Chemical characteristics of PM_{10} aerosols and airmass trajectories over Bay of Bengal and Arabian Sea during ICARB

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For the first time, chemical characterization of PM_{10} aerosols was attempted over the Bay of Bengal (BoB) and Arabian Sea (AS) during the ICARB campaign. Dominance of SO_4^{2-}, NH_4^+ and NO_3^- was noticed over both the regions which indicated the presence of ammonium sulphate and ammonium nitrate as major water soluble particles playing a very important role in the radiation budget. It was observed that all the chemical constituents had higher concentrations over Bay of Bengal as compared to Arabian Sea. Higher concentrations were observed near the Indian coast showing influence of landmass indicating that gaseous pollutants like SO_2, NH_3 and NO_x are transported over to the sea regions which consequently contribute to higher SO_4^{2-}, NH_4^+ and NO_3^- aerosols respectively. The most polluted region over BoB was 13°–19°N and 70°–90°E while it was near 11°N and 75°E over AS. Although the concentrations were higher over Bay of Bengal for all the chemical constituents of PM_{10} aerosols, per cent non-sea salt (nss) fraction (with respect to Na) was higher over Arabian Sea. Very low Ca^{2+} concentration was observed at Arabian Sea which led to higher atmospheric acidity as compared to BoB. Nss SO_4^{2-} alone contributed 48% of total water soluble fraction over BoB as well as AS. Ratios SO_4^{2-}/NO_3^- over both the regions (7.8 and 9 over BoB and AS respectively) were very high as compared to reported values at land sites like Allahabad (0.63) and Kanpur (0.66) which may be due to very low NO_3^- over sea regions as compared to land sites. Air trajectory analysis showed four classes: **(i)** airmass passing through Indian land, **(ii)** from oceanic region, **(iii)** northern Arabian Sea and Middle East and **(iv)** African continent. The highest nss SO_4^{2-} was observed during airmasses coming from the Indian land side while lowest concentrations were observed when the air was coming from oceanic regions. Moderate concentrations of nss SO_4^{2-} were observed when air was seen moving from the Middle East and African continent. The pH of rainwater was observed to be in the range of 5.9–6.5 which is lower than the values reported over land sites. Similar feature was reported over the Indian Ocean during INDOEX indicating that marine atmosphere had more free acidity than land atmosphere.

1. Introduction

Atmospheric aerosols play a very important role in various atmospheric phenomena such as cloud formation and radiative transfer, etc. These processes depend on the size and chemical composition of aerosols. Increased consumption of fossil fuel and biofuel as energy sources is necessary for the increasing populations but contributes pollutants like sulphate and carbon aerosols (Reddy and Venkataraman 2002; Gadi *et al* 2003; Menon *et al* 2002). It is well established that accumulation

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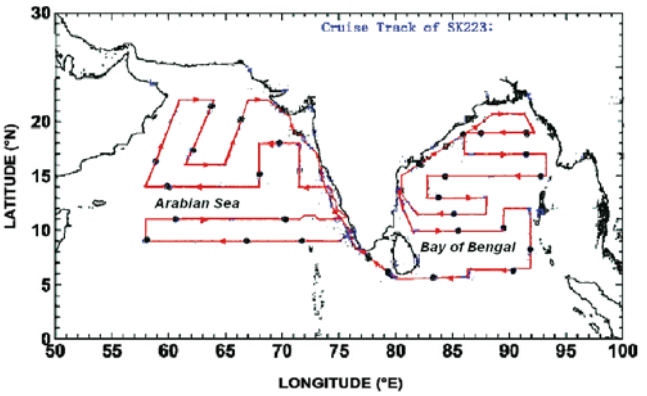

Figure 1. Cruise track showing ship movement and sampling points.

