



Anthropogenic fine aerosols dominate the wintertime regime over the northern Indian Ocean

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ABSTRACT

This study presents and evaluates the most comprehensive set to date of chemical, physical and optical properties of aerosols in the outflow from South Asia covering a full winter (Nov. 2014 – March 2015), here intercepted at the Indian Ocean receptor site of the Maldives Climate Observatory in Hanimaadhoo (MCOH). Cluster analysis of air-mass back trajectories for MCOH, combined with AOD and meteorological data, demonstrate that the wintertime northern Indian Ocean is strongly influenced by aerosols transported from source regions with three major wind regimes, originating from the Indo-Gangetic Plain (IGP), the Bay of Bengal (BoB) and the Arabian Sea (AS). As much as $97 \pm 3\%$ of elemental carbon (EC) in the PM_{10} was also found in the fine mode ($PM_{2.5}$). Other mainly anthropogenic constituents such as organic carbon (OC), non-sea-salt (nss) $-K^+$, $nss-SO_4^{2-}$ and NH_4^+ were also predominantly in the fine mode (70–95%), particularly in the air masses from IGP. The combination at this large-footprint receptor observatory of consistently low OC/EC ratio (2.0 ± 0.5), strong linear relationships between EC and OC as well as between $nss-K^+$ and both OC and EC, suggest a predominance of primary sources, with a large biomass burning contribution. The particle number-size distributions for the air masses from IGP and BoB exhibited clear bimodal shapes within the fine fraction with distinct accumulation ($0.1 \mu m < d < 1 \mu m$) and Aitken ($0.025 \mu m < d < 0.10 \mu m$) modes. This study also supports that IGP is a key source region for the wider South Asia and nearby oceans, as defined by the criteria that anthropogenic AODs exceed 0.3 and absorption AOD > 0.03 . Taken together, the aerosol pollution over the northern Indian Ocean in the dry season is dominated by a well-mixed long-range transported regime of the fine-mode aerosols largely from primary combustion origin.

Keywords: Aerosol optical depth, air pollution, black carbon, chemical composition, South Asia

1. Introduction

Aerosol loadings in the South Asian atmosphere are much higher than the global average, largely due to anthropogenic activities. Black Carbon aerosols (BC, also known as soot) originate from incomplete combustion of biomass and fossil fuels. BC is an important component of Atmospheric Brown Clouds (ABC), which are visible from space, spreading out from India, and other key source regions. Given the relatively short atmospheric lifetime of BC (~ 1 –2 weeks), the climate effects of BC are more regional compared to those of long-lived gases such as CO_2 (atmospheric lifetime 100 – 1000s of years). It is therefore important to investigate BC in the regions where the climate

effects are the highest. In addition to having a strong regional impact on the South Asian climate (e.g., Satheesh and Ramathan, 2000; Lawrence and Lelieveld, 2010; Bond et al., 2013; Boucher, et al., 2013), BC particles also have adverse effects on human health (Janssen et al., 2012; WHO, 2016), agriculture and precipitation patterns (e.g., Ramanathan et al., 2001), and the melting of snow and ice in the Himalaya-Tibetan plateau (e.g., Menon et al., 2010; Kaspari et al., 2013; Li et al., 2016).

While the climate implications of BC and other anthropogenic aerosols in South Asia are considered to be of global importance, there are still many large uncertainties. BC is co-emitted with a variety of other aerosols and aerosol precursor gases. Thus, the knowledge about particle number and size distribu-

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tion, chemical composition and mixing ratios of different species is essential for developing the basic understanding of atmospheric processes that affect the composition and hence the effects of South Asian aerosols.

There is only one earlier report with a comprehensive coverage of core aerosol parameters through a winter campaign from the northern Indian Ocean (Ramanathan et al., 2007a). In addition to the need to compare with that pioneering 2004–2005 study, also held at MCOH, and to add a second set of observations, the current study is performed ten years later in this region with presumably rapidly changing emission scenarios. The current study also adds several additional dimensions compared to the 2004–2005 investigations. For instance, our new study provides information on the relative importance of fine ($PM_{2.5}$) versus coarse ($PM_{10-2.5}$) partitioning of organic carbon (OC), elemental carbon (EC), mass and inorganic aerosol constituents. Additionally, absorption aerosol optical depth (AAOD) and carbon monoxide (CO) serve as further markers of incomplete combustion. As espoused below, this study thus adds an important decennial revisit of this important aerosol regime expands with several new parameters and establishes both some similarities and some differences between the winter-long campaigns performed ten years apart.

The main objectives of the present study were to provide a comprehensive perspective on the physical, chemical and optical properties of aerosols over the northern Indian Ocean during the six-month-long dry season with highest regional aerosol optical depth (AOD), with specific focus on the homogeneity/regional mixing state, sources and distributional mode of aerosols – fundamental aspects needed to move towards an improved understanding of aerosol dynamics and effects in this high loading – less studied system.

2. Measurements and methods

2.1 The Maldives climate observatory Hanimaadhoo

The location of the MCOH was selected, in the wake of the Indian Ocean Experiment (INDOEX) programme, to intercept the outflow from continental S. Asia with minimal local influence. MCOH is located in the northernmost atoll of the Maldives, on the northernmost of its major inhabited islands, and at the northern point of the 6000 x 700 m sized island of Hanimaadhoo (6.77 °N, 73.18 °E) (Fig. 1).

The winter monsoon is typically active from November to April during which NE and NW winds generally bring pollutants from the S. Asian continent to the Indian Ocean (Fig. 1). The MCOH laboratory is located 1.5 m above sea level and is surrounded by ocean within 100 m on the W, N and E sides. MCOH has a 15 m atmospheric tower that rises above the local vegetation to allow unobstructed aerosol sampling and solar radiation measurements at the top platform. Various additional

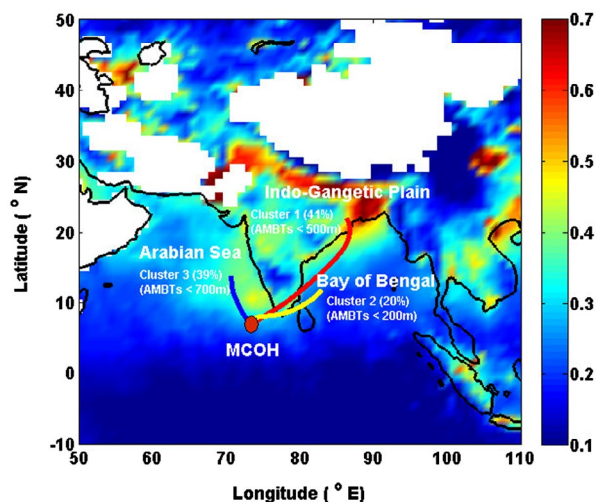


Fig. 1. This map depicts a MODIS satellite image, illustrating the typical high loadings of aerosols (aerosol optical depth-AOD at 550 nm, November 2014 to April 2015) over South Asia. Clustered 6-hourly, 7-day air mass back trajectories (AMBTs) means for Hanimaadhoo (total $n = 724$). The cluster means lines coloured in map indicate three different source regions: Indo Gangetic Plain (red), Bay of Bengal (yellow) and Arabian Sea (blue).

on-line measurements and samples are taken in the temperature- and humidity-controlled instrument room through a 15 m long PM_{10} sampling inlet. More details about the MCOH sampling site and operation of the station are described elsewhere (e.g., Corrigan et al., 2006; Budhavant et al., 2015a).

