariate statistical methods, and comprehensive knowledge of emission characteristics can help overcome the limitations associated with haphazard source identification and provide more robust insights into air pollution sources in Bangladesh.

6 Sources of PM and Associated PTEs in Bangladesh

Although both natural and anthropogenic activities contribute to the atmospheric particulate burden, their composition and toxicity strongly depend on the emission source(s). Identifying the source of airborne particulates is crucial for emissions management and protecting human and environmental health (Elmes & Gasparon, 2017). Various anthropogenic sources of PM have been identified in Bangladesh, including traffic emissions, biomass burning, coal and wood-burning in brick kilns, construction activities, smelters, and other industrial emissions and petrochemicals. Natural sources are derived primarily from re-entrained soil and road dust, forest fires, biological particles, such as bacteria, fungi and pollens, and sea salt. The major anthropogenic sources causing the degradation of air quality by emitting PM loaded with PTEs are described below.

6.1 Traffic Emissions

Traffic emissions contribute to the atmospheric PM through both combustion and non-combustion processes. One significant source of PM from traffic is the combustion of lubricating oil in two-stroke engines, commonly found in motorcycles and scooters. This combustion has been identified as a major emitter of zinc (Zn) into the atmosphere (Aucélio et al., 2007; Begum et al., 2006, 2013). Additionally, non-exhaust traffic emissions play a significant role, as Zn is used during the vulcanization process in tire production (Kumar et al., 2020). Approximately 1.5% of the tire's weight is attributed to Zn (Ali et al., 2019). Moreover, the wear and tear of tires also contribute to the atmospheric burden of cadmium (Cd), copper (Cu), chromium (Cr), cobalt (Co), mercury (Hg), molybdenum (Mo), and nickel (Ni) (Ali et al., 2019). Pb may also be derived from traffic emissions. Although its use as a fuel additive has been banned, it is used in several vehicle parts, such as batteries, rims, and weights for wheel balancing (Hwang et al., 2016).

6.2 Brick Industries

The brick industry has experienced rapid growth in Bangladesh, with around 1000 kilns operating in the vicinity of Dhaka city alone. These kilns collectively emit an estimated 23,300 tons of $PM_{2.5}$ and 6000 tons of black carbon per year (Guttikunda et al., 2013). It has been speculated that brick kiln operations and traffic emissions are the two main sources of PM and associated element pollution in the air in Dhaka city (Salam et al., 2008). The exhaust from brick kilns is a major source of atmospheric As, derived from the combustion of fossil fuels containing As compounds (Kumar et al., 2021). Na, Ca, Mg, Mn, and Ni can also be emitted as a byproduct of coal combustion in brick industries (Shammi et al., 2021).

6.3 Soil and Dust

This source comprises emissions from resuspended soil and dust, road dust, unpaved roads,

and construction activities (Hopke et al., 2020). Al, Si, Fe, Mn, Ca, Mg, Ti, Rb, Sr, and Sc are emitted from resuspended soil, road dust, and unpaved roads (de Miranda et al., 2018; Rahman et al., 2019a). The presence of these elements in the air suggests an origin from the resuspension of dust by road-tire interactions (Gustafsson et al., 2008). Although Fe in the atmosphere is primarily derived from crustal sources, anthropogenic sources of Fe include fossil fuel combustion and biomass burning (Matsui et al., 2018), industrial and metallurgical processes (Wang et al., 2015), exhaust emissions (Salazar et al., 2020), and tire and brake wear (Wahid, 2018).

6.4 Biomass Burning

This category has multiple emission sources, including agricultural residue, wood, dung and municipal waste burning. Biomass burning is the major contributor to K in the air (Zhang et al., 2013). In Bangladesh, biomass and wood burning in brick kilns were identified as the dominant source of K emission (Rahman et al., 2019a). Sulfur (S) is also emitted from biomass burning. However, the source apportionment of S can be problematic as it is also emitted during energy production, refuse incineration, traffic exhaust, and from ships using heavy oil (Rahman et al., 2019a). Large-scale open biomass burning across the Indo-Gangetic Plain during winter significantly increases $PM_{2.5}$ concentrations in Bangladesh (Rahman et al., 2020).

6.5 Industrial Emissions

Industrial activities contribute significantly to the release of various toxic gases and metals into the atmosphere, with specific emissions depending on the nature of industrial operations. For example, Cd is released into the urban air from various mechanical processes performed in different industries, including battery, electroplating, and metal refining (Mondol et al., 2015). In European cities, ten potential sources of Cd were identified, including, but not limited to, coal, wood, gasoline and oil combustion, and cement production (Pacyna et al., 2009). Steel production and the use of phosphate fertilizer in agricultural lands also contribute to atmospheric Cd emissions (Hayat et al., 2019).

Chromium (Cr) is primarily originated from industrial processes (Rahman et al., 2014; Saha et al., 2016). Metal industries such as chrome plating and steel production, cement production, incineration of municipal refuse and sewage sludge, and emission from chromium-based automotive catalytic converters are the major sources of Cr emissions (ATSDR, 2012). In Bangladesh, industries involved in dye and pigment production, leather and wood preservation, cooling tower water treatment, drilling muds, textiles, and toner production for copying machines have been identified as sources of atmospheric Cr (Rahman et al., 2019a).

Calcium (Ca) is a good indicator of construction activities due to its widespread use in concrete production and as the principal ingredient of the wall putty (calcium carbonate) used for finishing walls before painting (Akther et al., 2019).

PM in industrial areas is enriched with Fe, Zn, Pb, Mn, Cr, Cu, Ni, Sn, Sb, As, Cd, Ag, and Co, which was attributed to scrap handling and iron-steel production (Kara et al., 2015). The co-presence of As, Pb, Sb, and Sn has been observed in relation to metallurgical works, fossil fuels burning, incineration of solid waste, and the ceramics and glass industry (Geagea et al., 2007; Kulkarni et al., 2006).

6.6 Sea Salt Spray

Wind stress at the ocean surface results in bubble bursting and jet drops, which are responsible for forming sea salt aerosols. About 80% of the PM mass in coastal areas is attributed to sea salt spray (Van Dingenen et al., 2004). Sea salt spray is the major source of Na and Cl in the air. However, Cl may originate from municipal waste incineration, while the co-presence of Cl, P, and Pb in the air indicates pyrogenic emissions (Rahman et al., 2019a). Additionally, Ni might come from sea salt sprays. However, vehicular repair workshops, paints and varnishes, and the recycling of telephone cables and electrical equipment may also contribute significantly to the source of atmospheric Ni (Salam et al., 2012).

6.7 Municipal Waste Incineration

Municipal waste includes a large variety of substances, for example, organic waste, batteries, packaging materials, paper, metal containers, glass, plastics, clothes, electronics, and furniture (Brindha & Schneider, 2019). Based on the composition of municipal waste, several elements may be emitted during incineration, though in general, municipal waste incineration emits Ni, Mo, Cr, and Sb (Khillare & Sarkar, 2012; Tian et al., 2012).

6.8 Lead-Acid Batteries as a Potential Source of Pb

Although the use of leaded gasoline in Bangladesh was banned since 1999, a substantial concentration of Pb remains in the atmosphere, contributing to health concerns. A report by the United Nations International Children's Emergency Fund (UNICEF) in 2020 revealed Bangladesh has the world's fourth highest death rate due to Pb exposure.

Approximately 80% of global lead consumption goes into the production of lead-acid batteries used not only in the traditional and electric vehicles but in energy storage for green technologies such as photovoltaic systems and wind turbines (Ericson et al., 2017). Due to the increased demand from the transport sector, the use of lead-acid batteries in Bangladesh has risen significantly, 97% of which are made from recycled batteries and scrap metal (Islam et al., 2019). The number of battery recycling operations throughout the country has been estimated between 1,100 (Ericson et al., 2017) and 12,000 per year (Ahmad et al., 2014). Unfortunately, many of these operations are unregulated, with few or no pollution controls. Other air toxins, such as As, Sb, Ba, Cd, and toxic gases including sulfur dioxide, chlorine, dioxins, and dibenzofurans, can also be released during the recycling process (WHO, 2017). Apart from automobile repair shops, battery manufacturers and recycling facilities near residential areas (Akther et al., 2019), the use of lead-based paint applied on walls, toys, and furniture (Lin et al., 2009) may contribute to the enrichment of Pb in the air (Laidlaw et al., 2012; Srithawirat & Latif, 2015; Zahran et al., 2013).