Table 1. *Filter blank values (μg/ml).*

	Whatman 41 filter	PTFE Filter
Cl^-	1.9	0.4
Br^-	nd	nd
NO_3^-	0.4	nd
SO_4^{2-}	3.3	0.7
Na^+	0.1	0.3
NH_4^+	nd	0.1
K^+	0.2	0.2
Ca^{2+}	0.4	0.4
Mg^{2+}	nd	nd

(nd = not detected).

mode sulphate aerosols contribute to negative radiative forcing resulting in global cooling of the atmosphere. On the other hand, black carbon aerosols which are absorbing in nature have positive radiative forcing (Ramanathan *et al* 2001). Considering the importance of atmospheric aerosols, Indian Space Research Organization–Geosphere Biosphere Programme (ISRO-GBP) launched a campaign called ICARB (**I**ntegrated **C**ampaign of **A**tmospheric **A**erosols, gases, and **R**adiation **B**udget) which focused on aerosol properties and radiative budget over India and adjoining oceans, impact of long range transport and its influence on regional radiative forcing. Out of several groups, our group participated in ocean and land segments to carry out measurements on the chemistry of aerosols. Onboard ship measurements were carried out over Bay of Bengal and Arabian Sea during cruise SK 223A and 223B. In this paper, we report for the first time, the chemical composition of PM_{10} aerosols over BoB and AS along with latitudinal and longitudinal variations of major components. Air trajectory analysis has also been attempted to understand the levels and sources of various chemical constituents of aerosols in relation to wind direction.

2. Experimental

2.1 *Cruise track of ICARB*

The ocean segment of ICARB had two cruises, i.e., SK 223A and SK 223B which were completed using the ORV *Sagar Kanya*. The cruise tracks for both SK 223A and 223B have been shown in figure 1. The SK 223A cruise started on 18 March 2006 from Chennai and ended at Kochi (9.96°N, 76.3°E) on 13 April 2006, whereas SK 223B cruise started on 18 April 2006 from Kochi and terminated at Goa on 11 May 2006. The arrow heads in figure 1 show the direction of movements of the ship and solid circles denote the average position where samples were collected during the cruise.

2.2 *Sample collection, preparation and analysis*

Samples were collected onboard ship *Sagar Kanya* during cruise SK 223A and 223B. These two cruise tracks covered 20.5°N to 5.6°N latitude and 80.4°E to 93.4°E longitude over BoB while 9.0°N to 22°N latitude and 58°E to 77.3°E longitude over AS. PM_{10} aerosol sampler (Envirotech 460 NL) was placed on 'B' deck of the ship at around 11 m height from sea level. Samples were collected on Whatman 41 filters ($20*25\,cm^2$) on 8 h basis during daytime (0900–1700 hrs). The collected samples were stored in polythene bags and kept in a refrigerator till analysis. A field blank was also collected in the same manner. The samples were brought to the lab for further extraction and analysis.

For the extraction, a $4 \times 4\,cm^2$ size portion was cut from Whatman 41 filter and transferred into 7 ml deionized distilled water. The water soluble fraction was extracted by using ultrasonic system for 75 min.

Cations and anions were determined by using Ion Chromatography with conductivity detector (Metrohm 792, Basic system). For anions, separation was done using Metrosep A supp5-100 column. A mixture of 3.2 mM Na_2CO_3 and 1 mM $NaHCO_3$ was used as eluent at a flow rate of 0.7 ml/min. Cation separation was done using Metrosep C2-250 column using a mixture of 4 mM tartaric acid and 0.75 mM pyridine 2, 6 dicarboxylic acid as eluent at a flow rate of 1.0 ml/min.

2.3 *Filter blank and extraction efficiencies*

For quality control in analysis, filter blanks were prepared and analyzed in the same manner as samples. Filter blank values are given in table 1. These

Table 2. *Extraction efficiency of sample preparation method.*

	1st extraction (μg/ml)	2nd extraction (μg/ml)
Cl^-	14.4	–
NO_3^-	36.2	–
SO_4^{2-}	35.5	–
Na^+	2.3	0.3
NH_4^+	55.2	–
K^+	4.5	0.5
Ca^{2+}	9.7	–
Mg^{2+}	0.8	–

values were used for final calculation of sample concentration. Similarly, as a quality check in sample preparation, extraction efficiencies were calculated by extracting the samples two times. As given in table 2, for most of the ions, the extraction efficiencies were found $>99\%$.