2.2 Instrumentation

This study comprises aerosol data from MCOH for six months (November 2014 – April 2015). The instrumentation at the measurement site that is relevant to this work is described below.

We measured the particle number size distributions in a size range of 0.007 – 0.80 μm with a 10-min time resolution using a Differential Mobility Particle Sizer (DMPS). The size-selected particles were detected with a condensation particle counter (CPC, TSI Inc., model 3010) after passage through a 28.5 cm long differential mobility analyzer (DMA). Size selected particles were measured in 35 increments. A neutralizer was used to obtain a steady-state charge distribution of the particles. In addition, total particle number concentrations were measured with another CPC (TSI Inc., model 3772). The integrated total number concentration from the DMPS agreed well ($\pm 5\%$) with the concentration measured by the standalone CPC.

Concentrations of carbon monoxide (CO) was measured continuously with an APMA-370 Analyzer (Horiba Instruments). The device employs an independent, internal dry-method to achieve a high level of sensitivity and accuracy ($\pm 1\%$ of full scale). The sample is pumped through an oxidizing catalyst

that converts the CO to CO₂ followed by non-dispersive infrared detection. This sampler has a detection limit of 0.02 ppm (3 times the standard deviation) and the calibration is auto-adjusted regularly with an external CO reference gas.

We also analysed the columnar aerosol optical depth (AOD), single scattering albedo (SSA, ratio of scattering efficiency to total extinction efficiency) and absorption aerosol optical depth (AAOD = [1 - SSA] × AOD) with data obtained from the sun photometer, which is part of the AERONET (AErosol RObotic NETwork) program. Here, we used the pre- and post-deployment calibration to attain the quality assured level 2.0 data (al-mucantar retrieval data); calibrated sky radiance measurements typically have an uncertainty less than 5% (Holben et al., 1998, 2001). The AOD is representative of columnar aerosol loading, and was here assessed both temporally through a sun photometer placed at the MCOH throughout the campaign, as well as average conditions for Nov 2014–April 2015 over the whole region (Fig. 1).

Additionally, a total number of 52 aerosol filter samples (26 of PM_{2.5} and 26 of PM₁₀) were collected in parallel using two identical high-volume samplers (Digital AG, Switzerland; model DH-77) that were set up on the top platform of the 15 m MCOH tower, and operating at 500 LPM. All samples were collected on pre-weighed and pre-combusted (5 h at 450 °C) 150 mm diameter quartz filters. Approximately one-week-long collections were performed to enable several elemental, molecular and isotopic analyses in this and forthcoming studies. All the filter handling and analysis of the carbonaceous aerosols followed the quality-controlled analytical protocols described earlier (e.g., Gustafsson et al., 2009; Sheesley et al., 2012; Bosch et al., 2014; Budhavant et al., 2015a).

2.3 Chemical analyses

We determined the mass concentrations of elemental carbon (EC) and organic carbon (OC) on the thermal-optical transmission (TOT) based Carbon Analyser from the Sunset Laboratory (Tigard, OR, USA) using the National Institute for Occupational Safety and Health (NIOSH) 5040 method (Birch and Cary, 1996). All reported OC results were blank subtracted using an average field filter blank (n = 6). The OC blank was 3% of the average filter loading. No EC was detected in the field blanks. Before the analysis of each batch of samples, the instruments were calibrated in the range 10–50 µgC with sucrose solutions for assessing the accuracy of TC (i.e., OC + EC) and is found to be within 5%. Further, we also analyse a sample which is traceable to the NIST RM-8785 reference material, to ensure that the split between OC and EC is properly estimated. Triplicate analysis was done roughly every sixth filter. The average relative standard deviation of triplicate analysis was 4% for EC and 5% for OC.

An aliquot of each aerosol sample was also extracted with ultrapure deionized water (Milli-Q; Specific resistivity > 18.2 MΩ-cm) and analysed for water-soluble inorganic

ionic species (cations: NH₄⁺, Na⁺, K⁺, Mg²⁺, and Ca²⁺; anions: Cl⁻, NO₃⁻ and SO₄²⁻) using a DIONEX Ion Chromatograph (IC; ICS-2000 model). Analytical quality checks were performed with external standards/reference material both with the Sunset OCEC (NIST-SRM urban dust, and gravimetrically prepared solutions of sucrose and potassium hydrogen phthalate) and with the IC (high-purity inorganic salt solutions containing the measured water-soluble inorganic constituents). Random error for the IC analyses, measured with a synthetic sample in each batch, given as one standard deviation, was 10% for cations and 5% for anions. To investigate the effect of marine influences on the aerosol composition, sea salt ratios were calculated using Na⁺ as the reference element (Keene et al., 1986).

2.4 Back-trajectory analysis

Air mass back trajectories were used to identify the pollutant source regions and subsequent transport pathways to the measurement site. The NOAA HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory (Stein et al., 2016) model was used to calculate seven-day air mass back trajectories (at 06:00 h, local time) at an arrival height of 100 m over MCOH (Fig. S1). The trajectory calculations were based on data from the Global Data Assimilation System (GDAS, 1 degree) of the US National Weather Service's National Centre for Environmental Prediction (NCEP), (Kanamitsu, 1989). Transport on local scales (<10 km) surrounding a site is better represented by local surface wind observations, which were obtained by a Vaisala weather station on Hanimaadhoo (Fig. S2). The back trajectories and local measured wind direction showed good agreement with predominantly northeasterly winds. We have also computed the average concentration and diagnostic mass ratios of specific chemical constituents, weighted according to their mean back trajectory paths over MCOH using the concentration-weighted-trajectory analysis (CWT), following a mathematical expression similar to that described earlier (Sheesley et al., 2012; Kirillova et al., 2013; Bikkina et al., 2016).

$$C_{SRj} = \frac{\sum C_i w_{ji}}{\sum w_{ji}}$$

where w_{ji} is the fraction wind originating from source cluster 'j' for sample number 'i', C_i denotes the concentration of chemical constituents in PM, and C_{SRj} is the mean concentration for cluster 'j'.

The local meteorological conditions (wind speed and direction, temperature, relative humidity and rainfall), recorded by the Maldives Meteorological Services (MMS) at the Hanimaadhoo airport (1.5 km to the south of the MCOH station), associated with these periods are shown in Figs. S2 and S3. The daily mean relative humidity was 80% and varied in the range 70–93%. The daily mean temperature was 28 °C and varied in the range 25–33 °C.

3. Results and discussion

3.1 Atmospheric transport

The cluster analysis of the HYSPLIT air mass back trajectory files yielded that the air parcels sampled over MCOH during November 2014 – April 2015 could be grouped into three classes. These originated from the Indo-Gangetic Plain (hereafter referred as IGP-origin, red colour), the Bay of Bengal (BoB-origin, yellow) and the Arabian Sea (AS-origin, blue) (Fig. 1 and Fig. S1). About 40% each of the received air parcels at MCOH were classified as IGP-origin and AS-origin with the remaining 20% occurrences were BoB-origin (Fig. 1). The spatial distribution of the period-average AOD also demonstrates that these air masses are instrumental in spreading out the aerosol pollution from these key continental source regions to the northern Indian Ocean (Fig. 1).

