Study on Mercury in PM₁₀ at an Urban Site in the Central Indo-Gangetic Plain: Seasonal Variability and Influencing Factors

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ABSTRACT

Mercury (Hg) is among the most toxic metals possessing a major threat to human health and aquatic ecosystems over the globe. However, measurement of Hg concentrations and seasonal variability remain poorly understood over the Indo-Gangetic Plain (IGP) in northern India. In this study, we present one-year data of particulate-bound mercury (HgP) in aerosol samples (PM₁₀) collected from Kanpur to understand seasonal variability and factors influencing concentration, as well as dry deposition flux. The HgP concentration exhibit a large temporal variability and ranged between 100 (on 14 June 2007) to 4340 pg m⁻³ (on 4 March 2007) with an annual average concentration of HgP is 776 ± 846 pg m⁻³. The HgP concentrations and HgP/PM₁₀ ratios showed a marked seasonality with the highest in winter (Dec-Feb) followed by post-monsoon (Oct–Nov) and summer (April–June) seasons. HgP and HgP/PM₁₀ were positively correlated ($r^2 = 0.77$, $p < 0.05$, $N = 58$) during the sampling period and the estimated dry deposition flux of HgP was 104.7 µg m⁻² y⁻¹. Although this study provides a comprehensive data set on HgP in an urban atmosphere of the IGP revealing high levels of HgP, measurement of gaseous Hg is needed for estimation of the total Hg budget. Therefore, future studies should focus on identification of different sources as well as emission characteristics of all forms of Hg (organic and inorganic forms) for better mitigation strategy to prevent health risks associated with toxic Hg in the region.

Keywords: Particulate mercury; Aerosol; Dry deposition flux; Indo-Gangetic Plain; South Asia.

INTRODUCTION

Mercury (Hg) is a highly toxic trace metal that poses a major threat to human health and aquatic ecosystems (Keeler *et al.*, 1995; Li *et al.*, 2017; Wright *et al.*, 2018). Emitted mercury can be transported and reach remote areas where it can be deposited in glaciers, water, and soil and accumulate (Lindberg *et al.*, 2007; Qie *et al.*, 2018; Tripathi *et al.*, 2019b). Sources of Hg emission can be natural, such as volcanoes and forest fires, or anthropogenic, such as coal-fired power plants, nonferrous metal smelters, waste incinerators, and cement production plants (Pacyna *et al.*, 2010; Chou *et al.*, 2018). These sources emit approximately

1230–2890 t of Hg per year globally (UNEP, 2008).

Particulate-bound mercury (HgP) is the fraction of total Hg associated with atmospheric aerosols and derived from various physical and photochemical processes in the atmosphere (Ariya *et al.*, 2015; Zhang *et al.*, 2019). The residence time for HgP ranges from hours to weeks; therefore, substantial amounts of Hg can be found in various urban, pristine, and remote environments. In pristine and remote areas, HgP concentrations are typically less than 10 pg m⁻³ (Liu *et al.*, 2010; Das *et al.*, 2016). However, in polluted urban-industrial regions and during atmospheric Hg depletion events, HgP concentrations can exceed 300 pg m⁻³ (Lindberg *et al.*, 2007; Das *et al.*, 2016). Moreover, HgP in industrialized regions is mostly anthropogenic and can account for up to 40% of atmospheric Hg (Xiao *et al.*, 1991).

Studies employing field measurements and multiscale model analysis (Bieser *et al.*, 2014), in polluted regions in Asia (Fu *et al.*, 2011; Xu *et al.*, 2015; Zhang *et al.*, 2017),

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Europe (Li *et al.*, 2008; Pyta and Rogula-Kozłowska, 2016), and the United States (Song *et al.*, 2009; Gratz *et al.*, 2013; Jiang *et al.*, 2013) have reported significant spatiotemporal variability in atmospheric HgP concentrations. For example, HgP concentrations are considerably higher in areas with abundant refineries, iron and steel manufacturing, power plants, chemical plants, coal-fired utility plants, and road traffic (Landis *et al.*, 2002; Zhang *et al.*, 2015; Siudek *et al.*, 2016; Cheng *et al.*, 2017; Li *et al.*, 2017; Lewandowska *et al.*, 2018). Moreover, emissions from biomass burning are source of atmospheric Hg and a crucial factor driving seasonal atmospheric Hg variation (Friedli *et al.*, 2009; De Simone *et al.*, 2017). In addition, studies have noted the associations of particulate matter and Hg with health, including cancer, respiratory symptoms, and mortality (Dockery and Pope, 1994; Kumari and Kulshrestha, 2018). In addition to several studies on chemical composition (Ram *et al.*, 2010; Sen *et al.*, 2016; Srivastava *et al.*, 2016), some research has recently been conducted on the isotopic ratio of carbon, both stable ^{13}C and radiocarbon ^{14}C , to characterize the sources of carbonaceous aerosols (i.e., biomass burning, coal combustion and other fossil fuels at Delhi (Sawhani *et al.*, 2019), Kanpur (Ghosh *et al.*, 2014; Bikkina *et al.*, 2017) and Lucknow and Varanasi in the IGP region (Sharma *et al.*, 2015, 2017; Sen *et al.*, 2018). Lead (Pb) isotopic compositions of aerosols have been used to study the combustion of fossil fuel and high-temperature industries to evaluate Pb contamination at Kanpur (Sen *et al.*, 2016). Although, HgP has been studied in various regions, and researchers have considerable temporal and spatial information; however, studies measuring Hg over South Asia remain limited (Guo *et al.*, 2017; Kumari and Kulshrestha, 2018; Tripathi *et al.*, 2019a). The only studies conducted at a rural site (Mahasar, Haryana) near

