Impact of Biomass Burning Plumes on the Size-Segregated Aerosol Chemistry over an Urban Atmosphere at Indo-Gangetic Plain

Abhinandan Ghosh¹, Arindam Roy¹, Abhijit Chatterjee^{1,2*}, Sanat K Das¹, Sanjay K Ghosh^{2,3}, Sibaji Raha^{1,2,3}

¹ Environmental Sciences Section, Bose Institute, Kolkata 700054, India

² National Facility on Astroparticle Physics and Space Science, Bose Institute, Darjeeling 734101, India

³ Center for Astroparticle Physics and Space Science, Bose Institute, Kolkata 700091, India

ABSTRACT

A study was conducted on the impact of biomass burning plumes transported from the Eastern Ghats region on the chemical characteristics of size-segregated aerosols over Kolkata, a tropical megacity situated in the eastern part of the Indo-Gangetic Plain. A Micro Orifice Uniform Deposit Impactor (MOUDI) was used to collect size-segregated aerosols affected and unaffected by the influence of transported biomass burning plumes during the pre-monsoon season (March–May) in 2016. Satellite observations revealed the presence of a thick polluted aerosol layer over Kolkata during the fire episodes, whereas a clean marine aerosol layer was observed during the normal episodes. Transported plumes enhanced the fine-mode aerosols, whereas the coarse-mode aerosols remained unaffected. $PM_{1.0-0.1}$ and $PM_{0.1}$ were greatly affected by the transported plumes, with a threefold increase during the fire episodes. K^+ , NH_4^+ and SO_4^{2-} showed significant increases in the accumulation-mode ($PM_{1.0-0.1}$) under the influence of biomass burning plumes, whereas NO_3^- showed an increase in the coarse-mode aerosols. The transported plumes did not change the mass-size distribution patterns of K^+ , NH_4^+ and SO_4^{2-} (in which the unimodal fine-mode was dominant), but NO_3^- showed a change from a bimodal to a unimodal coarse-mode distribution under the influence of the plumes. It was observed that the biomass burning plumes interacted strongly with the accumulation- and superfine-mode ($PM_{0.1}$) sea-salt particles, leading to the depletion of chloride. The maximum chloride depletion (~80%) was observed in the size range of 0.56–0.32 μm . It was observed that non-sea- SO_4^{2-} was the key constituent responsible for the chloride loss from sea-salt particles. The biomass burning plume was found to have the potential to shift the dominance from coarse- to fine-mode particles in the mass-size distribution pattern of the total aerosols.

Keywords: Shifting cultivation; Size-segregated aerosols; Biomass burning; Eastern Ghats.

INTRODUCTION

Natural forest fire and human induced burning of agricultural wastes and woods are the major contributors of biomass burning aerosols. Depending upon the vegetation types, a wide variety of aerosols like black carbon, organic carbon and various inorganic ionic species like nitrate, sulfate, potassium etc., emit from biomass burning (Andreae and Merlet, 2001). The consequences of biomass burning to the earth's climate is not well understood as their direct radiative effect or cloud modification properties depend on the types of fuel, combustion phase, atmospheric aging and several other factors (Koppmann *et al.*, 2005; Andreae and

Rosenfeld *et al.*, 2008; Lack *et al.*, 2012; Bond *et al.*, 2013; Lack *et al.*, 2013; Liu *et al.*, 2013). Van der Werf *et al.* (2006) showed that biomass burning accounts for 2.5 Pg C year⁻¹ through a long-term (1997–2004) global scale study and the amount is almost 50% of CO₂ emission from fossil fuel combustion (Bowman *et al.*, 2009). Recently a 20 year transient simulation study shows that the biomass burning may cause a net global warming of ~0.4 K (Jacobson, 2014). In addition to the climatic impact, emission of carcinogenic polycyclic aromatic hydrocarbons like benzo(a)pyrene and high concentration of alveoli penetrating fine particulates from biomass burning can cause severe pulmonary disorders like asthma, obstructive pulmonary disease, bronchitis etc. (Shusterman *et al.*, 1993; Asgharian *et al.*, 2001; Hedberg *et al.*, 2002).

Most of the aerosols emitted from biomass burning remain confined into the fine fraction and hence remain suspended in the atmosphere for several days (Freitas *et al.*, 2005). Intercontinental transport of biomass burning

* Corresponding author.

Tel.: +91-33-2569-3282; Fax: +91-33-2355-3886

E-mail address: abhijit.boseinst@gmail.com

plumes from Africa was found to adversely affect the air quality of clean remote regions of New Zealand (Edwards *et al.*, 2006). Tsai *et al.* (2012) observed high increase in fine-mode aerosols over a coastal station in Taiwan due to the transport of biomass burning plume from south-east Asia. Very high loading of sulfate aerosols was observed over a hill station, Sonia, in Vietnam because of the advected biomass burning plume from southern China and Indo-China peninsula (Lee *et al.*, 2016). A recent study conducted by Li *et al.* (2017) observed the uplift of the biomass burning aerosol plume to the altitude of around 2 km over Indo-China peninsula and transported towards the western Pacific at an altitude of around 4 km. Various laboratory and field measurements have shown the potential of biomass burning plume to influence the concentration, size distribution pattern and other atmospheric heterogeneous reactions of aerosols during their transport (Reid *et al.*, 2005b; Capes *et al.*, 2008; Hennigan *et al.*, 2011, 2012). Wildfires in the southern Siberia and crop residue burning in Kazakhstan and south Russia played a major role in the formation of haze over the Arctic (Warneke *et al.*, 2009). Real *et al.* (2010) showed that the enhancement of ozone in the mid troposphere over the Atlantic was due to the transport of biomass burning plume from central Africa. Popovicheva *et al.* (2016b) showed that how the hydration properties and the inorganic components increased the hygroscopicity of biomass burning smoke over Siberia. The intensity of the biomass burning plume not only changes the aerosol composition but also changes their microstructure as observed in north-west Vietnam (Popovicheva *et al.*, 2016a). Souza *et al.* (2014) observed that how the long-range transport of sugar cane burning plume increased fine-mode aerosol and the water soluble constituents over an urban atmosphere in Brazil. A recent study showed that wildfire and agricultural burning significantly degraded the air quality over Mexico City (Tzompa-Sosa *et al.*, 2016).