The Indo-Gangetic Plain (IGP) is a 255-million-hectar fertile plain encompassing most of the densely populated and industrialized northern and northeastern India, the eastern parts of Pakistan, and virtually all of Bangladesh. This back-trajectory cluster category represents air with very-likely influence of anthropogenic and other continental sources from Southern India and Sri Lanka. Atmospheric transport from the IGP, the BoB, and Southern India/Sri Lanka to MCOH belongs to this category, with typical transport times of 5 to 10 days until arrival at the site. The curved nature of the cluster trajectory through the Bay of Bengal is due to the persistent anticyclone circulation over the Indian subcontinent during the NE monsoon season.

Bay of Bengal (BoB) forms the northeastern part of the Indian Ocean. It is bordered mostly by India and Sri Lanka to the west, Bangladesh to the north, and Myanmar to the east. For the BoB-origin, air mass back trajectories mostly transect over the BoB and then through the southern part of India or Sri Lanka to reach MCOH.

Arabian Sea (AS) is located on the west side of the northern Indian Ocean, surrounded by Pakistan and Iran to the north and by NE Somalia and the Arabian Peninsula on the west, and by India on the east. Air masses of the AS-origin is particularly common at MCOH during the NE Monsoon and inter-monsoon periods.

3.2 Particulate mass and total particle number concentrations

Particle size is a fundamental property of an aerosol particle. In addition, the aerosol number, aerosol mass and chemical composition of the atmospheric aerosols are all contributors to the diverse processes that continuously determine how the ever-changing aerosol population evolve. These aerosol properties will be successively described for the northern Indian Ocean winter regime in the following sections starting with mass concentrations and total particle number concentrations.

Table 1. Statistical details of aerosol number, aerosol mass, various ionic concentrations and CO measured at MCOH over the full winter period November 2014 to April 2015.

Source region	Particle (#/cm ³)	Aerosol mass (µg/m ³)		EC (µg/m ³)		OC (µg/m ³)		Nss SO ₄ ²⁻ (µg/m ³)		Nss K ⁺ (µg/m ³)		CO (ppb)
		PM _{2.5}	PM ₁₀	PM _{2.5}	PM ₁₀	PM _{2.5}	PM ₁₀	PM _{2.5}	PM ₁₀	PM _{2.5}	PM ₁₀	
All	1300 ± 700	18.1 ± 8.26	28.7 ± 7.93	0.59 ± 0.28	0.60 ± 0.29	1.22 ± 0.71	1.40 ± 0.72	2.96 ± 2.90	3.14 ± 3.07	0.24 ± 0.13	0.24 ± 0.13	196 ± 64
IGP	1700 ± 650	22.9 ± 7.43	32.6 ± 5.54	0.74 ± 0.17	0.74 ± 0.17	1.72 ± 0.56	1.93 ± 0.54	5.92 ± 3.40	5.82 ± 3.63	0.29 ± 0.07	0.29 ± 0.08	232 ± 54
Bay of Bengal	1500 ± 450	19.5 ± 6.32	32.1 ± 7.35	0.72 ± 0.35	0.73 ± 0.35	1.23 ± 0.67	1.33 ± 0.68	1.56 ± 1.48	2.50 ± 2.40	0.30 ± 0.17	0.30 ± 0.15	188 ± 29
Arabia Sea	650 ± 330	12.2 ± 5.98	22.8 ± 6.69	0.38 ± 0.20	0.39 ± 0.19	0.66 ± 0.41	0.86 ± 0.43	0.42 ± 0.37	0.53 ± 0.52	0.15 ± 0.11	0.16 ± 0.11	145 ± 61

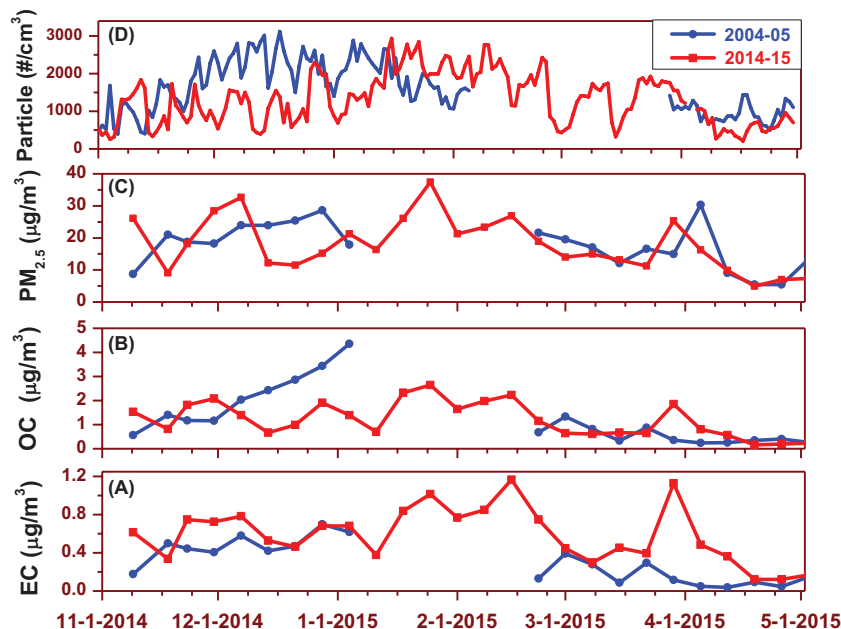


Fig. 2. Comparison of EC, OC, $PM_{2.5}$ (mass) and a daily average of total particle number concentration measured at Maldives Climate Observatory-Hanimaadhoo in full winter seasons ten years apart. The 2004–05 data are from Ramanathan et al. (2007a).

The highest mean concentration of total mass in $PM_{2.5}$ ($23 \pm 7 \mu\text{g m}^{-3}$) and in PM_{10} ($33 \pm 6 \mu\text{g m}^{-3}$) were observed at MCOH in the air masses of IGP-origin (Table 1 and Fig. 2). In contrast, the lowest mean values of $PM_{2.5}$ ($12 \pm 6 \mu\text{g m}^{-3}$) and of PM_{10} ($23 \pm 7 \mu\text{g m}^{-3}$) mass concentration were seen when the air masses came from over AS. The current study establishes that the fine fraction measured in $PM_{2.5}$ also made up an overwhelming part also of the total PM_{10} for all three air masses. On average, $PM_{2.5}$ mass of PM_{10} accounted for $\sim 54\%$ in the AS-origin, $\sim 61\%$ in the BoB-origin and $\sim 70\%$ in the IGP-origin. This observation suggests that the proportions of $PM_{2.5}$ contained within PM_{10} were quite high, compared to those documented over the Bay of Bengal by Budhavant et al. (2010) while more similar to those reported by Bikkina et al. (2011) (i.e., $PM_{2.5}$ accounted for $\sim 65\%$ of PM_{10} over the BoB for the air masses from IGP-origin). The somewhat higher bulk mass for PM_{10} is likely related to sea salt aerosols. The partitioning into the fine $PM_{2.5}$ fraction was greater for the carbonaceous components of aerosols as espoused below. This holds true also for non-sea-salt major ion components, as also shown below.

The atmospheric lifetime of fine mode aerosols in the IGP outflow is estimated to be about a week. This assessment is based on two independent approaches. First, the washout ratios of chemical constituents in the simultaneously collected rainwater and aerosol samples (Budhavant et al., 2010; Granat et al., 2010); and secondly, on the measured ^{210}Pb activity in the $PM_{2.5}$ from the IGP (Bikkina et al., 2015). The suggested 7–10 day timescale allows for long-range transport of a significant fraction of aerosols from as far as the N. India region

to MCOH. However, coarse mode particles, mainly produced by mechanical processes from both natural and anthropogenic sources, are apparently scavenged before reaching MCOH. At MCOH, sea salt aerosols are therefore expected to be the dominant contributor to the coarse mode. A comparison of total aerosol mass concentrations from the same site as well as other locations in the S. Asian region is presented in Table 2. Across S. Asia, $PM_{2.5}$ mass concentrations have been reported between 17 and $277 \mu\text{g m}^{-3}$ (Table 2). In summary, the total particle mass was partitioned between both coarse and fine modes, with the coarse mode likely dominated by sea salt aerosols, and the fine mode likely hosting most of the anthropogenic aerosols.

