



Seasonal variability in atmospheric black carbon at three stations in South-Asia

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ABSTRACT

Filter-based optical measurements of black carbon in air, a constituent of soot, have been determined with a 528 nm light source during the period from 1 June 2005 to 31 May 2009 on samples taken at Godavari in Nepal, Sinhadag in India and Hanimaadhoo in the Maldives. In order to reduce systematic errors due to the light scattering of non-absorbing particles co-deposited on the filter, such as inorganic salts and mineral dust, an additional sensor recording backscattered light was implemented. Two protocols of corrections (optical and chemical) were applied to the samples collected at the observatories. The Indian monsoon circulation with its two annual phases in combination with the location of the combustion sources and their contribution relative to other non-anthropogenic sources dominated the observed patterns of black carbon at two of the observatories: in India and the Maldives. The observatory in Nepal was however mainly influenced by combustion sources all year around concealing possible variability related to the monsoon circulation. At the receptor observatory in the Maldives, peak values in the black carbon absorption coefficient occurred during the winter season (December to April) when air was transported from the polluted Indian subcontinent out over the Indian Ocean. A close to two orders of magnitude lower values were recorded in air that had spent more than 10-days over the Indian Ocean during the monsoon season (July to September), suggested to be dominated by particulate matter from remote marine biogenic sources and not by combustion sources.

Keywords: soot, BC, light absorption, light scattering, South-East Asia, monsoon circulation

1. Introduction

Anthropogenic Black carbon (BC), a constituent of soot, is getting increasing attention as climate warming agent of comparable strength as anthropogenic CO₂ (IPCC, 2013) albeit with higher mitigation potential (Ramanathan et al., 2007). Besides the climatic effects, polyaromatic compounds in soot are known to cause serious health effects (UNEP, 2002). Both, climate and health effects change with residence time of BC in the atmosphere because of atmospheric transformations. Consequently, it is essential in both climate and public health discussions to know not only the origin of the BC component of the aerosol and the emitted amounts of BC and other light absorbing anthropogenic particles but also their life cycle in the atmosphere.

Whereas major sources of BC have been estimated (Gustafsson et al., 2009) much less is known about its change of morphology and different states of BC mixture for a given mass fraction of BC during ageing in the atmosphere. In the atmosphere, 'fresh' BC spheres are normally aggregated together in chain-like structures that have an opened and branched structure (Coz and Leck, 2011, Li et al., 2016). These chain-like structures

basically consist of elemental and organic carbon. During ageing in the atmosphere these structures will collapse to become a compact and closed structure. The structure of the primary spherules of BC, either in chain-like open structures, or in more compact and closed aggregates frequently form dislocations that allow the incorporation of other matter such as sulphuric acid, inorganic salt or any organic compound with surface active properties (Coz and Leck, 2011; Hede et al., 2013), that will influence not only particle dispersion, transport and removal from the atmosphere but also modify particle absorption and scattering of light. Data on atmospheric particle morphology and quantification of how it affects all these processes are quite complex and still largely unknown.

Moreover, our understanding of the sampling and analytical techniques concerning BC and its effects on the estimates of the light absorption are far from satisfactory. A fundamental and serious drawback of the well-established filter-based optical measurements of aerosol absorption coefficients is that the light absorbing particulate matter is co-deposited with non-absorbing or low absorbing particles on a filter matrix prior to the light absorption determination.

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A number of studies have suggested that the presence organic carbon (OC) co-deposited on the filter-matrix could have a significant effect on aerosol light absorbing measurements (Schnaiter et al., 2005, Subramanian et al., 2007, Cappa et al., 2008; Lack et al., 2008). The light beam will pass the transparent organic coating, which enables the beam to be focused on the absorbent ‘soot’-core. The absorbing core will intercept more of the incoming light with a reduction in the transmitted light as a result. Therefore, a larger apparent absorption will be detected. Quinn et al. (2002) reported a mass fraction of <15% for OC and up to 7% for BC in the outflow from the Indian sub-continent. In the same area Mayol-Bracero et al. (2002) found a mass fraction of 35% for OC and 14% for BC. So even if we allow for a reduction in the transmitted light by internally mixed OC, non-absorbing constituents other than OC make up more than 65% of the non-absorbing mass. Additional, empirical corrections are therefore required to reduce optical effects of e.g. sulphate, nitrate, pollution-dust and sea salt.

In a study by Engström and Leck, (2011) optical corrections of light scattering by the non-absorbing inorganic material were applied based on the chemical quantification of the material co-deposited on the filter matrix. The correction protocol was applied to aerosol samples collected at the Maldives Climate Observatory Hanimaadhoo (MCOH) during episodes with either continentally influenced air from the Indian/Arabian subcontinents (winter season) or pristine air from the Southern Indian Ocean (summer monsoon). The chemical correction lowered the particle light absorption coefficient of BC by 61%, for data sourced from the Arabian Sea. The corresponding value for the South Indian Ocean data was 97%. Thus in general *in situ* measurements of direct climate forcing parameters such as aerosol light extinction (sum of scattering and absorption) are still in their infancy (Thompson et al., 2003) and require additional chemical information preferably determined on the same samples in order to attribute the climate effect to specific sources.

The present study report on filter-based optical measurements of BC in air collected at three baseline-monitoring observatories: Godavari (Nepal), Sinhadag (India) and Hanimaadhoo (the Maldives) from 1 June 2005 to 31 May 2009. An advantage in using a filter-based optical method is that it can be implemented in the field under primitive conditions and at low cost. This choice of method therefore enables researchers with small economical means to perform monitoring at remote locations, especially in the Asia where it is much needed (Ramanathan et al., 2007). A photometer was used to measure light transmission at a wavelength of 528 nm on particulate matter accumulated on polycarbonate membrane (PCMB) filters (Heintzenberg, 1988). The PCMB filters were selected to avoid high chemical blank values characteristic for the otherwise commonly used glass fibre filters (Bond et al., 1999). In order to reduce systematic errors due to the light scattering of non-absorbing particles co-deposited on the filter, such as inorganic salts and mineral

dust, an additional sensor recording backscattered light was implemented. Two protocols of corrections (optical and chemical) based on Engström and Leck (2011) were applied to the samples collected at the observatories.

Key questions addressed were: (1) Is there a seasonal variability in light absorbing particulate matter observed in air at three monitoring sites located in Nepal, India and the Maldives, and if so, what is the cause of the variability? (2) Would the particulate light absorption coefficient change from urban Indian/Nepal sources to the receptor region in the Maldives?

2. The sites and methods of measurement

2.1. Sites, setups and observing periods

2.1.1. Godavari, Nepal. The Godavari observatory (longitude E85°23'20", latitude N27°35'31") is located on a north-facing mountain slope 1600 m above sea level (asl) in central Nepal (Fig. 1). The mountain slope is a part of a narrow valley that opens up to the west and the Kathmandu basin. The distance to the city of Kathmandu in the north-west is around 10 km. The vegetation on the mountain slope is mixed deciduous and evergreen broad-leaved forest, and the soil is clay rich with varying content of sand. No strong emission sources are in the immediate surroundings and the closest village is 1 km away (International Center for Integrated Mountain Development, ICIMOD, 2010). The most important emission source is Kathmandu (total population of about 2 million), and other large emission sources are present at longer distances in Nepal, Indian and China. Measurements were performed from 1 October 2005 to 31 December 2007.

2.1.2. Sinhadag, India. Sinhadag (longitude E73°45'17", latitude N18°21'58") is an old fortress built on a mountain-top at a height of 1300–1400 m asl in Western Ghats in India (Fig. 1). The observatory at Sinhadag is located at the highest position of the mountain-top that is a flat area of 0.5 km². No strong emission sources are in the immediate surroundings, and the closest city is Pune (total population of about 3 million), 18 km to the north-east. Few people live at the summit and tourists visit the area by foot. Cars and buses cannot get closer than 1 km (Momin et al., 2005). Data was collected from 1 August 2005 to 8 June 2008.

2.1.3. Hanimaadhoo, Maldives. The Hanimaadhoo observatory is located on the northern point of Hanimaadhoo, an island in the northern part of the Maldives (longitude E73°10'59", latitude N6°46'34"), and stretching 4 km north to south and 1 km east to west (Fig. 1). Hanimaadhoo has a population of around 1200 and is free from influence of larger landmasses, population centres and industries. The observatory's location, at the northern cape of the island with

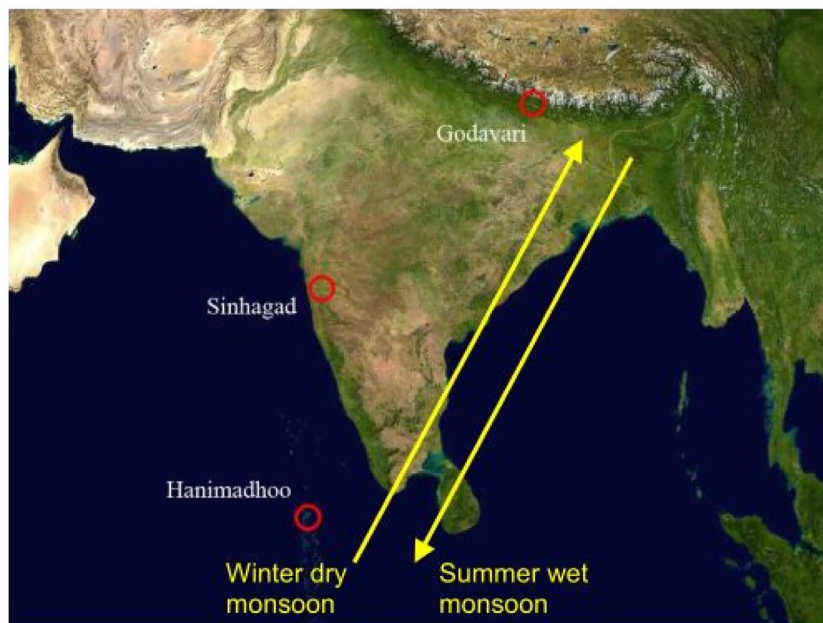


Fig. 1. The location of the ABC observatories in Nepal (Godavari, longitude E85°23'20", latitude N27°35'31"), India (Sinhagad, longitude E73°45'17", latitude N18°21'58") and the Maldives (Hanimaadhoo, longitude E73°10'59", latitude N6°46'34"). Also shown is the main direction of the monsoon wind.

ocean on three sides at most 100 m away, ensures that the prevailing wind comes from over the ocean. Thus the site has no significant local BC emissions upwind from the station and the composition of the air and rain is dominated by the regional sources (Corrigan et al., 2006). Samples were collected during the period 1 June 2005 to 31 May 2009.

