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Characteristics, seasonality and sources of carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri¹, K. Kawamura¹, S. G. Aggarwal^{1,2}, and T. Swaminathan³

¹Institute of Low Temperature Science, Hokkaido University, N19 W8, Kita-ku, Sapporo 060-0819, Japan

²National Physical Laboratory (Council of Scientific and Industrial Research), Dr. K. S. Krishnan Road, New Delhi 110012, India

³Department of Chemical Engineering, Indian Institute of Technology Madras, Chennai 600036, India

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Correspondence to: K. Kawamura (kawamura@lowtem.hokudai.ac.jp)

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Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

To better characterize South and Southeast Asian aerosols, PM₁₀ samples collected from tropical Chennai, India (13.04° N; 80.17° E) were analyzed for carbonaceous and water-soluble ionic components. Concentration ranges of elemental carbon (EC) and organic carbon (OC) were 2.4–14 μg m⁻³ and 3.2–15.6 μg m⁻³ in winter samples whereas they were 1.1–2.5 μg m⁻³ and 4.1–17.6 μg m⁻³ in summer samples, respectively. Concentration of secondary organic carbon (SOC) retrieved from EC-tracer method was 4.6 ± 2.8 μg m⁻³ in winter and 4.3 ± 2.8 μg m⁻³ in summer. SO₄²⁻ (8.8 ± 2.5 μg m⁻³ and 4.1 ± 2.7 μg m⁻³ in winter and summer, respectively) was found as the most abundant ionic species (57% on average, *n* = 49), followed by NH₄⁺ (15%) > NO₃⁻ > Cl⁻ > K⁺ > Na⁺ > Ca²⁺ > MSA⁻ > Mg²⁺. The mass fractions of EC, organic matter (OM) and ionic species varied seasonally, following the air mass trajectories and corresponding source strength. Based on mass concentration ratios of selected components and relations of EC and OC to marker species, we found that biofuel/biomass burning is the major source of atmospheric aerosols in South and Southeast Asia. The high concentrations of SOC and WSOC/OC ratios (ave. 0.45; *n* = 49) as well as good correlations between SOC and WSOC suggest that the secondary production of organic aerosols during long-range atmospheric transport is also significant in this region. This study provides the baseline data of carbonaceous aerosols for southern part of the Indian subcontinent.

1 Introduction

Elemental carbon (EC) and particulate organic matter (POM) contribute to 5–9% and 20–50% of the continental fine (PM_{2.5}) aerosol mass in mid-latitudes, respectively (Sillanpää et al., 2005). In tropical forested areas, POM alone contributes up to 90% (Kanakidou et al., 2005). EC is formed by fossil fuel combustion and biofuel/biomass burning (anthropogenic sources). POM, a complex mixture of numerous

ACPD

11, 3937–3976, 2011

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



organic compounds, is directly emitted from anthropogenic sources or secondary formed through atmospheric oxidation of reactive organic gases and subsequent gas-to-particle conversion (Yu et al., 2004). In contrast, inorganic materials comprise up to 70% of the total aerosol mass emitted from both natural and anthropogenic sources (Ali-Mohamed, 1991).

The atmospheric aerosols have an impact on climate as well as hydrological cycle and human health at local, regional and global scales (Ackerman et al., 2000; Menon et al., 2002; Ramanathan et al., 2001). The impacts of aerosols largely depend on their chemical composition. Elemental carbon (EC) is a strong absorber of solar radiation and causes both positive (warming) and negative (cooling) radiative forcing at top of the atmosphere and the surface, respectively (Haywood and Ramaswamy, 1998; Jacobson, 2001). In contrast, organic carbon (OC) and sulfate (SO_4^{2-}) are highly reflective to light and thus cause cooling effect (Haywood and Ramaswamy, 1998; Kiehl and Briegleb, 1993). Further, water-soluble OC (WSOC) including dicarboxylic acids enhances the capability of aerosols as cloud condensation nuclei (CCN) (Saxena et al., 1995; Sun and Aria, 2006).

In South and Southeast Asia, pollutants are highly emitted to the atmosphere due to growing economies, particularly India (World Bank, 2000), and due to biomass burning including forest fires as well as widespread use of biofuels (dung cake, wood and agricultural waste) for cooking (Reddy and Venkataraman, 2002a, b; Tata Energy Research Institute, 1997). Conversely, heavily polluted atmospheres have been more often observed for last decades (Lelieveld et al., 2001), which cover a long-term dimming in the region that extends over the equatorial Indian Ocean (Ramanathan et al., 2001). For example, high mass loading ($\sim 6 \mu\text{g m}^{-3}$) of organics have been reported in the atmospheric aerosols over the northern Indian Ocean whereas their concentrations were negligible south of the Intertropical Convergence Zone (ITCZ) (Lelieveld et al., 2001). Further, a drastic decrease in mean clear-sky solar radiative heating has also been observed in 1998- and 1999-winter at the Indian Ocean surface (Satheesh and Ramanathan, 2000) as a consequence of high loading of aerosols in this region.

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Novakov et al. (2000) reported that fossil fuel combustion and biomass burning would contribute about 80% and 21% of carbonaceous aerosols over the tropical Indian Ocean, respectively. On the other hand, Guazzotti et al. (2003) found that the predominance of submicron, chemically mixed particles contain abundant carbon and potassium (K^+ , a tracer for biofuel/biomass burning; Andreae and Merlet, 2001; Chow et al., 1994), implying a biomass burning origin of aerosols in this region. Recently, the relative contributions of fossil fuel combustion, open biomass burning and biofuel combustion to BC in India were estimated to be 25%, 33%, and 42%, respectively, whereas those to OC were to be 13%, 43%, and 44%, respectively (Venkataraman et al., 2005). Stone et al. (2010) also estimated small contribution from diesel combustion (4%) and large contribution from biomass burning (21%) to OC in the Himalayas.

However, chemical compositions of carbonaceous aerosols have rarely been studied in South Asia (Ram et al., 2008 and references therein) and no measurements of carbonaceous components have been reported from southern part of the Indian subcontinent, although BC was studied by using optical methods (Babu and Moorthy, 2002; Babu et al., 2002; Latha and Badrinath, 2003). Further, the studies on inorganic aerosols are also sparse in southern India (Khemani et al., 1982; Mouli et al., 2006), although they have been better documented in South and Southeast Asia (Ali-Mohamed, 1991; Negi et al., 1987; Parmar et al., 2001; Sharma and Patil, 1992).

In order to clarify chemical characteristics of the atmospheric aerosols in South and Southeast Asia and specify their sources, we studied the tropical Indian aerosol (PM_{10}) samples collected from Chennai, India in winter and summer, 2007 for carbonaceous (EC and OC) and water-soluble ionic components. Here, we discuss the contribution of biofuel/biomass burning and secondary production of carbonaceous aerosols during long-range transport from distant source regions in South and Southeast Asia based on mass concentration ratios and linear relations of selected ionic species, EC, OC and organic marker species (levoglucosan) together with backward air mass trajectory analyses. The results of carbonaceous components of this study would become as baseline data for southern part of the Indian subcontinent. Water-soluble organic species

including dicarboxylic acids, ketoacids and α -dicarbonyls in the Chennai aerosol samples have been reported elsewhere (Pavuluri et al., 2010a).

2 Experimental

2.1 Aerosol sampling

5 The tropical Indian aerosol (PM₁₀) samples were collected in a mega-city, Chennai (13.04° N; 80.17° E) located on the south east coast of India (see Fig. 1) on day- (approximately 06:00–18:00 LT) and night-time (18:00–06:00 LT) basis in winter, 2007 (23 January to 6 February, $n = 29$) and summer (22–31 May, $n = 20$). The geographical features of sampling site were described elsewhere (Pavuluri et al., 2010a). Sampling was performed on the rooftop of the Mechanical Sciences building (~ 18 m above the ground level), Indian Institute of Technology Madras (IITM), Chennai using a high volume air sampler (Envirotech APM 460 DX, India) and pre-combusted (450 °C, 4 h) quartz fiber filters (Pallflex 2500QAT-UP, 20 × 25 cm). The sample filter was placed in a pre-heated glass jar with a Teflon-lined screw cap before and after sampling. After
15 sampling, the filters were stored in darkness at –20 °C prior to analysis.

2.2 Chemical analyses

2.2.1 Carbonaceous components

EC and OC were determined using OC/EC Analyzer (Sunset Laboratory Inc., USA) following Interagency Monitoring Protected Visual Environments (IMPROVE) thermal/optical evolution protocol and assuming carbonate carbon to be negligible. A filter disc (1.4 cm in diameter) was placed in a quartz boat inside the thermal desorption chamber of the analyzer and then stepwise heating was applied in a He flow at first and then after the initial ramp, He gas was switched to He/O₂ mixture (Wang et al.,
20

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

2005). The evolved CO₂ at each temperature step was measured directly by a non-dispersive infrared (NDIR) detector system and the transmittance of light (red 660 nm) through the filter punch was used for setting up OC/EC split point and thereby OC correction. The analytical errors in duplicate analysis were within 1.2% for OC and 1.7% for EC.