As discussed above, the same element can entrain into the atmosphere from multiple sources, making the source identification process, using either the receptor-based model or mass balance approach, complex and ambiguous. However, a promising approach for accurate source identification is through isotope fingerprinting. By analyzing the unique isotopic signature of toxic elements, it becomes possible to trace their origins, even in scenarios where multiple sources of a particular element are mixed together. Surprisingly, the utilization of isotopic signature for source identification of toxic elements associated with particulate matter (PM) has not been reported in Bangladesh.

7 Particulate Matter Pollution in Bangladesh

Descriptive statistics of the reported concentrations for $PM_{2.5}$, PM_{10}, TSP, and $PM_{2.5–10}$ in different cities across Bangladesh have been compiled in Supplementary Table S1 and visually depicted in Fig. 3. In addition, the PM concentrations have been categorized into five different zones: residential, semi-residential, traffic, commercial, and industrial. These divisions are presented in Supplementary Table S1 and Fig. 3. It is worth mentioning that most of the studies were conducted in Dhaka, with only a few considering rural and other urban areas. This is due to the higher number of industries and traffic in Dhaka relative to other parts of the country.

According to the Bangladesh National Ambient Air Quality Standards (BNAAQS), the guideline value of $PM_{2.5}$ in ambient air is 65 μg m^{-3} for a 24-h period and 15 μg m^{-3} on an annual average (BNAAQS, 2005). In contrast, the WHO Air Quality Guidelines (WHOAQG) value of $PM_{2.5}$ is 15 μg m^{-3} for a 24-h period and 5 μg m^{-3} for an annual period (WHO, 2021). Based on our analysis, $PM_{2.5}$ concentrations in about half of the studies exceeded the BNAAQS guideline value of 65 μg m^{-3}. The exceedance percentage increased to ~ 98% when considering the WHOAQG suggested guideline value of 25 μg m^{-3}. The $PM_{2.5}$ concentrations in all of the cities and different functional areas in Bangladesh, except, Saidpur, were about 4–11 times higher than the WHOAQG recommended value of 15 μg m^{-3}. This indicates that Bangladesh is one of the most PM-polluted countries in the world. Among the cities, Gazipur had the highest $PM_{2.5}$ concentration of 257.3 μg m^{-3}, with a historical average concentration of 114 ± 68.6 μg m^{-3}. Gazipur is a highly industrialized region adjacent

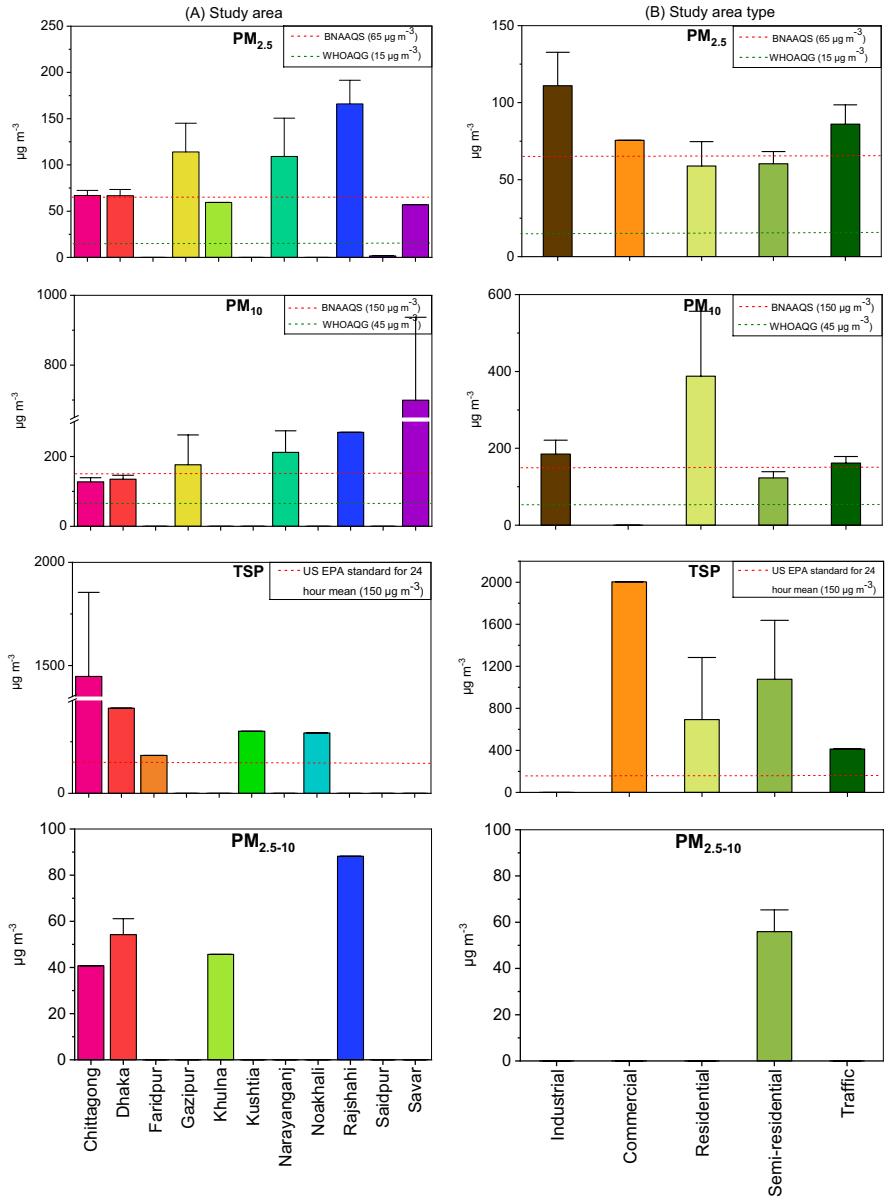

Fig. 3 Variations of different PM size fractions in different **A** study areas and **B** study area types (including megacity Dhaka and other cities) in Bangladesh (BNAAQS is the Bangladesh National Ambient Air Quality Standards and WHOAQG is the World Health Organization Air Quality Guidelines)

to the capital Dhaka city (Rahman et al., 2019b). In Dhaka city, the $PM_{2.5}$ concentration ranged from 17 to 191.8 μg m^{-3} with an average concentration of 66.9 ± 41.4 μg m^{-3}, which is slightly higher than the BNAAQS value for the 24-h mean and about four times higher than the WHOAQG value. The highest concentration of $PM_{2.5}$ (191.8 μg m^{-3}) in Dhaka city was reported at the Dhaka University campus, which is situated in an area with heavy traffic load (Salam et al., 2015). On the other hand, the lowest concentration (17 μg m^{-3}) was found at the Atomic Energy Centre in a semi-residential area (Begum & Hopke, 2018). The average $PM_{2.5}$ concentrations in other cities (except Khulna, Saidpur, and Savar) were also higher than the BNAAQS value for a 24-h period. The lowest $PM_{2.5}$ concentration of 1.7 μg m^{-3} was found in Saidpur. Generally, the residential and semi-residential areas of Dhaka city and divisional cities in Bangladesh showed a lower content of $PM_{2.5}$ in ambient air compared to the traffic and industrial areas, with the exception of Rajshahi city in north-western Bangladesh

(Fig. 3). This difference is pollution levels may be attributed to the transboundary transportation of PM from the Indo-Gangetic region of India, since Rajshahi shares a border with India. Additionally, the sampling period was winter, i.e., during December–February (Begum et al., 2015). Besides, a combination of meteorological conditions, long-range transport during the winter, and local sources result in PM concentrations remaining much higher than the BNAAQS (Begum et al., 2013, 2015). However, the Kruskal-Wallis test (non-parametric one-way ANOVA) indicated no significance difference of $PM_{2.5}$ level among the Bangladeshi cities and area types. Nevertheless, a pairwise comparison among area types indicated a significant difference in $PM_{2.5}$ concentration between semi-residential and traffic areas ($p = 0.019$) and semi-residential and industrial zones ($p = 0.002$) in Bangladesh.