2.1.4. Sampling set up at the three observatories. The collection of aerosol particles was simultaneously operated with two parallel PCMB filter (0.4 μm pore size, 37 mm diameter) based samplers with identical geometry: (1) discrete sampling using a filter cassette and subsequent post sampling determinations with a BC-photometer instrument located at the Department of Meteorology, Stockholm University, MISU, (more details in Section 2.2.1), (2) continuous recording (1 s time resolution) with an online Particle Soot Absorption Photometer (PSAP-MISU) as described in Section 2.2.2.

At Godavari and Sinhagad the sampling inlets for atmospheric aerosol particles were 4 m long and made of stainless steel tubing (50 mm inner diameter). At Godavari the inlet was 1 m away from the building and on level with the roof-top. The air inlet at the Sinhagad observatory was positioned 1 m above the roof-top at the end of the stainless steel tubing. The air at the Hanimaadhoo observatory was sampled from a 15 m high tower. A cyclone at the top of the sampling line (15 m long stainless steel tubing with an inner diameter of 20 cm) excluded particles smaller than 10 μm in equivalent aerodynamic diameter at a laminar flow of 300 $\text{dm}^3 \text{min}^{-1}$. To avoid wall effects, an aluminium tubing (diameter 15 cm, concentric head) selected

air in the middle of the inlet. At all three observatories the samplers were installed at surface level in the building in a temperature and humidity controlled environment to avoid condensation of water inside the instruments due to lower temperatures (Corrigan et al., 2006). Through isokinetic take off lines from the inlet pipe a cyclone with a size cut of $\text{Dp}_{50} = 2 \mu\text{m}$ was mounted upstream of the samplers. Sampling flow was set to 2.0 $\text{dm}^3 \text{min}^{-1}$. As the three stations were located at different heights above the sea level, Sinhagad 1300–1400 m, Godavari 1600 m and Hanimaadhoo at 0 m the decrease of air density with height demanded a flow correction in the conversion from air mass to air volume. The sampling flows were therefore monitored with mass flow controllers and logged. The discrete filter cassette samples were collected, for 24 or 48 h depending on real-time concentration levels. After sampling, the cassettes were sealed with para-film and stored until subsequent determination at MISU.

2.2. Methods of recording the light absorption coefficient common to all observatories

2.2.1. The BC-photometer instrument. For post-sampling determinations of the light absorption coefficient (σ_{ap}) of BC for the discrete filter cassette samples, a BC-photometer instrument located at MISU was used. To optimize the analytical conditions (increase the signal to noise ratio) in the BC determinations the exposed PCMB Nuclepore® filter surface was masked to 8 mm sampling diameter (0.5 cm^2 area) and referred to as the sample spot (Hansson et al., 1987). The BC-photometer instrument was

constructed according to the set-up by Heintzenberg (1988) and used a 528 nm LED light source (Fig. 2). A sensor behind the PCMB Nuclepore® filter measured the transmitted light intensity through the BC sample spot on the filter. A second sensor measured the transmitted light through an unexposed part of the filter. Behind both the sample and reference spot a Teflon® plate acting as a Lambertian diffuser was placed. The difference in intensity of transmitted light between exposed (I_2) and unexposed (I_1) filter surface was used to calculate the optical density (Od) (Equation 1). σ_{ap} is defined as Od per meter air column and is calculated by multiplying Od with the PCMB filter sample spot area (A) and dividing by the volume of air sampled (V) (Equation 2).

$$Od = \ln \left[\frac{I_1}{I_2} \right] \quad (1)$$

$$\sigma_{ap} = \frac{A}{V} \ln \left[\frac{I_1}{I_2} \right] \quad (2)$$

To correct for light scattering by the non-absorbing matter co-deposited on the PCMB filter the BC-photometer had a third sensor to measure the light backscattered at an angle of 40° from the filter surface. Two protocols of corrections (optical and chemical) based on Engström and Leck (2011) were applied to the discrete filter cassette samples. More details in Section 2.4.

Using PCMB filters over a range of BC loadings, corresponding to ambient levels, the standard deviation (1σ) was established to be 0.0004 in units of Od and 0.0014 Mm^{-1} in units of σ_{ap} (in relative measures better than 7, 0.03 and 0.009% for $\sigma_{ap} > 0.02$, 5 and 15 Mm^{-1} , respectively). The overall coefficient of variation for the sample collection and determination added up to 7% (Engström and Leck, 2011).

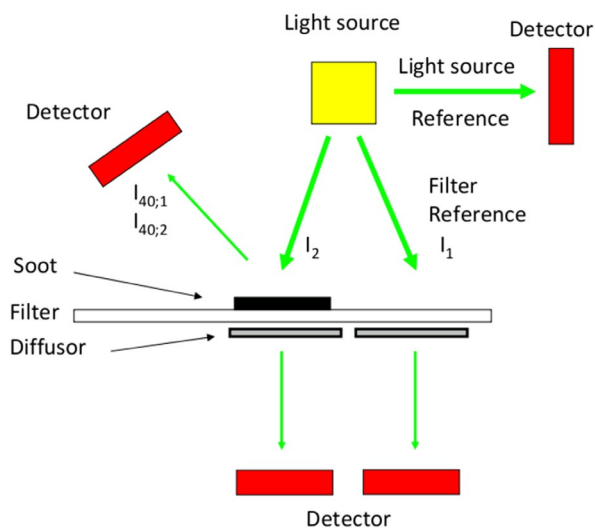


Fig. 2. The principle of the BC-photometer instrument.

2.2.2. *The online particle soot absorption photometer (MISU-PSAP).* At each of the three observatories, a PSAP (Bond et al., 1999), designed at MISU continuously measured the σ_{ap} of BC by monitoring the change in transmittance across a PCMB Nuclepore® filter, supported by a Whatman® filter paper as a diffuser, with a similar 528 nm LED as light source as in the BC-photometer (Section 2.2.1.). The MISU-PSAP was also equipped with an additional sensor recording backscattered light at 40° from the filter surface. The recorded (logged every second) transmitted light through the PCMB filter together with the light scattered by the deposited particles were extracted as minute median values and integrated over the total sampling periods of the discrete filter cassettes, 24–48 h. Subsequently, as in the case with the cassette samples, an optical correction for light scattering by the non-absorbing matter co-deposited on the filter was performed (Section 2.4). No chemical correction for the non-absorbing matter was applied.

Through laboratory tests with filtered air (free from particles) the standard deviation (1σ) of the MISU-PSAP were established to be 0.0015 in units of Od and 0.005 Mm^{-1} as σ_{ap} (in relative measures better than 25, 0.1 and 0.03% for $\sigma_{ap} > 0.02$, 5 and 15 Mm^{-1} , respectively). The limits of detection at 3σ were 0.015 Mm^{-1} . Similar to the BC photometer instrument the MISU-PSAP overall coefficient of variation was 6% (Engström and Leck, 2011).

2.3. Chemical analyses of the Nuclepore® filters using ion chromatography

After exposure, the PCMB Nuclepore® filters (ambient and blank samples) were changed in a glove box. Still inside the glove box (free from particles, sulphur dioxide and ammonia) the filter samples were extracted with 5 cm^3 deionized water ($18 \text{ M}\Omega \text{ cm}$). For sufficient extraction the filter solutions were sonicated for 60 min and subsequently analysed for major cations, anions and weak anions by chemically suppressed ion chromatography (IC, Dionex ICS-2000). The average particulate sodium (Na^+), ammonium (NH_4^+), potassium (K^+), magnesium (Mg^{2+}), calcium (Ca^{2+}), chloride (Cl^-), nitrate (NO_3^-), sulphate (SO_4^{2-}), and methane sulphonate (MSA), blank concentrations were <5%, <3%, <1%, <0.2%, <0.3%, <6%, <32%, <0.2% and 0% of the sample, respectively. Details are given in Engström and Leck (2011). Non-sea-salt (nss)- SO_4^{2-} concentrations were calculated by using sodium concentrations and seawater composition taken from Stumm and Morgan (1981).

2.4. Correction of light scattering of non-absorbing particles co-deposited on the PCMB filters

Two protocols (optical and chemical) described in Engström and Leck (2011) for correction for light scattering of the recorded transmitted light through the PCMB Nuclepore® filters were applied. The optical correction protocol was applied to both the discrete filter cassette samples with post sampling

determinations with the BC-photometer and the MISU-PSAP continuous BC recordings. The method behind this protocol made practical use of the recorded signal from both the detector measuring the transmittance and the back scattering of light derived by comparing with a Multi Angle Absorption Photometer (MAAP) (Petzold et al., 2005). The chemical correction protocol was applied only to the filter cassettes.

The chemical correction method made use of the chemical quantification of the non-absorbing material co-deposited on the filter. This was made possible by the relatively low chemical blank value of the PCMB filters used in this study compared to the otherwise so commonly used fibre glass filters (Bond et al., 1999). The chemical protocol used a mass scattering efficiency of $3.8 \text{ m}^2 \text{ g}^{-1}$ (Clarke et al., 2002) for samples sourced over the Arabian Sea and Indian Ocean. A mass scattering efficiency of $2.5 \text{ m}^2 \text{ g}^{-1}$ (Hand and Malm, 2007) was used for samples mainly sourced over the continent.

A comparison of the BC σ_{ap} (corrected) from discrete filter cassettes and the continuous recordings differed on average with up to 28% ($r^2 = 0.59$), with the discrete sampling giving the higher values.

2.5. Trajectory analysis

To evaluate possible source regions for the air sampled, three-dimensional back trajectories were calculated for the three different receptor sites used in this study, Godavari, Sinhagad and Hanimaadhoo at 1650, 1450 and 50 m asl, respectively. The trajectories were calculated backward for 10 days (expected life time of the collected particles) using the HYSPLIT2 model, version 4.8, (Draxler and Hess, 1997). More information about the GDAS (Global Data Assimilation System) data-set may be found at Air Resources Laboratory (ARL), NOAA (<http://ready.arl.noaa.gov/>), where meteorological data also can be downloaded. Back trajectories have several sources of uncertainty, which generally grows with the length of the trajectory. Most uncertain is transport in the vicinity of strong gradients, such as frontal zones while within a single air mass the trajectory calculations are likely more reliable. According to Stohl (1998), in consistency with other studies, an average position error for trajectories is 20–30% (200–300 km/day).

The HYSPLIT2 model was also used to calculate trajectory clusters. Initially every trajectory is defined to be a cluster. For the first iteration the clusters are compared two by two, and

for every combination of trajectory pairs the spatial variance is calculated. The variance is defined as the sum of the squared distances between the endpoints of the clusters component trajectories and the mean of the trajectories in that cluster. Then the sum of the spatial variance for all the clusters is calculated. The pairs of clusters combined are the combination that results in the lowest increase in total spatial variance. Then the procedure is repeated with the new-defined clusters. The operator will make the decision on the final number of clusters, based on the relative change in total spatial variance for each number of them.