Water-soluble organic carbon (WSOC) was determined as described in Wang et al. (2005). Briefly, an aliquot of filter (3 discs with 2 cm in diameter) was extracted with organic free Milli-Q water under ultrasonication for 20 min. The extracts were then filtered using a syringe filter (Millex-GV, 0.22 µm, Millipore) and then WSOC was measured using TOC analyzer (Shimadzu 5000A).

The sum of EC and OC was considered as TC. The difference between OC and WSOC was considered as water-insoluble OC (WIOC).

2.2.2 Water-soluble ionic species

The water-soluble ionic species (methanesulfonate: MSA⁻ (CH₃SO₃⁻), Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺ and Ca²⁺) were measured using a part of extracts prepared for WSOC by ion chromatograph (761 Compact IC, Metrohm, Switzerland). A mixture of 1.8 mM Na₂CO₃ and 17 mM NaHCO₃ and 40 mM H₂SO₄ were used as eluent at a flow rate of 1.2 mL min⁻¹ and suppressor for anion measurement, respectively. The 4 mM H₃PO₄ was used as eluent at a flow rate of 1.0 mL min⁻¹ for cation measurement. A calibration curve was evaluated for each sequence by the analyses of a set of authentic standards. The nss-SO₄²⁻, nss-K⁺ and nss-Ca²⁺ were calculated using Na⁺ as a reference tracer for sea-salt correction.

All the concentrations of carbonaceous and ionic components reported here are corrected for field blanks.

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

2.3 Estimated organic matter (OM) and secondary organic carbon (SOC)

Atmospheric concentrations of OM are normally estimated by multiplying the measured OC concentrations with a conversion factor (1.6 ± 0.2 ; Turpin and Lim, 2001). In this work, OM was estimated by using a factor of 1.7, as higher ratios are possible for aerosols influenced by wood smoke (Turpin and Lim, 2001). In fact, Mayol-Bracero et al. (2002) used a factor of 1.7 for organic aerosols over the Indian Ocean, which were considered to be transported from the South and Southeast Asian countries.

Primary OC (POC) and SOC were estimated following the EC-tracer method (Castro et al., 1999; Turpin and Huntzicker, 1995; Yu et al., 2004). EC is a good tracer of primary combustion-generated carbon. Primary OC/EC ratio is assumed to be relatively constant for a given area, season and local meteorology because EC and POC typically have the same sources. Hence, the minimum value of OC/EC ratios can be used to estimate the amount of SOC in the atmospheric aerosol for a specific region of interest (Castro et al., 1999).

In this work, we assume the following equations,

$$[\text{POC}] = [\text{OC/EC}]_{\min} \times [\text{EC}] + c \quad (1)$$

$$[\text{SOC}] = [\text{OC}]_{\text{meas}} - [\text{POC}] \quad (2)$$

where $[\text{OC/EC}]_{\min}$ is the minimum OC/EC ratio observed during the sampling period, c is a parameter to account for non-combustion sources contributing to the POC, and $[\text{OC}]_{\text{meas}}$ is the measured OC concentration. Here, the POC contribution from non-combustion sources was assumed to be negligible.

2.4 Meteorology

According to meteorological station of IITM (sampling site), the ambient temperature and relative humidity were varied from 14.2 to 34.9°C (ave. 23°C) and 38% to 89% (ave. 68%) in winter, whereas in summer their ranges were 28.3–41°C (ave. 32°C) and

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



31%–81% (ave. 60%), respectively. No rainfall was observed during the campaigns. Due to a strong land/sea thermal gradient, a clear diurnal oscillation was observed in wind speed and wind direction (Pavuluri et al., 2010a) but such a wind contrast decreases with height and disappears above 1 km (Srinivas et al., 2006).

2.5 Backward air mass trajectories

Ten-day backward air mass trajectories that arrive in Chennai at an altitude of 500 m for every 6 h were computed using Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model of the National Oceanic and Atmospheric Administration (NOAA) (http://ready.arl.noaa.gov/HYSPLIT_traj.php). The obtained plots showed three major transport pathways with three different source regions during the campaigns (see Fig. 1). In winter (23 January to 6 February), the air masses that arrived in Chennai originated from the Middle East and/or the Indian mainland (ME/IN) in January and Southeast Asia (SEA) in February passing over the Bay of Bengal, except for few cases. Based on these trajectories, winter is further classified into two seasonal categories; early- (23–28 January) and late-winter (29 January to 6 February). However, the trajectories of 31 January (night) and 1 February fall in the category of early winter. Meanwhile, the results of 29 January (day), 30 January (night), 31 January (day), and 2 February (day) were not included in either categories and handled as such, because their air masses originated from mixed regions. In summer (22–31 May), the air masses originated from the Arabian Sea and/or the Indian Ocean (AS/IO) passing over southern part of the Indian subcontinent.

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

3 Results and discussion

3.1 Characteristics and temporal variations of carbonaceous and water-soluble ionic components

Table 1 presents the statistical summary of concentrations of carbonaceous and water-soluble ionic components in the tropical Indian aerosol (PM₁₀) samples from Chennai. Their temporal variations are shown in Figs. 2 and 3, respectively. Concentrations of EC and OC ranged from 1.1 to 14 µg m⁻³ (ave. 4.5 µg m⁻³) and 3.2 to 17.6 µg m⁻³ (ave. 9.4 µg m⁻³) (*n* = 49), respectively. EC concentrations showed higher values in winter than summer (see Table 2). But OC did not show significant difference between winter and summer (see Table 2). On the other hand, WSOC and WIOC concentration ranges were 1.7–7 µg m⁻³ (ave. 3.9 µg m⁻³) and 1.5–12.9 µg m⁻³ (ave. 5.5 µg m⁻³) (*n* = 49), respectively. On average, both WSOC and WIOC did not show significant variation between winter (3.8 ± 1.3 µg m⁻³, 5.4 ± 3.3 µg m⁻³, respectively) and summer (4.1 ± 0.9 µg m⁻³, 5.6 ± 3.3 µg m⁻³, respectively). Similarly, SOC in winter (4.6 ± 2.8 µg m⁻³) was also comparable to that in summer (4.3 ± 3.8 µg m⁻³). However, all the components, except EC in most of summer samples, showed a clear diurnal variation (see Table 1; Fig. 2; Sect. 3.4).

On average (*n* = 49), SO₄²⁻ was found as the most abundant species (6.9 µg m⁻³) followed by NH₄⁺ (1.7 µg m⁻³). They account for 57% and 15% of total ionic mass, respectively. NO₃⁻ (0.78 µg m⁻³) was found as the third most abundant species followed by Cl⁻ (0.74 µg m⁻³), K⁺ (0.61 µg m⁻³), Na⁺ (0.46 µg m⁻³), Ca²⁺ (0.37 µg m⁻³), MSA⁻ (0.22 µg m⁻³) and Mg²⁺ (0.13 µg m⁻³) (*n* = 49). The contributions of nss-SO₄²⁻, nss-K⁺ and nss-Ca²⁺ to the total SO₄²⁻, K⁺ and Ca²⁺ concentrations were on average 97.7%, 96.5% and 93% (range, 93–99.6%, 91–99.3% and 78–98.6%), respectively, demonstrating the dominant contributions from anthropogenic sources compared to marine sources. Seasonal distributions of ionic species showed peak concentrations in wintertime (Fig. 3). The average concentrations of ionic species, except NO₃⁻ and Na⁺,

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

were 2 to 5 times higher in winter than in summer (Table 1). However, NO_3^- showed a little higher average concentration in winter ($0.89 \mu\text{g m}^{-3}$) than summer ($0.61 \mu\text{g m}^{-3}$). Furthermore, most ionic species, except for MSA^- , Cl^- , NO_3^- and NH_4^+ , showed a clear diurnal variation with daytime maxima (see Table 1; Fig. 3; Sect. 3.4).

Average equivalent ratios of total cations (Na^+ , NH_4^+ , K^+ , Mg^{2+} and Ca^{2+}) to anions (Cl^- , NO_3^- and SO_4^{2-}) were 0.85 (range, 0.67–1.07) in winter and 1.21 (range, 1.01–1.58) in summer. The linear regression lines for total cations and anions are shown in Fig. 4 for day- and night-time samples in summer and winter. A significant cation deficiency was found in Fig. 4 due to the exclusion of H^+ , suggesting that the Chennai aerosols are more acidic in winter than summer. Ammonia is a dominant alkaline gas in the atmosphere, which neutralizes the acidic particles. However, $\text{NH}_4^+/\text{SO}_4^{2-}$ equivalent ratios were only 0.63 in winter and 0.81 in summer. The acidic nature, particularly in winter, and the low $\text{NH}_4^+/\text{SO}_4^{2-}$ equivalent ratios suggest that H_2SO_4 and NH_4HSO_4 were present in addition to $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and NH_4Cl in Chennai aerosols.