The reported PM_{10} concentration in all the previous studies in Bangladesh surpassed the yearly WHOAQG value of 20 μg m^{-3} (WHO, 2021). Only a few studies recorded the PM_{10} concentrations lower than the 24-h guideline value of 45 μg m^{-3} recommended by WHO. Approximately 37% of the studies also exceeded the BNAAQS value of 150 μg m^{-3} for a 24-h period (BNAAQS, 2005). Analysis of historical average concentrations indicated that the highest PM_{10} was found in a kitchen in the Savar district (average, 699.4 μg m^{-3}; range, 133–1086 μg m^{-3}), which is more than 4 times higher than the BNAAQS value of 150 μg m^{-3} for 24 h. Such a high concentration of PM_{10} found in a kitchen was attributed to the combustion of wood as fuel for daily cooking activities. Poor ventilation in households was also suggested as a major factor for higher PM_{10} content in rural areas (Begum et al., 2009b). Historical average data also indicated a relatively high PM_{10} content in industrial and residential areas in Bangladesh (Fig. 3). The lowest PM_{10} concentrations (43 μg m^{-3}) were observed in a semi-residential area in Dhaka city (Mahmud et al., 2008). Similar to the $PM_{2.5}$, there was no significance difference in PM_{10} concentration among different locations and area types in Bangladesh. However, pairwise comparison indicated a significant difference between semi-residential and traffic areas ($p = 0.011$) and semi-residential and residential areas ($p = 0.025$), which is also reflected from historical average PM_{10} concentration in these areas.

Limited studies in Bangladesh have reported TSP concentrations, mostly from semi-residential areas in the Chittagong district, which limits the application of statistical analysis. However, compiled data showed that the highest concentration of TSP was found in semi-residential regions (1076 ± 838 μg m^{-3}) and Chittagong city (1447 ± 665.3 μg m^{-3}), which was about 7 and 10 times, respectively, higher than US-EPA recommended concentration of 150 μg m^{-3}. In addition, all the study areas in Bangladesh surpassed the guideline value of TSP for 24 h (USEPA, 2021). The lowest $PM_{2.5–10}$ content (40.7 ± 31.7 μg m^{-3}) was reported in a semi-residential area in Chittagong city (Begum et al., 2014), which is slightly lower than the WHOAQG value of 45 μg m^{-3} for 24 h of $PM_{2.5}$ level (WHO, 2021). The highest $PM_{2.5–10}$ content was 88.2 μg m^{-3} in Rajshahi city (Begum et al., 2014), which was about twice of WHOAQG value of 45 μg m^{-3} for 24 h of $PM_{2.5}$ level.

It should be highlighted that only a limited number of studies provided specific sampling dates, making it challenging to analyze temporal trends in PM concentrations. However, based on the available information, the recent studies documented a relatively higher presence of PM in the air. This surge could be attributed to the escalating levels of traffic and industrial activities, as well as the expanding population in Bangladesh.

8 Particulate Matter-Associated Black Carbon in Bangladesh

Incomplete burning of fossil fuels, vehicular exhaust, and biomass burning result in the release of black carbon (BC) into the atmosphere. Black carbon contributes a substantial mass of particulate matter, which is supported by the strong correlations between long-term (2003–2019) concentrations of BC and PM (Fig. 4). $PM_{2.5}$ contains relatively higher concentrations of BC than PM_{10}, possibly due to the higher surface area (Fig. 4). The BC concentration in $PM_{2.5}$, PM_{10}, and $PM_{2.5–10}$ fractions of residential, semi-residential, and traffic areas in Bangladesh is presented in Supplementary Table S2. It has been observed that the BC content was relatively higher in the $PM_{2.5}$ fraction in residential areas, possibly due to using biomass as cooking fuel in mud stoves. In rural and peri-urban residential areas of Bangladesh, low-grade solid biomasses (such as wood,

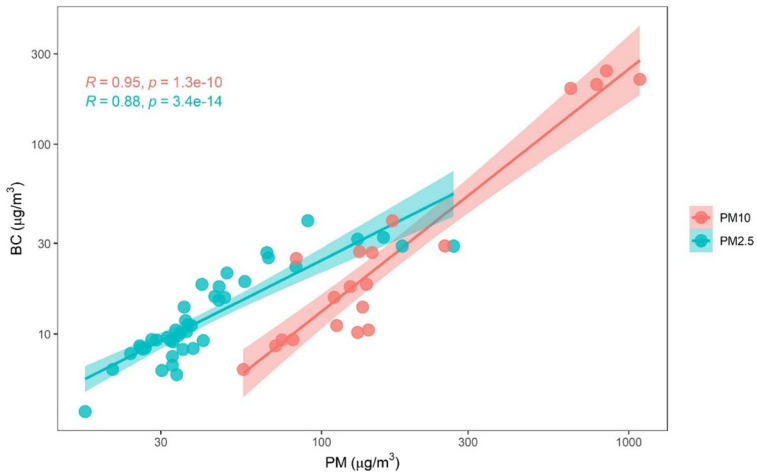

Fig. 4 Relationship between BC with $PM_{2.5}$ and PM_{10} in Bangladesh (The complied data from 2003 to 2019 were used to generate the correlation)

agri-wastes, and cattle dung) are still commonly used for cooking, which may trigger the release of a higher amount of BC per unit of energy due to low combustion efficiency. A study by Begum et al., (2009a, 2009b) reported that the concentration of BC associated with PM_{10} emitted from a kitchen using liquified petroleum gas as fuel in Savar was 27.2 μg m^{-3}. In comparison, the emission of BC from a kitchen in the same area using wood and cow dung as fuel has substantially increased to 244 μg m^{-3}. A recent study conducted in Dhaka city also reported a reduction in BC in 2016 compared to the level observed in 2001–2002 in the Farmgate area. This decline was attributed to the conversion of motor vehicles from petrol to CNG engines after 2003 (Rahman et al., 2021a). Additionally, the presence of BC in the atmospheric particles is influenced by seasonal variations and transboundary air transport. The higher BC levels observed during the winter period in Dhaka city were associated with minimal rainfall and increased resuspension of dust particles (Begum et al., 2013). Moreover, the movement of air parcels from north and northwest directions originating in India was identified as a potential contributor to the BC inventory in Bangladesh. During the monsoon season in Dhaka, reduced BC concentrations were described by wet scavenging and the washout of air particles following continuous precipitation.

9 Comparative Analysis of PM Pollution Between Bangladesh and Global Megacities

PM concentrations ($PM_{2.5}$, PM_{10}, and TSP) in different megacities in the world were compared with the average concentration of all compiled data in Dhaka, Bangladesh (Fig. 5, Table S3). There was no significance difference of respective $PM_{2.5}$, PM_{10}, and TSP concentrations among global megacities indicated by the Kruskal–Wallis test, although there was an apparent variability in their respective concentration across megacities.

Based on the compiled data, Asian megacities, particularly in Tehran, Iran (525 μg m^{-3}); Lahore, Pakistan (522 μg m^{-3}); and Kolkata, India (313 μg m^{-3}) exhibited very high average $PM_{2.5}$ concentrations. Elevated levels of $PM_{2.5}$ were also observed in Chengdu, China (165.1 μg m^{-3}); Sao Paulo, Brazil (142.5 μg m^{-3}); Lagos, Nigeria (122.4 μg m^{-3}); and Tianjin, China (120.3 μg m^{-3}). In other world megacities, the average $PM_{2.5}$ concentration ranged between 5.85 μg m^{-3} in Maringa, Brazil, and 86.2 μg m^{-3} in Cairo, Egypt. However, $PM_{2.5}$ concentrations in a few megacities were lower than the WHOAQG value of 15 μg m^{-3}, for example, Londrina, Brazil (7.47 μg m^{-3}); Maringa, Brazil (5.85 μg m^{-3}); Los Angeles, USA (13.8 μg m^{-3}); and New York, USA