Delhi during winter and summer (Kumari and Kulshrestha, 2018) have revealed a high concentration of HgP (annual average: $756.7 \pm 436.3 \text{ pg m}^{-3}$ in particulate matter (PM_{10})). Hg emissions in South Asian countries, including India, are expected to increase because of economic and industrial growth and fossil fuel consumption (Burger Chakraborty *et al.*, 2013). However, systematic and long-term studies measuring Hg in India are lacking (Mukherjee *et al.*, 2009). Therefore, studies on atmospheric Hg in the Indo-Gangetic Plain (IGP) region are necessary for understanding Hg abundance and emission sources to mitigate health risks associated with toxic Hg in the region.

In this study, we measured HgP in aerosol samples (PM_{10}) collected from Kanpur from January 2007 to January 2008, excluding the monsoon period (July–September), to understand HgP abundance, seasonal variation and influencing factors. We further estimated dry deposition fluxes of HgP and investigated the relationships between HgP and chemical constituents (organic species and inorganic ions) to identify possible sources of HgP in Kanpur.

MATERIALS AND METHODS

Sampling Site Description

The sampling site, Kanpur (26.5°N, 80.3°E, 142 m asl), is in the central part of the IGP and highly urbanized (Fig. 1). Major cities in the IGP are characterized by high concentrations of anthropogenic aerosols because of rapid urbanization and emissions from industrial areas, thermal power plants, metal smelting sites, brick kilns, residential areas, and biomass-burning sites. Studies have suggested the high concentrations of PM in this region as the main reason for poor air quality, reduced visibility, and fog and haze

Fig. 1. Sampling site location, Kanpur, shown with red dot and HgP concentrations over surrounding regions.

formation during wintertime (Ram *et al.*, 2012; Ghosh *et al.*, 2014). Biomass burning significantly affects regional air quality in the IGP, particularly in the winter (Chen *et al.*, 2017). Major sources of biomass-burning emissions are crop harvesting and the common practice of wood burning for domestic use (Bikkina *et al.*, 2017). Kanpur has a mainly humid subtropical climate characterized by hot summers and cold winters (Ghosh *et al.*, 2014). In addition, the region contains numerous coal-burning thermal power plants. The major coal-burning thermal power plant nearest the sampling site is approximately 3 km to the southeast (Kanawade *et al.*, 2014) and hosts several brick kilns. In addition to anthropogenic emissions, long-range transport of aerosols by westerly and northwesterly winds assisted by high ambient temperature and low surface pressure occurs in summer (Singh *et al.*, 2005). Thus, high levels of PM and HgP in the atmosphere worsen air quality and pose a high health risk across the region.

The wind during the sampling period was mainly northwesterly and occasionally from the northeast or southwest (Figs. S1 and S2). The average relative humidity was approximately 60%, and the winds during winter and post-monsoon were typically weaker ($< 0.8 \text{ m s}^{-1}$) than those during summer.