Slash and burn cultivation or shifting cultivation is an old practice of crop cultivation amongst the large number of tribal communities in tropics and act as a backbone of their sustenance (Rasul *et al.*, 2003). The global mean CO₂ emission from shifting cultivation is estimated to be ~741 Tg year⁻¹ and Asia alone contributes 30% of the global average (Silva *et al.*, 2011). This practice of shifting cultivation is very common over the Eastern Ghats region, the discontinuous mountainous region along India's eastern coast covering different states like Odisha, Andhra Pradesh, Tamilnadu and some parts of Karnataka. A recent study on the emissions of air pollutants from all the states in India reported that 5.29 Mt yr⁻¹ crop residue was burnt in Andhra Pradesh, 1.31 Mt yr⁻¹ in Orissa, 3.37 Mt yr⁻¹ in Tamil Nadu and 5.93 Mt yr⁻¹ in Karnataka in the year of 2008–2009 (Jain *et al.*, 2014). Shifting cultivation over Eastern Ghats starts with the clearance of mixed deciduous forest in winter (January–February) followed by burning of dry biomass in summer (March–April) and cropping in monsoon (Singh *et al.*, 2016). Previous studies over this region indicated dense active fire locations, elevated concentration of trace gasses, high aerosol optical depth and presence of smoke layer in

the atmosphere during the burning period (Gupta *et al.*, 2001; Prasad *et al.*, 2008; Vadrevu *et al.*, 2015). Venkataraman *et al.* (2006) estimated crop-waste burning as 116 (58–289) Tg yr⁻¹ in India. They also estimated the forest and shrubland biomass burning as 32 (16–61) Tg yr⁻¹ with the maximum burning occurring in central India followed by eastern and north-eastern part of India.

Kolkata, an overcrowded megacity on lower Indo-Gangetic Plain (IGP), experienced severe air pollution problem since recent past. The lack of open space for dispersion of the pollutants makes the condition even worse for the city (Gurjar *et al.*, 2016). With the increasing vehicular density and industrial activities, air pollution has become an emerging challenge and serious concern for the city (Gurjar *et al.*, 2016). Previous studies on source apportionment of PM₁₀ and PM_{2.5} aerosols over this city shows dominance of aerosol from fossil fuel (especially vehicular) emission, biomass and coal burning, solid waste dumping and transported carbonaceous and dust (Karar *et al.*, 2007; Chatterjee *et al.*, 2012). Size segregated aerosol sampling in several sites over Kolkata reveals the dominance of finer-mode aerosol and threefold higher loading of aerosol than the National Ambient Air Quality Standards or NAAQS (Das *et al.*, 2005). The lower IGP region including Kolkata city experiences huge aerosol loading (with high aerosol optical depth in the range of 0.6–0.7) during pre-monsoon months because of local and transported aerosols (Tiwari and Singh, 2013).

The present study is an attempt to address the following questions. 1) How the size-segregated concentrations and the mass-size distributions of aerosols and the ionic components change under the influence of the biomass burning plumes transported from Eastern Ghats and adjacent regions? 2) Whether and to what extent marine and polluted aerosols interact reaching over Kolkata during pre-monsoon (March–May) in 2016? This would help us to better understand the effect of the advection of the pollution plumes on the air quality of Kolkata, which is already a major and serious concern at the current scenario.

STUDY AREA

Kolkata (22.33°N and 88.20°E) is the financial capital of eastern India and one of the major cities of south-east Asia. Situated in the lower Indo-Gangetic Plain, the city consists of 4.5 million of the population (2011 census report) and stands second after Mumbai in the country. The city is neighboured by the Hooghly River to its west and north-west. The largest coastal wetland of the world and listed among the natural wonder Sundarban is just 90 km away from the city. The interior area of the city is leveled with the average elevation of 6 m above the mean sea level. Figs. 1(A) and 1(B) show the sampling site and the adjacent regions. The sampling site can be classified as a residential cum commercial area. The site is surrounded by the major roads by all four directions with the average distance of 100 m with a high density of all types of vehicles (heavy, medium and light). The Eastern Ghats region and the biomass burning plumes from a part of this region are shown in Figs. 1(C) and 1(D).

Fig. 1. (A) Position of the study area along with the Eastern Ghats region in India, (B) Study site and the adjacent regions, (C) Eastern Ghats regions, and (D) Representative satellite image (on 13 April, 2016) showing burning plumes along with the fire spots over a part of the Eastern Ghats region retrieved from Worldview.

METHOD

Collection of Samples

An eleven stage micro-orifice uniform deposit impactor (MOUDI™ II, Model 120, MSP Corporation, USA) was used for the collection of size-segregated aerosols over Kolkata. The impactor consists of a large number of micro-orifice nozzles to reduce the jet velocity and pressure drop, minimize particle bounce and enhance the collection efficiency. The impactor consists of rotating stages below the impaction plate for the uniform deposition of particles. Detail description of the instrument can be found elsewhere (Marple *et al.*, 2004, 2014). Samples were collected using pure thin aluminium foil (47 mm in diameter, MSP Corporation) as

impaction substrates in all the stages. The nominal cut-off sizes of the stages are 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18, 0.10 and 0.056 μm with the inlet cut-off diameter of 18 μm . The impactor was placed on the rooftop of the campus of Bose Institute, ~15 m above the ground and 2 m above the rooftop level. The impactor was run continuously for 72 hours with the flow rate of 30 L min^{-1} during each sampling event. Samples were kept in a refrigerator at 4°C for prior to gravimetric and chemical analysis.

Gravimetric and Chemical Analysis

The mass concentration of aerosol was measured by the gravimetric process. The impaction substrates were put in desiccators (Secador, Model code 401090) for 24 hrs before

and after the sampling to remove the absorbed water and weighed in a controlled environmental chamber using an analytical balance (Sartorius, Model ME 235 P). The mass concentrations of aerosols for each of the size range (impaction substrate) was calculated by dividing the mass (μg) of substrates (difference of the initial and the final weight of the impaction substrate) by the total volume of air (m^3) drawn.