The total particle number concentration measured with the CPC is shown in Fig. 2. During the measurement period, the daily particle number concentration varied between 200–3000 cm^{-3} (av. $1300 \pm 700 \text{ cm}^{-3}$). Rapid increases in daily particle number concentration at MCOH were mainly associated with air masses carrying continental pollutants from the IGP region ($1700 \pm 650 \text{ cm}^{-3}$). The daily average aerosol number concentrations for the BoB and AS aerosols were 1500 ± 450 and $650 \pm 330 \text{ cm}^{-3}$, respectively. The measured variability of aerosol mass and number concentration at MCOH thus clearly depends on meteorological conditions.

The aerosol number concentration was mostly affected by wind speed ($r = -0.82$) and to some extent by precipitation (or rainfall) events (Table S2). The rain plays an important role in modulating the aerosol size distribution as the rainfall more efficiently scavenges larger particles (Hyvärinen et al., 2011; Budhavant et al., 2012). Occasionally, very high particle num-

Table 2. Aerosol mass and carbonaceous species ($\mu\text{g}/\text{m}^3$) from different sites in South Asia and adjacent seas and regions.

Station	Period	Size	Mass	EC	OC	OC/EC	nss SO_4^{2-}	nss K^+	NO_3^-	Reference
Hanimaadhoo, Maldives	Nov'14–Apr'15	$\text{PM}_{2.5}$	18.1	0.58	1.22	2.0	3.38	0.24	0.08	This study
Hanimaadhoo, Maldives	Jan'13–Apr'13	$\text{PM}_{2.5}$	19.5	0.49	1.31	2.7	na	na	na	Budhavant et al. (2015a)
Hanimaadhoo, Maldives	Nov'13–Dec'13	$\text{PM}_{2.5}$	17.0	0.27	1.12	6.1	na	na	na	Ramnathan et al. (2007a)
Male, Maldives	Nov'13–Dec'13	$\text{PM}_{2.5}$	21.2	0.76	1.88	2.4	na	na	na	Budhavant et al. (2015a)
Bay of Bengal	Jan'09	$\text{PM}_{2.5}$	22	1.2	2.6	2.0	7.44	0.47	na	Bikkina and Sarin, (2014)
Bay of Bengal	Jul'09–Aug'09	$\text{PM}_{2.5}$	33.8	na	na	na	2.30	0.09	0.72	Budhavant et al. (2010)
Dhaka, Bangladesh	Feb'10–Feb'11	$\text{PM}_{2.5}$	83	22.8	23.9	1.0	na	na	na	Begum et al. (2012)
Kuala Lumpur, Malaysia	2002–2011	$\text{PM}_{2.5}$	25	na	na	na	420	16.0	12.8	Rahman et al. (2015)
Karachi, Pakistan	Mar'09–Apr'09	$\text{PM}_{2.5}$	75	1.9	9.7	5.2	5.1	0.28	1.5	Shahid et al. (2016)
Lahore, Pakistan	Nov'05–Jan'06	$\text{PM}_{2.5}$	190	14.7	76	5.2	12.7	3.96	20	Raja et al. (2009)
Delhi, India	Nov'10–Feb'11	$\text{PM}_{2.5}$	277	46.3	104.4	2.3	25.9	3.81	32.8	Pant et al. (2015)
Kanpur, India	Oct'08–Nov'08	$\text{PM}_{2.5}$	133	6.2	36.6	5.9	13.5	2.1	2.10	Ram et al. (2014)
Hanimaadhoo, Maldives	Nov'14–Apr'15	PM_{10}	29.3	0.59	1.38	2.2	2.92	0.30	0.64	This study
Bay of Bengal	Jul'09–Aug'09	PM_{10}	80.9	na	na	na	3.31	0.29	1.24	Budhavant et al. (2010)
Karachi, Pakistan	Mar'09–Apr'09	PM_{10}	437	4.7	21.8	4.6	13.1	1.06	5.7	Shahid et al. (2016)
Delhi, India	Jan'13–Dec'14	PM_{10}	233	na	na	na	15.2	6.55	12.0	Saxena et al. (2017)
Lahore, Pakistan	March 2010	PM_{10}	406	21	61	2.9	na	na	na	Alam et al. (2014)
Ahmedabad, India	Dec'06–Jan'07	PM_{10}	171	4.4	29.8	7.5	13.6	0.90	7.2	Rengarajan et al. (2011)
Kanpur, India	Dec'08–Feb'09	PM_{10}	225	6.2	50	7.8	19.3	2.4	20.6	Ram et al. (2012)
Chennai, India	Jan'07–Feb'07	PM_{10}	na	6.5	9.1	1.5	–	–	–	Pavuluri et al. (2011)

Note: na = not available.

ber concentrations were found during midday hours. This is perhaps linked to new particle formation events over the ocean surface. The earliest reported aerosol number concentration data from MCOH (Ramanathan et al., 2007a) also show a similar kind of daily average variation ($400\text{--}3100\text{ cm}^{-3}$) for the dry season 2004–2005 as shown in Fig. 2.

3.3 Aerosol number-size distributions

Particle number size distribution and its variations were explored to investigate sources/formation and properties of the aerosol regime. During the Indian Ocean Experiment (INDOEX), the majority of detected particles with aerodynamic diameters between 0.3 and 1.0 μm were classified as carbon-containing particles by an aerosol time-of-flight mass spectrometer (Guazzotti et al., 2003). To explore the size distribution also of sub-micron particles ($d < 1\text{ }\mu\text{m}$), we measured aerosol size distributions in the range 0.007–0.8 μm using the DMPS. Size distributions were plotted separately for the three previously described source regions and presented as an average for the

entire study period (Fig. 3). The averaged size distributions of all the three clusters showed a modal peak close to 0.09 μm , indicative of aged and processed aerosols. The tails of the size distributions into larger sizes is strongly suggestive of cloud processing. The bi-modal nature of the distributions, typical to marine cloud-processed aerosol, becomes even more evident in Figs. S4–S6. They also show that the Arabian Sea cluster is the most diverse and IGP cluster is the most homogeneous. The Arabian Sea cluster is characterized by large variability in both concentrations and size distributions (Fig. S6). In the IGP cluster (Fig. S4), the Hoppel minimum (Hoppel et al., 1986) in the size distribution (about 0.15–0.20 μm) is indicative of the minimum size of aerosols that are activated. This minimum is the result of a selective activation of larger aerosol particles in the cloud forming process. Upon cloud evaporation, larger aerosol particles that had been activated create an accumulation-size mode that is distinct from the smaller aerosol particles that have not increased in mass by aqueous phase chemistry. Aqueous phase chemistry, the oxidation of SO_2 to sulphate adds solute mass. The bimodal distribution of the aged aerosol