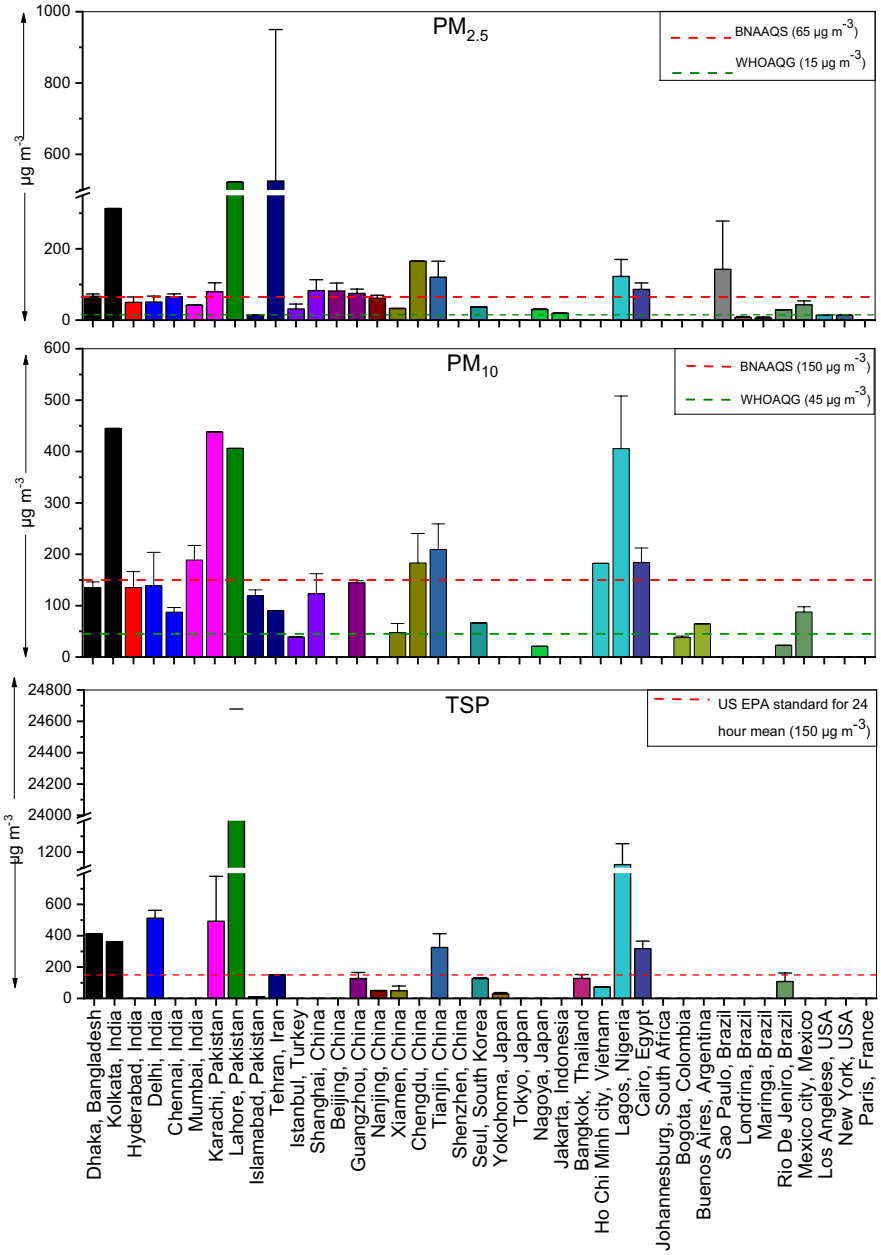

Fig. 5 $PM_{2.5}$, PM_{10}, and TSP concentrations (μg m^{-3}) in ambient air in world megacities based on the data included in this study (BNAAQS is the Bangladesh National Ambient Air Quality Standards, WHOAQG is the World Health Organization Air Quality Guidelines, and US EPA standard is the United States Environmental Protection Agency Standard Values)

(13.4 μg m^{-3}). In Jakarta and Islamabad, the $PM_{2.5}$ values of 19.3 and 15.1 μg m^{-3}, respectively, were slightly higher than the WHOAQGV guideline. In contrast to these megacities, $PM_{2.5}$ content in others was about 2–35 times higher than the guideline value of 15 μg m^{-3} for 24 h. Most of the global megacities reported lower $PM_{2.5}$ concentrations than Dhaka, which has a historical average concentration of more than 4 times higher than that of the WHOAQG value of 15 μg m^{-3} and slightly higher than the BNAAQS value of 65 μg m^{-3}. Some megacities, however, showed similar $PM_{2.5}$ concentrations to Dhaka, such as Nanjing, China (61.4 μg m^{-3}); Chennai, India (65.7 μg m^{-3}); Guangzhou, China (75 μg m^{-3}); and Karachi, Pakistan (79.5 μg m^{-3}) (Fig. 5). Nevertheless, recent studies in Dhaka indicated a very high level of $PM_{2.5}$ concentration that surpassed the recommended guideline values and exceeded

those observed in several megacities, for example, 191.8 μg m^{-3} in the Dhaka University campus (Salam et al., 2015), 175.8 μg m^{-3} in Darussalam area (Rahman et al., 2019b), 159.07 μg m^{-3} in Farmgate region (Rahman et al., 2019a), and 172.23 μg m^{-3} in Mirpur-10 area (Hossain et al., 2019). These locations in Dhaka are highly urbanized with congested traffic zones and a very high anthropogenic load that might have contributed to such a high level of $PM_{2.5}$ in the air.

Most of the global megacities show average PM_{10} contents higher than the WHOAQG of 45 μg m^{-3}, except Nagoya in Japan (21.18 μg m^{-3}), Rio De Janeiro in Brazil (22.9 μg m^{-3}), Istanbul in Turkey (39.1 μg m^{-3}), and Bogota in Colombia (37.8 μg m^{-3}). An extremely high PM_{10} concentration was recorded in Lagos, Nigeria (405.6 μg m^{-3}); Lahore, Pakistan (406.2 μg m^{-3}); Karachi, Pakistan (438 μg m^{-3}); and Kolkata, India (445 μg m^{-3}), which are about 3 times higher than Dhaka, Bangladesh. In Dhaka city, the historical average PM_{10} content was 134.8 μg m^{-3}, which is about 3 times higher than the WHOAQG of 45 μg m^{-3}, but slightly lower than the BNAAQS value of 150 μg m^{-3} for 24-h average. Some of the Asian megacities reported similar PM_{10} concentrations to Dhaka, for example, Hyderabad, India (135.1 μg m^{-3}); Delhi, India (138.6 μg m^{-3}); Islamabad, Pakistan (119.5 μg m^{-3}); Shanghai, China (123.3 μg m^{-3}); Guangzhou, China (144.4 μg m^{-3}) (Table S3).

Among the studied world megacities, the TSP concentrations in Lahore city of Pakistan showed an extreme level, which ranges from 16,690 to 26,650 μg m^{-3}, while the lowest content was found in Nanjing, China (5.0 μg m^{-3}) (Table S3). Compared to Dhaka, few megacities showed relatively higher TSP in the air, while most of the cities showed much lower TSP than Dhaka, Bangladesh. The US-EPA guideline value for TSP in the air is 150 μg m^{-3} and historical average TSP concentrations in Lahore, Pakistan (21,122.5 μg m^{-3}); Lagos, Nigeria (1119.7 μg m^{-3}); Delhi, India (512.5 μg m^{-3}); Karachi, Pakistan (492.7 μg m^{-3}); Dhaka, Bangladesh (413 μg m^{-3}); Kolkata, India (362.4 μg m^{-3}); Tianjin, China (324.6 μg m^{-3}); Cairo, Egypt (317.5 μg m^{-3}); and Tehran, Iran (151.6 μg m^{-3}) surpassed the guideline value. The highest TSP level in Lahore might originate from a number of sources, for example, metallurgy and other industrial operations, traffic exhaust, and biomass burning along with a huge anthropogenic load in the city (Jalees and Asim 2016). Additionally, climatic variables might have influenced the heavy load of aerosols in the air of these cities (Mushtaq et al. 2022).

10 PM-Bound Elements in Bangladesh

There is a considerable variation in the reported major and trace elements and their content in different PM fractions. However, the most frequently reported elements in Bangladesh include Al, Ca, Fe, K, Mg, Mo, Na, P, S, and Si (major elements) and As, Cd, Cr, Cl, Cu, Mn, Ni, Pb, Ti, and Zn (trace elements). Supplementary Table S4 provides descriptive statistics of their content in $PM_{2.5}$, PM_{10}, TSP, and $PM_{2.5–10}$ in Bangladesh, while Fig. 6 illustrates the concentration of major and trace elements in various PM fractions.