Higher concentrations of EC and some ionic species (e.g. SO_4^{2-}) suggest that contributions from anthropogenic sources are larger in winter than in summer. Comparable concentrations of OC between summer and winter suggest that the organic aerosols have an additional source(s) in summer. These seasonal distributions are consistent with the origins of air masses that arrived in Chennai; ME/IN and SEA in winter and AS/IO in summer. The air masses originated from ME/IN and SEA, where anthropogenic emissions including forest fires (Streets et al., 2003) are larger (Lelieveld et al., 2001), may be enriched with EC and other pollutants. In contrast, the air masses originated from AS/IO passing over southern part of the Indian subcontinent, where the burning of cow-dung (Reddy and Venkataraman, 2002b) and livestock emissions (e.g. CH_4 ; Garg et al., 2001) are significant, may be enriched with OC rather EC. Marine biota and livestock emissions are the significant sources of OC. Cow-dung combustion is known as more important source of OC than EC (Stone et al., 2010).

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

The average concentration of NH_4^+ that contributes from biomass burning as well as animal excreta (Olivier et al., 1998) was twice higher in winter ($2.1 \mu\text{g m}^{-3}$) than in summer ($1.3 \mu\text{g m}^{-3}$) whereas NO_3^- that emits from fossil fuel combustion (Olivier et al., 1998) did not show significant seasonal variation (Table 1). This suggests that bio-fuel/biomass burning might be significant sources in South and Southeast Asia rather than fossil fuel combustion. Furthermore, the average concentrations of MSA^- and Cl^- , which are generally considered to be of marine origin, were higher in winter ($0.3 \mu\text{g m}^{-3}$ and $1.1 \mu\text{g m}^{-3}$) than in summer ($0.1 \mu\text{g m}^{-3}$ and $0.2 \mu\text{g m}^{-3}$) by a factor of 3 and 5, respectively, although the air masses have not been originated from oceanic region in winter. In fact, biomass burning produce dimethyl sulfide (DMS) (Meinardi et al., 2003), a precursor for MSA^- (von Glasow and Crutzen, 2004), as well as Cl^- (Keene et al., 2006; Watson et al., 2001; Yamasoe et al., 2000).

3.2 Comparison of EC and OC in Chennai with previous studies in South Asia

Table 2 compares the concentrations of EC and OC and mass concentration ratios of OC to EC and EC to TC in Chennai aerosols together with those from different locations in South Asia including the Bay of Bengal and the Indian Ocean. In winter, the average concentration ($6.5 \mu\text{g m}^{-3}$) of EC in Chennai is higher than those reported in Hisar, Manora Peak and over the Indian Ocean but lower than that reported in Mumbai (Table 2). In summer, EC concentration in Chennai is comparable to that reported in Kathmandu valley, higher than those reported in remote Himalayas and over the Bay of Bengal and lower than that reported in Dhaka (Table 2). In contrast, the average concentrations of OC in Chennai in both winter and summer are lower than that reported in Hisar in winter and in Kathmandu valley in summer (Table 2). These comparisons to other locations are similar to the case of EC (Table 2).

The major sources of carbonaceous aerosols in Kathmandu valley, remote Himalaya and Godavari are expected to be biofuel combustion (cow-dung cake, wood and agricultural waste) and biomass burning (Carrico et al., 2003; Stone et al., 2010). Similarly,

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

the major contributions of carbonaceous aerosols in Hisar, Manora Peak and Mt. Abu as well as over the Bay of Bengal were attributed to be biomass burning (Ram et al., 2008; Rengarajan et al., 2007; Sudheer and Sarin, 2008). In contrast, fossil fuel combustion is expected to be the major source of EC and OC in mega-cities; Mumbai (Venkataraman et al., 2002), Dhaka (Salam et al., 2003) and Lahore (Smith et al., 1996). Further, emissions from fossil fuel combustion (60%–80%) and biomass burning (20%–40%) in South and Southeast Asia are considered as the sources of EC and OC over the Indian Ocean (Mayol-Bracero et al., 2002).

The comparability of EC and OC concentrations in Chennai aerosols to those reported in Khathmandu valley, remote Himalaya, Godavari, Hisar (EC only) and Manora Peak suggests a significant contribution of biofuel/biomass burning to Chennai aerosols. The lower concentrations of EC and OC than those reported in Mumbai, Dhaka and Lahore indicate that the contribution from fossil fuel combustion, which emits significantly high amounts of carbonaceous components, is minor.

3.3 Diurnal variations of carbonaceous and ionic components: land/sea breeze circulation

All carbonaceous and ionic components showed a clear diurnal distribution in both winter and summer, except for few cases (Figs. 2 and 3; Table 1). The diurnal distributions of EC and OC were similar with nighttime maxima, except for few cases, but opposite on 28, 30 and 31 May (Fig. 2a and b). WSOC and WIOC showed day- and night-time maxima, respectively, in both winter and summer (Fig. 2c and d). Interestingly, SOC also showed nighttime maxima in opposite to that of WSOC (Fig. 2e). Concentrations of MSA^- , SO_4^{2-} , K^+ , Mg^{2+} and Ca^{2+} showed daytime maxima whereas those of Cl^- and NO_3^- showed opposite trend, except for few cases, in both winter and summer (Fig. 3). In contrast, Na^+ showed daytime maxima in winter but daytime minima in summer (Fig. 2e) whereas NH_4^+ showed an opposite trend to that of Na^+ (Fig. 2f).

These diurnal changes may be caused by land/sea breeze circulation because wind speed and wind direction on the near surface showed a clear diurnal oscillation during

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the campaigns (Pavuluri et al., 2010a). The onset of sea breeze in daytime causes the onshore flow of marine air masses (Lu and Turco, 1994; Miller et al., 2003) and develops the thermal internal boundary layer (TIBL) extending up to 50–100 m above ground level under the atmospheric boundary layer (ABL) at the coast in Chennai region (Srinivas et al., 2007) that restricts the vertical distribution of pollutants. In contrast, the TIBL disappears and the ABL moves down after the onset of the land breeze in nighttime. In addition, the oxidant concentration may stay high under nighttime smog condition at surface level (Miller et al., 2003) that may promote the in-situ oxidation processes.

The onset of land breeze that transports the inland air masses, enriched with anthropogenic emissions, to the sampling site may increase the EC, OC and WIOC concentrations in nighttime, whereas in daytime, their concentrations might be reduced by dilution with aloft marine air masses. The daytime maxima of EC in summer, for few cases, is probably due to mixing of polluted inland air masses as the winds prevailed from southwest (Pavuluri et al., 2010a). The daytime maxima of WSOC are likely as most of WSOC considered to be secondary that may be produced under strong solar radiation in daytime by in-situ photochemical processes. Further, the long-range transported marine aerosols may also be enriched with WSOC. In contrast, the nighttime maxima of SOC (some SOC compounds may not be water-soluble; Saxena and Hildebrand, 1996) might be due to larger contributions of long-range transported aerosols because vertical mixing of the aged air masses that were accumulated between TIBL and ABL is significant once TIBL disappears by the onset of land breeze in nighttime.

On the other hand, the onset of sea breeze in daytime may uplift aged aerosols that are accumulated over oceanic region together with inland air masses transported offshore in previous night. The aged aerosols may be enriched with secondary species such as MSA^- , SO_4^{2-} and WSOC as well as primary pollutants; K^+ , Mg^{2+} and Ca^{2+} . The former two species are produced by photooxidation of DMS emitted either from oceanic phytoplankton and/or biomass burning, whereas K^+ is emitted from biomass burning and Mg^{2+} and Ca^{2+} are from crustal sources. Thus, MSA^- , SO_4^{2-} , K^+ , Mg^{2+} and Ca^{2+} might become predominant in daytime, and hence aerosols become

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

relatively acidic in daytime than nighttime (Fig. 4). Conversely, Cl^- showed lower concentrations in daytime than nighttime, because HCl is released from NaCl by the reaction with acidic sulfur species and is attached to aerosols after the reaction with NH_3 (Hara et al., 2004). Local emissions, such as fossil fuel combustion, might be the major source of NO_3^- and hence, it showed a peak in nighttime due to land breeze.