Ion chromatography was used for the analysis of water soluble ionic species of aerosols. The impaction substrates were soaked in 20 mL Milli-Q water (18.2 M Ω resistivity) for 30 min and ultrasonicated for 30 minutes. The solutions were then kept in pre-cleaned polypropylene bottles. Water-soluble ionic species (Na^+ , NH_4^+ , K^+ , Cl^- , NO_3^- and SO_4^{2-}) were determined by using ion chromatograph (861 Advanced Compact IC, Metrohm, Switzerland). Detail of eluent and column specifications used for the analysis of anionic and cationic species are given elsewhere (Roy *et al.*, 2016). Field blanks were also analyzed following the same process. It was found that the concentration of the water soluble species in the field blank was negligible and below detection limit. The limit of detection for the water soluble ionic species were 0.05, 0.06, 0.08, 0.06, 0.08 and 0.07 ppm for Na^+ , NH_4^+ , K^+ , Cl^- , NO_3^- and SO_4^{2-} respectively. The analytical precision was also calculated from repeated measurements running standard samples of ionic species. The precision was found to be 3% for SO_4^{2-} and 2% for Cl^- , Na^+ , NO_3^- , K^+ and NH_4^+ .

Meteorological Parameters

Meteorological parameters were recorded continuously during the sampling period using automated weather station (AWS) installed and collocated with MOUDI. The weather station was equipped with the temperature and relative humidity sensors (Rotronic, Switzerland), tipping bucket rain gauge (R.M. Young, U.S.A), cup anemometer (Vaisala, Finland), pyranometer (Kipp & Zonen, Netherlands) and barometer (R.M. Young, U.S.A). The data was recorded continuously during the sampling period with the interval of 1 min.

Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) Observation

Since June 2006, the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) aboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) (Winker *et al.*, 2009) satellite provides the scientific society continuous and vertically resolved observations of the global atmospheric aerosol distribution. Polarized light at 532 and 1064 nm emits from an elastic back scatter LIDAR in CALIOP. Backscattered light at 532 nm is split into a parallel and a cross-polarized signal with respect to the plane of polarization of the emitted laser light to allow the depolarization profile and total backscattering signal is detected at 1064 nm. The instrument architecture and performance is stated by Hunt *et al.* (2009) while the abridgement of the data retrieval algorithms is stated by Winker *et al.* (2009). The analysis of measurements with elastic-backscatter LIDAR requires a prior knowledge of

the aerosol-type-specific LIDAR ratio (Klett, 1981; Fernald, 1984; Ansmann and Müller, 2005; Müller *et al.*, 2007). The CALIPSO aerosol retrieval gives this information distinguishing one out of six different aerosol types. The aerosol types used in the CALIPSO aerosol model were identified from cluster analysis of AERONET measurements and include smoke (biomass burning), polluted dust (mixtures of dust and smoke), dust, polluted continental, clean continental and clean marine (Omar *et al.*, 2005, 2009).

Retrieval of MODIS Active Fire Count and HYSPLIT Model

Detection of active fire has been performed using a contextual algorithm (Giglio *et al.*, 2003) that exploits the strong emission of mid-infrared radiation from fires. Brightness temperature of 4 μm and 11 μm channel is the key input in this algorithm. The fire count data used in this study is a special subset of MODIS collection 6 data (combined product from Terra and Aqua, MCD14ML) taken from Fire Information for Resource Management System (FIRMS). The improved algorithm of latest MODIS collection 6 fire products has a new hot-spot type attribute with high precision in small fire detection and reduction in a false alarm. Only high confidence ($> 80\%$) fire pixels are taken from the dataset.

5-day air-mass back trajectories, ending at 500 m amsl over Kolkata, have been calculated for all the days on which aerosol samples were collected using Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT4) model developed by National Oceanic and Atmospheric Administration (NOAA), USA (Draxler *et al.*, 2016). Trajectories are classified according to their height and plotted using open source GIS software Meteoinfo (Wang, Y.Q., 2014). Four trajectories were run per day at 00:00, 06:00, 12:00 and 18:00 UTC.

Classification of Aerosols

We have classified the total aerosol (PM_{10}) based on the size segregation. Aerosols with the aerodynamic diameter from 18 to 10 μm (PM_{18-10}) have been classified as non-respirable particulate matter or NRPM as they are not respirable in nature. Aerosols with the aerodynamic diameter below 10 μm (PM_{10}) and below 1.8 μm ($\text{PM}_{1.8}$) have been classified as respirable particulate matter or RPM and fine particulate matter or FPM; from 1.8 to 10 μm ($\text{PM}_{10-1.8}$) as respirable coarse particulate matter or RCPM; from 1.8 to 1.0 μm ($\text{PM}_{1.8-1.0}$) as superfine particulate matter or SFPM; from 1.0 to 0.1 μm ($\text{PM}_{1.0-0.1}$) as accumulation particulate matter or APM and less than 0.1 μm ($\text{PM}_{0.1}$) as ultrafine particulate matter or UFP.

Sampling Schedule

A total of ten samples were collected during the entire study period, 15 March–28 May 2016. Samples were collected on 15–18 March, 24–27 March, 2–5 April, 12–15 April, 19–22 April, 26–29 April, 4–7 May, 11–14 May, 18–21 May and 25–28 May in 2016. All the sampling events were started at 10:00 hrs (local time) and ended at 10:00 hrs (local time) after 72 hours.

RESULTS AND DISCUSSION

Classification of Sampling Episodes

The entire sampling period was divided into two episodes; normal and fire episodes. The fire episodes correspond to the period of 15 March–29 April 2016 and the period 4–28 May 2016 corresponds to the normal episodes. Thus, a total of six (06) samples were collected during the fire episodes and four (04) samples were collected during the normal episodes. The classification of the episodes was made mainly based on the number of fire pixels (MODIS) from the biomass burning over Eastern Ghats region, backward trajectory analysis and the identification of polluted aerosol layer over Kolkata using CALIPSO.

Air Mass Back Trajectories and Fire Pixels

Air mass back trajectories (using the HYSPLIT model) ending at the height of 500 m over the sampling site and the fire pixels (from MODIS) over the Eastern Ghats and the other regions have been plotted and shown in Fig. 2 for the sampling events in normal and the fire episodes. The number of fire pixels per day identified over the Eastern Ghats region was ~42 during the six sampling events in the fire episodes which was much higher than that during the four sampling events in the normal episodes (10).