Among the major elements, concentrations of Ca, Fe, K, and Mg were reported in all PM fractions in Bangladesh. In general, the concentrations of these elements were higher in PM_{10} fraction (Ca = 19.42, Fe = 7.43, K = 10.53, and Mg = 6.14 μg m^{-3}), except Fe in TSP reported at Purana Paltan crossing, Dhaka (maximum of 29.1 μg m^{-3}), followed by Motijheel, Dhaka (maximum 26.1 μg m^{-3}) (Salam et al., 2003). In the PM_{10} fraction, the highest Fe content was found in Tejgaon industrial area, Dhaka (10.8 μg m^{-3}) (Rahman et al., 2013). In $PM_{2.5}$, the highest Fe concentration was reported at Farmgate, Dhaka (3.41 μg m^{-3}) (Rahman et al., 2019a). These elements are soil-forming elements, which might be why their higher abundance was found in coarser PM than the fine ($PM_{2.5}$) fraction.

In contrast to geogenic elements, the finer fraction ($PM_{2.5}$) containing higher S (average, 4.97 μg m^{-3}; range, 0.18–48.5 μg m^{-3}) was recorded at Farmgate, Dhaka, which is one of the busiest traffic areas in Dhaka city (Rahman et al., 2019a).

The highest Cl concentration was reported in $PM_{2.5}$ (average, 5.36; range, 0.08–35.01 μg m^{-3}) in the Saidpur district, which was attributed to biomass burning in traditional mud stoves for cooking (Chowdhury et al., 2012). Apparently, the Cl content was relatively higher in $PM_{2.5}$ than in $PM_{2.5–10}$. In $PM_{2.5–10}$, the highest Cl concentration was 1.14 μg m^{-3} found at the Atomic Energy Center, Dhaka (Begum et al., 2011). However, in Chattogram, Cl was higher in $PM_{2.5–10}$

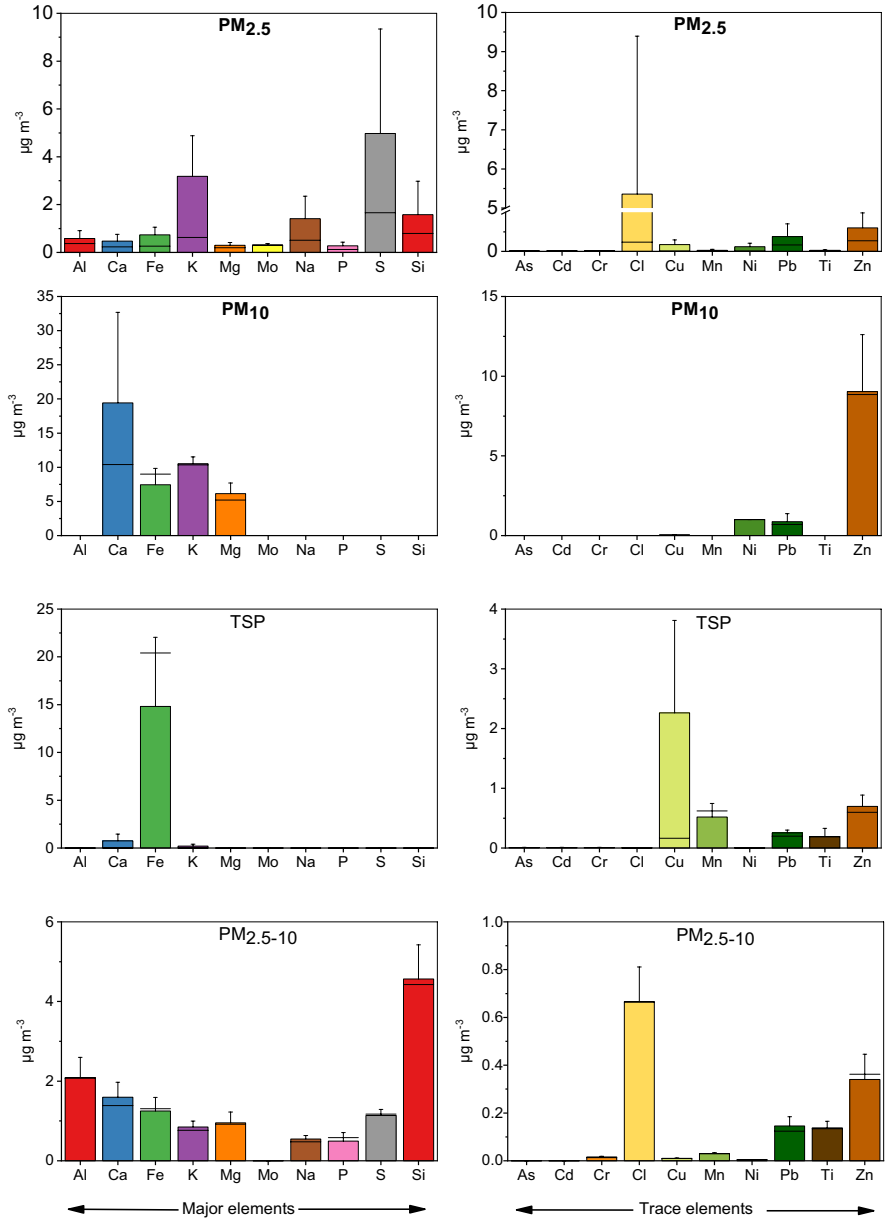

Fig. 6 Particulate matter (PM)-bound major and trace element concentrations (μg m^{-3}) in Bangladesh

(0.80 μg m^{-3}) than in $PM_{2.5}$ (0.24 μg m^{-3}), where a maximum of 4.2% was contributed by sea salt (Begum et al., 2009a). Probably, the degree of contribution of Cl from different sources is the reason for the difference in Cl content in different PM fractions.

Concentrations of As, Cd, Cr, Cu, and Pb in the air are a matter of concern due to their possible carcinogenicity even at minute concentrations. Data for As concentrations in Bangladeshi air was scarce. However, the As content in $PM_{2.5}$ ranges between 0.004 and 0.015 μg m^{-3}, averaging 0.008 μg m^{-3} (Table S4). The highest As concentration was found at Saidpur, principally released from stoves due to biomass and wood-burning (Chowdhury et al., 2012). A slightly higher average concentration of As was found in TSP (average, 0.009; ranges, 0.003–0.012 μg m^{-3}). Gladly, the airborne As concentration is much lower than the guideline value of 10 μg m^{-3} of inorganic arsenic. Basically, the United States Occupational Safety and Health Administration (OSHA) and National Institute

of Occupational Safety and Health (NIOSH) mandate of 10 μg m^{-3} inorganic As averaged over any 8-h period for a 40-h workweek, as the permissible limits for arsenic occupational exposure (NIOSH, 2005).

The Cd concentrations in PM range between 0.0002 and 0.035 μg m^{-3}, where most of the studies quantified Cd content in $PM_{2.5}$ or TSP (Fig. 6). The highest Cd content in $PM_{2.5}$ (0.035 μg m^{-3}) was found at a Pharmaceutical company (Tejgaon industrial area, Dhaka), while the lowest concentration (0.0002 μg m^{-3}) was around Tower Bhavan of Dhaka University (semi-residential area in Dhaka) (Salam et al., 2008). In TSP, the highest Cd concentration was 0.024 μg m^{-3} at Newmarket, Chittagong (Ahmed et al., 2012), while the lowest (0.0008 μg m^{-3}) was observed in the semi-residential area of Nilkhet, Dhaka city (Salam et al., 2003).

The Cr content in PM in Bangladesh ranges from 0.004 to 0.08 μg m^{-3} at Saidpur and Farmgate, Dhaka, respectively, emitted from mud-stoves. Based on the average concentration, the highest Cr was found in $PM_{2.5–10}$ (average, 0.02; range, 0.01–0.03 μg m^{-3}), whereas the lowest was found in TSP (average, 0.009; range, 0.005–0.013 μg m^{-3}).

In Bangladesh, atmospheric Cu content was reported for $PM_{2.5}$, TSP, and $PM_{2.5–10}$. Only one study reported Cu (0.028 μg m^{-3}) in PM_{10} in Dhaka, Bangladesh (Mahmud et al., 2008). Overall, the Cu concentration ranges from 0.0006 to 14.48 μg m^{-3} (Table S4). The highest Cu content was reported for TSP in the GEC circle area in Chittagong city, while the lowest was for $PM_{2.5}$ at the Atomic Energy Centre, Dhaka (Boman et al., 2005).