The daytime maxima of Na^+ in winter are caused by sea breeze that brings marine air masses. Although its nighttime maxima in few cases during summer (Fig. 3e) are ambiguous, we assume that the sea-salt particles might significantly accumulate between TIBL and ABL during development of the TIBL and their atmospheric level become higher due to vertical dispersion of pollutants, after TIBL disappears in nighttime. As described in Pavuluri et al. (2010b), animal excreta and biofuel/biomass burning are two major sources of NH_4^+ . Hence, the air masses uplifted by land breeze in nighttime during winter may be enriched with NH_4^+ and those by sea breeze in daytime during summer might become enriched with NH_4^+ as they passed over south Indian subcontinent where cow-dung combustion and animal excreta are more important than in central India (Reddy and Venkataraman, 2002b; Garg et al., 2001).

3.4 Seasonal changes in mass fractions of EC, OM and ionic species in PM_{10} : role of source regions

Distributions of mass fractions of EC, OM and ionic species in PM_{10} for three seasons; early winter, late winter and summer, as categorized by the source regions of air masses that arrived in Chennai during the campaigns (see Sect. 3.1 and Fig. 1), are given by box-and-whisker plots in Fig. 5. The mass fractions of EC, SO_4^{2-} and K^+ were higher in late winter than early winter when the air masses originated from SEA and ME/IN (Fig. 1). They are lowest in summer when air masses arrived in Chennai from AS/IO (Fig. 1). Meanwhile, mass fractions of OM and NH_4^+ were higher in late winter than in summer and in early winter. However, MSA^- , Cl^- and NO_3^- did not show

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

any significant difference between early- and late-winter. In contrast, mass fractions of Mg^{2+} and Ca^{2+} were higher in early winter and Na^+ fraction was higher in summer.

It is well known that biomass burning including forest fires are commonly occurred in South and Southeast Asia but the amount of vegetation burned in Southeast Asia (331 Tg dry matter) are almost twice large compared to those of South Asia (178 Tg; Streets et al., 2003). Similarly, the consumption of bio-fuel, in particular cow-dung (Reddy and Venkataraman, 2002b), and livestock emissions (e.g. CH_4 ; Garg et al., 2001) are higher in southern part of India than any other parts of the Indian subcontinent. Thus the origins of the air masses and their source strength should have played a vital role in changing the concentrations of carbonaceous and ionic components from season to season. These considerations also indicate that biomass burning is a significant source of EC, OC, SO_4^{2-} and NH_4^+ as well as MSA^- , Cl^- and NO_3^- . The less abundant EC in summer is likely because biofuel combustion less contributes to EC compared to OC (Stone et al., 2010).

3.5 Assessment of sources: biofuel/biomass burning and secondary production during long-range atmospheric transport

3.5.1 Mass concentration ratios of selected components

Figure 6 shows the mass concentration ratios of selected components in Chennai aerosols. The mass concentration ratios of OC/EC were higher in summer than winter whereas EC/TC were higher in winter than summer (Table 2). OC/EC ratios did not show day- to night-time variation in winter, whereas in summer, they were higher in nighttime than daytime (Fig. 6). It is apparent that the OC/EC ratios for most of the urban sites around the world fall in the range between 1.0 and 4.0 and the OC/EC ratios exceeding 2.0 have been used to indicate the important presence of secondary organic aerosols (Cao et al., 2003). On the other hand, it has been inferred that wood burning generally produces a small fraction of EC in TC, typically around 10% or less (Andreae

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



and Merlet, 2001), whereas vehicular emissions especially from diesel engines contain a larger fraction (about 50%) of EC in TC (Gillies et al., 2001).

OC/EC ratios in Chennai winter aerosols are similar to those reported over the Indian Ocean (Table 2) and comparable to those (< 4.0) reported for urban locations (Table 2; Cao et al., 2003). In addition, EC/TC ratios are also similar to those reported over the Indian Ocean (Table 2) and comparable to those (0.56) reported in road tunnel aerosols (Gillies et al., 2001). EC mass fraction in TC was also always higher than 10%, a typical value for wood burning (Andreae and Merlet, 2001). These results and comparisons suggest that the contributions from fossil fuel combustion are highly significant in winter. However, this is not the actual case.

The air masses that arrived in Chennai originated from ME/IN and SEA in winter (Fig. 1) where biofuel/biomass burning including forest fires are significant (Streets et al., 2003). EC emission from biomass burning depends on burn rate (kg h^{-1}) and type of biomass (Habib et al., 2008; Stone et al., 2010) that can lead the OC/EC and EC/TC ratios to be comparable to those of fossil fuel combustion emissions. Habib et al. (2008) found a significant negative correlation ($r^2 = 0.5$) between EC/PM_{2.5} ratio and burn rate. Stone et al. (2010) reported extremely high EC/OC ratio (1.68) for mango wood burning at low burn rate compared to that (0.24) at high burn rate. In fact, most of cooking energy in South and Southeast Asia come from biofuel (cow-dung, wood and agricultural waste) combustion, generally at low rate, that could contribute a larger amount of EC to the atmosphere. Interestingly, OC/EC ratios were higher and EC/TC ratios were lower in summer than winter, which is mainly driven by EC, although the air masses originated from AS/IO passed over southern part of the Indian subcontinent where anthropogenic emissions are significant. Further, the OC/EC ratios are highly comparable to those reported in rural and mountain sites where the biofuel/biomass burning was expected as a major source(s) (Table 2).

On the other hand, nss-K⁺/EC and nss-K⁺/OC ratios in Chennai aerosols ranged from 0.07 to 0.20 (0.13 ± 0.04) and 0.05 to 0.16 (0.09 ± 0.03) in winter and 0.11–0.33 (0.21 ± 0.05) and 0.01–0.06 (0.04 ± 0.01) in summer, respectively. They did not show

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

any significant diurnal variation (Fig. 6). These ratios are comparable to K/BC and K/OC ratios calculated from emission factors reported for tropical forest (0.44 and 0.06, respectively) and the burning of agricultural residues (0.19 and 0.04, respectively; Andreae and Merelet, 2001). In addition, $\text{nss-K}^+/\text{EC}$ ratios in Chennai aerosols are much higher than that (ave. 0.05) reported in Christchurch, New Zealand, where wood and coal burning was considered as major sources of domestic energy (Wang et al., 2005). These comparisons clearly imply that biofuel/biomass burning are the major sources of Chennai aerosols.

The ratios of $\text{nss-SO}_4^{2-}/\text{EC}$, $\text{nss-SO}_4^{2-}/\text{OC}$, $\text{nss-Ca}^{2+}/\text{EC}$ and $\text{nss-Ca}^{2+}/\text{OC}$ in Chennai aerosols were found to be 1.5 ± 0.59 , 1.1 ± 0.53 , 0.14 ± 0.06 and 0.11 ± 0.05 in winter and 2.6 ± 1.7 , 0.44 ± 0.3 , 0.39 ± 0.17 and 0.07 ± 0.02 in summer, respectively, with daytime maxima in winter and summer (Fig. 6). These ratios are several times higher than those (0.09, 0.1, 0.03 and 0.03, respectively) calculated from road tunnel aerosols (Gillies et al., 2001). Although NO_3^-/EC and NO_3^-/OC ratios (0.14 ± 0.06 and 0.11 ± 0.05) found in Chennai winter aerosols as well as NO_3^-/OC ratios in Chennai summer aerosols are comparable to those (0.08 and 0.1, respectively) obtained for road tunnel aerosols (Gillies et al., 2001), NO_3^-/EC ratios (0.39 ± 1.7) in Chennai summer aerosols are quite different. These comparisons also suggest that the Chennai aerosols have been seriously influenced by biofuel/biomass burning rather than fossil fuel combustion. Hence, we presume that EC might have also been contributed from biomass burning at significant level. Recently, Gustafsson et al. (2009) found a much larger contribution (46 and 68%) of different fractions of black carbon from biomass burning in South Asia based on radiocarbon analyses ($\Delta^{14}\text{C}$), which further confirms that our assumption is reasonable.

WSOC/OC ratios ranged from 0.19 to 0.73 (ave. 0.45) in winter and 0.23–0.61 (ave. 0.45) in summer. We found that the ratios are higher in daytime than nighttime (Fig. 6). The WSOC/OC ratio has been proposed as a measure of photochemical processing or aging during long-range transport (Aggarwal and Kawamura, 2009; Yang et al., 2004). The higher WSOC/OC ratios found in Chennai aerosols suggest that they were

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

subjected for photochemical processing during long-range atmospheric transport and thus the contribution from secondary production is also significant. On the other hand, diurnal variations of WSOC/EC ratios were not significant in both winter and summer, suggesting that local emissions of EC (and OC) as well as in-situ photochemical production of WSOC may also be significant to certain extent in daytime.