During both the episodes, most of the trajectories were originated from the Bay of Bengal and passed over the Eastern Ghats regions and along the eastern coast before arriving at the sampling site. However, during the fire episodes, the number of trajectories passing through the Eastern Ghats regions was comparatively higher than the normal episodes. Some trajectories were also found to be originated from the arid and semi-arid regions of western India and passing through the burning regions in the central part of India, too, during the fire episodes. Another important difference between the two episodes is that the number of lower altitude or near-surface trajectories (< 500 m AGL) was found to be higher during the fire episodes whereas the number of higher altitude trajectories (500–2000 m AGL) was higher during the normal episodes. Thus we can say that the air masses could well collect the biomass burning pollutants from Eastern Ghats and the adjacent regions before arriving at the sampling site during the fire episodes. This is true that not all the trajectories passed exactly over the fire regions (as can be seen from Fig. 2) and they passed over the Bay of Bengal but adjacent to the fire regions. Thus the greater impact of biomass burning is well expected during the fire episodes with the higher number of fire pixels and the higher number of near-surface trajectories. However, as the air masses were originated from the Bay of Bengal, it is well expected that the air masses were laden with sea-salt aerosols in addition to the biomass burning/anthropogenic aerosols during both the episodes.

Observation of Aerosol Layer over Kolkata by CALIPSO

The Lidar ratios corresponds to the polluted dust and smoke aerosols are 65 and 70 respectively for 532 nm channel and 30 and 40 for 1064 nm channel (Omar *et al.*; 2009). The polluted dust in CALIPSO aerosol classification scheme

refers to the biomass burning smoke mixed with the dust particles (Omar *et al.*, 2009). In this study, we have identified thick layer of smoke and polluted dust aerosols over Kolkata up to 5 km from the ground during three days (27 March, 12 April and 28 April 2016) in the fire episodes (Figs. 3(A)–3(C)). Biomass burning/smoke plumes transported from Eastern Ghats regions could get well mixed with the dust aerosols transported from western part of India after arriving at the sampling site. Vadrevu *et al.* (2015) observed high altitude smoke and polluted dust layer (1–5 km) using CALIPSO over biomass burning regions of Indonesia, Thailand and India. Cloud free observations of CALIPSO were not available over Kolkata during the normal episodes in 2016. Hence we have made the CALIPSO observation during the same period (May) in 2015 and shown in Fig. 3(D). No such polluted smoke or dust aerosol layer was observed during the normal episodes rather the clean marine aerosol layer was identified over Kolkata suggesting the advection of sea-salt particles from the Bay of Bengal driven by the marine air masses. Thus the presence of the polluted aerosol layer and the clean marine aerosol layer over Kolkata made the distinct difference between the two episodes.

Synoptic Meteorology

The statistical analysis of the meteorological parameters during the normal and the fire episodes have been performed and shown in Table 1. It was observed that the mean ambient temperature, wind speed, solar radiation and mixed layer depth during the fire episodes were higher ($p < 0.05$) than the normal episodes. Usually, higher ambient temperature and solar radiation in the month of May (late summer/pre-monsoon) compared to the month of March and April (early summer/pre-monsoon) is expected over a tropical station in India like Kolkata. But the reverse was found to be true in 2016, as observed in the present study. Two of the sampling events during normal episodes were cloudy (daytime) which could result to the lowering of mean solar radiative flux compared to the fire episodes when all the sampling events were cloud free and sunny (daytime). During April, the south and south-western part of the state West Bengal including Kolkata experienced strong heat waves which could enhance the mean ambient temperature during the fire episodes. The episodes of heat wave can be observed from the Meteorological and Oceanographic Satellite Data Archival Centre (MOSDAC) of Indian Space Research Organization (ISRO) (https://www.mosdac.gov.in/data/heatwave_archival_2016.jsp). However the relative humidity during the normal episodes was higher ($p < 0.05$) which could be due to the advection of more moisture from the Bay of Bengal driven by marine air masses. No rainfall was recorded during both the episodes.

Concentrations of Size Segregated Aerosols and Chemical Components

The concentrations of size segregated aerosols and the chemical components are shown in Table 2. PM_{10} , PM_{18} and $PM_{1.8}$ showed significantly higher loading during the fire episodes relative to the normal episodes. $PM_{1.8}$ was found to contribute ~70% to the total aerosol loading

during the fire episodes indicating the dominance of fine-mode aerosols whereas the contribution was ~50% during the normal episodes indicating almost equal dominances of

fine and coarse-mode aerosols. The concentration of PM_{18-10} during the normal episodes was higher than the fire episodes indicating that the NRPM over Kolkata was of local origin

Fig. 2. Active fire pixels retrieved from MODIS and air mass back trajectories from HYSPLIT model during (a) fire and (b) normal episodes.

Fig. 3. Aerosol layers over Kolkata (black circles) and the satellite paths (inset) retrieved from CALIPSO during (A)–(C) fire and (D) normal episodes.

Table 1. Meteorological Parameters during the normal and fire episodes.

Episode	Temperature (°C)			Relative humidity%			Wind speed (m s ⁻¹)			Solar radiation (W m ⁻²)			Mixed layer depth (m)		
	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD
Normal	25.2	37.1	30.1 ± 0.9	68.9	98.2	85.8 ± 10.3	0.3	3.1	2.1 ± 0.9	29.4	732.2	347.5 ± 51.2	640	973	807 ± 52
Fire	29.4	41.2	35.1 ± .07	48.2	95.6	66.9 ± 11.4	0.3	8.1	3.5 ± 1.9	50.1	756.3	394.1 ± 49.3	761	1281	1037 ± 120

Table 2. Mean concentrations of aerosol and ionic species during the normal and fire episodes.