2.6. Data coverage

Monthly numbers of collected and determined samples at the three observatories are given in Table 1. The geographical location of the observatories and the local facilities gave crucial conditions for the maintenance of the instruments. For example the Sinhagad observatory was located on the top of a remote mountain with no direct access by motor vehicles. When computers, samplers and/or pumps stopped functioning, there was usually a couple of days delay before the sampling could be re-established. The observatory at Hanimaadhoo is, on the other hand, well facilitated for maintenance with trained operators to solve complicated problems themselves. Nevertheless interruptions in the sampling sequence have occurred also at the Hanimaadhoo observatory. The longest stop lasted from May 2008 to February 2009. During this period no resident scientist was stationed at the observatory. In addition to the maintenance matter, the filter holder sometimes leaked with both diffuse and irregular sample spot as a result. This reduced the optimized analytical conditions in the BC determinations. Samples identified with a smeared out BC spot were removed.

3. Aerosol inorganic characteristics assigned to trajectory clusters at each of the three observatories

3.1. The Godavari observatory

At Godavari the dry winter season (December–April) is dominated by a westerly flow following the Himalaya mountain range. During the wet monsoon season (July–September) the

Table 1. Sampling frequency and yield for the three observatories.

Observatory	Sampling start	Sampling stop	Nr of days	Soot photo-meter		PSAP	
				Collected samples	Analysed samples	Collected samples	Analysed samples
Godavari	1 October 2005	31 December 2007	821	688	233	692	412
Sinhagad	1 August 2005	8 June 2008	882	342	119	302	–
Hanimaadhoo	1 June 2005	31 May 2009	1460	821	497	932	545

Note: Analysed samples are checked for correct air sampling volume and black carbon spot area.

mean wind direction from south-east to south-west is associated with marine air from the southern hemisphere Indian Ocean with large amounts of precipitation deposited over India and the surrounding regions as a result. The average annual rainfall is 2000 mm with 80% of precipitation during the monsoon season. Before reaching Godavari these air masses accumulate not only moisture over the Indian Ocean but also pollutants as they move northwards over India. The temperature on an annual basis ranges from 12 to 22 °C, with a mean of 16 °C (ICIMOD).

The trajectory cluster analysis for this site resulted in two clusters (shown in Fig. 3). They were named: 'southerly transport' and 'westerly transport'. The 'southerly transport' cluster represented air arriving at Godavari during the wet monsoon thus bringing a mixture of aerosol particles of marine and continental origin. The trajectories within the 'westerly transport' (dry winter season) cluster spent relatively long time over land (North Africa, Middle East and Indian subcontinent) prior to arrival at Godavari. The inorganic composition of the collected particulate matter for the 'westerly transport' cluster was dominated by nss-SO_4^{2-} and NH_4^+ (Fig. 4) with influences of K^+ , which all are typical fingerprints of combustions sources (fossil fuel and biomass burning). For the 'southerly transport' (wet monsoon) cluster the composition was similarly characterized by nss-SO_4^{2-} and NH_4^+ , but with a total mass concentration of $1.6 \mu\text{g m}^{-3}$, less than half of the 'westerly transport' cluster ($3.8 \mu\text{g m}^{-3}$). Relative to the samples within the 'westerly transport' cluster a slight marine influence was suggested in the 'southerly transport' cluster by the higher contribution of Cl^- and Na^+ to the inorganic mass determined. Shrestha et al. (2010) measured the chemical composition of aerosol particles during the monsoon season (May–June) of 2009 at Kathmandu University located in the neighbour valley to the Godavari observatory. They also reported SO_4^{2-} and NH_4^+ to be the major inorganic aerosol constituents collected. A long-term study from February 2005 to July 2008 at the Manora Peak (1950 m asl) in central Himalaya (Ram et al., 2010) found indications of stronger biomass burning emissions during the wintertime (December–February) supported by higher K^+ concentrations ($0.6 \mu\text{g m}^{-3}$) compared to the summer season ($0.3 \mu\text{g m}^{-3}$). A similar pattern, although lower values, is seen in the K^+ concentrations of this study with 0.2 and $0.09 \mu\text{g m}^{-3}$ for the 'westerly transport' and 'southerly transport' cluster, respectively.

3.2. The Sinhagad observatory

At the Sinhagad observatory the meteorology in general is dominated by marine sourced air and precipitation during the wet monsoon season and during the dry winter season by air influenced by land based sources (Momin et al., 2005). The main wind direction is south-westerly during the monsoon season. As a result, the meteorology is in general dominated by precipitation as the air is being advected from the Indian Ocean. During

the dry winter season the wind pattern at Sinhagad is more complex with variable directions mostly from over land. The monsoon season experiences moderate rainfall (about 180 mm per month) and temperatures ranging from 10 to 28 °C. The day-time temperature during the winter season remains near 28 °C while night temperature is below 10 °C for most of December and January.

The trajectory clustering resulted in three clusters called: 'Indian Subcontinent', 'Arabian Sea' and 'Indian Ocean' (Fig. 5). The inorganic composition of the aerosol samples that were collected at the Sinhagad observatory showed a dominance of nss-SO_4^{2-} and Ca^{2+} (Fig. 6). A noticeable contribution came also from K^+ and Mg^{2+} . Surprisingly the relative contribution of the various constituents to the inorganic mass didn't change significantly between the three trajectory clusters. However differences in total mass were observed, with the highest total particle median mass concentration determined in the samples belonging to the 'Indian Subcontinent' cluster ($9.7 \mu\text{g m}^{-3}$). Least inorganic aerosol mass was detected within the 'Indian Ocean' trajectory cluster ($0.96 \mu\text{g m}^{-3}$). A median aerosol mass concentration of $3.7 \mu\text{g m}^{-3}$ resulted for the 'Arabian sea' cluster.

The significant contribution of nss-SO_4^{2-} together with Ca^{2+} , for all trajectory clusters, pointed to a substantial nss-SO_4^{2-} contribution from soil dust (Kulshrestha et al., 1998). The significant contribution of NO_3^- for the 'Indian Ocean' trajectory cluster indicated local traffic to be another possible source of the nss-SO_4^{2-} concentrations observed. In addition, the presence of K^+ together with NO_3^- and nss-SO_4^{2-} suggested both a local and a long-range transport of combustion-derived aerosol from biomass burning (Andreae, 1983, Fabian et al., 2009).

3.3. Hanimaadhoo observatory

Hanimaadhoo experiences a distinct seasonal cycle of the atmospheric circulation with south-westerly winds and precipitation (monthly average rainfall is 200 mm) during the wet monsoon season and a dry (monthly average rainfall as low as 50 mm) winter season with north-easterly winds. The monthly average air temperature is around 28 °C in the monsoon season and peaks at 30 °C during the dry season (MDM, Maldives Department of Meteorology, 2010).

The samples collected at the Hanimaadhoo observatory contained the most complete data series covering close to four full winter seasons and three monsoon seasons. Three major source regions, shown in Fig. 7, resulted from the trajectory cluster analysis. They are referred to as: 'Indian Subcontinent' and 'Arabian Sea' in being representative of the dry winter season and 'Indian Ocean' representing the wet monsoon season. The chemical composition determined on the samples collected at Hanimaadhoo was similar for the 'Indian Subcontinent' and the 'Arabian sea' clusters (Fig. 8). The dominating constituents were nss-SO_4^{2-} and NH_4^+ . Similar to the observations at Godavari, significant contribution also came from K^+ as an

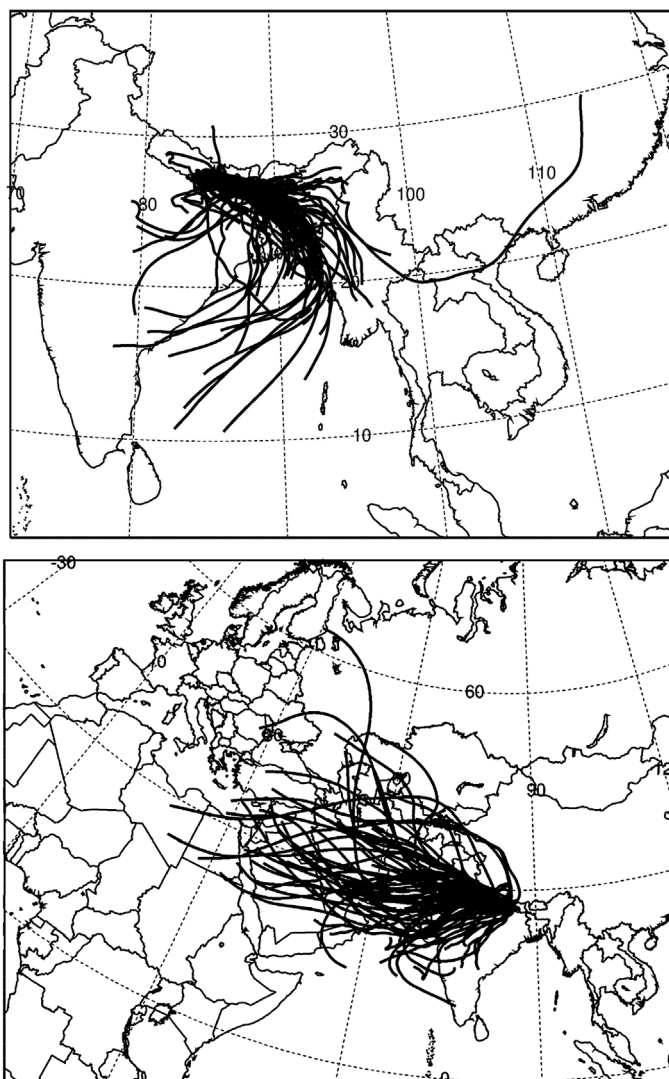


Fig. 3. Five days trajectory clusters arriving at Godavari. Only days with quality insured samples are included. The panels show trajectories in the clusters 'Southerly Transport' (upper, 53%), 'Westerly Transport' (lower, 47%). In total 237 trajectories were clustered. Latitude and longitude notations are inside the panels.

indicator of combustion sources from fossil fuel and biomass burning. The continental trajectory clusters also showed a minor contribution to total mass from Mg^{2+} and Ca^{2+} , being most elevated for the 'Arabian Sea' cluster. This was possibly due to soil dust, shown to be a large component for the samples collected at the Sinhagad observatory. The Cl^-/Na^+ molar ratio in the 'Arabian Sea' cluster was found to be lower than 1.2, which is the molar ratio of pure seawater sodium chloride (NaCl) particles introduced into the atmosphere. However, once airborne displacement of chlorine could be acquired through aqueous oxidation of sulphur oxide or condensation of sulphuric acid to form nss-SO_4^{2-} (Keene et al., 1998).