3. Results and discussion

3.1 *Variation of anions over Bay of Bengal and Arabian Sea*

The concentrations of major anions and cations in PM_{10} aerosols over Bay of Bengal (BoB) and Arabian Sea (AS) during ICARB campaign period with a comparison at a land site in Allahabad have been given in table 3. Both over BoB and AS, among anions, SO_4^{2-} concentrations were observed to be the highest followed by Cl^-, NO_3^- and Br^-. Over AS, the level of most of the anions were lower than BoB. Figure 2(a–d) shows latitudinal and longitudinal variation of NO_3^- and SO_4^{2-} over BoB and AS. It shows that concentrations of SO_4^{2-} and NO_3^- were the highest between 13°N and 19°N and 70°E and 90°E which represents the area near the Indian coast. Over BoB, this was the region of high concentrations for most of the pollutants while over AS, the highest SO_4^{2-} was observed at around 11°N and 75°E. In this study, mean PM_{10} SO_4^{2-}/NO_3^- ratios over BoB and AS were 7.8 and 9 respectively while at Allahabad it was 0.63 (table 3) and at Kanpur, 0.66 (Chinnam *et al* 2005). The higher SO_4^{2-} concentration may be due to preferable gas to particle conversion of SO_2 over sea regions. This is corroborated by higher correlation coefficient ($r = 0.48$) of SO_4^{2-} with relative humidity (RH). In the presence of sufficient humidity over the ocean, SO_2 gets converted into H_2SO_4 which may further react with atmospheric NH_3 resulting in $(NH_4)_2SO_4$ aerosols. Simplest pathway of sulphuric acid formation may be written as follows (Stockwell and Calvert 1983):

$$SO_2 + OH \rightarrow HOSO_2, \quad (1)$$

$$HOSO_2 + O_2 \rightarrow HO_2 + SO_3, \quad (2)$$

$$SO_3 + H_2O \rightarrow H_2SO_4, \quad (3)$$

which further reacts with atmospheric NH_3 as follows:

$$H_2SO_4 + 2NH_3 \leftrightarrow (NH_4)_2SO_4. \quad (4)$$

During the campaign, wind data show passing of air parcels from Indian land regions towards SE in BoB. Higher levels of all chemical constituents observed over BoB may be due to strong influence from the Indian continental airmasses (Moorthy *et al* 2006). Higher Aerosol Optical Depth (AOD) were observed over BoB as compared to AS during the same campaign (Kedia and Ramachandran 2006). Higher loadings have been reported in the northern BoB and lowest in eastern BoB and AS regions (Nair *et al* 2006). More anthropogenic influence over BoB than over AS has been reported by Rao *et al* (2006). Surface ozone concentrations were also observed to be higher over BoB than over AS (Gupta *et al* 2006). These observations indicate that BoB has significant influence of landmass and various anthropogenic activities. Very high nss SO_4^{2-} concentrations associated with landmass trajectories indicate that long range transport of SO_2 significantly alters the oceanic air composition over BoB. The high concentrations of nss SO_4^{2-} can have a significant role in scattering further influencing the radiation budget.

3.2 *Variation of major cations over Bay of Bengal and Arabian Sea*

The mean concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+} and NH_4^+ over BoB and AS are shown in table 3. The order of cation concentration observed over Bay of Bengal was $K^+ > Na^+ > NH_4^+ > Ca^{2+} > Mg^{2+}$ while $NH_4^+ > Na^+ > Ca^{2+} > Mg^{2+} > K^+$ over AS. NH_4^+ was observed to be the main cation over both the regions. Figure 2(e–j) shows the latitudinal and longitudinal variations of NH_4^+, Ca^{2+} and K^+ over BoB and AS. The figure also shows that the highest NH_4^+ concentrations were observed at around 13–19°N and 85°E, where it is almost twice as much as those seen at other positions. However, NH_4^+ concentrations were lower than these reported

Table 3. *Mean ($\mu g/m^3$) and standard deviation (SD) of major ions in PM_{10} aerosols.*