Analytical Procedures

A high-volume aerosol sampler (Envirotech, Nagpur, India; model: 460 NL) was set up on the third floor of the Environmental Engineering Laboratory (approximately 15 m above ground level) at the Indian Institute of Technology Kanpur campus. Samples were collected from January 2007 to January 2008, excluding months during the monsoon season (July–September 2007). Ambient aerosol (PM₁₀) samples were collected on pre-combusted (550°C for approximately 6 h) tissuquartz filters (2500 QAT-UP, Pallflex, Putnam, CT, USA; size: $20.0 \times 25.4 \text{ cm}^2$). The sampler operated at a flow rate of $1.0 \pm 0.1 \text{ m}^3 \text{ min}^{-1}$. All samples were collected during the daytime and integrated over approximately 8–10 h. The samples were collected every fifth day during January and February and October–December. However, the sampling frequency was higher, twice per week, during March–June, the months in which mineral dust and mixed aerosols are more abundant in the region. The total PM mass (μg) was obtained gravimetrically, and the average PM mass concentration ($\mu\text{g m}^{-3}$) was calculated as the quotient of the net accumulated mass divided by the total volume of air (m^3). The net accumulated mass for each filter was calculated by subtracting the weight of the blank filter from that of the filter after sampling. The collected filters were sealed with aluminum foil and stored at -4°C before lab analysis. Approximately 1.77-cm^2 punches of filter samples were used to analyze HgP concentrations through cold vapor atomic absorption spectrometry after calcination in an O₂ stream and amalgamation on Au trap using a Leeman Hydra-II_C Direct Hg Analyzer (Leeman Lab Hydra, Teledyne Leeman Laboratories, Hudson, NH, USA; (Wang *et al.*, 2006; Guo *et al.*, 2017). This analysis was performed in the State Key Laboratory for Cryospheric Science at the Chinese Academy of Sciences (CAS) in Lanzhou,

China. HgP concentrations (pg m^{-3}) were obtained using HgT (Total mercury) data, and total air volume (m^3) in each sample was considered. Method blanks, field blanks, and standard reference materials were analyzed to ensure the quality of sample analysis. Each filter sample was analyzed three times, and the standard deviation of 10 replicate measurements from a blank quartz-fiber filter ($0.06\text{--}0.52 \text{ pg m}^{-3}$) with an average of 0.31 pg m^{-3} at the flow rate employed in our study for calculation of the method detection limit (MDL).

Calibration and accuracy of measurement was ascertained by measuring Hg concentrations in a series of standard solutions (0.01 mL) were prepared by diluting the Hg_(II) standard of 1000 mg L^{-1} . The correlation coefficient of calibration curve was 0.9999 for calibration standards of 0.05 mg L^{-1} , 0.1 mg L^{-1} , 0.2 mg L^{-1} , 0.4 mg L^{-1} and 0.8 mg L^{-1} . The detection limit of the instrument was 0.0006 ng . The Chinese geochemical standard reference material (GSS-9) (lake sediment, $[\text{Hg}] = 32 \text{ ng g}^{-1}$, from the China National Center for Standard Reference Materials, (GBW07423)) and lobster hepatopancreas standard reference material for Trace Metals TORT-2 ($[\text{Hg}] = 270 \text{ ng g}^{-1}$, National Research Council of Canada, Ontario, Canada) were interspersed with every 20 samples for external quality control. The recovery percentage for Hg ranged from 93% to 106% for GSS-9 and 98% to 107% for TORT-2, illustrating good analytical precision and accuracy of Hg determination (Huang *et al.*, 2016). The HgP concentrations in the field blanks were calculated below the average MDL to ensure minimal contamination during sampling, storage, transport, and treatment. The data on major ions, organic carbon (OC), elemental carbon (EC), and water-soluble organic carbon for the samples used for correlation analysis in this study were from previous works, and details on the analysis of these chemical species are described elsewhere (Ram *et al.*, 2010).

RESULTS AND DISCUSSION

Atmospheric HgP Concentrations at Kanpur

The concentration of HgP fluctuated widely from approximately 100 to 4340 pg m^{-3} , averaging $776 \pm 846 \text{ pg m}^{-3}$ during the sampling period (Table 1; Fig. 2(b)). High HgP concentrations were detected during the winter months, with the highest concentration in March (4340 pg m^{-3} on March 4) and lowest during summer (June 14, 100 pg m^{-3} on June 14; Figs. 2(a) and 2(b)). This variability indicates that the primary Hg emission sources varied considerably over the study period. However, in addition to variable emission sources, meteorological parameters contributed to the seasonal variation in HgP (this is discussed in Section 3.3).

The average atmospheric HgP concentration in Kanpur is similar to that in other South Asian urban areas such as Mahasar, India, and Kathmandu, Nepal (Guo *et al.*, 2017; Kumari and Kulshrestha, 2018). However, the annual average HgP concentration in Kanpur is approximately half those in Chinese megacities such as Beijing and Nanjing (Wang *et al.*, 2006; Zhu *et al.*, 2014); therefore, heavy HgP pollution likely plagues such megacities. Other Chinese urban areas such as Shanghai, Jinan, Qingdao, and Guiyang have atmospheric HgP concentrations nearly an order of magnitude lower than

Table 1. Concentrations of HgP in urban Kanpur and other urban sites worldwide.