Furthermore, WSOC/OC ratios in Chennai aerosols are comparable to those found in other rural and some urban sites where the contributions from biogenic/biomass burning emissions and/or photochemical processing are significant. The ratios are higher than those reported in some other urban sites where the fossil fuel combustion emissions were expected to be significant. On average WSOC accounted for 66% (range, 38–72%) in fine ($D_p < 1.5 \mu\text{m}$) aerosols collected from a rural site that situated on a forest clearing on the Great Hungarian Plain (Kiss et al., 2002), and 44% (range, 23–69%) in Sapporo aerosols (TSP), which were considered as photochemically aged (Aggarwal and Kawamura, 2008). At an urban site in Nanjing, China, WSOC contributed 30% of OC in $\text{PM}_{2.5}$ in winter (Yang et al., 2005). Miyazaki et al. (2006) reported that the median WSOC/OC ratios in Tokyo by semicontinuous measurements were 0.19 in winter and 0.35 in summer and fall where fossil fuel combustion was expected as the major source. These comparisons again indicate that the biofuel/biomass burning emissions as well as photochemical processing during long-range atmospheric transport are significant in South and Southeast Asia.

On average, WIOC/OC ratio was 0.55 in both winter and summer whereas WIOC/EC ratios were 0.8 and 3.6 in winter and summer, respectively, which driven by EC concentrations. Diurnal variations of WIOC/OC were significant in both winter and summer whereas WIOC/EC did not (Fig. 6). The higher WIOC/OC ratios in nighttime than daytime are opposite to that of WSOC/OC but the distributions of WIOC were similar to that of SOC. This suggests that the origin of WIOC and SOC may be similar and most of the SOC may be water-insoluble (Saxena and Hildemann, 1996). The WIOC/EC ratios in summer were higher by a factor about 3 than those (range, 0.9–1.31) reported in Tokyo by semicontinuous measurements, where fossil fuel combustion was expected

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

as the major source (Miyazaki et al., 2006), suggesting that WIOC might be contributed from other source (biofuel/biomass burning) rather fossil fuel combustion. However, the WIOC/EC in Chennai winter aerosols (0.8 ± 0.32) were comparable to those reported in Tokyo (Miyazaki et al., 2006) that may be due to differences in emission factors of EC and OC depending on type of biomass and/or burn rate (Habib et al., 2008; Stone et al., 2010).

3.5.2 Linear relations of EC and OC with nss-K^+ and levoglucosan

We now examine the linear relations of EC and OC with marker species; nss-K^+ and levoglucosan, in order to confirm the important contributions of biofuel/biomass burning to Chennai aerosols. K^+ and EC are significantly emitted from biomass burning (Andreae, 1983) but the former does not contribute from fossil fuel combustion and hence K^+ can be used as marker for biomass burning. Levoglucosan is also an excellent marker for biomass burning as it is produced by pyrolysis of cellulose during biomass burning. Scatter plots of EC and OC with nss-K^+ and levoglucosan are shown in Fig. 7. EC showed medium correlations ($r^2 \leq 0.5$) with nss-K^+ in both winter and summer (Fig. 7a and b) whereas OC showed good correlation ($r^2 \sim 0.7$), except in summer nighttime (Fig. 7c and d). These correlations suggest that carbonaceous components in Chennai aerosols might be mainly originated from biofuel/biomass burning. The weak correlation between OC and nss-K^+ in summer nighttime indicates an additional source, probably higher plant emissions, which only contribute to OC.

The relations of EC and OC were positive with levoglucosan in both winter and summer (Fig. 7). However, the plots between EC and levoglucosan were highly scattered (Fig. 7e and f). It is likely that the emission factors of EC and levoglucosan are highly dependent on type of biomass and burn rate, especially in Asian biofuels as discussed earlier (Stone et al., 2010). In contrast, OC showed a strong correlation ($r^2 = 0.83$) in winter daytime and medium correlations in rest of the campaigns OC ($r^2 \geq 0.3$), suggesting a larger fraction of OC (may be EC too) is derived from biomass burning rather than fossil fuel combustion.

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

3.5.3 Relationships between selected carbonaceous components

As shown in Fig. 8, EC and OC correlate well in winter nighttime and summer daytime and weakly in winter daytime but no correlation was found in summer nighttime, suggesting a significant contribution of EC and OC from common sources; biofuel/biomass burning. However, the weak and no correlation in winter daytime and summer nighttime, respectively, indicate an additional source(s) for either. As noted earlier, land/sea breeze circulation causes the flow of marine and inland air masses onshore and offshore in day- and night-time, respectively. This may weaken the correlation in winter daytime. Marine sources contribute to OC only, but not EC (except ship exhaust), whereas inland air masses may be enriched with both. In fact, the higher ends of EC in winter nighttime were almost twice to that of daytime whereas those of OC were just about 10% (see Table 1) that indicate the significant contribution of OC from marine sources in daytime.

In summer daytime, EC contributions may be as significant as nighttime although sea breeze uplifts marine air masses. The significant mixing of inland air masses is very likely as the winds prevail from southwest in summer (Pavuluri et al., 2010a), passing over southern part of the Indian subcontinent, and hence the influence of marine air masses on EC and OC distributions may be insignificant. Interestingly, EC concentrations were almost constant in summer day- and night-time whereas the higher ends of OC were almost 45% to those of daytime (see Table 1), indicating a larger contribution of OC in nighttime. In fact, emission of VOCs from tropical plants in India is higher in summer than winter (Padhey and Varshaney, 2005). Pavuluri et al. (2010a) also found high concentrations of long-chain (C_9 – C_{12}) diacids in summer nighttime, which are likely formed by oxidation of unsaturated fatty acids that are emitted from higher plants. Hence, we presume that production of secondary organic aerosol by the photochemical oxidation of VOCs emitted from higher plants might be significant in inland region during daytime and thus the inland air masses become enriched with OC that are transported to sampling site in nighttime by land breeze.

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

It is generally believed that SOC is nearly equivalent to WSOC because secondarily produced OC preferably contains the oxygenated chemical functional groups that are water-soluble, although this has not been experimentally demonstrated for ambient air. Kondo et al. (2007) reported very high correlations ($r^2 = 0.87\text{--}0.93$) between WSOC and total carbon concentrations of oxygenated organic aerosols (defined as OOC). Miyazaki et al. (2006) found good correlations ($r^2 = 0.61\text{--}0.79$) between SOC and WSOC in Tokyo aerosols, where fossil fuel combustion was expected to be the major source. The scatter plots between SOC and WSOC in Chennai aerosols in winter and summer are shown in Fig. 9. We found high and medium correlations between SOC and WSOC in winter daytime and in winter and summer nighttime, respectively, but weak in summer daytime (Fig. 9). Further, the correlation coefficients are close to those reported for Tokyo aerosols (Miyazaki et al., 2006). Hence, the retrieved SOC from EC-tracer method can be considered as reasonable in this work although this method tends to overestimate SOC (Yuan et al., 2006).

Slopes (0.13–0.39) of the correlations between SOC and WSOC in Chennai aerosols were lower in nighttime than daytime in both winter and summer (Fig. 9). They were twice to five times lower than those (0.61–0.74) reported in Tokyo aerosols (Miyazaki et al., 2006). These low slope values in Chennai aerosols indicate that most of the SOC is not water-soluble. It is likely because some SOC compounds are considered to have large carbon-hydrogen chain ($\geq C_9$) and functional groups, leading to water-insolubility (Saxena and Hildemann, 1996). Interestingly, the increase in WSOC was insignificant with increasing SOC in summer nighttime aerosols, suggesting that SOC originated from biofuel/biomass burning and terrestrial plant emissions are less water-soluble. Thus, the mass concentration ratios and linear relations of selected ionic species, carbonaceous components and organic marker species indicate that Chennai aerosols are mainly derived by biofuel/biomass burning and secondary production during long-range atmospheric transport from distant source regions in South and Southeast Asia.

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

4 Summary and conclusions

The tropical Indian aerosol (PM₁₀) samples collected at Chennai on day- and night-time bases in winter and summer, 2007 were studied for carbonaceous and ionic components. EC and ionic components showed higher concentrations in winter than summer, but OC was rather constant in both seasons. EC and OC showed similar diurnal variations with nighttime maxima in winter but EC showed daytime maxima for few cases in summer. SO₄²⁻ was the most abundant ion followed by NH₄⁺. MSA⁻, SO₄²⁻, K⁺, Mg²⁺ and Ca²⁺, showing daytime maxima. In contrast, Cl⁻ and NO₃⁻ showed nighttime maxima in both winter and summer, except for few cases. On average, estimated SOC was found to be similar in both winter and summer.

The seasonal changes in mass fractions of EC, OM and ionic species and the comparisons of EC and OC concentrations with those reported for aerosols from other locale in South Asia together with air mass trajectories suggest that the biofuel/biomass burning is a major source of South and Southeast Asian aerosols. The mass concentration ratios of selected components, particularly nss-K⁺, nss-SO₄²⁻, nss-Ca²⁺ and NO₃⁻ to EC and OC and the relations between EC, OC and marker species (nss-K⁺ and levoglucosan) also inferred that the biofuel/biomass burning as the major source of atmospheric aerosols in this region. The high concentrations of SOC, WSOC/OC ratios and positive correlations between SOC and WSOC indicated that secondary production of organic aerosols during long-range transport is also an important source of the tropical Indian aerosols. The data sets reported here could serve as baseline observation of carbonaceous aerosols for southern part of the Indian subcontinent, which could contribute to regional climate and air quality models.