The 'Indian Subcontinent' trajectory cluster had the highest total median mass concentration of the Hanimaadhoo clusters,

$11 \mu\text{g m}^{-3}$, while it was slightly lower for the 'Arabian sea', $9.0 \mu\text{g m}^{-3}$. This was probably due to stronger aerosol sources in the eastern part of the Indian Subcontinent. During the wet monsoon season the 'Indian Ocean' cluster showed that the air had been advected over the South Indian Ocean for more than 10 days prior to sampling at the observatory (Fig. 7). The chemical composition of the 'Indian Ocean' cluster turned out markedly different than the 'Indian Subcontinent' and 'Arabian Sea' clusters, with a strong contribution from Na^+ and Cl^- . However, the molar ratio of Cl^-/Na^+ for these marine influenced clusters (0.35 for the 'Indian Ocean' cluster and 0.17 for the 'Arabian Sea' cluster) were clearly lower than 1.2. We attribute the displacement of chlorine to be acquired through the presence of sulphuric acid or nss-SO_4^{2-} . This result was in agreement with

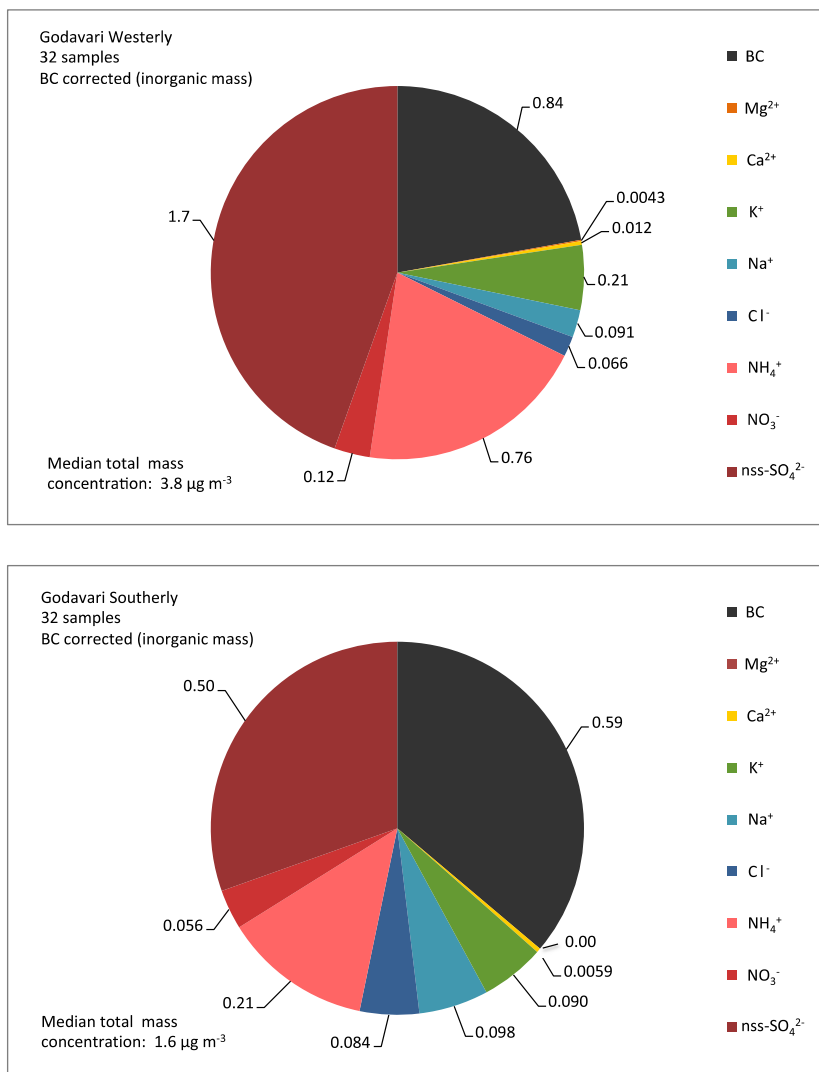


Fig. 4. Chemical composition of aerosol particles collected at Godavari for the Westerly Cluster (upper) and the Southerly cluster (lower).

Norman et al. (2002) that found a strong contribution of nss-SO_4^{2-} from the marine biogenic source of dimethyl sulphide in air sourced over the Indian Ocean. The relative contribution of Ca^{2+} and Mg^{2+} to total median aerosol mass ($1.6 \mu\text{g m}^{-3}$) was higher compared to the 'Arabian Sea' and 'Indian Subcontinent' clusters. This argues for a possible contribution of nss-Ca^{2+} from marine polymer gels (polysaccharides inter bridged with divalent ions preferable Ca^{2+}) shown to be an important component of the tropical marine aerosol (Bigg and Leck, 2008; Leck and Bigg, 2008; Granat et al., 2009).

Common for the three observatories was the presence of indicators of combustion sources in the inorganic chemical composition, such as nss-SO_4^{2-} , NH_4^+ and K^+ . For some of the clusters, especially for Sinhgad, clear contribution from Ca^{2+} indicated influence of soil dust, while marine influenced clusters showed relatively larger mass contributions from Na^+ and

Cl^- . The total median mass concentrations for the continental influenced clusters ranged from 1.6 to $11 \mu\text{g m}^{-3}$. The marine influenced clusters in general showed lower total median mass concentrations ranging between 0.96 and $1.6 \mu\text{g m}^{-3}$.

4. Seasonal variability of the BC in air at the three observatories

4.1. Data selection

The optical protocol for correction of light scattering by the non-absorbing inorganic material was applied both to the PSAP-MISU sampler (Table 2) and to the discrete filter cassette samples (Table 3). The chemical correction protocol was used for the latter only (Table 3) and represents a subpopulation of the former. In order to get the statistically best representation

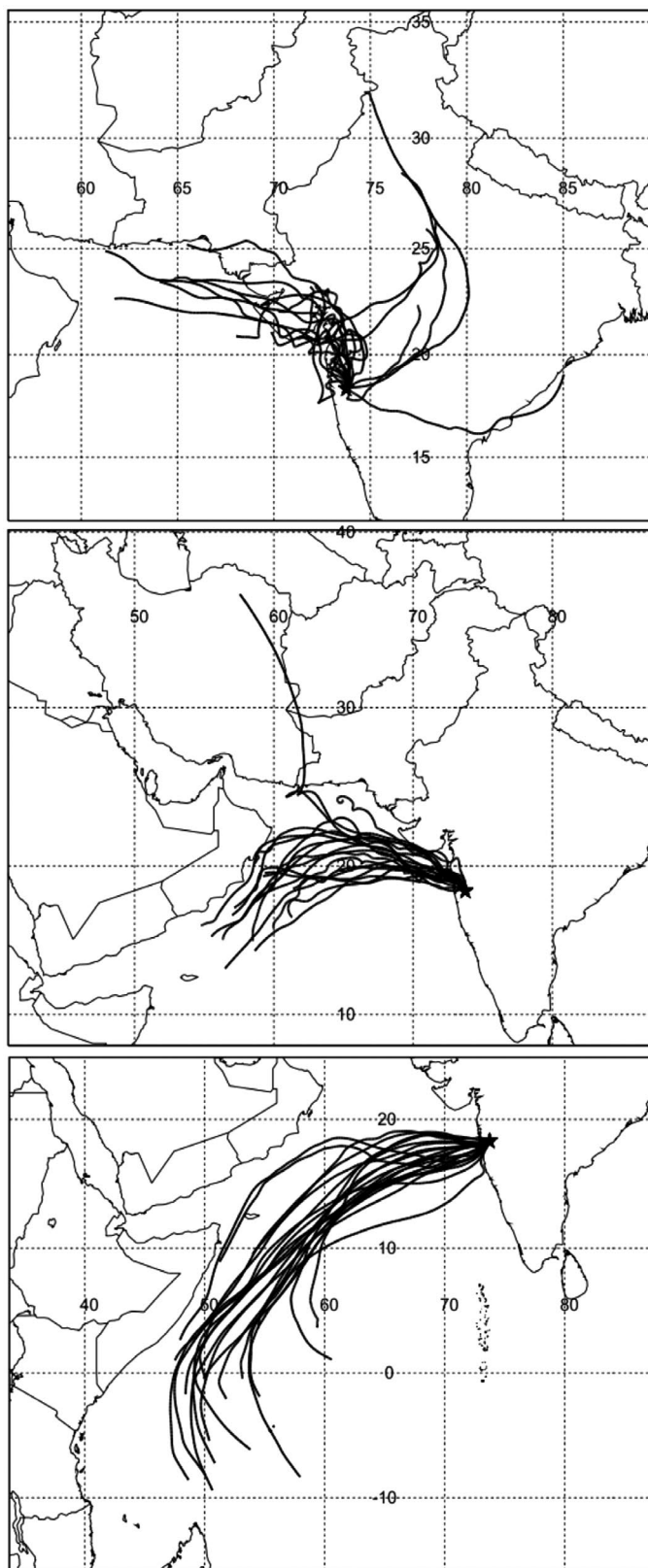


Fig. 5. Five days trajectory clusters arriving at Sinhgad. Only days with quality insured samples are included. The panels show trajectories in the clusters 'Indian Subcontinent' (upper, 32%), 'Arabian Sea' (middle, 32%) and 'Indian Ocean' (lower, 36%). In total 57 trajectories were clustered. Latitude and longitude notations are inside the panels.