Sites	Cutoff size	Period	HgP Concentration (pg m ⁻³)	PBM/PM (ng g ⁻¹)	References
Kanpur	PM ₁₀	2007–2008	776.4 ± 845.5	6554.8 ± 9731.5	This study
Mahasar, India	PM ₁₀	2014–2015	756.7 ± 436.3	1620 ± 1100	(Kumari and Kulshrestha, 2018)
Kathmandu, Nepal	TSP	2013–2014	850.5 ± 926.8	2586.0 ± 2072.1	(Guo <i>et al.</i> , 2017)
Lhasa, China	TSP	2013–2014	224 ± 14		(Huang <i>et al.</i> , 2016)
Beijing, China	TSP	2003–2004	1180 ± 82	1400 ± 917	(Wang <i>et al.</i> , 2006)
Shanghai, China	TSP	2004–2007	560 ± 220		(Xiu <i>et al.</i> , 2009)
Nanjing, China	PM ₁₀	2011–2012	1100 ± 57		(Zhu <i>et al.</i> , 2014)
Qingdao, China		2008–2011	194 ± 12; 304 ± 23		(Zhang <i>et al.</i> , 2015)
Xiamen, China	PM _{2.5}	2012–2013	174.4 ± 280.6		(Xu <i>et al.</i> , 2015)
Guiyang, China		2001–2002	368 ± 676		(Fu <i>et al.</i> , 2011)
Jinan, China	PM _{2.5}	2014–2015	508.5 ± 402.7		(Li <i>et al.</i> , 2017)
Tokyo, Japan		2000–2001	98 ± 51		(Sakata and Marumoto, 2002)
Seoul, Korea	TSP	2007–2008	65.4 ± 47.8		(Nguyen <i>et al.</i> , 2016)
Zabrze, Poland	PM ₁₀	2013	63.6 ± 53.0		(Pyta and Rogula-Kozłowska, 2016)
Goteborg, Sweden	PM _{2.5}	2005	12.5 ± 5.88		(Li <i>et al.</i> , 2008)
Chicago, USA	PM _{2.5}	2007	9 ± 20		(Gratz <i>et al.</i> , 2013)
Mississippi, USA	PM _{2.5}	2011–2012	4.58 ± 3.40		(Jiang <i>et al.</i> , 2013)
Toronto, Canada	PM _{2.5}	2003–2004	21.5 ± 16.4		(Song <i>et al.</i> , 2009)

that in Kanpur (Xiu *et al.*, 2009; Fu *et al.*, 2011; Zhang *et al.*, 2015; Li *et al.*, 2017). By contrast, much lower HgP levels (approximately fourfold that of Kanpur) can be found in coastal Chinese cities. Xiamen has a cleaner air mass than the inland Chinese cities (Xu *et al.*, 2015). Nonetheless, the HgP level in Kanpur exceeds those in East Asian cities, such as Seoul and Tokyo (Sakata and Marumoto, 2002; Nguyen *et al.*, 2016), European cities (Li *et al.*, 2008; Pyta and Rogula-Kozłowska, 2016), and North American cities (Song *et al.*, 2009; Gratz *et al.*, 2013; Jiang *et al.*, 2013) by more than one or two orders of magnitude. Atmospheric HgP concentrations and HgP/PM₁₀ ratios in Kanpur are compared with those from previous studies at other urban locations in Table 1. Although the comparisons are not absolutely accurate because of methodological differences (e.g., extraction/digestion methods, thermal-reduction methods, PM sizes, and sampling times), they reveal considerable spatiotemporal variation in HgP. These variabilities are governed mainly by differences between India and other developed countries in the characteristics of emissions resulting from the energy and industrial sectors and pollution control technologies (Kaskaoutis *et al.*, 2013; Li *et al.*, 2017).

HgP/PM₁₀ ratio can be helpful for identifying Hg sources if we have Hg content values for natural and anthropogenic emissions. The HgP/PM₁₀ ratio varied widely from 978 to 42,294 ng g⁻¹, with an average of 6555 ± 9732 ng g⁻¹ during the study period (Fig. 2(a)), suggesting that Hg is generally associated with PM₁₀. Furthermore, comparisons with HgP/PM₁₀ ratios from natural sources (e.g., dust and topsoil; 56–300 ng g⁻¹; Schleicher *et al.*, 2016) and background values from Himalayan topsoil (3.82–105.7 ng g⁻¹; Tripathi *et al.*, 2019b) reveal that the HgP/PM₁₀ ratio in Kanpur is higher than that of natural sources, suggesting an anthropogenic origin

of the Hg. A study reported that the Hg content in Indian coal varied from 1800 to 6100 ng g⁻¹, and the Hg concentration in the soil at different locations around a steel plant in Bhilai varied from 3590 to 16,580 ng g⁻¹ (Koshle *et al.*, 2008). Therefore, Hg content in Kanpur aerosols can be inferred to be mostly anthropogenic.