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Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

References

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ACPD

11, 3937–3976, 2011

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- Kanakidou, M., Seinfeld, J. H., Pandis, S. N., Barnes, I., Dentener, F. J., Facchini, M. C., Van Dingenen, R., Ervens, B., Nenes, A., Nielsen, C. J., Swietlicki, E., Putaud, J. P., Balkanski, Y., Fuzzi, S., Horth, J., Moortgat, G. K., Winterhalter, R., Myhre, C. E. L., Tsigaridis, K., Vignati, E., Stephanou, E. G., and Wilson, J.: Organic aerosol and global climate modelling: a review, *Atmos. Chem. Phys.*, 5, 1053–1123, doi:10.5194/acp-5-1053-2005, 2005.
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Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 1. Statistical summary of carbonaceous and ionic components ($\mu\text{g m}^{-3}$) in the tropical Indian aerosol (PM_{10}) samples collected on day- and night-time bases in winter (23 January to 6 February) and summer (22–31 May) 2007 from Chennai, India.

Components	Winter						Summer					
	Daytime			Nighttime			Daytime			Nighttime		
	Range	Ave./Med. ^a	SD ^b	Range	Ave./Med.	SD	Range	Ave./Med.	SD	Range	Ave./Med.	SD
Carbonaceous components												
EC	3.8–7.5	5.8/5.7	1.3	2.4–14.0	7.1/6.8	3.5	1.1–2.5	1.7/1.7	0.4	1.1–1.9	1.5/1.6	0.3
OC ^c	4.4–13.9	8.5/7.4	3.2	3.2–15.6	9.8/10.6	4.1	6.4–9.8	8.4/8.5	1.2	4.2–17.6	11.0/10.9	4.8
WSOC ^d	2.6–7.0	4.2/4.0	1.4	1.7–5.1	3.3/3.1	1.0	3.4–6.0	4.4/4.4	0.8	2.2–4.8	3.8/3.8	0.9
WIOC	1.6–8.0	4.3/3.0	2.2	1.5–11.6	6.5/5.8	3.9	2.9–5.4	3.9/3.9	0.7	2.0–12.9	7.3/6.8	4.1
SOC	0.0–9.2	4.4/3.7	2.9	0.84–8.9	4.9/4.9	2.7	0.51–3.8	2.7/2.9	0.9	0.0–13.1	5.8/5.5	4.8
Ionic components												
MSA [−]	0.18–0.48	0.30/0.30	0.09	0.13–0.64	0.30/0.22	0.17	0.07–0.16	0.11/0.10	0.04	0.04–0.18	0.08/0.09	0.04
Cl [−]	0.12–2.0	0.63/0.45	0.56	0.32–3.1	1.6/1.7	0.81	0.05–0.35	0.15/0.12	0.10	0.06–0.70	0.26/0.22	0.21
NO ₃ [−]	0.22–1.26	0.66/0.66	0.27	0.46–3.1	1.1/0.95	0.67	0.39–1.0	0.58/0.50	0.21	0.21–1.06	0.65/0.58	0.29
SO ₄ ^{2−}	6.9–15.3	9.6/8.8	2.3	4.2–11.9	7.8/8.2	2.4	1.5–11.8	4.8/4.8	3.1	0.96–8.9	3.3/2.6	2.1
Na ⁺	0.18–1.40	0.51/0.42	0.32	0.18–0.66	0.38/0.38	0.16	0.25–0.76	0.46/0.46	0.15	0.13–0.93	0.48/0.47	0.20
NH ₄ ⁺	0.38–2.79	1.82/1.96	0.66	0.29–3.9	2.3/2.5	1.2	0.48–3.5	1.4/1.3	0.97	0.34–3.1	1.1/0.93	0.76
K ⁺	0.44–1.1	0.80/0.78	0.20	0.49–1.0	0.77/0.79	0.20	0.28–0.62	0.40/0.38	0.11	0.14–0.46	0.31/0.32	0.09
Mg ²⁺	0.04–0.82	0.24/0.18	0.22	0.01–0.34	0.14/0.13	0.11	0.03–0.09	0.05/0.04	0.03	0.01–0.08	0.04/0.04	0.02
Ca ²⁺	0.12–2.0	0.61/0.39	0.52	0.07–0.55	0.28/0.23	0.17	0.15–0.80	0.37/0.33	0.19	0.08–0.29	0.14/0.13	0.06
nss-SO ₄ ^{2−}	6.8–15.1	9.5/8.7	2.3	4.1–11.8	7.8/8.1	2.4	1.4–11.6	4.7/4.7	3.1	0.92–8.7	3.2/2.5	2.1
nss-K ⁺	0.42–1.08	0.78/0.76	0.20	0.47–1.02	0.75/0.78	0.20	0.26–0.60	0.38/0.36	0.11	0.14–0.43	0.29/0.30	0.09
nss-Ca ²⁺	0.10–1.9	0.60/0.38	0.50	0.06–0.53	0.27/0.22	0.16	0.13–0.78	0.36/0.31	0.19	0.08–0.25	0.12/0.11	0.05

^a Ave. = average; Med. = Median

^b SD = Standard Deviation

^c OC data from Fu et al. (2010)

^d WSOC data from Pavuluri et al. (2010a).

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Table 2. Concentrations of EC and OC and mass ratios of OC to EC and EC to TC^a in atmospheric aerosols from different locations in South Asia.

Location	Sampling period	Size	Concentrations ($\mu\text{g m}^{-3}$)		Concentration ratios		Reference
			EC	OC	OC/EC	EC/TC	
Chennai, India	January–February (winter) 2007	PM ₁₀	6.5 ± 2.7	9.1 ± 3.7	1.5 ± 0.47	0.42 ± 0.08	This study
Chennai, India	May (summer) 2007	PM ₁₀	1.6 ± 0.37	9.7 ± 3.7	6.2 ± 3.0	0.15 ± 0.04	This study
Hisar, India	December 2004	TSP	3.8 ± 1.4	33.0 ± 17.9	8.47 ± 2.19	NA ^b	Rengarajan et al. (2007)
Manora Peak, India	December 2004	TSP	0.89 ± 0.33	4.9 ± 1.3	5.96 ± 1.92	NA	Rengarajan et al. (2007)
Manora Peak, India	February 2005–November 2006	TSP	8.7	1.1	8.4 ± 2.8	NA	Ram et al. (2008)
Mt. Abu, India	May 2005–February 2006	TSP	3.7	0.5	6.1 ± 2.0	NA	Ram et al. (2008)
Mumbai, India	January–February 1999	PM ₁₀	12.4 ± 5.1	37.3 ± 10.5	3.1 ± 0.5	NA	Venkataraman et al. (2002)
Kanpur, India	January 2007–March 2008	PM ₁₀	3.8 ± 2.3	25.8 ± 16.1	7.4 ± 3.5	NA	Ram et al. (2010)
Kathmandu valley, Nepal	February–May 1999, 2000	PM ₁₀	1.5 ± 0.9	14.4 ± 15.6	9.6	NA	Carrico et al. (2003)
Himalaya, Nepal	February–May 1999, 2000	PM ₁₀	0.48 ± 0.38	3.4 ± 4.2	7.1	NA	Carrico et al. (2003)
Godavari, Nepal	January–December 2006	PM _{2.5}	1.0 ± 0.8	4.8 ± 4.4	4.8	NA	Stone et al. (2010)
Dhaka, Bangladesh	March–April 2001	TSP	22.0	45.7	2.08	0.32	Salam et al. (2003)
Lahore, Pakistan	September 1992–October 1993	TSP	17.6 ± 10.2	76.9 ± 39.1	4.37	NA	Smith et al. (1996)
Bay of Bengal	March–April 2006	TSP	0.4	1.9	7.4	NA	Sudheer and Sarin (2008)
Indian Ocean	February–March 1999	PM _{2.5}	2.5 ± 1.4	3.4 ± 2.0	1.36 ± 1.1	0.42 ± 0.09	Mayol-Bracero et al. (2002)

^a EC = Elemental carbon; OC = Organic carbon; TC = total carbon (EC + OC)

^b NA = Not Available.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



[Back](#)

Close

Full Screen / Esc

[Printer-friendly Version](#)