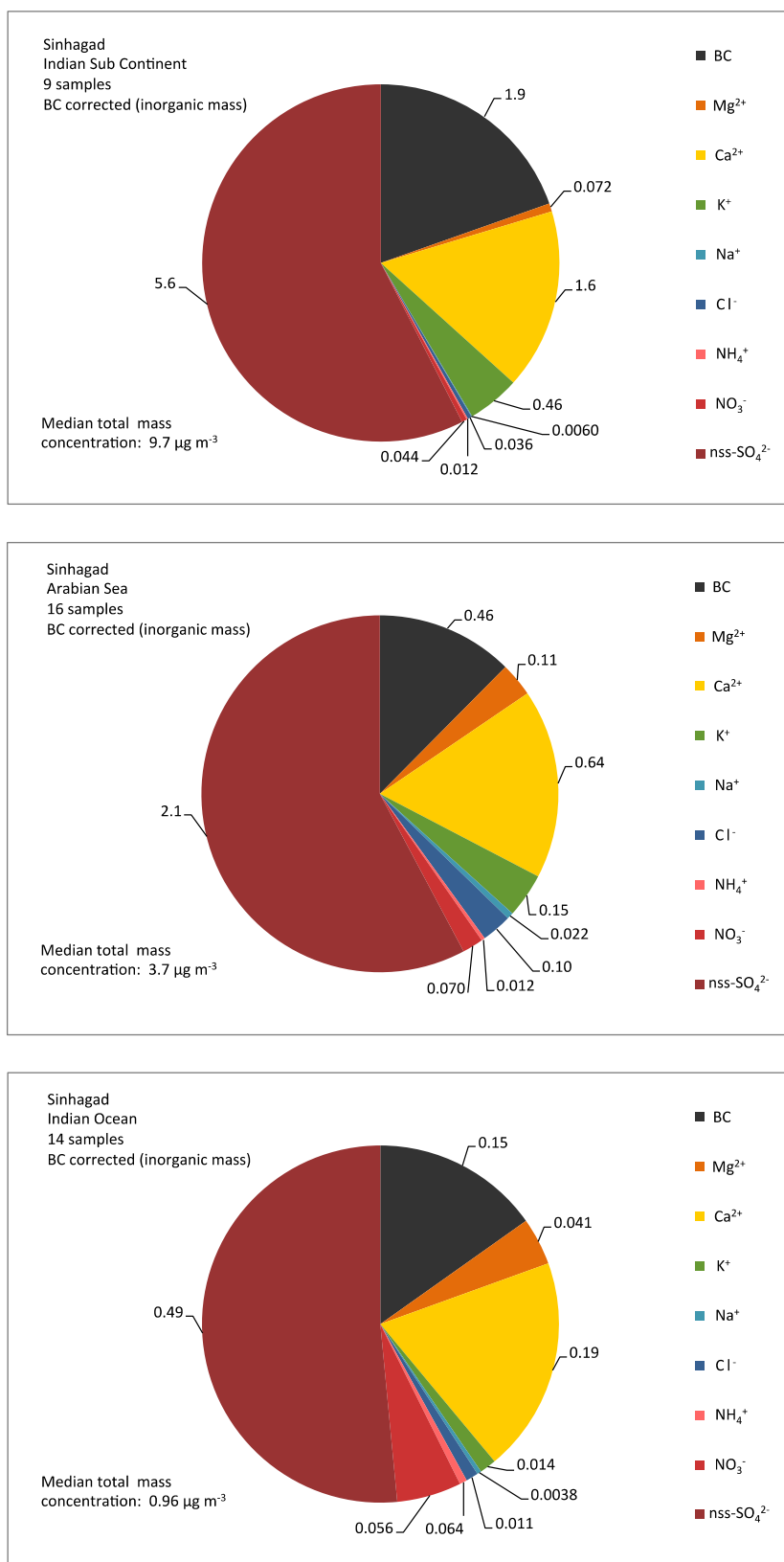


Fig. 6. Chemical composition of aerosol particles collected at Sinhagad for the Indian Sub Continent cluster (upper), Arabian Sea (middle) and Indian Ocean (lower).

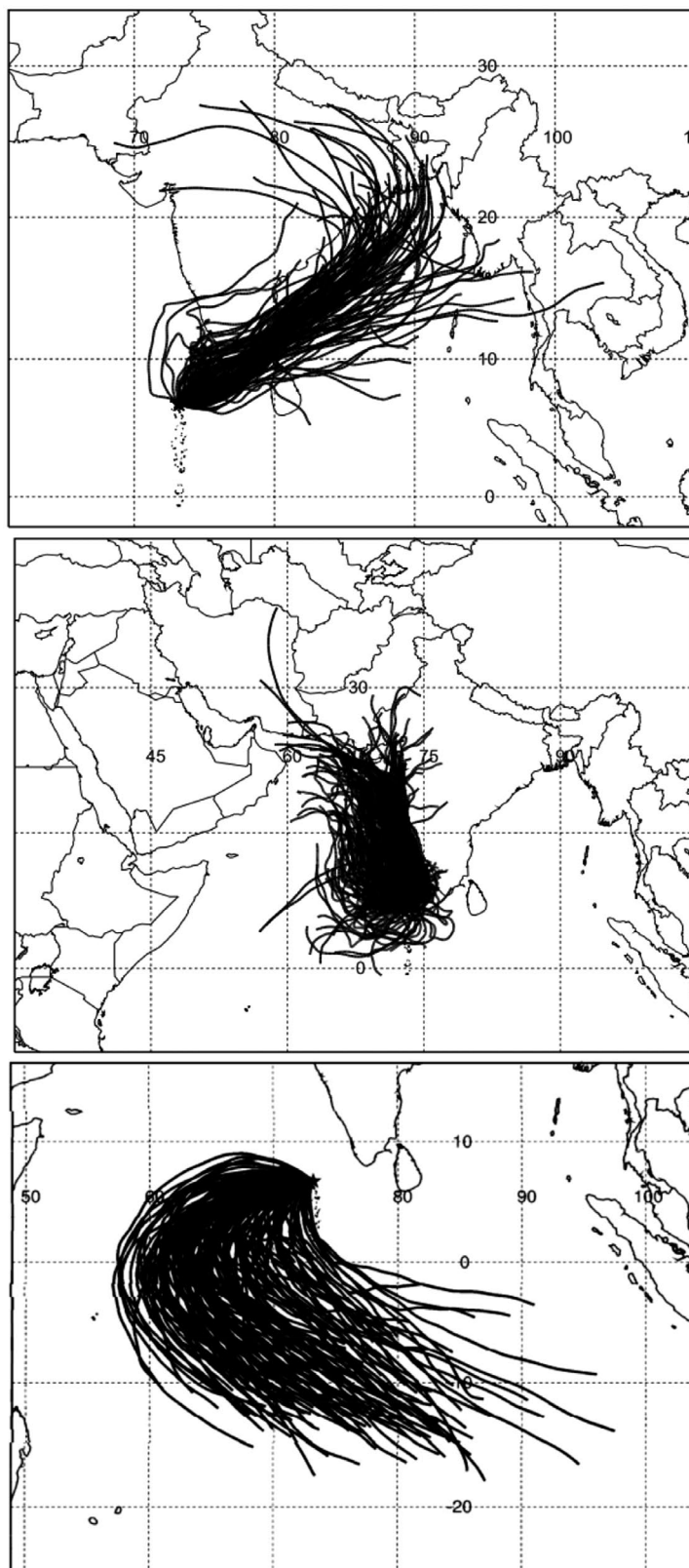


Fig. 7. Five days trajectory clusters arriving at Hanimaadhoo. Only days with quality insured samples are included. The panels show trajectories in the clusters 'Indian Subcontinent' (upper, 19%), 'Arabian Sea' (middle, 42%), 'Indian Ocean' (lower, 39%). In total 623 trajectories were clustered. Latitude and longitude notations are inside the panels.

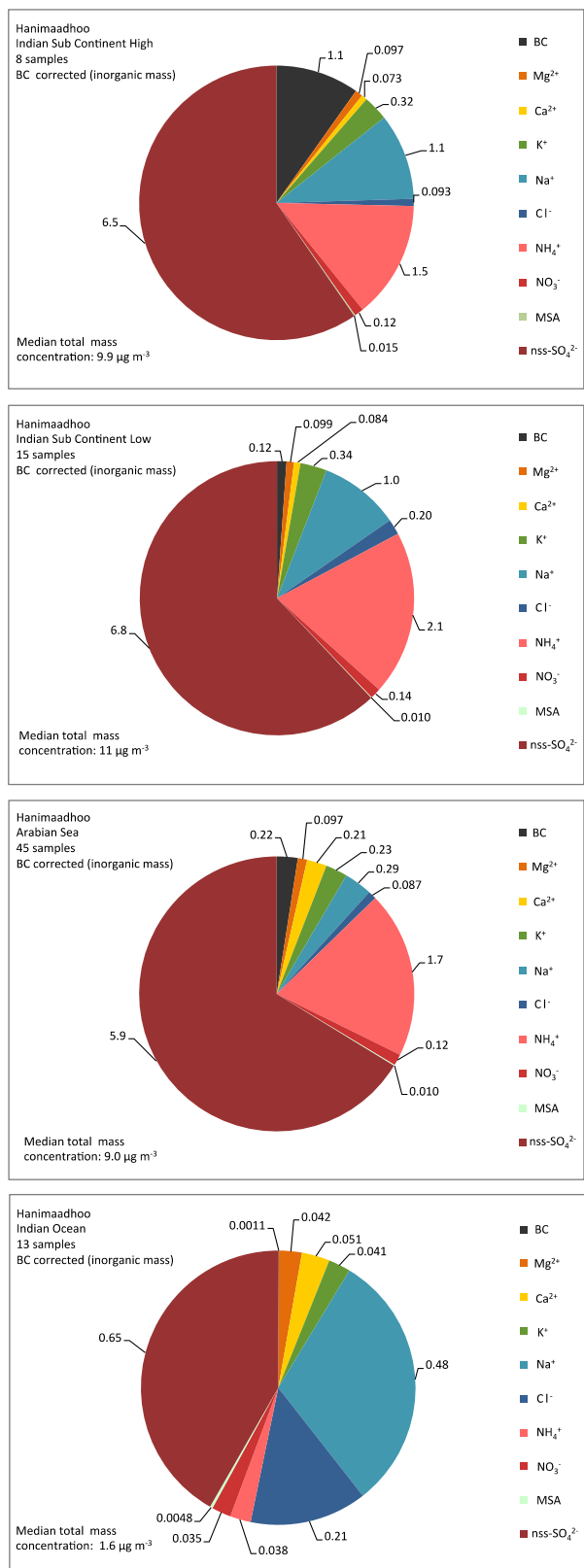


Fig. 8. Chemical composition of aerosol particles collected at Hanimaadhoo for the Indian Sub Continent high cluster (top), the Indian Sub Continent low cluster (upper middle), Arabian Sea (lower middle) and Indian Ocean (bottom).

of the observations, the BC σ_{ap} values discussed in the sections to follow used the optically corrected continuous PSAP-MISU recordings (Section 4.2–4.4). In the discussion in Section 4.5 of the BC contribution to aerosol total mass concentration the chemically corrected BC values were used. Data was evaluated for the period from 1 June 2005 to 31 May 2009 (Table 1 and Fig. 9).

4.2. Godavari observatory

The over two years monitoring of BC σ_{ap} values at Godavari ranged from 1 to 10 Mm^{-1} but did not show any distinct seasonal pattern (Fig. 9, upper panel). The median σ_{ap} of BC for the ‘southerly transport’ and ‘westerly transport’ clusters were $2.5 \pm 0.2 \text{ Mm}^{-1}$ and $4.1 \pm 2.2 \text{ Mm}^{-1}$, respectively (at 95% confidence interval) (Table 2 and Fig. 10). The trajectories belonging to the latter cluster spent relatively long time over North Africa and Middle East and Indian Subcontinent prior to arrival at Godavari and represent long-range transport of pollution. The relatively lower median σ_{ap} value of the ‘southerly transport’ cluster confirms the suggestion in Section 3.1 of a cause and effect relationship to the length of time the air spent over the continent influenced by combustion sources before arriving at the Godavari observatory. This result is also an indication of that BC is not efficiently dry or wet deposited during 5–10 days of transport.