The HgP/PM₁₀ ratios obtained in Kanpur are significantly higher than those reported in other megacities such as Beijing and Kathmandu. The higher Hg concentration and HgP/PM₁₀ ratios in Kanpur might result from the greater prevalence of activities causing anthropogenic emissions (e.g., automobile driving, coal combustion, waste incineration, biomass burning, chemical production, and brickmaking); (Mukherjee *et al.*, 2009; Kanawade *et al.*, 2014; Kumari and Kulshrestha, 2018). A positive relationship between HgP concentration and HgP/PM₁₀ ratio ($r^2 = 0.77$, $p < 0.05$, $N = 58$) was observed for all samples in Fig. 3. However, when high-HgP-content samples (such as those from March) are excluded, the correlations between HgP and HgP/PM₁₀ ratio increases ($r^2 = 0.80$, $p < 0.05$, $N = 50$). The air quality in the IGP is influenced not only by the emissions from various anthropogenic sources (e.g., wood/crop residue burning and fossil fuel combustion) but also transport from the upwind regions in western India (Chinnam *et al.*, 2006; Ram *et al.*, 2010; Sharma *et al.*, 2016; Bikkina *et al.*, 2017). In addition to changes in meteorological changes, the boundary layer dynamics and emission sources have a strong impact on aerosol chemical composition and overall aerosol budget in the IGP (Ram *et al.*, 2010; Bikkina *et al.*, 2017). Although, Indian Meteorological Department defines four seasons as winter (December–February), summer/pre-monsoon season (April–June), monsoon season (late June–September), and post-monsoon season (October and November) (Table S2).

Fig. 2. Time series plots of (A) daily HgP/PM₁₀, (B) daily atmospheric HgP concentrations, (C) PM₁₀ mass concentrations and (D) monthly atmospheric HgP concentrations and HgP/PM₁₀ during the sampling period.

Further, daily average meteorological information and wind rose plots during the sampling period are shown in Figs. S1 and S2. In this regard, the month of March represents a transition period between winter and summer; wherein biomass-burning emissions exhibit a drastic change (Ram *et al.*, 2010; Bikkina *et al.*, 2017). We do find very different data of Hg during March (Fig. 2(a)); therefore, owing to changes in emission source patterns as well as to be consistent with the earlier study (Ram *et al.*, 2010), we discuss this data separately. In contrast, PM₁₀ mass concentrations were the lowest in March and highest during winter, suggesting that HgP concentration is dependent on not PM but Hg concentration (Fig. 2(c)). The weak association of HgP with aerosol particles during this period reflects a variety of sources and chemical compositions of anthropogenic particles (Zhang *et al.*, 2015). This result suggests that Hg content is not influenced by the concentration of particles during that period. The results of this study are supported by a recent study of the urban atmosphere of Beijing, where PM_{2.5} and PM₁₀ were not related to HgP (Tang *et al.*, 2019).

Seasonal Characteristics of Atmospheric HgP

The temporal variation in HgP composition was evaluated. Here, we examined monthly and daily changes in HgP concentration during the study period, except for during the monsoon season, when no samples were collected. The daily and monthly variations in HgP concentration, PM₁₀ mass, and HgP/PM₁₀ ratios are displayed in Figs. 2(a)–2(d). Average HgP concentrations and HgP/PM₁₀ ratios were the highest in March, followed by winter, post-monsoon and summer seasons (Table S1). The observations of high Hg levels in March are similar to the reports of chemical constituents, particularly carbonaceous species, in Kanpur. Ram *et al.* (2010) reported high levels of carbonaceous aerosols (OC and EC from fossil fuels) in March. Although OC/EC ratios were high at other times of year (winter: 8.7 ± 3.9 , summer: 7.3 ± 3.2 , post-monsoon season: 7.5 ± 1.4), OC/EC ratios varied minimally (between 2.4 and 3.5) in March, with an average value of 2.9 ± 0.5 (Ram *et al.*, 2010). Low OC/EC ratios in March indicate higher contributions from fossil fuel than in other parts of the year (Ram *et al.*, 2012). It should

Fig. 3. Regression between Hg content ($\text{HgP}/\text{PM}_{10}$) and particulate Hg (HgP).