Interactive Discussion



Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

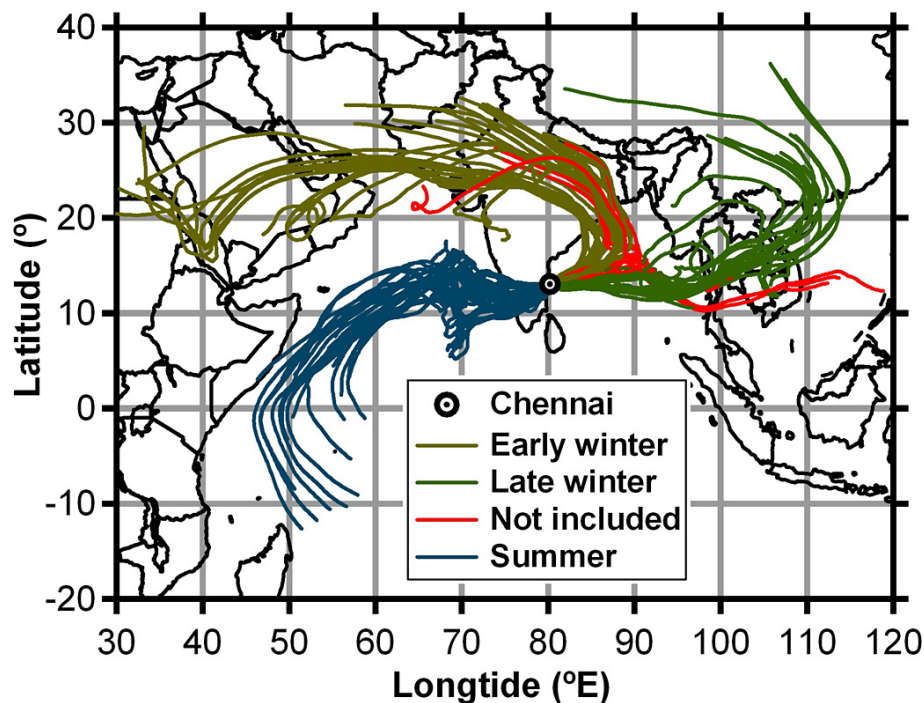


Fig. 1. A map of South and Southeast Asia with sampling site, Chennai (13.04° N; 80.17° E), India together with plots of 10-day backward air mass trajectories arriving at an altitude of 500 m over Chennai in winter (23 January to 6 February) and summer (22–31 May), 2007 showing three major pathways reflecting early winter, late winter and summer.

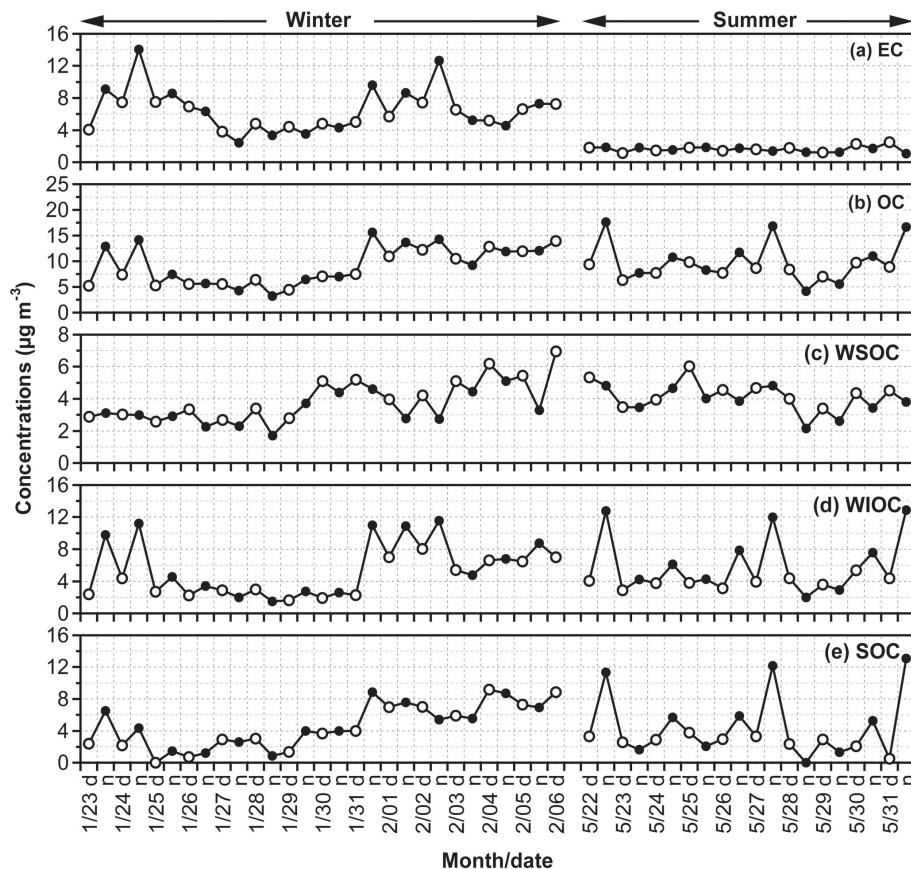


Fig. 2. Temporal variations of carbonaceous components in the tropical Indian aerosol (PM_{10}) samples ($n = 49$) collected on day- and night-time bases in winter (23 January to 6 February) and summer (22–31 May), 2007 from Chennai, India. Open and solid circles show the day- and night-time concentrations, respectively.

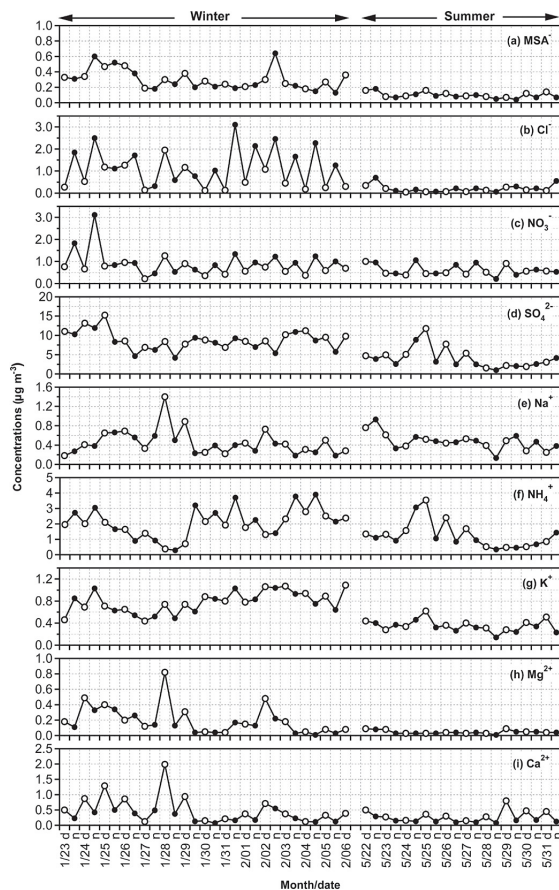


Fig. 3. Temporal variations of water-soluble ionic components in the tropical Indian aerosol (PM_{10}) samples ($n = 49$) collected on day- and night-time bases in winter (23 January to 6 February) and summer (22–31 May), 2007 from Chennai, India. Open and solid circles show the day- and night-time concentrations, respectively.

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

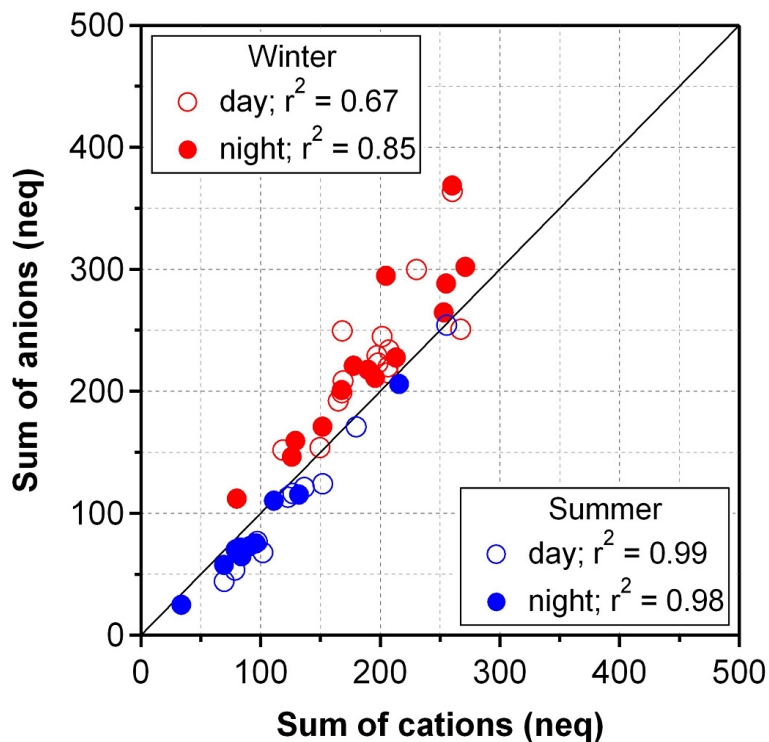


Fig. 4. Linear regression plots between sum of cation equivalents (neq) and anion equivalents (neq) in the tropical Indian aerosol (PM_{10}) samples collected on day- and night-time bases in **(a)** winter (23 January to 6 February) and **(b)** summer (22–31 May), 2007 from Chennai, India.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