In summary, the trajectory analyses suggested that air sampled at the Godavari observatory was influenced mainly by combustion sources all year around, while the about 1.5 times stronger BC signal during the dry winter season could have resulted from a longer path over areas with frequent combustion and weak deposition processes.

4.3. The Sinhadad observatory

Although being relatively few in number the data available exhibited, when comparing with the measurements at the Godavari observatory, a much more distinct temporal variability in the BC σ_{ap} , with peak values during the winter months and up to one order of magnitude lower values during summer (Fig. 9, center).

Both the ‘Indian Subcontinent’ and ‘Arabian Sea’ clusters were dominated by the dry winter season. However, the ‘Arabian Sea’ cluster was to a minor extent also influenced by the wet monsoon during the month of September. The ‘Indian Ocean’ cluster represented the wet monsoon season with a median BC σ_{ap} of $0.7 \pm 0.3 \text{ Mm}^{-1}$ (Table 2 and Fig. 11). About a factor of 3 higher values ($2.5 \pm 0.9 \text{ Mm}^{-1}$, Fig. 10) was obtained for the ‘Arabian Sea’ cluster. Neither the continental sources over land areas north and/or west of the Arabian Sea nor from the Bombay city up wind the observatory seemed to have had a significant influence on the observed BC σ_{ap} when comparing with the 3–11 times higher median levels ($8.0 \pm 3.3 \text{ Mm}^{-1}$) observed for the ‘Indian Subcontinent’ cluster. This was however not totally surprising since both former clusters were sourced

over the ocean for at least four days before arriving to the Sinhadag observatory.

In summary, the readily distinguishable seasonal variability in light absorbing matter observed in air at the Sinhadag observatory was suggested to be causally related to the Indian monsoon circulation in combination with the location of both a local (traffic) and a long-range transport of combustion-derived aerosol from biomass burning as suggested by the inorganic aerosol constituents.

4.4. Hanimaadhoo observatory

The distinct temporal variability in the σ_{ap} of BC observed at the Sinhadag observatory, was at the Hanimaadhoo observatory further amplified with between one to two orders of magnitude

Table 2. The light absorption coefficient of black carbon (Mm^{-1}) in air at the three observatories during 1 June 2005 to 30 May 2009 from optically corrected MISU-PSAP values.

Observatory	Godavari		Sinhadag			Hanimaadhoo		
Trajectory cluster ^a	W	S	ISC	AS	IO	ISC	AS	IO
Nr of samples	41	70	11	19	20	82	229	174
Average	4.7	2.6	8.3	3.2	0.7	5.8	3.4	0.8
Median	4.1	2.5	8.0	2.5	0.7	6.0	2.9	0.6
Percentile 90	8.0	3.8	14.8	5.2	1.3	9.7	6.1	1.9
Percentile 75	5.6	3.1	10.1	3.4	1.1	7.9	4.4	1.1
Percentile 25	3.3	2.1	5.2	2.2	0.4	3.5	2.1	0.3
Percentile 10	2.6	1.5	3.1	1.2	-0.2	1.6	1.4	0.2

^aWesterly = W, Southerly = S, Indian Subcontinent = ISC, Arabian Sea = AS, Indian Ocean = IO.

difference between the winter and the monsoon seasons (Fig. 9, bottom).

The ‘Indian Subcontinent’ cluster represented air sampled at the observatory with not only the highest median σ_{ap} levels of BC ($6.0 \pm 0.6 Mm^{-1}$) but also with the largest spread (Table 2 and Fig. 10). On average 53% lower BC σ_{ap} levels ($2.9 \pm 0.2 Mm^{-1}$) resulted for the ‘Arabian Sea’ trajectory cluster. This cluster contained marine samples influenced by combustions sources from the Indian Subcontinent as a result of that the air was transported southwards over the Arabian Sea before reaching the Hanimaadhoo observatory. The chemical signature of the ‘Indian Ocean’ cluster suggested that the air being sampled during the monsoon season was not influenced by continental emission sources. As a result this cluster had the lowest BC σ_{ap} ($0.6 \pm 0.1 Mm^{-1}$) compared to the other two clusters at Hanimaadhoo (Fig. 11). The BC σ_{ap} value was also very close to the BC σ_{ap} value reported in Section 4.3 for the ‘Indian Ocean’ cluster at Sinhadag, which indicates that the two air masses had similar source regions. The trajectory clusters shown in Figs. 5 and 7 verified this.

After a closer look at the trajectories in the ‘Indian Subcontinent’ cluster, with shown peak BC σ_{ap} values, they all passed over the Indo-Gangetic plain area, which is one of the most populated areas on Earth (900 million people). We therefore attribute the BC σ_{ap} peak values observed at Hanimaadhoo observatory to strong man-made combustion sources in the Indo-Gangetic plain area. The relatively lower median BC σ_{ap} levels within this cluster were associated with either advection from less polluted areas or relatively longer transport from the areas influenced by combustion sources before arriving at the observatory. The BC σ_{ap} levels for the ‘Indian Ocean’ cluster were 4 to 8 times lower compared to the ‘Arabian Sea’ and ‘Indian Subcontinent’ clusters

Table 3. Quality insured filter cassette samples (Section 2.4.1) collected in air at the three observatories during 1 June 2005 to 30 May 2009. Data shown are the total inorganic mass analysed, the mass ratio of BC equivalent to total inorganic analysed mass, the optically and chemically corrected light absorption coefficient of BC (σ_{ap}) and the corresponding reduction of the uncorrected σ_{ap} ($\sigma_{apuncorr}$). Also shown are values of the scattering coefficient (σ_{sp}) calculated for the samples.

Observatory	Godavari		Sinhadag			Hanimaadhoo			
Trajectory cluster ^a	W	S	ISC	AS	IO	ISC High	ISC Low	AS	IO
Inorganic mass ($\mu g m^{-3}$)	3.0	1.0	7.8	3.2	0.8	9.9	10.9	8.8	1.6
σ_{sp} (Mm^{-1})	8.0	2.9	19.4	8.1	2.2	28.1	25.3	33.5	7.2
$\sigma_{apuncorr}$ (Mm^{-1})	19.1	9.0	22.5	6.7	2.0	18.3	5.0	11.2	0.8
<i>Optical corrected</i>									
σ_{apcorr} (Mm^{-1})	5.6	2.9	8.5	2.6	0.8	7.0	1.9	4.2	0.3
Reduction of $\sigma_{apuncorr}$ (%)	71	68	62	62	62	62	62	63	62
BC eq./Inorganic mass (%)	19	29	11	8	10	7	2	5	2
<i>Chemical corrected</i>									
σ_{apcorr} (Mm^{-1})	8.4	5.9	7.1	4.2	1.6	10.8	1.2	2.2	0.0*
Reduction of $\sigma_{apuncorr}$ (%)	56	34	68	38	19	41	76	81	100
BC eq./Inorganic mass (%)	28	59	9	13	20	11	1	3	0

Note: The calculations were based on a subpopulation of the samples reported in Table 2, but analysed on the BC photometer. The Indian Sub Continent cluster were split in high and low BC content for the chemically BC correction. All values represent median values (50% percentile).

^aSoutherly = S, Westerly = W, Arabian Sea = AS, Indian Ocean = IO, Indian SubContinent = ISC.

*Not different from zero at the 25 percentile level.

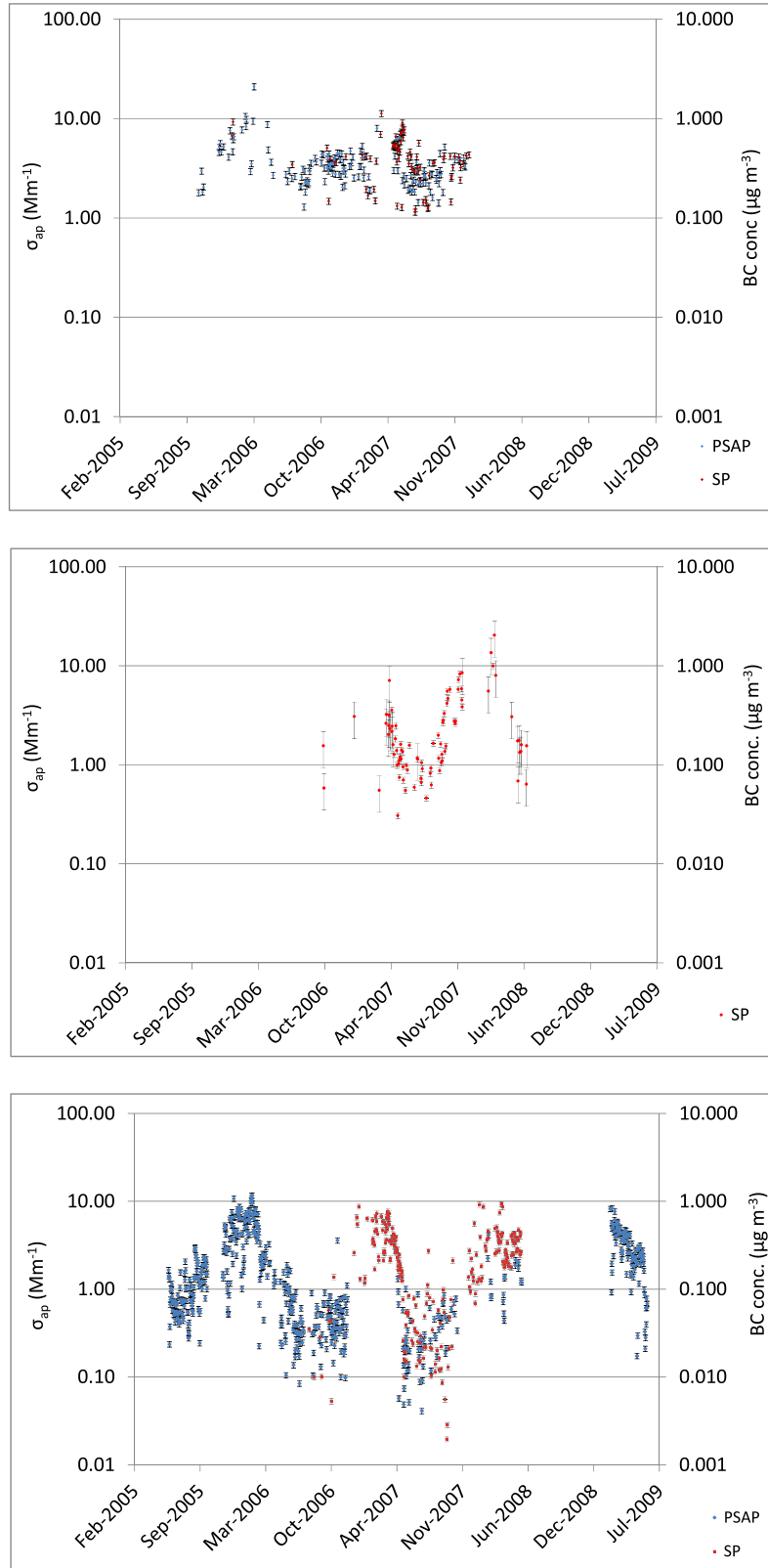


Fig. 9. The observed light absorption coefficient (σ_{ap}) at 528 nm of BC (black carbon) and corresponding equivalent BC mass concentration in air at Godavari, Nepal (upper), Sinhagad, India (middle) and Hanimaadhoo, Maldives (lower). Blue markers indicate PSAP and red soot photometer (SP). Error bars show the coefficient of variation. The larger variation for the Sinhagad data is due to uncertainties in sampling volume.