Pb in ambient air in Bangladesh ranges between 0.001 and 5.51 μg m^{-3} at different locations. The highest Pb content was reported for $PM_{2.5}$ at Farmgate (a highly traffic-congested area in Dhaka), while the lowest was reported for $PM_{2.5}$ at Atomic Energy Centre (a semi-residential place in Dhaka) (Boman et al., 2005; Rahman et al., 2019a). Historical data shows that during the dry months spanning from November 1994 and January 1996 in Dhaka city, the $PM_{2.5}$-associated Pb content reached 0.463 μg m^{-3}, making Dhaka the most heavily Pb-polluted city in Bangladesh (Khaliquzzaman et al., 1997). Although several studies indicated a decreasing trend in Pb pollution in the air, this review found several times higher Pb levels than 90 s studies reported in Dhaka city's ambient air (Fig. S4). For example, Rahman et al. (2019a, 2019b) reported that Pb in $PM_{2.5}$ was 5.51 μg m^{-3} at Farmgate, and Akther et al. (2019) found that Pb in PM_{10} were 3.3, 1.2, 0.7, and 0.7 μg m^{-3} at Mirpur, Tongi, Khilkhet, and Rampura in Dhaka city, respectively. While Pb was phased out from gasoline in 1999, a substantial background concentration of Pb persists, contributing to its elevated presence in the air. The atmospheric Pb levels in Bangladesh are closely related to traffic emissions. Additionally, lead battery recycling and lead-based paints are the noteworthy sources of Pb pollution in the atmosphere of Dhaka city.

The range of Ti content in Bangladeshi air ranged between 0.001 μg m^{-3} in $PM_{2.5}$ in the outdoor environment of a kitchen in Saidpur and 0.28 μg m^{-3} in TSP in a semi-residential area in Dhaka city (Chowdhury et al., 2012; Khaliquzzaman and Biswas, 1992).

The principal component analysis of PM-associated PTEs in Bangladesh was performed to evaluate inter-correlation of PTEs among different PMs (Fig. 7, Table S11). The first two components explained about 60% of the total variance, where the first and second components explained about 37% and 22% of the variance, respectively (Fig. 7). The first component has higher loadings on Al, Ca, Cr, Cl, Cu, K, Mg, P, Pb, S, Si, and Zn as well as PM_{10} samples, indicating these elements are in higher concentrations in PM_{10} than other PM fractions. Precisely, major elements are strongly associated with PM_{10} as it is also inferred from concentration data, described earlier. Similarly, trace elements, like As, Cd, Co, and Ni, have higher positive loading on the second principal component along with $PM_{2.5}$ samples. Exceptionally, Ni, P, and S have higher loadings on both components, implying their significant presence in both PM_{10} and $PM_{2.5}$ in Bangladeshi air. Further, the PCA results imply a mixed and intriguing origin of PM and associated PTEs in Bangladesh.

11 PM-Bound Elements in Global Megacities

This section outlined the current status of the major and trace elements in different fractions of atmospheric PM ($PM_{2.5}$, PM_{10}, and TSP) in megacities worldwide (Table S5-S10 and Figs. S4-S9). In general, concentrations of major elements were higher than the trace elements in all types of PM fractions, with some exceptions. Al, Ca, Fe, K, Mg, Mn, Mo,

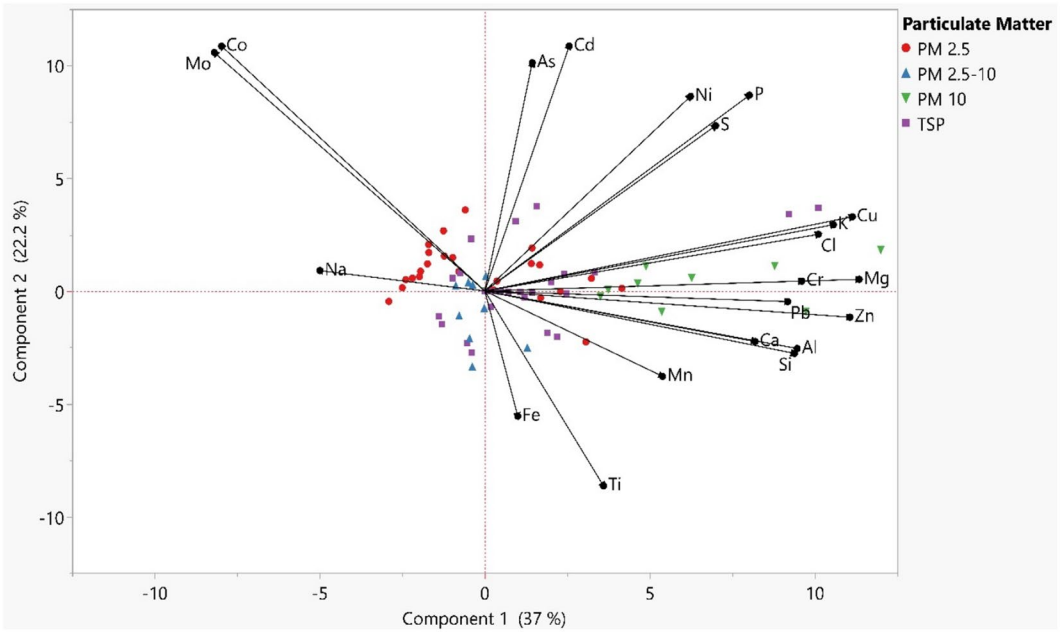

Fig. 7 Principal component analysis (PCA) biplot of particulate matter-associated PTEs in Bangladesh

Na, S, Si, and Zn concentrations were relatively higher than other elements in PM. It is important to note that the elemental concentrations vary significantly worldwide. This variation is due to the difference in geographical location, population density, variable pollution sources, and different degrees of vehicular and industrial activities.

11.1 Elements Bound to $PM_{2.5}$ Fraction

Apparently, among the $PM_{2.5}$-bound trace elements, the concentration of Cl was higher followed by Zn in ambient air of world megacities (Fig. S5). However, $PM_{2.5}$-associated Cl concentrations in the world megacities were about 6–46 times lower than its historical average concentration in Dhaka (1.83 μg m^{-3}). An exceptionally higher Cl concentration of 20.79 μg m^{-3} was reported in Cairo, which is ~ 11 times greater than Dhaka (Fig. S5, Table S5). Such high Cl in Cairo's air was attributed to sea salt and secondary ammonium chloride origin (Abu-Allaban et al., 2007). Additionally, concentrations of Zn in Dhaka (0.73 μg m^{-3}) were relatively higher than most megacities worldwide. However, Zn levels in Kolkata (0.54 μg m^{-3}), Delhi (0.65 μg m^{-3}), Guangzhou (0.59 μg m^{-3}), Nanjing (0.56 μg m^{-3}), Chengdu (0.82 μg m^{-3}), Bangkok (0.80 μg m^{-3}) and Lagos (0.67 μg m^{-3}) were close to Dhaka (Table S5). Contrarily, Zn concentrations in Karachi (2.48 μg m^{-3}), Lahore (7.29 μg m^{-3}) and Tehran (2.28 μg m^{-3}) were about 3–10 times higher than in Dhaka. The higher Zn content in Lahore might have originated from tire wear and burning of fuels in vehicles (Klimaszewska et al., 2007).

Among the $PM_{2.5}$-bound major elements, the concentration of S in global megacities was much lower than in Dhaka except in Bangkok (17.05 μg m^{-3}), which was about three times higher. In Bangkok, K concentrations (7.68 μg m^{-3}) were also very high. The burning of agricultural waste was suggested as a source of K origin (Pongpiacha et al., 2014). Overall, most of the major elements in Bangkok showed higher concentrations than other global megacities, indicating severe air pollution (Fig. S6). Several natural and anthropogenic sources, including resuspension of road dust, sea salt aerosols from the Gulf of Thailand, crustal sources, and construction dust have contributed to such a high major elemental concentration in Bangkok (Pongpiachan et al., 2017). Among the soil forming elements, for example, Al, Ca, Fe, Mg, and K, have an elevated content in some Asian megacities like Karachi and Tehran that might

have originated from soil/road dust, industrial emissions, vehicular emissions, sea salt originated from the Arabian Sea and secondary aerosols (Mansha et al., 2012). Nevertheless, concentrations of both $PM_{2.5}$-bound trace and major elements in American megacities were comparatively very low, with a few exceptions. For example, Ti (0.18 μg m^{-3}), Fe (1.39 μg m^{-3}), and Si (2.30 μg m^{-3}) in Mexico City; S (1.39 μg m^{-3}) in New York; Cu (0.18 μg m^{-3}), Ti (0.13 μg m^{-3}), Zn (0.23 μg m^{-3}), Fe (4.04 μg m^{-3}), P (0.17 μg m^{-3}), and S (2.46 μg m^{-3}) in Sau Paolo were relatively higher or close to those in Asian megacities.