Aerosol Mass	K ⁺			NH ₄ ⁺			SO ₄ ²⁻			NO ₃ ⁻			Na ⁺			Cl ⁻		
	Fire	Normal	Fire	Normal	Fire	Normal	Fire	Normal	Fire	Normal	Fire	Normal	Fire	Normal	Fire	Normal	Fire	Normal
PM ₁₀	100 ± 10.4	58.7 ± 9.2	0.8 ± 0.4	0.2 ± 0.1	2.8 ± 0.6	1.2 ± 0.5	11.6 ± 4.6	8.2 ± 4.1	2.6 ± 1.2	2.2 ± 1.1	2.1 ± 0.6	1.6 ± 0.5	2.4 ± 1.1	2.2 ± 0.9	2.4 ± 1.1	2.2 ± 0.9	2.4 ± 1.1	2.2 ± 0.9
PM _{10-1.8}	95.0 ± 9.7	52.9 ± 8.9	0.8 ± 0.4	0.2 ± 0.1	2.8 ± 0.6	1.2 ± 0.5	11.5 ± 4.5	8.1 ± 4.0	2.5 ± 1.1	2.0 ± 1.0	2.0 ± 0.6	1.4 ± 0.5	2.3 ± 1.0	2.0 ± 0.1	2.3 ± 1.0	2.0 ± 0.1	2.3 ± 1.0	2.0 ± 0.1
PM _{1.8}	70.6 ± 12.3	28.2 ± 7.4	0.8 ± 0.4	0.2 ± 0.1	2.8 ± 0.6	1.2 ± 0.5	10.3 ± 3.8	7.5 ± 3.7	1.0 ± 0.4	1.2 ± 0.8	0.9 ± 0.8	0.8 ± 0.3	0.8 ± 0.4	1.2 ± 0.9	0.8 ± 0.4	1.2 ± 0.9	0.8 ± 0.4	1.2 ± 0.9
PM _{1.8-1.0}	5.0 ± 0.9	5.8 ± 1.1	BDL	BDL	BDL	BDL	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1	0.2 ± 0.1	0.1 ± 0.1	0.2 ± 0.1	0.1 ± 0.1	0.2 ± 0.1	0.1 ± 0.1	0.2 ± 0.1	0.1 ± 0.1	0.2 ± 0.1
PM _{1.0-1.8}	24.4 ± 6.5	24.7 ± 6.2	BDL	BDL	BDL	BDL	1.2 ± 1.2	0.6 ± 0.2	1.5 ± 1.3	0.8 ± 0.5	1.1 ± 0.7	0.6 ± 0.4	1.5 ± 1.3	0.8 ± 0.4	1.5 ± 1.3	0.8 ± 0.4	1.5 ± 1.3	0.8 ± 0.4
PM _{1.8-1.0}	9.7 ± 0.4	6.2 ± 0.8	BDL	BDL	BDL	BDL	0.5 ± 0.4	0.3 ± 0.1	0.6 ± 0.1	0.5 ± 0.4	0.5 ± 0.2	0.4 ± 0.1	0.4 ± 0.1	0.4 ± 0.5	0.4 ± 0.1	0.4 ± 0.5	0.4 ± 0.1	0.4 ± 0.5
PM _{1.0-0.1}	52.1 ± 15.6	19.2 ± 7.8	0.8 ± 0.5	0.2 ± 0.1	2.7 ± 0.6	1.1 ± 0.4	9.1 ± 5.1	6.8 ± 4.8	0.4 ± 0.3	0.6 ± 0.6	0.4 ± 0.2	0.4 ± 0.2	0.3 ± 0.2	0.7 ± 0.6	0.3 ± 0.2	0.7 ± 0.6	0.3 ± 0.2	0.7 ± 0.6
PM _{<0.1}	8.8 ± 2.6	2.8 ± 0.8	BDL	BDL	0.1 ± 0.1	0.1 ± 0.1	0.7 ± 0.2	0.4 ± 0.2	BDL	0.1 ± 0.1	0.0 ± 0.0	0.0 ± 0.0	0.1 ± 0.1	0.2 ± 0.2	0.1 ± 0.1	0.2 ± 0.2	0.1 ± 0.1	0.2 ± 0.2

All units are in µg m⁻³; BDL: Below Detection Limit.

and not influenced by transported biomass burning plumes. This could also be transported sea-salt particles driven by the marine air masses originating from Bay of Bengal and remaining uninvolved with biomass burning pollutants. The same arguments could be made for $\text{PM}_{10-1.8}$ or RCPM which showed almost equal loadings during the normal and fire episodes. While the concentration of $\text{PM}_{1.8-1.0}$ or SFPM was 60–70% higher during the fire episodes, the concentrations of $\text{PM}_{1.0-0.1}$ and $\text{PM}_{0.1}$ were increased by threefold during the fire episodes suggesting that the APM and UFPM were greatly influenced by the transported biomass burning plumes from Eastern Ghats regions. Alonso-Blanco *et al.* (2014) observed that the number concentration of aerosols in the size range of 0.1–0.2 μm was enhanced by sevenfold because of the transported biomass burning plume over a rural site in Spain. PM_{10} measured in the present study was found to be much lower compared to the wintertime PM_{10} concentration of 330 $\mu\text{g m}^{-3}$ as observed by Das *et al.* (2006), summertime PM_{10} concentration of 110 $\mu\text{g m}^{-3}$ as observed by Karar *et al.* (2006) and annual average PM_{10} concentration of 140 $\mu\text{g m}^{-3}$ as observed by Karar *et al.* (2007) over Kolkata. However, Chatterjee *et al.* (2012) observed the $\text{PM}_{2.5}$ loading of 73 $\mu\text{g m}^{-3}$ in the atmosphere over Kolkata during March–April 2005 which is almost equal to the $\text{PM}_{1.8}$ concentration during March–April 2016 (fire episode) as measured in the present study.

The mean concentration of K^+ in $\text{PM}_{1.8}$ during the fire episodes ($0.8 \pm 0.4 \mu\text{g m}^{-3}$) was fourfold higher than the normal episodes ($0.2 \pm 0.1 \mu\text{g m}^{-3}$). Interestingly all the K^+ over Kolkata was found to be confined into $\text{PM}_{1.0-0.1}$ or accumulation-mode during both the episodes. Similarly, NH_4^+ shows ~100% contributions from $\text{PM}_{1.0-0.1}$ or APM irrespective of the normal and fire episodes. However, NH_4^+ during the fire episodes ($2.7 \pm 0.6 \mu\text{g m}^{-3}$) was significantly

higher than the normal episodes ($1.1 \pm 0.4 \mu\text{g m}^{-3}$). It was observed that 80–85% of SO_4^{2-} was confined into accumulation-mode aerosols or $\text{PM}_{1.0-0.1}$ during both the episodes with higher loading during the fire ($9.1 \pm 5.1 \mu\text{g m}^{-3}$) than the normal episodes ($6.8 \pm 4.8 \mu\text{g m}^{-3}$). NO_3^- shows comparable concentrations in $\text{PM}_{1.8}$ during both the episodes. It was observed that the fire episodes enhanced NO_3^- loading mostly in $\text{PM}_{10-1.8}$ or respirable coarse-mode. Overall, coarse-mode NO_3^- dominated (55–60%) during both the episodes. We observed that the fire episodes increased the coarse-mode Na^+ and Cl^- and decreased the fine-mode Cl^- more than the normal episodes. However, the fine-mode Na^+ remained unchanged during both the episodes. The concentrations of the total aerosol mass as well as its ionic components for each of the sampling event during the fire and the normal episodes are given in Table S1 (see supplementary).