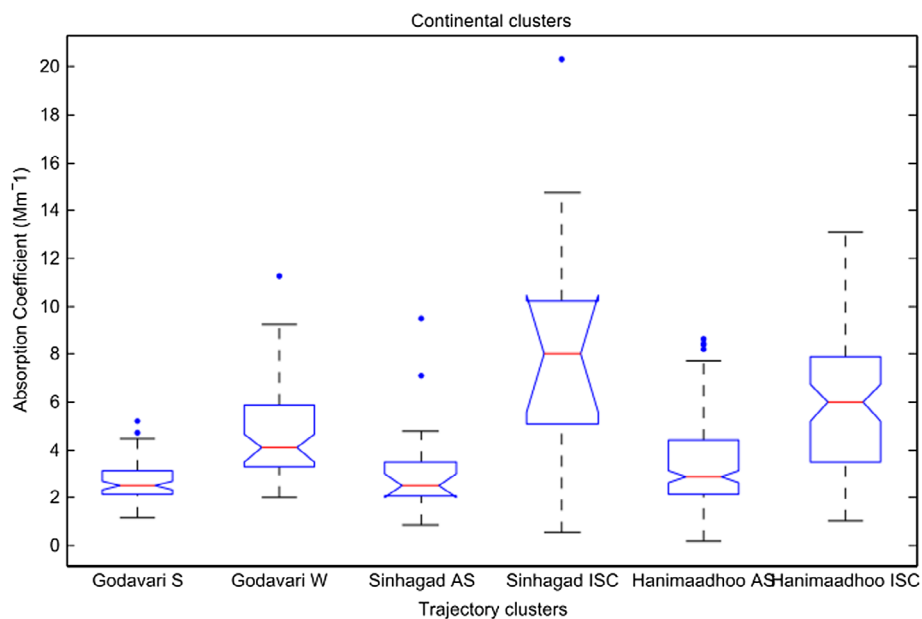


Fig. 10. Median light absorption coefficients of BC (red lines) for continentally influenced clusters at Godavari, Sinhagad and Hanimaadhoo, associated with the clusters shown in Figs. 3–5. Boxes indicate 75 and 25 percentile values. Bars have the length of the inter-quartile range times 1.5. Blue dots are values outside the inter-quartile range. If the notches around the median values do not overlap, the true median values do differ with 95% confidence. Southerly = S, Westerly = W, Arabian Sea = AS, Indian Subcontinent = ISC.

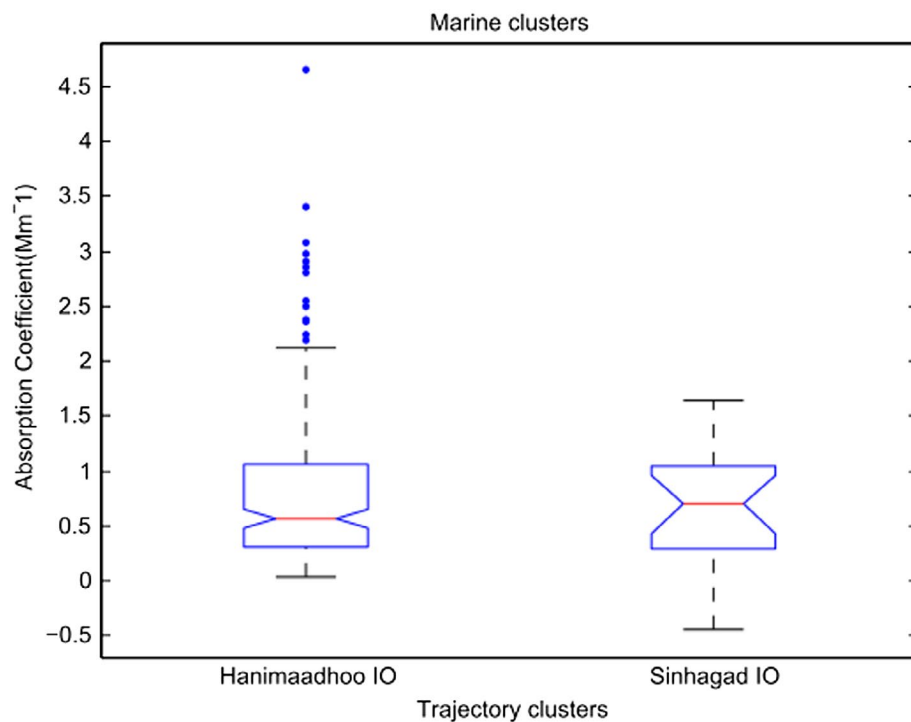


Fig. 11. Median light absorption coefficients of BC (red lines) for marine influenced clusters at Sinhagad and Hanimaadhoo associated with clusters shown in Figs. 3–5. Boxes indicate 75 and 25 percentile values. Bars have the length of the inter-quartile range times 1.5. Blue dots are values outside the inter-quartile range. If the notches around the median values do not overlap, the true median values do differ with 95% confidence. Indian Ocean = IO.

(Table 2). Consistent with these very low median BC σ_{ap} levels 0.6 Mm^{-1} and with more than 10-days advection over the ocean, the samples collected at the observatory were expected to be dominated by non-absorbing particulate matter such as biogenic sulphur (Norman et al., 2002), sea salt and marine polymer gels (Leck and Bigg, 2008; Bigg and Leck, 2008), as discussed in Section 3.3.

In summary, we observed a strong seasonal variability in the light absorbing particulate matter in air at the Hanimaadhoo receptor observatory, with peak values during the winter season associated with urban Indian combustion sources from fossil fuel and biomass burning suggested by the aerosol inorganic chemical composition. During the monsoon season 4 to 8 times lower median BC σ_{ap} levels were observed. In this case the collected aerosol was dominated by remote marine non-light absorbing particulate matter.

4.5. Correction of the BC light absorption coefficient by light scattering of the non-absorbing matter

Also the chemical correction of the BC σ_{ap} for the light scattering of inorganic material co-deposited on the PCMB Nuclepore® filters (Table 3) was evaluated. For this study only air sampled with the filter cassettes could be used due to the need to avoid chemical contaminations of the samples.

After the chemical protocol was applied the median BC σ_{ap} was lowered by on average 57%, ranging from 19 to 100%, both seasons included (Table 3). The most efficient reduction resulted for samples collected in air at the receptor observatory at Hanimaadhoo. The samples collected in air at the Godavari observatory were the least effected by non-absorbing particles co-deposited on the filters. The significant higher relative mass proportion of BC determined for these samples probably served as an explanation.

As a result of the large spread in BC σ_{ap} values (Section 4.4, Table 2) of the ‘Indian Subcontinent’ cluster at the Hanimaadhoo observatory they were divided further into two clusters (Table 3 and Fig. 8), ‘high BC’ (7.0 Mm^{-1}) and ‘low BC’ (1.9 Mm^{-1}). This close to four times difference in median BC σ_{ap} was not in proportion to the total median aerosol mass concentration observed: $9.9 \mu\text{g m}^{-3}$ for the ‘high BC’ sub-cluster and $10.9 \mu\text{g m}^{-3}$ for the ‘low BC’ sub-cluster. The BC σ_{ap} levels for the ‘Arabian Sea’ and ‘Indian Subcontinent’ clusters were in the same range as the BC σ_{ap} values observed at the Godavari observatory all year round and at the Sinhgad observatory during the winter season when the trajectories passed over the Indian Subcontinent. For the ‘Indian Ocean’ cluster at Hanimaadhoo, median BC σ_{ap} showed to be very low, mostly not significant different from zero, and constituted not more than 0.6% of the total analysed inorganic aerosol mass. This result was in agreement with the reported light absorption coefficients of BC determined by Engström and Leck (2011) at the same location and season.

When in general comparing the correction of light scattering on the recorded BC σ_{ap} values by the optical and chemical

protocols applied to the filter cassettes, the chemical correction was generally weaker than the optical correction (Table 3). The optically corrected BC σ_{ap} values from the MISU-PSAP (Table 2) and the optically corrected BC σ_{ap} values from the BC-photometer (Table 3) resulted in similar values for each of the corresponding trajectory clusters at the three observatories. Some minor difference can be seen and can be explained by the filter cassettes in Table 3 being a subpopulation of the samples in Table 2.

5. Comparing calculated mass concentrations of BC with other studies in South-Asia

5.1. Godavari

Table 4 gives a comparison of the equivalent BC (eBC) median mass concentrations derived in this study with other studies performed over the Indian subcontinent and the Indian Ocean. The eBC mass concentrations were derived from the chemically corrected BC σ_{ap} values listed in Table 4 using a mass absorption efficiency of $10 \text{ m}^2 \text{ g}^{-1}$ (Heintzenberg, 1982). The eBC mass concentration ($0.84 \mu\text{g m}^{-3}$) was larger in the ‘westerly transport’ cluster although the mass ratio was lower (0.28) due to the higher total mass concentration (Tables 3 and 4). The eBC mass concentration for the ‘southerly transport’ cluster was $0.59 \mu\text{g m}^{-3}$, which resulted in a eBC mass ratio of 0.59. The mass ratio of eBC was in agreement with the 12 to 56 per cent measured in central Kathmandu by Sharma et al. (2012). On the other hand, Sharma et al. reported a monthly mean of BC concentration from $3.0 \mu\text{g m}^{-3}$ in July to $14.9 \mu\text{g m}^{-3}$ in January. The one order of magnitude difference in BC concentrations of the studies is most possibly a consequence of that Sharma et al. performed their sampling in central Kathmandu with emission sources all around and not at Godavari located some 10 km south-east of the city. The sampling period of May 2009 to April 2010 also did not overlap with the present study, which could have included different meteorological conditions in between the years. Closer to this study’s eBC concentration, between 0.84 and $0.59 \mu\text{g m}^{-3}$, was the average elemental carbon (EC) concentration of $1.69 \pm 0.62 \mu\text{g m}^{-3}$ observed at the Kathmandu University by Shrestha et al. (2010) for the monsoon season of 2009.