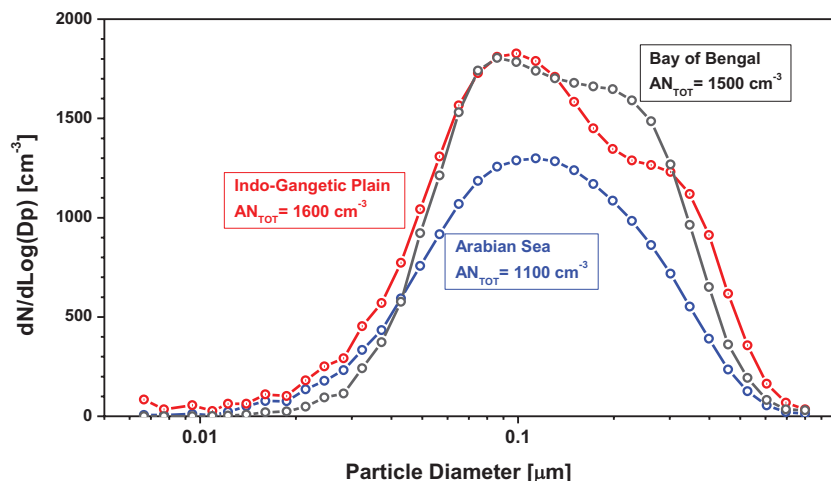


Fig. 3. The average particle number size distributions using Differential Mobility Particle Sizer (DMPS), resulting from grouping the data into three source regions. The numbers in coloured boxes present the average total number concentration for each of the sources.

is therefore most likely the result of atmospheric processing of regional haze particles during several days.

Most of the particle numbers found at MCOH during the study period are in the ‘accumulation mode’ (0.05 to 0.40 μm). These accumulation mode particles are removed from the atmosphere mainly by wet deposition. As aerosol particles in accumulation mode can remain in the troposphere for a long time (7–10 days) due to their slow removal, especially during dry season, they can be transported long distances from their sources. We have also used the HYSPLIT trajectory model to calculate rainfall to estimate and see wet deposition effects during transport. We found that the wet deposition effect was negligible, especially when trajectories come from the IGP (Figs. S7 and S8).

A unimodal size distribution was observed when the trajectories had not passed over land for the last 7–10 days. Accumulation mode particles over MCOH most likely originate from coagulation and condensational growth of Aitken mode particles advected to the site through long-range transport from South Asia. In addition, cloud processing is also likely to have grown particles into the Accumulation mode. Previous research in this region shows that the low frequency of rainfall during the wintertime Indian NE monsoon period allows for the build-up of aerosol pollution due to strong sources and limited advection (e.g., Budhavant et al., 2015a), which subsequently can be observed as a brown cloud over extensive spatial scales of South Asia and N. Indian Ocean (e.g., Ramanathan et al., 2001; Corrikan et al., 2006; Gustafsson and Ramanathan, 2016).

3.4 Aerosol optical depth (AOD)

The AOD gives a columnar integrated picture of aerosol abundance at the MCOH sampling location. The AOD at MCOH varied between 0.1 and 0.8 with an average of 0.36 ± 0.22 for

this period. The increase in columnar AOD and decrease in SSA were attributed to the arrival of air masses carrying more continental pollutants from IGP and the western margin of India that were characterized by relatively high concentrations of submicron absorbing aerosols (Fig. 4). In INDOEX aircraft experiment data (Ramanathan et al., 2001; 2007b), most of the contribution to the increase in AOD comes from levels below 3 km. During the sampling period, columnar AAOD varies from 0.01 to 0.07 with an average of 0.03 (Fig. 4). The highest values in AAODs normally match those regions that are heavily influenced by either regional forest fires or enhanced use of biomass fuels in densely populated developing countries (Laskin et al., 2015). This also supports that South Asia-IGP is a key regional hot spot source region defined by the criteria that anthropogenic AODs should exceed 0.3 and AAOD > 0.03 (Ramanathan et al., 2007a; Laskin et al., 2015). Experimental and modelling studies (e.g., Ramanathan et al., 2007b; Ten Hoeve et al., 2012; Lin et al., 2014) have recognized significant contributions from the Brown Carbon component (BrC, the light-absorbing organic carbon) to the observed AOD and AAOD values, and emphasized the critical need for a quantitative breakdown of the relative BrC and BC contributions as well as the need for advanced characterization of BrC, for improved climate predictions.

3.5 Aerosol chemical mass closure

The chemical mass closure was studied by comparing the mass derived from the chemical analysis to the gravimetric mass obtained by weighing the filter samples. PM mass closure applies multipliers to several of the measured species to estimate unmeasured components (e.g., Neususs et al., 2000; Chow et al., 2015). In this work, the mass of chemical components was calculated by summing up all measured ions, EC and organic matter (OM). Further, the chemical components were divid-

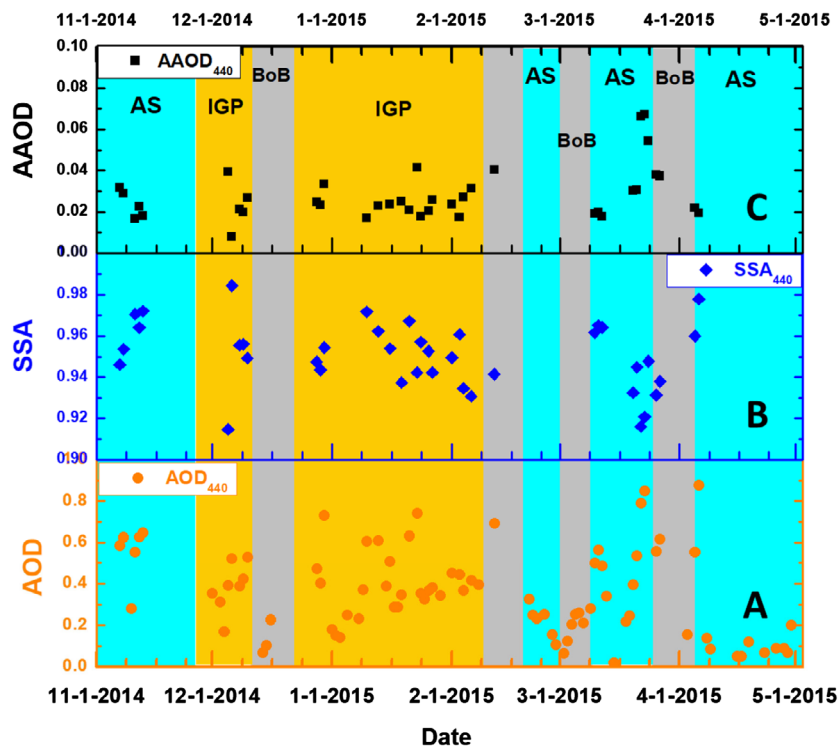


Fig. 4. Daily average aerosol optical depth (AOD), single-scattering albedo (SSA) and absorption aerosol optical depth (AAOD) measured at Maldives Climate Observatory-Hanimaadhoo using CIMEL Sun photometer during November 2014 to April 2016. Vertical colour shaded bars indicate the three different source regions: Arabian Sea (Cyan), Indo Gangetic Plain (Canary) and Bay of Bengal (Grey).