Mass-Size Distribution of Aerosols

Fig. 4 shows the mass-size distribution of aerosols over Kolkata during the fire and normal episodes. Both the episodes show a bimodal distribution with two prominent peaks. The first peaks came up in the accumulation-mode (0.32–0.18 μm) where the maximum peak-height was observed for the fire episodes. The second peak came up in the respirable coarse-mode (5.6–3.2 μm) for the normal episodes whereas it was found to shift towards the smaller size ranges (3.2–1.8 μm) during the fire episodes. The accumulation-mode peak over Kolkata during the normal episodes could be due to the local anthropogenic activities and the transported dust particles from Indo-Gangetic Plain (IGP) as well as the arid and semi-arid regions of western India and beyond (Chatterjee *et al.*, 2012). On the other hand, the coarse-mode peak could be associated with the road dust, sea-salt aerosols, soil-derived particles etc. We

Fig. 4. Mass-size distribution of the total aerosols during normal and fire episodes.

discussed earlier that the coarse-mode aerosols were not influenced by the transported biomass burning plumes and mainly of local origin. Generally, the entire IGP region experiences intense solar heating during summer months leading to the elevated mixing layer depth and favourable dispersion conditions for aerosols (Deshmukh *et al.*, 2013). Under this condition, fine-mode aerosols could be dispersed or uplifted easily leading to the coarse-mode dominant environment (Carbone, *et al.*, 2010) as observed from the mass size distribution pattern during the normal episodes. Although the ambient temperature, wind speed, solar radiative flux and the mixing layer depth during the fire episodes were higher than the normal episodes (Table 1), the massive influx of biomass burning aerosol plumes from Eastern Ghats regions could additionally feed the fine-mode aerosols in spite of favourable dispersion conditions. The size distribution pattern during the fire episodes thus became fine-mode dominant. Hence we can say that the transported biomass burning plumes could have the potential to alter the mass-size distribution pattern of aerosols over Kolkata.

Mass-Size Distribution of Ionic Components

The mass-size distributions of water soluble ionic components over Kolkata are shown in Fig. 5. K^+ , SO_4^{2-} and NH_4^+ showed unimodal mass-size distributions during both the episodes with the peaks in accumulation-mode. For all of these three ionic species, maximum peak height was observed for the fire episodes. Several earlier studies (Echalar *et al.*, 1995; Duan *et al.*, 2004; Chatterjee *et al.*, 2010) have used fine-mode potassium as a biomass burning tracer. Li *et al.* (2003) reported high abundances of potassium sulfate and potassium nitrate in the aged smoke originated from biomass burning in a case study over South Africa. Li *et al.* (2010) observed the drastic increase in K^+ concentration over Shanghai due to agricultural burning during summer harvest-time. High increase in K^+ was also observed during biomass burning events over western Himalaya in India (Kumar and Attri, 2016) and an urban atmosphere in upper IGP regions (Rastogi *et al.*, 2015). However, the enhancement of accumulation-mode K^+ during the fire episodes in the present study strongly suggests that K^+ could be considered as the reliable biomass burning tracer species over the study region. In case of SO_4^{2-} , a peak shift was observed towards larger size range (0.56–0.32 μm) during the fire episodes. This could be due to the aging of sulphate aerosols during its transport from long distances. Under the conditions of stronger solar radiation and higher temperature (as observed in fire episodes), the gas-to-particle conversion of SO_2 to SO_4^{2-} aerosols could be the major pathway for the higher sulfate formation (Huang *et al.*, 2016). The high concentration of SO_2 , the precursor of SO_4^{2-} , during the fire episode is well expected (Popovicheva *et al.*, 2014). Gao *et al.* (2003) observed significant inclusion of SO_2 from savanna fire. Laboratory experiments have shown that the nucleation of sulfuric acid is enhanced in presence of organic acids (Zhang *et al.*, 2004). Maudlin *et al.* (2015) observed high emissions of organic acids from the wild fire in California. Though organic acids and SO_2 were not monitored in the present study, the transported biomass

burning plume could be enriched with these species which could promote the formation of sulphate aerosols under the favourable meteorological conditions. NH_3 usually originates from combustion, agricultural activity, livestock farming and biomass burning (Bouwman *et al.*, 1997). During the fire episodes, the enhanced supply of NH_3 and favourable meteorological conditions could accelerate the gas-to-particle conversion of NH_3 to NH_4^+ (Reid *et al.*, 1998; Gao *et al.*, 2003). During the normal episodes, the accumulation-mode NH_4^+ could be generated due to the local anthropogenic activities, domestic and industrial wastes etc. However, the unimodal accumulation-mode peaks of K^+ , SO_4^{2-} and NH_4^+ also indicate the probable formation of accumulation-mode K_2SO_4 and $(NH_4)_2SO_4$ over Kolkata.

NO_3^- shows bimodal distribution during normal episodes with a peak in accumulation-mode and relatively larger peak in respirable coarse-mode (3.2 μm). Under colder atmospheric conditions, nitrate usually reacts with ammonia to form NH_4NO_3 and gets accumulated in fine-mode aerosols. This reaction is reversible and the formation of ammonium nitrate is favored with the decrease in temperature (Tanner *et al.*, 1982). Thus the formation of ammonium nitrate during summer in a tropical city like Kolkata is unexpected. However, NO_3^- peak at accumulation-mode during normal episodes could be due to the local anthropogenic activities, e.g., fossil fuel emissions or transported from continental land masses. NO_3^- could also react with the soil derived or marine particles and accumulated in the coarse-mode aerosols (Wolff *et al.*, 1984). This results to the coarse-mode dominant distribution pattern of NO_3^- as observed in the present study during the normal episodes. Direct emissions from the controlled forest fire are responsible for the enhanced concentration of NO_3^- (Safai *et al.*, 1993; Posfai *et al.*, 2003; Wonaschutz *et al.*, 2011). Transported biomass burning plume during the fire episodes increased the peak height of NO_3^- at 3.2 μm . The increase in the peak height in the coarse-mode could be due to the mixing/reaction of nitrate aerosol or nitric acids or oxides of nitrogen etc. originating from biomass burning with the coarse-mode sea-salt aerosols (Wonaschutz *et al.*, 2011; Zauscher *et al.*, 2013). A case study in Brazil showed that most of the inorganic ions were confined in submicron mode but shifted towards the larger size due to ageing and mixing with other types of aerosol (Falkovich *et al.*, 2005). Similar kind of observation was also made over coastal California where Maudlin *et al.* (2015) reported enhanced concentration of nitrate in super-micron range during the fire activity. However, in a case study over Finland (Makkonen *et al.*, 2010), the result is contradicting from us where they found an increase in nitrate concentration only in the fine-mode during the fire episode. The significant increase in nitrate aerosols due to biomass burning was also reported by Diapouli *et al.* (2014) in Greece and Sillanpaa *et al.* (2005) in Finland. The mass-size distribution of NO_3^- shows similarity with that of Na^+ with unimodal large peaks at coarse-mode indicating interaction of NO_3^- with the coarse-mode sea-salt particles (NaCl) during the fire episodes when both NO_3^- and Na^+ concentrations were higher compared to the normal episodes.