During a study by Stone et al. (2009) aerosol particles were sampled at the very same site as the present study. The yearly average EC concentration measured was $1.0 \pm 0.8 \mu\text{g m}^{-3}$ and well in the range of this study. Also in the similar range was Ram et al. (2010), reporting an average EC concentration of $0.5 \mu\text{g m}^{-3}$ for the monsoon and $1.8 \mu\text{g m}^{-3}$ for the winter season at Manora Peak. But the average σ_{ap} from the study by Ram et al. (2010) was three times higher compared to this study (see Table 2) with 17 Mm^{-1} for the winter season and 7.4 Mm^{-1} for the monsoon. The dominating emission sources identified were

Table 4. BC (black carbon) mass concentrations ($\mu\text{g m}^{-3}$) in air measured with optical techniques at locations over the Indian subcontinent and the Indian Ocean.

Reference	Sampling site	Description	Method	BC concentration ($\mu\text{g m}^{-3}$) wet monsoon	BC concentration ($\mu\text{g m}^{-3}$) dry season
Corrigan et al. (2006)	Hanimaadhoo	Remote island	Aethalometer	0.087	1.17
Quinn et al. (2002)	Indian Ocean	Ship	PSAP	<LOD** (0.034)	0.5
This study*	Hanimaadhoo	Remote island	PSAP/Soot photometer	0.03	0.70
Budhavant et al. (2015)	Hanimaadhoo	Remote island	Mass spectrometer	0.06–0.40	0.07–10.8
Budhavant et al. (2015)	Sinhagad	Remote island	Mass spectrometer	0.03–0.54	0.12–8.24
Beegum et al. (2009)	Trivandrum	Urban	Aethalometer		1.8–5.7
Beegum et al. (2009)	Minicoy	Remote island	Aethalometer		0.065–0.47
Beegum et al. (2009)	Nainital	Remote mountain	Aethalometer		0.67–1.8
This study*	Sinhagad	Urban mountain	PSAP/Soot photometer	0.08	0.85
Safai et al. (2007)	Pune	Urban	Aethalometer	1.31	7.38
Badarinath et al. (2009)	Hyderabad	Urban	Aethalometer	12	
Badarinath et al. (2009)	Anantapur	Rural	Aethalometer	0.85	
Das et al. (2009)	Bhubaneswar	Suburban	Aethalometer	3.49	3.76
Shrestha et al. (2010)	Kathmandu University	Suburban	OC/EC analyzer	1.69	
Sharma et al. (2012)	Kathmandu	Urban	Aethalometer	3.0	14.9
Ram et al. (2010)	Manora Peak	Remote mountain	OC/EC analyzer	0.5	1.8
Stone et al. (2009)	Godavari	Suburban	OC/EC analyzer	0.54	1.19
This study*	Godavari	Suburban	PSAP/Soot photometer	0.29	0.56

*BC mass concentration was calculated applying a mass absorption efficiency of $10 \text{ m}^2 \text{ g}^{-1}$ (Heintzenberg, 1982).

**Limit of detection.

local traffic, residential biofuel use, agricultural waste burning and industrial fuel combustion (such as brick kilns) (Stone et al., 2009; Sharma et al., 2012).

Ram et al. (2010) also derived values of mass absorption efficiency ranging from 4.4 to $21.2 \text{ m}^2 \text{ g}^{-1}$, with a value of 12.2 ± 2.3 for the complete sampling time. The mass absorption efficiency of $10 \text{ m}^2 \text{ g}^{-1}$ used in this study to calculate BC mass concentrations from the determined σ_{ap} is within the uncertainty of the value reported by Ram et al. (2010).

Air sampling at the Nepal Climate Observatory-Pyramid, a high-altitude research station located in the Khumbu valley at 5079 m asl also showed a clear BC annual cycles with a maximum during the pre-monsoon season and a minimum during the monsoon (Marinoni et al., 2010) similar to the seasonal pattern reported by Sharma et al. (2012). At the Godavari site the seasonal cycle was not equally profound. This can be attributed to the surrounding mountains weakening the influence of the long-range transport at the same time as the local combustion emission sources were active more or less all year around.

5.2. Sinhagad

The air samples at Sinhagad were divided into the two clusters of ‘Arabian Sea’ and ‘Indian Subcontinent’. For the ‘Arabian Sea’ cluster, with prevailing wind direction from south-west and the Arabian Sea, the derived eBC mass concentration was $0.08 \mu\text{g m}^{-3}$ (Table 4). The ‘Indian Subcontinent’ cluster influenced by continental emission sources from north-east during the dry season had the derived eBC mass concentration of $0.85 \mu\text{g m}^{-3}$, one order of magnitude higher compared to the

‘Arabian Sea’ cluster. In a similar range were the reported BC mass concentrations by Budhavant et al. (2015): 0.03 to 0.54 for the wet monsoon and 0.12 – 8.24 for the dry season. Raju et al. (2010) reported BC mass concentrations for Sinhagad of $0.89 \mu\text{g m}^{-3}$ for the pre monsoon season (April–May 2009) and $1.5 \mu\text{g m}^{-3}$ for the post monsoon season period of December 2009 to January 2010. The values reported by Raju et al. (2010) were higher compared to the present study. Reasons for the apparent differences either are that the measuring periods were not overlapping or that Raju et al. (2010) instead used an aethalometer for the BC recordings or both. Safai et al. (2007) performed BC mass concentration measurements in nearby Pune and reported $1.31 \mu\text{g m}^{-3}$ for the monsoon season and $7.38 \mu\text{g m}^{-3}$ for the dry season. Higher BC concentrations are expected for measurement within an urban city, such as Pune.

5.3. Hanimaadhoo and other

Reported BC mass concentrations for the wet monsoon with air sourced over the Indian Ocean were both the relatively lowest reported and of resembling levels. The mass concentration of eBC of $0.03 \mu\text{g m}^{-3}$ ($0.017 \text{ } 1\sigma$) observed for the wet monsoon in this study was in concert with Quinn et al. (2002), which reported BC values under the detection limit ($0.034 \mu\text{g m}^{-3}$) of the instrument. A much larger spread was reported by Budhavant et al. (2015) (0.06 to $0.40 \mu\text{g m}^{-3}$). Corrigan et al. (2006) reported three time higher values than this study ($0.087 \mu\text{g m}^{-3}$) for the wet monsoon. The corresponding values reported for the dry winter monsoon in air collected at the Hanimaadhoo receptor observatory were also in concert with other studies and in general about 10 times higher than during the monsoon

season. For the dry season the present study reported a eBC mass concentration of $0.70 \mu\text{g m}^{-3}$ ($0.29 \, 1\sigma$). In comparison other studies reported a BC mass concentration of: $0.5 \mu\text{g m}^{-3}$ (Quinn et al., 2002), $0.07\text{--}10.8 \mu\text{g m}^{-3}$ (Budhavant et al., 2015) and $1.17 \mu\text{g m}^{-3}$ (Corrigan et al., 2006). Moreover, Beegum et al. (2009) reported BC mass concentrations from the remote island Minicoy in the range of $0.065\text{--}0.47 \mu\text{g m}^{-3}$.

In comparison with the rural and marine sites the BC mass concentration levels reported from various cities over the Indian Subcontinent were in general the highest as shown in Table 4. However the readily distinguishable seasonal variability seen in air sampled over the Indian Ocean area seemed to be concealed by combustion sources all year round. The high values reported from Bhubaneswar (Das et al., 2009) are likely a result of its location within the strongly polluted Indo-Gangetic plain area. Relative to these levels the even further elevated levels reported from Hyderabad (Badarinarath et al., 2009) and Pune (Safai et al., 2007) seem to be an overestimate that has not been explained.

6. Summary

When asked to make recommendations that emissions of major sources of BC over the Indo-Asia-Pacific should be reduced scientists are in the unfortunate situation of asking for financial commitments to reduce an effect whose magnitude and ramifications they cannot accurately project. This is due to non-realistic parameterizations of the atmospheric life cycle of BC particles in climate models. *In situ* measurements of direct climate forcing parameters such as aerosol light extinction (sum of scattering and absorption) are still in their infancy and are therefore indispensable for an appropriate parameterization of aerosol particles within aerosol models.

In an attempt to improve our knowledge of the light-absorbing aerosol in the atmosphere over Southern Asia the present study has presented filter-based optical measurements of BC performed during the period from 1 June 2005 to 31 May 2009 at three observatories at Godavari (Nepal), Sinhagad (India) and Hanimaadhoo (the Maldives).

The Indian monsoon circulation with its two annual phases in combination with the location of the combustion sources and their contribution relative to other non-anthropogenic sources dominated the observed patterns of black carbon at two of the observatories: in India and the Maldives. When comparing the about 10 times elevated median BC σ_{ap} levels observed during the dry winter season (December to April) at the Hanimaadhoo receptor observatory with the previously documented (INDO-EX, Lelieveld et al., 2001) temporal extent of pollution reaching from the Indian Subcontinent out over the Indian Ocean the situation seemed not have changed since 1998–1999. At the Sinhagad observatory the only slighter higher winter median levels of BC σ_{ap} (25%) measured in air suggested that BC

had not been efficiently removed from the atmosphere within 5–10 days. The dryness of the winter season in combination with hydrophobic aerosol properties (Granat et al., 2009) reduced the chances of wet scavenging of BC. At the observatory in Nepal any seasonal variability in light absorbing particulate matter was more or less concealed by the impact of regional/local combustion sources all year round. In comparison with other studies performed over the Indian Subcontinent some of them report on comparable BC levels to this study, while several others observed up to 10 times higher levels of BC. At present there is no obvious explanation at hand but the closeness to strong local emission sources and/or instrumental differences with no protocols for correction of non-absorbing matter applied can be a part of the explanation.

The up to two orders of magnitude lower BC σ_{ap} values recorded in air that had spent more than 10-days over the Indian Ocean suggested alternative sources of the absorbing particulate matter most likely being of marine biogenic origin.

To reduce errors, in assessing the radiative forcing by BC, dominated by the optical effects of non-absorbing particles, this study emphasises the results of Engström and Leck (2011) of the necessity to in addition to monitoring the scattered light provide a chemical quantification of the inorganic fraction of the non-absorbing material.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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