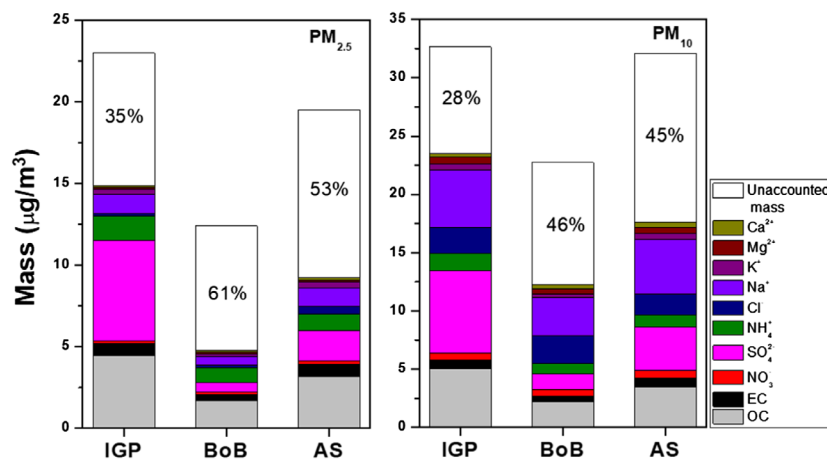


Fig. 5. Mass closure of particulate matter in $PM_{2.5}$ and PM_{10} measured at Maldives Climate Observatory-Hanimaadhoo from three different source regions: Indo Gangetic Plain (IGP), Bay of Bengal (BoB) and Arabian Sea (AS) during November 2014 to April 2015.

ed into four classes: secondary inorganic aerosols ($nss\text{-}SO_4^{2-}$, NO_3^- , and NH_4^+) (SIA), sea salt, dust and carbonaceous aerosols (EC + OM). The mass closure requires a multiplication factor to bring OC measurements to estimates of the mass of the total particulate OM. Bikkina and Sarin, (2014) recommended the conversion factor of 2.3 ± 0.6 for aged marine aerosols influ-

enced by air masses from IGP and south-east Asia. In the present work a factor of 2.3 was chosen to estimate OM.

The contributions of the various components to the average gravimetric PM from different source regions were calculated and are shown in Fig. 5. The identified aerosol chemical components accounted for a higher fraction of the total PM for the

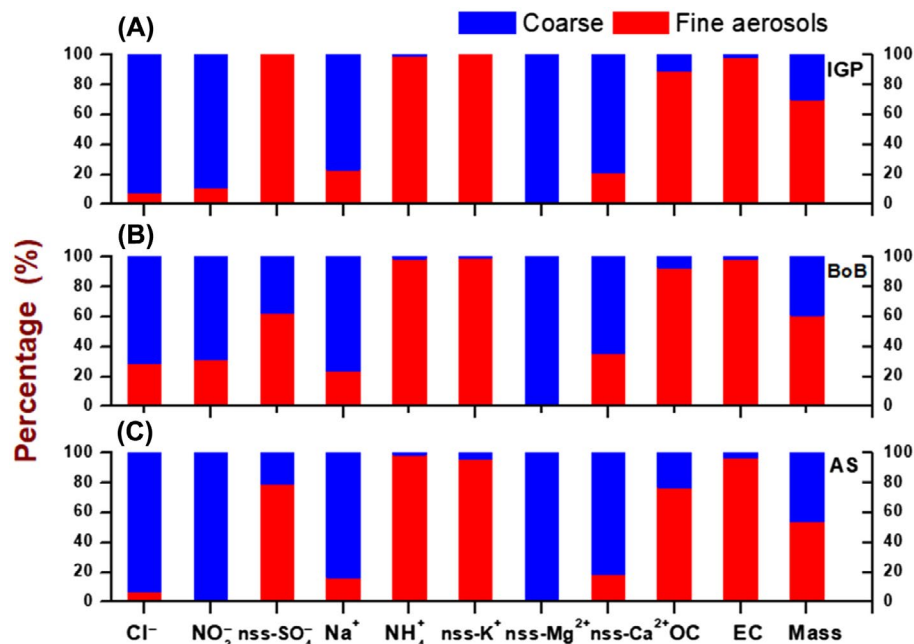


Fig. 6. Chemical composition and mass concentration of fine ($PM_{2.5}$, red) and coarse ($PM_{10-2.5}$ blue) aerosol from different source regions measured at MCOH. Panel A. Indo-Gangetic Plane (IGP), B. Bay of Bengal (BoB) and C. Arabian Sea (AS) during November 2014 to April 2015.

IGP outflow (72% for PM_{10} and 65% in $PM_{2.5}$) and for a lower fraction when the wind comes from the BoB (54% for PM_{10} and 38% in $PM_{2.5}$). SIA and carbonaceous aerosols were predominantly found in $PM_{2.5}$. On the other hand, sea salt and SIA were mainly present in PM_{10} . Particularly $nss-SO_4^{2-}$ concentrations were significantly higher in total sulphate during IGP outflow (95% in $PM_{2.5}$ and 82% for PM_{10}) compared to in air masses coming from over the AS and BoB (Table 1). The higher concentrations of $nss-SO_4^{2-}$ in the IGP outflow suggest a large input of non-biogenic sources. On the other side, the lower concentrations of $nss-SO_4^{2-}$ over the BoB and the AS representative of marine-biogenic sources.

Likely explanations in the unaccounted mass in $PM_{2.5}$ (54%) and PM_{10} (38%) include trace elements and aerosol-bound water. Another source of systematic error is the OC multiplication factor. In this study, the factor of 2.3 was applied for all the samples, although it is known that the particulate OC to OM ratio depends on the emission sources, atmospheric transport duration and meteorological conditions.

3.6 Characteristics of ionic aerosol components

The chemical characterization of aerosol particles is informative of their sources and transformation. $PM_{2.5}$ and PM_{10} were characterized in terms of major cations (NH_4^+ , Na^+ , K^+ , Mg^{2+} and Ca^{2+}) and major anions (Cl^- , NO_3^- and SO_4^{2-}) for the six-month sampling period (Fig. 5). Sulphate and ammonia dominate the $PM_{2.5}$ mass, indicating strong anthropogenic sources for the ambient fine aerosol over the N. Indian Ocean. In the fine mode,

SO_4^{2-} (20%) showed the highest concentration followed by NH_4^+ (6%), Na^+ (5%), K^+ (2%), Cl^- (1%), Mg^{2+} (1%); NO_3^- and Ca^{2+} were present in smaller amounts. In contrast, for the coarse fraction, Na^+ (28%) showed the highest concentration followed by Cl^- (17%), NO_3^- (4%), Mg^{2+} (3%), Ca^{2+} (2%), K^+ (2%) and NH_4^+ (negligible amount). These observations clearly showed that marine components like Na^+ , Cl^- and Mg^{2+} were dominant in coarse mode (48%) while their contribution was much less in the fine mode (7%). The major fraction of sea salt aerosols is generally found in the coarse size range (Heintzenberg et al., 1981; Porter and Clarke, 1997; Budhavant et al., 2016). The main source of sea salt aerosol over the open ocean is sea spray droplets (Blanchard and Woodcock, 1980; Martensson et al., 2003). In fact, the slope of the linear regression analysis of Na^+ versus Mg^{2+} ($R^2 = 0.99$; slope = 0.12) in PM_{10} is consistent with that typically observed for sea salt aerosols (Mg^{2+}/Na^+ ratio in Seawater: 0.12).