Fig. 5. Mass-size distributions of aerosol ionic components during normal and fire episodes.

Na^+ and Cl^- showed unimodal mass-size distribution during both the episodes. Both the species showed peaks at superfine-mode during the normal episodes indicating their common marine source, the Bay of Bengal. During the fire episodes, the peak of Na^+ was found to shift towards the coarse-mode ($3.2\text{--}1.8\text{ }\mu\text{m}$) whereas that of Cl^- shifted further to larger size ranges ($5.6\text{--}3.2\text{ }\mu\text{m}$). The significant decrease in Cl^- concentrations in superfine and accumulation ranges during the fire episodes could have made the distribution different (peak shift) from that of Na^+ . We observed that the Cl^-/Na^+ mass ratios in coarse-mode aerosol during the normal (1.4) and fire episodes (1.3) were almost equal and less than the ratio in standard seawater (1.81). On the other hand, the ratio in the fine-mode aerosol during the normal episodes was 1.9, i.e., closed to the standard ratio. It decreased to 0.8, i.e., much less than the standard ratio during the fire episodes. This could be discussed in the light of Cl^- depletion from sea-salt aerosols in the section given below.

Chloride Depletion from Sea-Salt Particles

The Cl^- depletion from sea-salt aerosols and the non-sea contribution of SO_4^{2-} (non-sea- SO_4^{2-}) were calculated considering Na^+ as the reference element for the seawater, 1.81 and 0.252 are the mass ratios of Cl^- to Na^+ and SO_4^{2-} to Na^+ , respectively (Chatterjee *et al.*, 2010).

$$\% \text{ of } \text{Cl}^- \text{ Depletion} = [(1.81 \times [\text{Na}^+] - [\text{Cl}^-]) / (1.81 \times [\text{Na}^+])] \times 100 \quad (1)$$

$$\text{Non-sea-}\text{SO}_4^{2-} = [\text{SO}_4^{2-}] - 0.252 \times [\text{Na}^+] \quad (2)$$

where $[\text{Na}^+]$, $[\text{Cl}^-]$ and $[\text{SO}_4^{2-}]$ are the observed/measured concentrations of Na^+ , Cl^- and SO_4^{2-} respectively. Fig. 6 shows the size-segregated chloride depletion (%) from sea-salt particles (NaCl) along with the concentrations of NO_3^- and non-sea- SO_4^{2-} during the normal and fire episodes. It can be observed from Fig. 6(a) that the fine-mode Cl^- was depleted to a greater extent during the fire episodes with maximum depletion ($\sim 80\%$) at $0.56\text{--}0.32\text{ }\mu\text{m}$. We observed significant Cl^- depletion at $< 0.056\text{ }\mu\text{m}$ during the fire episodes while no depletion was observed during the normal episodes. The same occurred at $1.0\text{--}0.56\text{ }\mu\text{m}$ where the fire episodes depleted Cl^- by $\sim 50\%$ but no such depletion was observed during the normal episodes. The association of the non-sea- SO_4^{2-} with the various anthropogenic activities (coal, biomass and fossil fuel burning etc.) over a tropical urban station like Kolkata is well expected and could be considered as the background concentrations during the normal episodes. However, the concentrations of non-sea- SO_4^{2-} were enhanced significantly during the fire episodes (Fig. 6(b)) at all the accumulation and ultrafine stages where significant chloride depletion was observed. For example, non-sea- SO_4^{2-} concentration was increased from $2.2\text{ }\mu\text{g m}^{-3}$ during normal episode to $4.5\text{ }\mu\text{g m}^{-3}$ during fire episode at the size of $0.56\text{--}0.32\text{ }\mu\text{m}$, where maximum chloride depletion was observed. Non-sea- SO_4^{2-} was enhanced by threefold during fire episodes, 0.1 to $0.35\text{ }\mu\text{g m}^{-3}$ at the size of $< 0.056\text{ }\mu\text{m}$. On the other hand, NO_3^- concentrations at $0.56\text{--}0.32$ and $< 0.056\text{ }\mu\text{m}$ were higher during normal

episodes than the fire episodes (Fig. 6(c)). This indicates that non-sea- SO_4^{2-} (and not NO_3^-) could be one of the key factors responsible for the chloride depletion from accumulation and ultrafine sea-salt particles. However, the chloride depletion at $1.0\text{--}0.56\text{ }\mu\text{m}$ during the fire episodes (which was insignificant during the normal episodes) could be due to both the NO_3^- and non-sea- SO_4^{2-} as they exhibited higher concentrations during the fire episodes. All the fine-mode stages (size below $1.8\text{ }\mu\text{m}$) showed higher chloride depletion during the fire episodes except at $0.18\text{--}0.1\text{ }\mu\text{m}$ where non-sea- SO_4^{2-} during the normal episodes was higher than fire episodes. Overall, we can say that the high increase in non-sea- SO_4^{2-} due to the transported biomass burning plume resulted to the depletion of chloride from the superfine- and accumulation-mode sea-salt particles but did not interact well with the non-respirable and respirable coarse-mode sea-salt particles. However, chloride depletion from the coarse-mode sea-salt particles during the fire episodes could be due to NO_3^- . Biomass burning also produces significant amount of particulate Cl^- in the atmosphere (Lobert *et al.*, 1999; Li *et al.*, 2003; Jing *et al.*, 2017). However, the high chloride depletion by non-sea- SO_4^{2-} and NO_3^- from sea-salt particles as observed from this study probably dominated over the chloride emission/enrichment due to biomass burning.