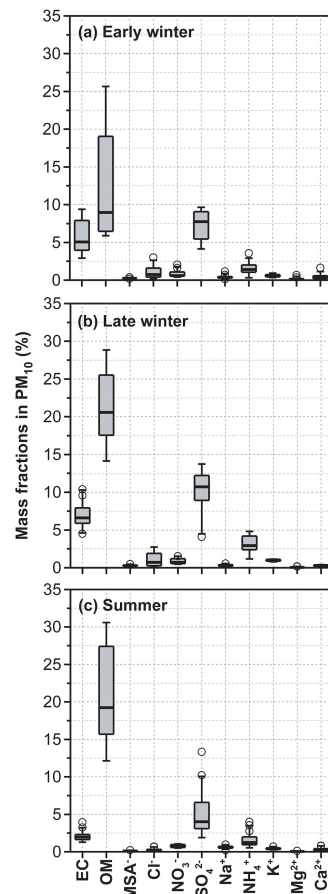


Fig. 5. Box-and whisker plots of mass fractions of EC, OM and ionic species (MSA⁻, Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺, K⁺, Mg²⁺ and Ca²⁺) in the tropical Indian aerosol (PM₁₀) samples collected from Chennai, India in **(a)** early winter, **(b)** late winter and **(c)** summer, 2007.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

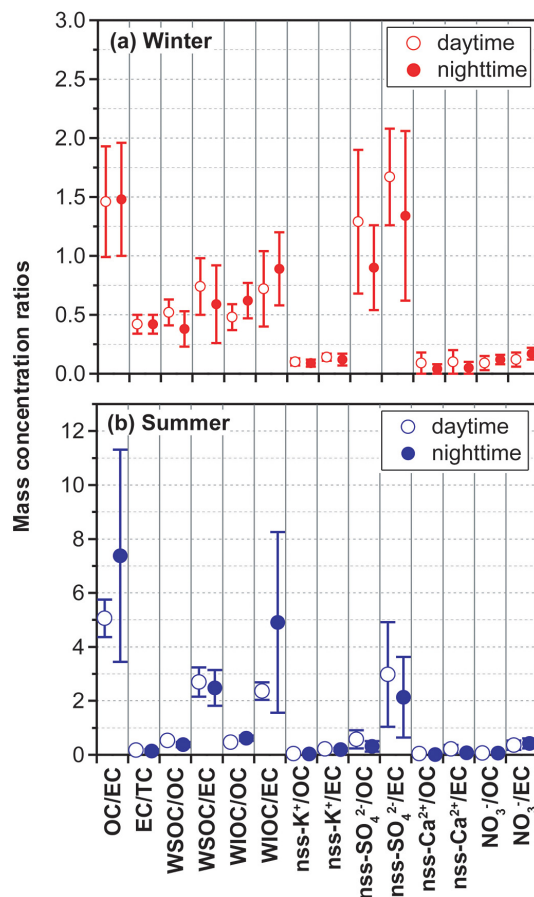


Fig. 6. Mass concentration ratios of selected components in the tropical Indian aerosol (PM_{10}) samples collected on day- and night-time bases in **(a)** winter (23 January to 6 February) and **(b)** summer (22–31 May), 2007 from Chennai, India.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Carbonaceous and ionic components in the tropical Indian aerosols

C. M. Pavuluri et al.

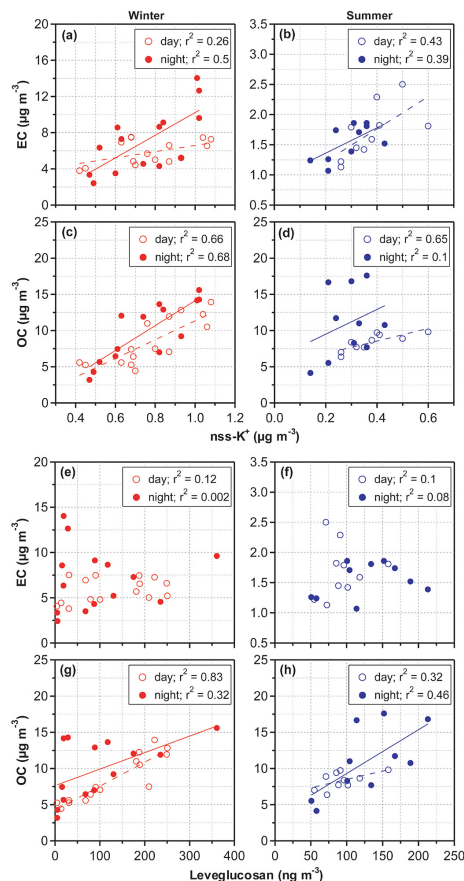


Fig. 7. Scatter plots of EC and OC with nss-K⁺ (a, b, c and d) and levoglucosan (e, f, g and h) in the tropical Indian aerosol (PM₁₀) samples collected on day- and night-time bases in winter (23 January to 6 February) and summer (22–31 May) 2007 from Chennai, India. Levoglucosan data is obtained from Fu et al. (2010).

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

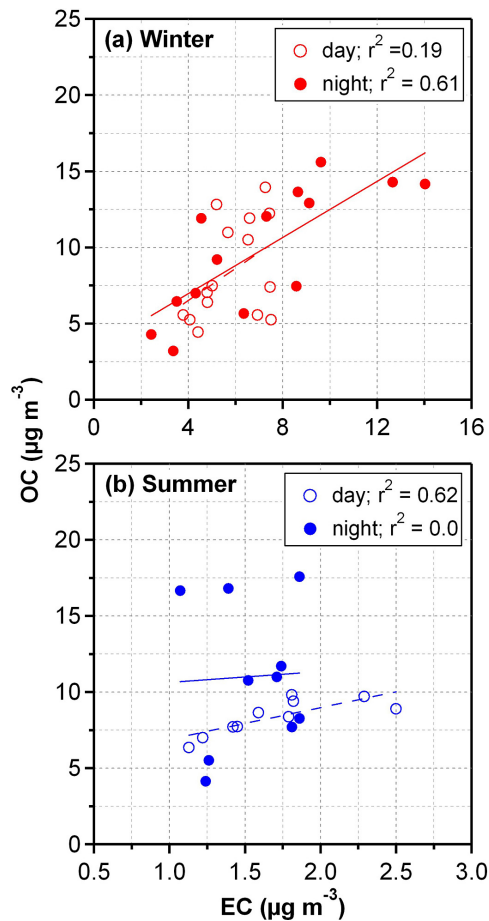


Fig. 8. Scatter plots between EC and OC in the tropical Indian aerosol (PM₁₀) samples collected on day- and night-time bases in **(a)** winter (23 January to 6 February) and **(b)** summer (22–31 May), 2007 from Chennai, India.

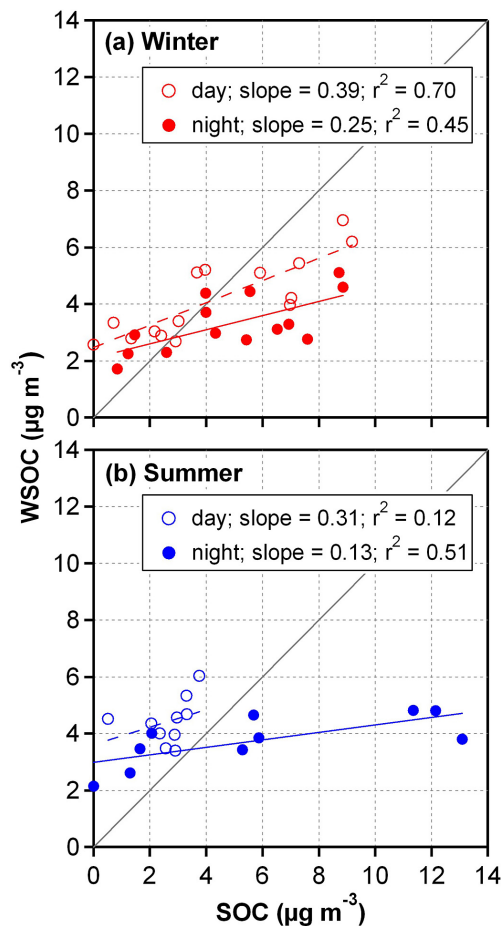


Fig. 9. Linear regression plots between SOC and WSOC in the tropical Indian aerosol (PM_{10}) samples collected on day- and night-time bases in **(a)** winter (23 January to 6 February) and **(b)** summer (22–31 May), 2007 from Chennai, India.