High concentrations of NO_3^- in the coarse mode at MCOH together with its clear linear relationship with Na^+ in PM_{10} ($R^2 = 0.40$; slope = 0.14) indicates the presence of $NaNO_3$ as a result of the reactive uptake of gaseous phase nitric acid on sea salts. Over the ocean, the size distribution of NO_3^- normally follows the surface area distribution of sea salt (Li-Jones and Prospero, 1998; Murphy and Thomson, 1997). The composition was thus quite different for the fine fraction. The contribution of SO_4^{2-} , NO_3^- and NH_4^+ together was much higher in the fine fraction (27%) than in the coarse fraction (5%). These are likely linked to long-range transport from anthropogenic emissions, originating primarily on the Indian subcontinent. By

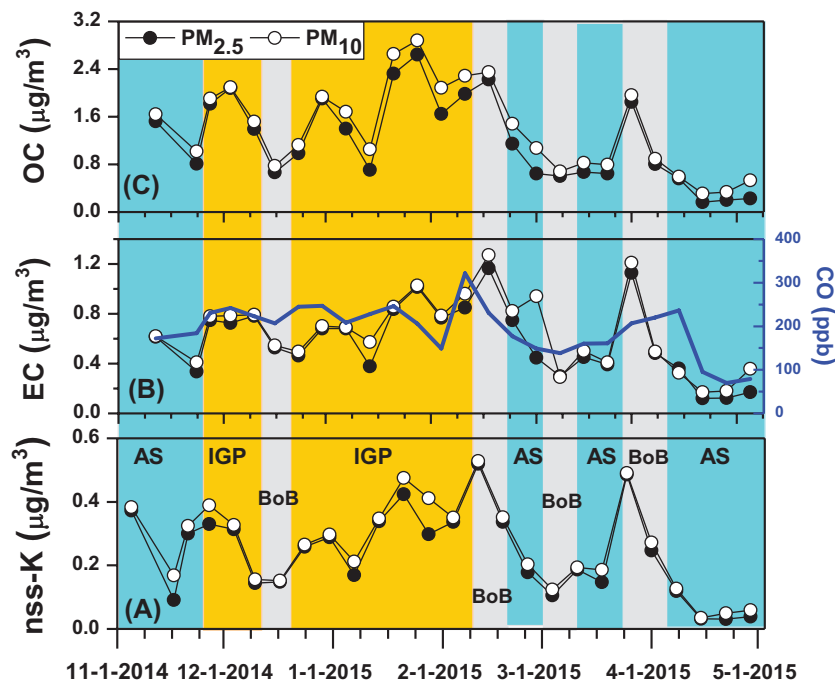


Fig. 7. Time series of nss K^+ , EC and CO, as well as OC in $PM_{2.5}$ and PM_{10} at Maldives Climate Observatory at Hanimaadhoo during November 2014 to April 2015. Vertical colour shaded bars indicate the three different source regions: Arabian Sea (Cyan), Indo Gangetic Plain (Canary) and Bay of Bengal (Grey).

enlargement, it was thus found that anthropogenic components (nss- SO_4^{2-} , nss- K^+ and NH_4^+) were predominantly partitioned to the fine-sized aerosols, which was similar to what was also found for the carbonaceous components.

3.7 Characteristics of carbonaceous aerosol components

The distribution and properties of the carbonaceous aerosol fraction are important as they have proportionally a much larger impact on the aerosol absorption of solar radiation and thus on net radiative effects. EC normally makes up 3–10% of total aerosol mass in S. Asia (e.g., Lelieveld et al., 2001; Bikkina and Sarin, 2014; Budhavant et al., 2015a). The mean concentration of EC was found to be $0.59 \pm 0.28 \mu\text{g}/\text{m}^3$ in $PM_{2.5}$ and $0.60 \pm 0.29 \mu\text{g}/\text{m}^3$ in PM_{10} (Table 1). The highest EC concentrations were observed for the IGP-origin, $0.74 \pm 0.17 \mu\text{g}/\text{m}^3$, whereas the lowest values were found for the Arabian Sea origin $0.38 \pm 0.20 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$. The EC concentration measured during this study is almost double that previously reported at MCOH during the dry winter 2004–05, $0.27 \mu\text{g}/\text{m}^3$ (Fig. 2). The temporal variations during winter 2014–2015 in concentrations of OC and EC (and the gaseous combustion marker CO) showed similar patterns (Fig. 7).

A significant correlation between OC and EC (slope = 2.28; $R^2 = 0.83$ for $PM_{2.5}$ and slope = 2.33; $R^2 = 0.81$ for PM_{10} ; Fig. 8) was observed during the study period, suggesting their com-

mon sources. EC is exclusively of primary origin as it is the product of incomplete combustion. Since EC undergoes minimal chemical transformation, it serves as a good tracer for anthropogenic aerosols. The co-variability of OC and EC together with their significant linear relationship and their near-exclusive partitioning to the fine $PM_{2.5}$ fraction (Fig. 2) suggests that the ambient concentration levels of not only EC but also of OC were controlled largely by primary emissions and atmospheric dispersion, rather than by any secondary OC formation. Table 2 shows the concentrations of total aerosol mass as well as of various aerosol chemical components measured at MCOH in comparison to previous studies at different locations in mainland South Asia. Locations in IGP such as Lahore, Delhi, Kanpur and Dhaka are the most polluted ($TC > 45 \mu\text{g m}^{-3}$). Lower but still high pollution ($TC < 15 \mu\text{g m}^{-3}$) has been observed in cities like Karachi, Ahmedabad and Kanpur.

The contribution of total carbonaceous aerosols (TCA) to the total particulate mass concentration was also estimated. The TCA accounted for a higher fraction of total PM for the IGP outflow (24% in $PM_{2.5}$ and 18% for PM_{10}) with lower values when the wind comes over the BoB (16% $PM_{2.5}$ and 11% for PM_{10}). The average TCA was $20 \pm 6\%$ of $PM_{2.5}$ and $14 \pm 5\%$ of PM_{10} at MCOH. These results are consistent with earlier reported values from the same station for $PM_{2.5}$ (Ramanathan et al., 2007a; Budhavant et al., 2015a). Bikkina and Sarin (2014) also reported similar TCA concentrations for the IGP source region ($22 \pm 6\%$, in $PM_{2.5}$, $16 \pm 3\%$ in PM_{10}).

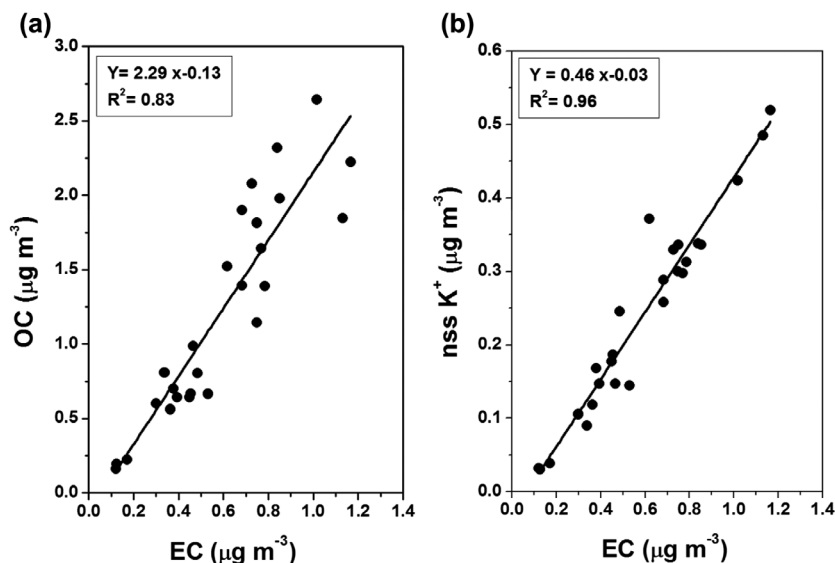


Fig. 8. Linear regression analysis between mass concentration of EC and (a) organic carbon, OC (b) non-sea-salt water-soluble potassium, (nss-K⁺) in PM_{2.5} collected over the MCOH during November 2014 – April 2015.