be noted that apart from biomass burning and vehicular emissions, there are coal-fired power plants and cement factories are located near the sampling site, which is characteristically very different. Generally, OC/EC ratios and ^{14}C are better deciphering biomass burning vs fossil fuels (Ram *et al.*, 2010; Bikkina *et al.*, 2017), provided they do not have much secondary organic aerosols. In addition, methods using OC/EC ratios and ^{14}C measurements fail to distinguish different sources contributing to biomass burning vs. fossil fuel category. Moreover, K^+ concentrations in March and summer were approximately a quarter of those during the post-monsoon season and winter, indicating lower biomass-burning emissions in March and summer. These observations are corroborated by a study that performed robust ^{14}C measurement for source apportionment (Bikkina *et al.*, 2017): the biomass-burning fraction decreased to 70%–75% in March–September, whereas it remained at 80%–85% in other months. Thus, high EC and Hg concentrations in March are likely attributable to an increase in fossil fuel combustion and lower biomass-burning fraction. There is a lack of more tracers to characterize and quantify these sources. Recently, Sen *et al.* (2016) have used a combination of trace metals and Pb isotopic composition of aerosols and suggested that industrial emissions were a major source of anthropogenic Pb at Kanpur. In addition, total power generation capacity (GW), annual coal consumption (in million tons) in different states of IGP from west to east of the sampling site is presented in Table S3. Kanpur is one of the cities of Uttar Pradesh, where considerable coal consumptions and power generation emissions occur (Guttikunda and Jawahar, 2018). Therefore, fossil fuel (e.g., coal-fired power plants, cement factories and vehicle emissions) and biomass burning (cow dung, leaves, wood fuels) all contribute to carbonaceous species and other pollutants, including Hg in the IGP.

Therefore, more focused researches on characterization of emission sources of all forms of Hg (i.e. organic and inorganic forms) are needed for source apportionment and better mitigation strategy to prevent health risks associated with toxic Hg in the region.

Recent studies have also reported higher $\text{NH}_4^+/\text{SO}_4^{2-}$ and $\text{K}^+/\text{Ca}^{2+}$ ratios in winter and post-monsoon samples than in those from spring and summer, indicating the strong influence of crop residue/wood-burning emissions (Singh *et al.*, 2005; Bikkina *et al.*, 2017). These emissions might be the result of long-range transport to Kanpur by upwind from source regions in northern India. However, increasing insolation during the summer results in the dispersal of ground-level emissions to considerable heights (typically approximately 1.5 km), which might explain the low HgP concentrations over Kanpur during the summer. In addition, in the Asian monsoon region, more rain events, which can scavenge aerosol particles and thus reduce aerosol concentrations, occur in summer than in other seasons (Tripathi *et al.*, 2017).

The HYSPLIT model was used for air mass back trajectory analyses of 5 days in each season. In addition, Moderate Resolution Imaging Spectroradiometer (MODIS)-derived fire spots were identified in each season and are illustrated in Fig. S3. MODIS Collection 6 Terra and Aqua fire products (Giglio *et al.*, 2016) and an 80% confidence level were used to retrieve MODIS fire. The cluster analysis showed that the imminent air mass at the receptor site Kanpur was mainly of continental type, i.e., from Punjab, Haryana, and Uttar Pradesh of IGP and surrounding areas. Similar trajectories were observed over other IGP sites (e.g., Delhi and Varanasi) in the previous studies (Sharma *et al.*, 2015, 2016, 2017). Saud *et al.* (2012) reported the higher EC emission from Uttar Pradesh (12.73%) emitted from different fuel woods, crop residues and dung cakes than in

other areas of IGP. The trajectories also showed that the air masses are mostly local during winter and post-monsoon, indicating the impact of biomass burning. Furthermore, the results revealed similar patterns in the winter and post-monsoon seasons, with air masses arriving mainly from the western and inland regions and increasing HgP concentrations by transporting biomass from these regions. IGP outflow observations revealed that a major contributor to HgP over Kanpur was large-scale open biomass burning in Punjab, Haryana, and Uttar Pradesh in northern India during winter, fall, and summer (March–May 2007). However, during March, the simultaneous decrease in the biomass-burning fraction (Fig. S3) with the increase in fossil fuel combustion was the most likely cause of the high Hg concentration.

Influences of Chemical Constituents on Atmospheric HgP

Hg, inorganic ions, and carbonaceous species have common sources in ambient air and can influence HgP concentrations (Xu *et al.*, 2013). Lindberg *et al.* (2007) noted that the biogeochemical cycling of Hg is identical to that of carbon (C) and nitrogen (N). Particle components are complex, and ions (e.g., NO_3^-) must compete to bond with Hg; this bonding can scavenge Hg adequately to form diverse mercuric species (Duan *et al.*, 2016). Therefore, correlations between HgP and other chemical constituents (e.g., OC, EC, Mg^{2+} , Ca^{2+} , and NO_3^-) were computed for four seasons because they have common sources that can influence their concentrations, as presented in Table 2. However, differentiating sources from receptors is challenging; chemical constituents affect ambient HgP in Kanpur because of the complex mixture of local and transported anthropogenic Hg. Contributions from mineral dust during the summer (April–June) due to long-distance transport from arid regions are prevalent in Kanpur (Chinnam *et al.*, 2006) and contribute to high PM_{10} mass (Kanawade *et al.*, 2014). Therefore, the close association of HgP with crustal ions (Ca^{2+} and Mg^{2+}) and PM_{10} mass during summer may be attributable to dust aerosols transported to the sampling site during that season.