11.2 Elements Bound to PM_{10} Fraction

Among PM_{10}-bound trace elements, concentrations of As and Cl were less reported than other elements in different world megacities (Fig. S7). Among the megacities, Hyderabad recorded very high concentrations of As at 0.87 μg m^{-3}, although this figure remains well below the permissible limits for occupational As exposure of 10 μg m^{-3} (NIOSH, 2005). The average Zn concentration (12.63 μg m^{-3}) was also higher in Hyderabad (India), which was slightly higher than that of Dhaka (9.04 μg m^{-3}) (Fig. S7). It was speculated that resuspended dust, vehicular emission, combustion processes, industrial emissions, and refuse burning were the major contributors to these elements in Hyderabad (Gummeneni et al., 2011). Besides, the highest PM_{10}-bound Cu was found in Karachi, (0.42 μg m^{-3}), followed by Ho Chi Minh City, (0.39 μg m^{-3}). Average Cu concentrations were also relatively higher in Kolkata (0.11 μg m^{-3}), Mumbai (0.14 μg m^{-3}), Lahore (0.23 μg m^{-3}), Islamabad (0.21 μg m^{-3}), Lagos (0.17 μg m^{-3}), and Dhaka (0.03 μg m^{-3}) (Fig. S7). Biomass burning, vehicular emissions, and resuspended dust contributed to high Cu in the air (Shahid et al., 2018).

Among the major elements in Dhaka, Ca, Fe, K, and Mg were mainly reported in the literature, and their concentrations increased in the following order: Mg (6.14 μg m^{-3}) < Fe (7.43 μg m^{-3}) < K (10.53 μg m^{-3}) < Ca (19.42 μg m^{-3}) (Table S8). The Ca content in megacity Dhaka (19.42 μg m^{-3}) was close to Lahore (18.36 μg m^{-3}), Tianjin (18.39 μg m^{-3}), and Cairo (14.52 μg m^{-3}) (Fig. S8). The concentration of Ca in Karachi (57.29 μg m^{-3}) was roughly three times higher than in Dhaka due to road/soil dust resuspension and biomass burning (Shahid et al., 2018). The PM_{10}-bound K concentration in Dhaka was the highest among the studied megacities (Fig. S8). As discussed earlier, biomass burning is the major contributor of K in Bangladeshi air. Overall, American and Chinese megacities, with the exception of Beijing, exhibit comparatively lower level of PM_{10}-bound elements when compared to megacities in other parts of the world.

11.3 Elements Bound to TSP Fraction

In Dhaka, the historical average concentration of TSP-bound trace elements was highest for Zn (0.64 μg m^{-3}), while Cl exhibited the lowest concentration (0.002 μg m^{-3}). Additionally, concentrations of Pb (0.23 μg m^{-3}) and Ti (0.19 μg m^{-3}) were relatively elevated in Dhaka (Table S9). The Cl content in the TSP was reported only for Dhaka (0.002 μg m^{-3}) and Ho Chi Minh City, Vietnam (1.27 μg m^{-3}), predominantly originating from sea salts (Phan et al., 2020). However, a significant variation in TSP-bound Zn content can be observed across other global megacities. Zn concentrations varied from 0.0008 μg m^{-3} in Tianjin (China) to 25.64 μg m^{-3} in a high-traffic area of Johannesburg (South Africa) (Fig. S9). Notably, Zn concentrations in Chinese and Japanese megacities were relatively lower than the other counterparts. However, certain megacities, including Johannesburg (25.64 μg m^{-3}), Lahore (12.64 μg m^{-3}), Mumbai (7.35 μg m^{-3}), Delhi (3.26 μg m^{-3}), Lagos (2.55 μg m^{-3}), and Bangkok (2.33 μg m^{-3}), displayed exceptionally high Zn content in the TSP fraction. All of these sites are located in urban and traffic areas and underscore the influence of traffic and urban context on Zn concentrations. The observed higher Zn concentration in the TSP fraction is possibly due to the larger particle size of this fraction relative to $PM_{2.5}$ and PM_{10}.

Among the major elements, the concentration of Fe (14.82 μg m^{-3}) was the highest in Dhaka, whereas the lowest was found for Mg (0.0004 μg m^{-3}). The concentrations of Al, Mo, P, S, and Si in TSP were not reported for Dhaka (Table S10). Examining the global megacities, the highest concentrations of Al (13.09 μg m^{-3}) and K (4.58 μg m^{-3}) were found in Delhi (India), Ca (35.26 μg m^{-3}) and P (0.65 μg m^{-3}) in Karachi (Pakistan), Na (37.43 μg m^{-3}) in Bangkok (Thailand), and Si (30.39 μg m^{-3}) in Tianjin (China). Interestingly, Johannesburg recorded the

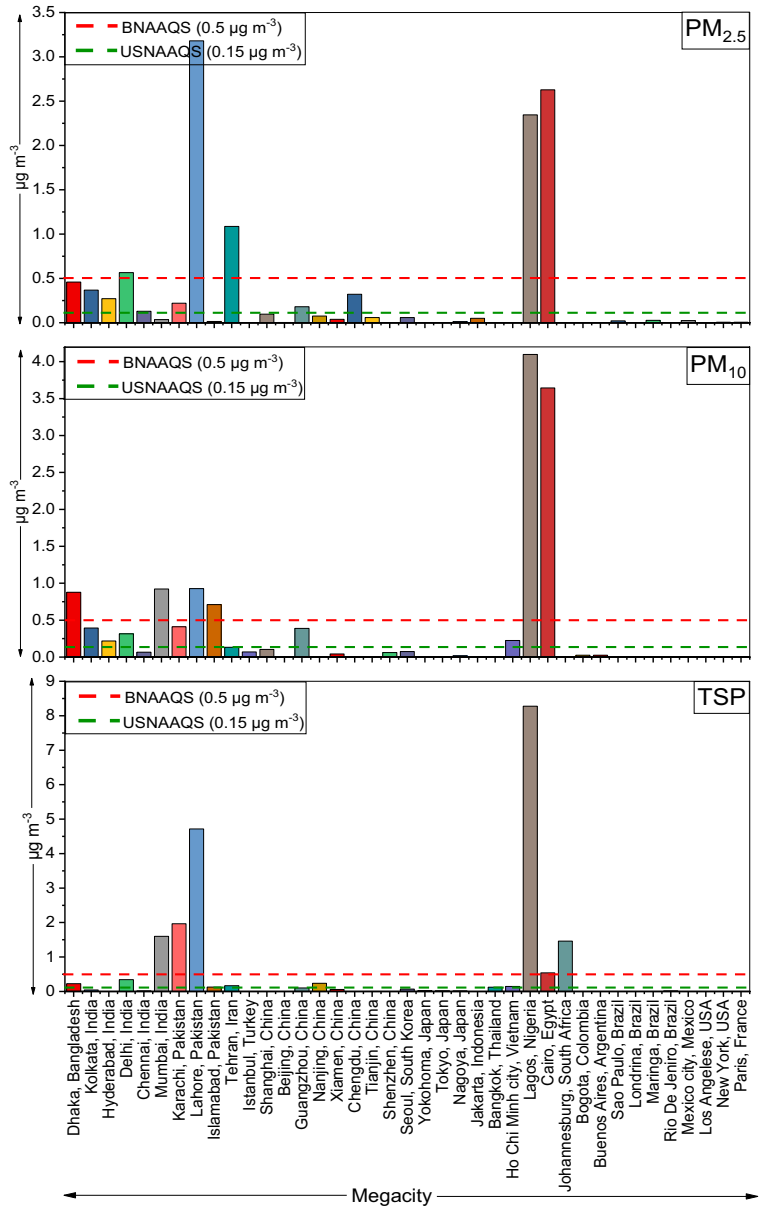

Fig. 8 Average concentrations (temporal and spatial average) of Pb (μg m.$^{-3}$) in world megacities based on the available data included in this study (BNAAQS is the Bangladesh National Ambient Air Quality Standards and USNAAQS is the United States National Ambient Air Quality Standards)

highest concentration of several elements, including Fe (67.06 μg m^{-3}), Mg (26.79 μg m^{-3}), Mo (12.56 μg m^{-3}), and S (13.98 μg m^{-3}) (Fig. S10).