3.8 Source categorization of chemical components of aerosols over northern Indian Ocean

Pinpointing the sources of carbonaceous aerosols is important in order to understand their atmospheric concentrations and processing, as well as to study their possible effects on climate and air quality. The mass ratio of OC to EC reflects multiple processes in the atmosphere (e.g., Kroll et al., 2011). The OC/EC ratios at MCOH ranged from 1.3 to 2.9 (avg. 2.0 ± 0.5) for PM_{2.5}. The highest values were observed for IGP-origin, 2.9 (avg. 2.3) for PM_{2.5}, whereas the lowest values were found for BoB-origin 1.3 (avg. 1.7) for PM_{2.5}. These OC/EC ratios are quite low in comparison with other reported studies based in mainland South Asia except some urban cities like Dhaka and Chennai (Table 2). Consistently low OC/EC ratios (2.0 ± 0.5), strong linear relationships between EC and OC, as well as the steady bimodal fine number-size distribution, combine to suggest a predominance of a common source for EC and OC.

We observed a significant linear relationship of not only EC with OC but also between EC and nss-K⁺ in the PM_{2.5} sampled at MCOH during November 2014–April 2015. The nss-K⁺ is a diagnostic proxy for identifying regional impact of biomass burning emissions (Andreae, 1983; Paris et al., 2010). Therefore, the former linear relationship indicates common source contributions and subsequent long-range atmospheric transport of carbonaceous species to MCOH, while the latter relationship emphasizes the significance of a biomass combustion source for EC and OC.

The MODIS active fire counts' data (Fig. S9) showed high incidences of fires over India and Bangladesh during Nov 2014 to April 2015, also suggesting high emissions of biomass burn-

ing aerosols. The major contributions of carbonaceous aerosols reported from S Asia in cities like Kanpur (Ram et al., 2014), Ahmedabad (Rengarajan et al., 2011) and Karachi (Shahid et al., 2016) have been suggested to stem from biomass burning. At the same time, fossil fuel combustion has been suggested to be the major source of EC and OC in mega-cities such as Lahore (Alam et al., 2014), Delhi (Saxena et al., 2017), Dhaka (Begum et al., 2012) and Chennai (Pavuluri et al., 2011). Research using source-diagnostic ¹⁴C fingerprinting of ambient EC shows that fossil and biomass combustion processes are about equally responsible for the emission of EC over South Asia (Gustafsson et al., 2009; Budhavant et al., 2015b).

Based on the Peking University emission inventory (Wang et al., 2014), using a box [South: long. 70° to 90°; lat 5° to 15°, North: long 70° to 90°; lat 20° to 30°], shows that the emissions from the northern India is ~5 times higher than for the southern part. However, dispersion and atmospheric lifetimes affect the concentrations observed at MCOH. As aerosols and air masses arriving at MCOH is predominantly of IGP origin, it is likely that the wood /crop-residue burning emissions over the Indo-Gangetic Plain could be a major source of carbonaceous aerosols in the atmosphere over the N. Indian Ocean (Fig. 9). This is further consistent with earlier studies of the diagnostic mass ratios of chemical constituents, organic molecular markers as well as radiocarbon source apportionment of aerosols that combine to suggest that postharvest crop residue and wood burning emissions contribute significantly to the S. Asian outflow (Ramanathan et al., 2007a; Rengarajan et al., 2007; Gustafsson et al., 2009; Bikkina and Sarin, 2014; Kirillova et al., 2014; Budhavant et al., 2015b).

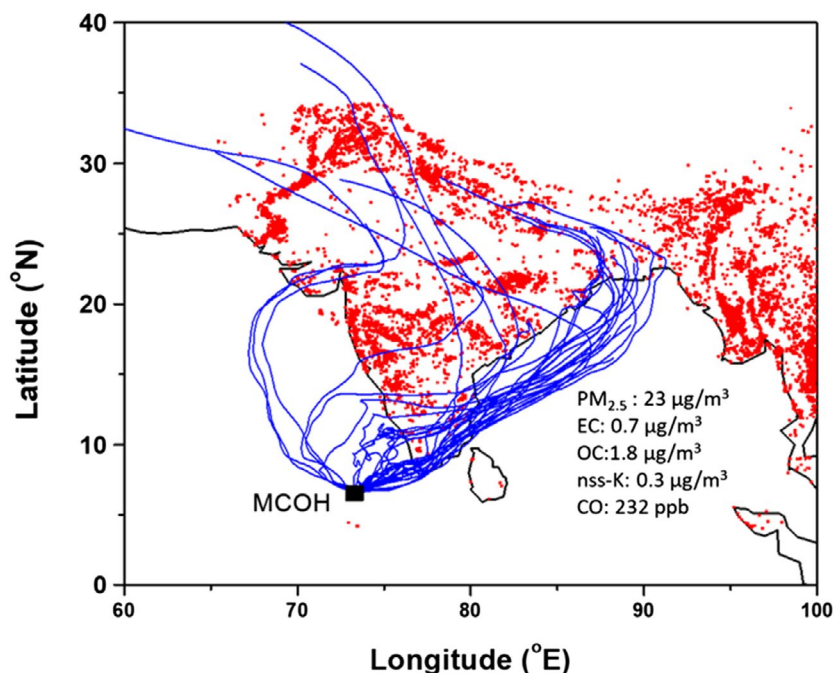


Fig. 9. Air mass trajectories for 7 days ending at MCOH at 100 m with MODIS /Aqua active fires during the Indo-Gangetic Plain (IGP) outflow.

4. Conclusions

The aerosol regime over the N. Indian Ocean during the November 2014–April 2015 period is largely influenced by South Asian pollution. The chemical and physical properties of the air masses transported from the dominant IGP, Bay of Bengal and Arabian Sea source regions showed remarkably little variation during the six-month period of intensive observations at the Maldives Climate Observatory – Hanimaadhoo (MCOH).

The coarse mode was influenced by both sea salt aerosols and long-range transported aerosols of largely anthropogenic influence. In contrast the much more abundant (in mass and number) fine ($PM_{2.5}$) aerosols were overwhelmingly dominated by anthropogenic particles, as indicated by the fingerprints of OC, EC, $nss-SO_4^{2-}$, $nss-K^+$, and NH_4^+ . The EC, as a tracer of incomplete combustion, was $97 \pm 3\%$ in the fine mode.

Consistently low OC/EC ratios (2.0 ± 0.5), strong linear relationships between EC and OC as well as between $nss-K^+$ (a proxy for biomass combustion) and both OC and EC, as well as the steady bimodal fine number-size distribution, combine to suggest a predominance of a common primary source, with a large biomass burning contribution.

Taken all together, the wintertime high-AOD, high-AAOD aerosol regime over N. Indian Ocean appears to be well homogenized, and dominated by the fine fraction for key components such as EC and OC, which in turn appear to be predominantly of primary origin with biomass burning as a putatively major source. These results for the sparsely studied, but high-AOD,

high-AAOD regime of the N. Indian Ocean facilitates our understanding of the sources and properties of its aerosol regime.

Supplementary material

Supplemental data for this article can be accessed here [10.1080/16000889.2018.1464871](https://doi.org/10.1080/16000889.2018.1464871).

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Disclosure statement

No potential conflict of interest was reported by the authors.

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