A significant correlation between HgP and EC was observed in winter and spring; this correlation should be studied further because EC may enhance heterogeneous reactions (Duan *et al.*, 2016). However, HgP was also strongly correlated with crustal ions (Mg^{2+} and Ca^{2+}) during winter; this result is attributed to local crustal dust influencing Hg sources (Tripathy *et al.*, 2019a). The strong relationship of HgP with OC is attributable to biomass burning during the

dry period (April and May). A close correlation between HgP and NO_3^- was observed during the summer and post-monsoon seasons because NO_3^- is generated by traffic sources (Xu *et al.*, 2013; Tripathy *et al.*, 2017) and gasoline vehicles contribute substantially to atmospheric Hg (Xu *et al.*, 2013). The results suggest mixed sources of HgP in the central IGP (Kanpur). Different seasons had different primary emission sources, and further long-term spatial scales studies are needed to develop dataset and understanding of Hg cycling for the region. Our study provides new insight into seasonal variations in the sources, destinations, and influential factors of PM_{10} -based Hg in the region, but further analysis is warranted.

Estimation of Dry Deposition of Atmospheric HgP

Atmospheric HgP can enter terrestrial and aquatic ecosystems through wet (precipitation scavenging) or dry (gravity) deposition (Marsik *et al.*, 2007). The deposition of atmospheric Hg is essential to understanding the biogeochemical cycling of Hg. However, because of the challenge of direct measurements, quantifying the deposition flux of HgP is difficult (Zhang *et al.*, 2012; Huang *et al.*, 2016). Therefore, we estimated HgP dry deposition flux with deposition velocity and measured concentrations of HgP. The database of dry deposition velocity (V_d) values still lacks accurate measurement (Zhang *et al.*, 2009), mainly because of a poor understanding of the size distribution of aerosols (Feddersen *et al.*, 2012). The following equation was used to estimate deposition flux:

$$F_d = V_d \times C_a \quad (1)$$

where, F_d refers to dry deposition flux, V_d refers to dry deposition velocity and C_a is the measured mass concentration of HgP in ambient aerosols.

V_d depends on several factors, including meteorological parameters and aerosol characteristics (Feddersen *et al.*, 2012). However, no significant associations were observed between HgP concentration and meteorological parameters. Therefore, uncertainties are present in flux estimation might result from slight changes in atmospheric Hg concentration or the selected deposition velocity (Guo *et al.*, 2017). However, estimated flux can serve as a first-order approximation for determining HgP deposition in IGP regions lacking data.

A representative deposition velocity of 0.5 cm s^{-1} was used in this study, following previous references regarding

Table 2. Correlation of HgP with chemical components.

Components	HgP			
	Winter	March	Summer	Post-monsoon
PM_{10}	0.04	0.31	0.46*	0.53*
OC	−0.09	0.26	0.44*	0.48*
EC	0.50*	0.79*	−0.23	0.22
Mg^{2+}	0.65**	0.24	0.52**	0.02
Ca^{2+}	0.66**	0.24	0.55**	0.37
NO_3^-	−0.02	0.24	0.52*	0.65*

* Correlations with $p < 0.05$.

** Correlations with $p < 0.01$.

the urban atmosphere (Huang *et al.*, 2016; Guo *et al.*, 2017). The percentage of estimated deposition flux in different seasons in Kanpur is displayed in Fig. 4. The atmospheric HgP dry deposition rates were the highest in winter (43.9 %) followed by the March (31.2 %), post-monsoon (12.7%), and summer periods (11.9%), and this pattern is nearly identical to that of atmospheric HgP concentrations. Although the highest HgP concentration occurred in March, the deposition rates were high in the winter because of the larger number of samples during the winter than in March.