We have compared our results with a recent study (Singh *et al.*, 2016) on size segregated aerosol chemistry over a semi-urban station, Patiala, at western IGP conducted during October 2012–September 2013. Autumn (October–November) was considered as the main season influenced by the paddy residue burning. The mass-size distribution of aerosol over Kolkata was found to be similar to that observed over Patiala (bimodal with fine ($< 1\text{ }\mu\text{m}$) and coarse ($3\text{--}7.2\text{ }\mu\text{m}$) mode peaks). K^+ , SO_4^{2-} and NH_4^+ showed similar distributions (unimodal with fine-mode dominant) and their concentrations were increased by several folds during the biomass burning episodes over both the stations. The distribution of NO_3^- (bimodal with fine and coarse-mode peaks) over Kolkata during the fire episodes was very much similar to that over Patiala during autumn. However, the concentration of PM_{10} during autumn over Patiala ($\sim 140\text{ }\mu\text{g m}^{-3}$) was much higher than that over Kolkata during the fire episodes ($\sim 61\text{ }\mu\text{g m}^{-3}$). Similarly, the concentrations of major biomass marker components K^+ and SO_4^{2-} showed higher concentrations over Patiala (~ 2.5 and $13\text{ }\mu\text{g m}^{-3}$ respectively) compared to Kolkata (0.8 and $9.8\text{ }\mu\text{g m}^{-3}$ respectively). The difference could arise due to the fact that Patiala itself is a source region of biomass/paddy residue burning emissions whereas the sampling site in Kolkata was influenced by the biomass burning plumes transported from the distant Eastern Ghats region. Overall we observed similarity in the pattern of the changes in aerosol chemistry under the influence of biomass burning between urban atmospheres at eastern and western IGP.

Implications of the Study

The present study has shown how the polluted aerosols transported from the Eastern Ghats regions resulted to the

Fig. 6. (a) Chloride depletion (%) at different aerosol size ranges during normal and the fire episodes, (b) Concentration of non-sea sulfate in different size ranges, and (c) Concentration of nitrate in different size ranges.

enhancement of aerosol loading in the atmosphere over a tropical megacity Kolkata at the eastern part of the Indo-Gangetic Plain. The city is one of the polluted cities in India where poor air quality and associated health disorders are the major concerns at the current scenario. The transported plume changed the mass-size distribution of aerosols from coarse-mode to fine-mode dominant. Thus the high increase

and the additional burden of submicron aerosols due to the transport of biomass burning plume could pose serious threat to human health over Kolkata. The city during pre-monsoon months remains mostly under the influence of marine air masses originating from the Bay of Bengal as observed in the present study (during normal episodes). The advection of the biomass burning plumes and the

interaction with the marine aerosols (as observed from high chloride depletion) change the aerosol chemistry. Because of this chloride loss, the physical and chemical nature of sea-salt particles (especially NaCl) change which could in turn affect the hygroscopic growth, activation efficiencies to cloud condensation nuclei and cloud microphysical properties. A close look at the air mass back trajectories (as analyzed in this study) show that before reaching Kolkata, the polluted biomass burning plume passes over Sundarban mangrove forest, the major ecosystem of biosphere in India. The intrusion of polluted aerosols into the clean marine environment could lead to a stressed chemical environment. This would certainly motivate the researchers to carry out the investigations of the impact of transported biomass burning plumes on the physical and chemical environment of this unique and ecologically important forest ecosystem too.

CONCLUSIONS

The present study was conducted in order to investigate the impact of biomass burning plumes transported from the Eastern Ghats region on the size-segregated aerosol chemistry over Kolkata, a tropical metropolis on the Indo-Gangetic Plain during the pre-monsoon season (March–May) of 2016. The major findings of the study are as follows:

- Trajectory analysis and satellite based observations have shown the transport of biomass burning plumes from the Eastern Ghats region and the formation of a thick polluted dust layer over Kolkata. On the other hand, during normal episodes, a layer of clean marine aerosols was observed, which may be driven by the marine air masses originating in the Bay of Bengal.
- It was observed that during the fire episodes, $PM_{1.8-1.0}$ was increased by 60–70%, whereas $PM_{1.0-0.1}$ and $PM_{0.1}$ increased two to threefold indicating that the accumulation and ultrafine aerosols over Kolkata were severely affected by the transported biomass burning plumes. However, the respirable and non-respirable coarse-mode aerosols ($PM_{10-1.8}$ and PM_{18-10}) remained unaffected by the transported plumes and thus could be either locally generated or transported sea-salt particles that do not interact with biomass burning aerosols.
- The transported biomass burning plume was found to have the potential to change the mass-size distribution pattern of aerosols over Kolkata. Under the influence of the transported plume, the pattern was changed from coarse-mode to fine-mode dominant fine-mode.
- The accumulation-modes of Accumulation-mode K^+ , NH_4^+ and SO_4^{2-} were greatly influenced, and their concentrations increased significantly due to the advection of biomass burning plumes. These three species showed a unimodal mass-size distribution with peaks in the accumulation-mode, and their significant enhancement during the fire episodes suggests that they can be considered reliable tracers of biomass burning activities in the source regions.
- The mass-size distribution patterns of all the ionic species except for NO_3^- remained the same under the influence of the transported biomass burning plume. During the normal episodes, NO_3^- showed a bimodal distribution

with peaks in the accumulation and coarse-modes. The biomass burning plume changed the pattern to a unimodal coarse-mode distribution dominated by the coarse-mode. Nitrate aerosols from biomass burning may interact with coarse-mode sea-salt particles, forming coarse-mode nitrate.

- The transported biomass burning plume interacted better with the accumulation and ultrafine-mode sea-salt particles than with the coarse-mode sea-salt particles, leading to the depletion of Cl^- . The maximum depletion (~80%) was observed in the range of 0.56–0.32 μm . Significant chloride depletion was observed in the ranges of 1.0–0.56 μm and < 0.056 μm during the fire episodes, whereas no such depletion was recorded during normal episodes. Non-sea- SO_4^{2-} was found to be the key constituent in the chloride depletion of accumulation and ultrafine sea-salt particles, whereas NO_3^- may have been responsible for chloride depletion from coarse-mode sea-salt particles.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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