12 Atmospheric Pb in the Global Megacities

The concentration of Pb in three different fractions of atmospheric particles in multiple world megacities is depicted in Fig. 8. South Asian and African megacities showed higher levels of Pb compared to other regions. Chinese megacities, except Chengdu, Guangzhou, and Shanghai, exhibited lower levels of Pb. North American megacities contained a lesser amount of Pb. Although the use of Pb in gasoline was banned in most countries, the legacy of past emission still contributes to the environmental Pb. In some recent studies, for example, Lahore, Pakistan, $PM_{2.5}$-bound Pb content was 3.18 μg m^{-3} (range, 1.78–18.1 μg m^{-3}), which was related to the emission from battery units, paint, alloy, plastics, and rubber industries (Ahmad et al., 2021). Hence,

both historical and present-day sources contribute to higher Pb levels in various PM fractions globally.

In Bangladesh, the national air quality standard for atmospheric Pb (BNAAQS) is set at 0.5 μg m^{-3} (BNAAQS, 2005), a benchmark generally met by the majority of global megacities. However, some megacities, including Delhi (0.57 μg m^{-3}), Lahore (3.18 μg m^{-3}), Tehran (1.09 μg m^{-3}), Lagos (2.35 μg m^{-3}), and Cairo (2.63 μg m^{-3}), surpassed the BNAAQS value for Pb in $PM_{2.5}$. While Dhaka's average $PM_{2.5}$-associated Pb content (0.46 μg m^{-3}) slightly fell below the BNAAQS value, it exceeded the US National Air Quality Standard (USNAAQS) value of 0.15 μgm^{-3} by more than three times (USNAAQS, 2016). Moreover, the PM_{10}-associated Pb in Dhaka (0.88 μg m^{-3}) significantly exceeded the guideline value. Similarly, other megacities like Mumbai (0.92 μg m^{-3}), Lahore (0.93 μg m^{-3}), Islamabad (0.71 μg m^{-3}), Lagos (4.10 μg m^{-3}), and Cairo (3.64 μg m^{-3}) also displayed higher Pb levels than the BNAAQS standard (Fig. 8).

The concentration of TSP-bound Pb was relatively higher compared to $PM_{2.5}$ and PM_{10} in certain megacities such as Mumbai (1.60 μg m^{-3}), Karachi (1.96 μg m^{-3}), Lahore (4.72 μg m^{-3}), Lagos (8.28 μg m^{-3}), Cairo (0.54 μg m^{-3}), and Johannesburg (1.46 μg m^{-3}). The TSP-bound Pb in those areas was also higher than the guideline value of 0.5 μg m^{-3} for a 24-h average (Fig. 8). This difference can be attributed to the larger size range of TSP, which tends to accumulate higher levels of Pb.

The high concentration of Pb in Dhaka primarily originates from the combustion of fossil fuels by numerous vehicles. Other significant contributors to Pb pollution in Dhaka include emissions from roads, windblown dust, brickfields and various industries (Salam and Salam, 2011). Although the use of leaded gasoline was phased out in Bangladesh during the 1990s (Biswas et al., 2003), Pb from previously combusted fuel may still persist in the soil and dust and complete elimination from the environment has not yet been achieved. This ongoing issue highlights the unfinished story of environmental Pb in Bangladesh and the prolonged decline. Accumulated lead from earlier depositions in dust becomes windblown and resuspended, contributing to elevated lead concentrations in the air. This phenomenon is reflected in the soil dust collected from different roads in Dhaka city (Ahmed and Ishiga, 2006; Rahman et al., 2022).

Furthermore, the uncontrolled use of Pb in paints and fugitive emissions from battery manufacturing and other industries continue to contribute to Pb levels. It is essential to reduce airborne Pb levels by addressing the use of lead in paints and controlling emissions from suspected industries, particularly those related to lead-based batteries (Begum and Biswas, 2008). The observation of elevated Pb levels in the dust from schools in Dhaka city suggests a mixed source of Pb in the area (Rahman et al., 2021b). Upgrading air quality measures and implementing stricter regulations are crucial steps towards effectively tackling Pb pollution in Dhaka.

13 Summary Points, Challenges, and Recommendations

Air pollution is a critical environmental problem, especially in underdeveloped and developing countries undergoing unregulated industrialization and population growth. The current review reveals that the air particle sampling stations are predominantly located in the capital Dhaka, limiting the representation of air conditions across other cities in Bangladesh. However, the PM contents are higher in traffic and industrial areas in Bangladesh. The residential and semi-residential areas in Dhaka and other divisional cities showed a relatively lower concentration of $PM_{2.5}$, though seasonal trans-boundary pollution increases PM levels in the north-western divisional cities. There was insignificant variation in atmospheric $PM_{2.5}$, PM_{10}, and TSP concentrations in global megacities; however, Asian and African megacities generally had higher $PM_{2.5}$ concentrations, while Japanese and American megacities had lower concentrations.

The metadata analysis revealed that the association of PTEs to the various size fractions of air particles was diverse, with no discernable clear pattern observed. Only a few studies highlighted As pollution in the atmosphere of Bangladesh, although its pollution in water, vegetables, and food items is well investigated. Notably, recent years have witnessed an upward trajectory of atmospheric Pb levels in Bangladesh. On a global scale, the Pb level is relatively higher in south Asian and African megacities than in Chinese and north American megacities. Variations in PM and associated PTEs depend on factors like

sampling location, time, season, regional meteorological conditions, and local anthropogenic influences, both in Bangladesh and other global megacities.

To improve air quality, it is important to identify and address the major sources contributing to PM and associated elemental pollution. Several methods, such as multivariate statistics, EF, PMF, PCA, PM size differentiation, and elemental composition analysis, have been used to identify trace element sources. These methods indicated that in addition to natural sources, anthropogenic sources including traffic, industrial, and fossil fuel combustion play as significant role in the generation of PM and associated PTEs in the air of Bangladesh (Rahman et al., 2021a). However, pinpointing the sources of PTEs in the atmosphere solely based on receptor-based models or mass balance using measured elemental concentrations in air particles is not straightforward, as specific elements can originate from multiple sources that mix together. A robust and emerging technique for source identification of PTEs is the utilization of metal stable isotope fingerprinting in airborne particulate matter. Applying multi-isotope fingerprints in Bangladesh can provide valuable insights for discriminating the sources of air pollutants. Besides, increasing the number of ground monitoring stations for air quality monitoring will be useful to understand the dynamic level of air pollution throughout Bangladesh. Further, satellite monitoring with geospatial analysis would broaden the scope of air pollution monitoring in Bangladesh (Shang et al. 2023; Zhao et al. 2023).

However, implementing effective mitigation strategies is crucial for improving air quality and achieving substantial environmental and health benefits. The efforts should come from citizens, enterprises and from governments for fruitful outcomes. Based on the current understanding on air pollution in Bangladesh, the following suggestions can contribute to these efforts (Ambade et al. 2023; IQAir 2020; Sofia et al. 2020; UNEP 2015):

a. Personal contribution may include using face masks rated to filter $PM_{2.5}$ (disposable respirators known as N-95 or P-100 respirators), limiting outdoor activities, introducing improved cooking stoves, ensuring high-quality biomass fuels, and exploring alternative renewable energies to replace fossil fuels and increase the use of public transport to prevent pollution.
b. Enterprises like various industries and transport sectors can incorporate advanced technologies into industrial processes, regulating fuel quality, and promoting low-emission technologies such as hybrid electric vehicles.
c. The government and respective organizations may increase public awareness of the adverse health impact of PM pollution; enforce environmental-friendly measures; reduce PM emissions by limiting the growth of petrol vehicles, constructing electric car charging stations, enacting stronger laws for polluting industries, and developing city action plans with mitigation and adaptation strategies; and promote zero-emission vehicles and alternative fuels, replacing aging diesel engines and encouraging the use of public transport and active transportation methods.
d. Additionally, improving air quality in Bangladesh requires ensuring easy access to air quality monitoring data and raising awareness among school children and teachers. Successful pollution control strategies, such as the ban on two-stroke engines and implementing national air quality policies, have shown positive results. By establishing new air quality regulations, identifying pollution sources, and adopting effective mitigation strategies, Bangladesh can maximize population benefits, reduce illnesses related to air pollution, and create a healthier environment for its citizens.

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Declarations

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