The estimated dry deposition flux in Kanpur was $105 \mu\text{g m}^{-2} \text{y}^{-1}$. The HgP dry deposition flux in Kanpur was slightly lower than that estimated in the Kathmandu valley ($134 \mu\text{g m}^{-2} \text{y}^{-1}$), and the wet deposition flux at Kathmandu was $35 \mu\text{g m}^{-2} \text{y}^{-1}$ (Tripathee *et al.*, 2019a), approximately one-fourth of the dry deposition flux. In addition, the HgP flux in Kanpur was three times higher than that estimated for Lhasa ($35 \mu\text{g m}^{-2} \text{y}^{-1}$), which was about four times the wet flux in Lhasa ($8 \mu\text{g m}^{-2} \text{y}^{-1}$; (Huang *et al.*, 2016). However, no data are available on Hg wet deposition flux in Kanpur. Therefore, we assumed that the dry deposition rate was higher than the Hg wet flux. These results highlight the importance of Hg dry deposition to biogeochemical Hg cycling in South Asia (Guo *et al.*, 2017). Consequently, HgP dry deposition may be an essential physical process for the atmospheric Hg loading in the IGP. Nevertheless, uncertainty in dry deposition flux estimates results from the aforementioned shortcomings. In addition, this study attempted the first estimations of the dry deposition flux of HgP, which is crucial to the ecology of the IGP. Still, more studies on the role of dry deposition in the biogeochemical cycling of Hg over critical regions in the IGP are necessary. For further detail on the IGP as a valuable region for studying Hg cycling and deposition, dry and wet deposition studies calculating the actual deposition flux of Hg in the region are merited.

Prospects and Implications for Hg Studies in the Region

Because of increasing coal combustion in Asia, anthropogenic emissions in Asian regions are expected to rise if not controlled (Streets *et al.*, 2009). The entire IGP

region has experienced severe air pollution in the past decades (Saikawa *et al.*, 2019). In addition, the data presented in this study is quite old (2007–2008) and in the recent years both emissions and sources (number of industries, power plants, brick kilns etc.) have significantly increased over the IGP region (Sawani *et al.*, 2019). This have given rise to prolonged haze over the region (Saikawa *et al.*, 2019), which calls for more studies on particulate pollution including Hg. Therefore, a quantitative analysis focusing on Hg emissions near their sources and measuring Hg in the environment with adequate spatial coverage is needed to understand the environmental consequences and risks associated with Hg and Hg compounds in the IGP. The present study measured HgP concentrations in ambient air only. Mercury speciation studies are necessary to characterize reactive gaseous mercury and Hg^0 to attain a quantitative understanding of Hg chemistry and its biogeochemical cycle (Tripathee *et al.*, 2019a, 2020). Although our data indicate a high association of Hg with PM_{10} , emission sources of Hg in Kanpur appeared mixed. Major anthropogenic sources of atmospheric Hg in India are industry, coal combustion in thermal power plants, and brick kilns (Mukherjee and Zevenhoven, 2006; Burger Chakraborty *et al.*, 2013; Kumari *et al.*, 2015). Nevertheless, atmospheric deposition is the most critical manner in which atmospheric Hg enters aquatic and terrestrial ecosystems (Tripathee *et al.*, 2019b), where it can be transferred to the food chain. High Hg levels in the food chain can damage ecosystems and increase human exposure to Hg in fish and other food products. Therefore, long-term studies measuring Hg and examining its sources and accumulation in ecosystems in the region through advanced techniques such as isotopic measurement are warranted. Moreover, coordinated regional Hg emission control and adaptation strategies are necessary to control Hg pollution in the IGP efficiently.

CONCLUSIONS

Concentrations of HgP in PM_{10} samples from Kanpur, an urban area in the IGP, were measured to study seasonal variation during 2007–2008. The average HgP mass

Fig. 4. Estimated HgP dry deposition flux (%) in different seasons.

concentration was $776 \pm 846 \text{ pg m}^{-3}$ during the sampling period. Although the HgP level obtained in Kanpur is similar to that in other South Asian cities, it is substantially higher than those in North American and European cities. Average HgP concentrations and HgP/PM₁₀ ratios were the highest in March compared to other months, whereas seasonal trends followed as winter > post-monsoon and summer. Nevertheless, the HgP/PM₁₀ ratios in this study were generally higher than to be expected from natural sources (crustal dust and soil values), suggesting an anthropogenic origin of most Hg content in Kanpur. The dry deposition flux was estimated to be approximately $105 \mu\text{g m}^{-2} \text{ y}^{-1}$, slightly lower than that estimated for other South Asian cities but nearly three times higher than that in Lhasa.

The present study is limited to Hg associated with particulate matter; therefore, future studies should be focused on characterizing all forms of Hg (organic and inorganic forms) for better mitigation strategy to prevent health risks associated with toxic Hg in the region. In addition, measurement of Hg isotopes for source characterization and understanding the mechanism of partitioning of Hg in gas/particulate would be essential for studying the biogeochemical cycling of Hg over the region.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <https://doi.org/10.4209/aaqr.2019.12.0630>

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