	Bay of Bengal (BoB) ($n = 17$)		Arabian Sea (AS) ($n = 13$)		Allahabad during LC-II ($n = 29$)	
	March–April 2006		April–May 2006		December 2004	
Period	Mean	SD	Mean	SD	Mean	SD
Cl^-	2.3	1.0	1.5	1.0	2.9	1.4
Br^-	0.1	0.08	0.02	0.04	Not reported	–
NO_3^-	1.1	0.7	0.5	0.2	25.0	13.9
SO_4^{2-}	8.6	6.2	4.5	2.6	15.8	9.6
Na^+	1.7	1.3	0.4	0.2	1.4	1.1
NH_4^+	1.5	1.1	1.3	0.6	13.8	8.3
K^+	2.2	2.3	0.02	0.07	2.0	0.8
Ca^{2+}	1.2	1.2	0.3	0.4	1.9	1.3
Mg^{2+}	0.2	0.35	0.04	0.1	0.1	0.1

at land sides. It is to be noted that NH_4^+ followed latitudinal and longitudinal variations similar to NO_3^- and SO_4^{2-}. The variation of Ca^{2+} was very similar to that of K^+ over both the regions indicating similar type of sources, possibly soil-dust or biomass burning. These observations suggest that PM_{10} chemical composition is dominated by SO_4^{2-}, NH_4^+ and NO_3^- indicating anthropogenic influence both over BoB and AS.

3.3 *Non-marine contribution*

To understand the contribution of major ions due to other sources like anthropogenic activities and crustal influence, non-marine fraction was calculated as follows:

$$\%\ \text{Sea salt fraction (\% ssf)} = (100 \times \text{Na}^+ \times r)/X \quad (5)$$

where X is component, r is ratio X/Na^+ in sea water

$$\%\ \text{nss fraction} = 100 - \text{ssf} \quad (6)$$

where r values were taken as 1.8, 0.25, 0.04, 0.04, 0.12 for Cl^-, SO_4^{2-}, K^+, Ca^{2+} and Mg^{2+} respectively (Keene *et al* 1986).

Table 4 gives non-sea salt fractions (nss) of chemical components over BoB and AS. Mg^{2+}, Cl^- and Na^+ were found almost originating from sea salt as their ratios represent standard sea salt ratios. Components like SO_4^{2-}, K^+, Ca^{2+}, were identified as non-marine components contributed by sources like anthropogenic activities or crustal influence. It is interesting to note that in spite of higher concentrations of components recorded over BoB, per cent nss-SO_4^{2-}, nss Ca^{2+} and nss K^+ concentrations were higher over Arabian Sea. nss-SO_4^{2-} over BoB was 92.8% while 97% over AS. nss Ca^{2+} was 84.3% over BoB as compared to 90.7% over AS. nss K^+ over BoB was 49.8% as compared to 55.2% over AS. Surprisingly, over BoB, all the Cl^- was observed to be of marine origin having Cl^-/Na^+ ratios representing sea-salt ratios while 42.9% of Cl^- was observed to be non-marine origin over AS.

Calculations of marine and non-marine contributions show that BoB as well as AS are highly influenced by non-marine sources (mostly anthropogenic). Very high nss SO_4^{2-} concentrations may be due to oxidation of transported SO_2 to H_2SO_4. The amount of free H_2SO_4 seems to be higher over AS as compared to BoB. pH of water soluble extract of aerosol samples also indicated higher acidity over AS as compared to BoB. Average pH of water soluble extract of aerosol over BoB was 4.7 whereas 4.5 over AS.

3.4 *Per cent contribution of different components in water soluble fraction*

Figures 3 and 4, show per cent contribution of different components in water soluble extract of PM_{10} aerosols over BoB and AS respectively. nss SO_4^{2-} was found to be a major ion of water soluble extract. Over BoB and AS, nss SO_4^{2-} contributed 48% fraction of total water soluble ions. NH_4^+ contribution was higher over AS (14%) as compared to BoB (9%). NO_3^- showed almost equal per cent influence over both the regions while nss Ca^{2+} contribution was higher over BoB than AS.

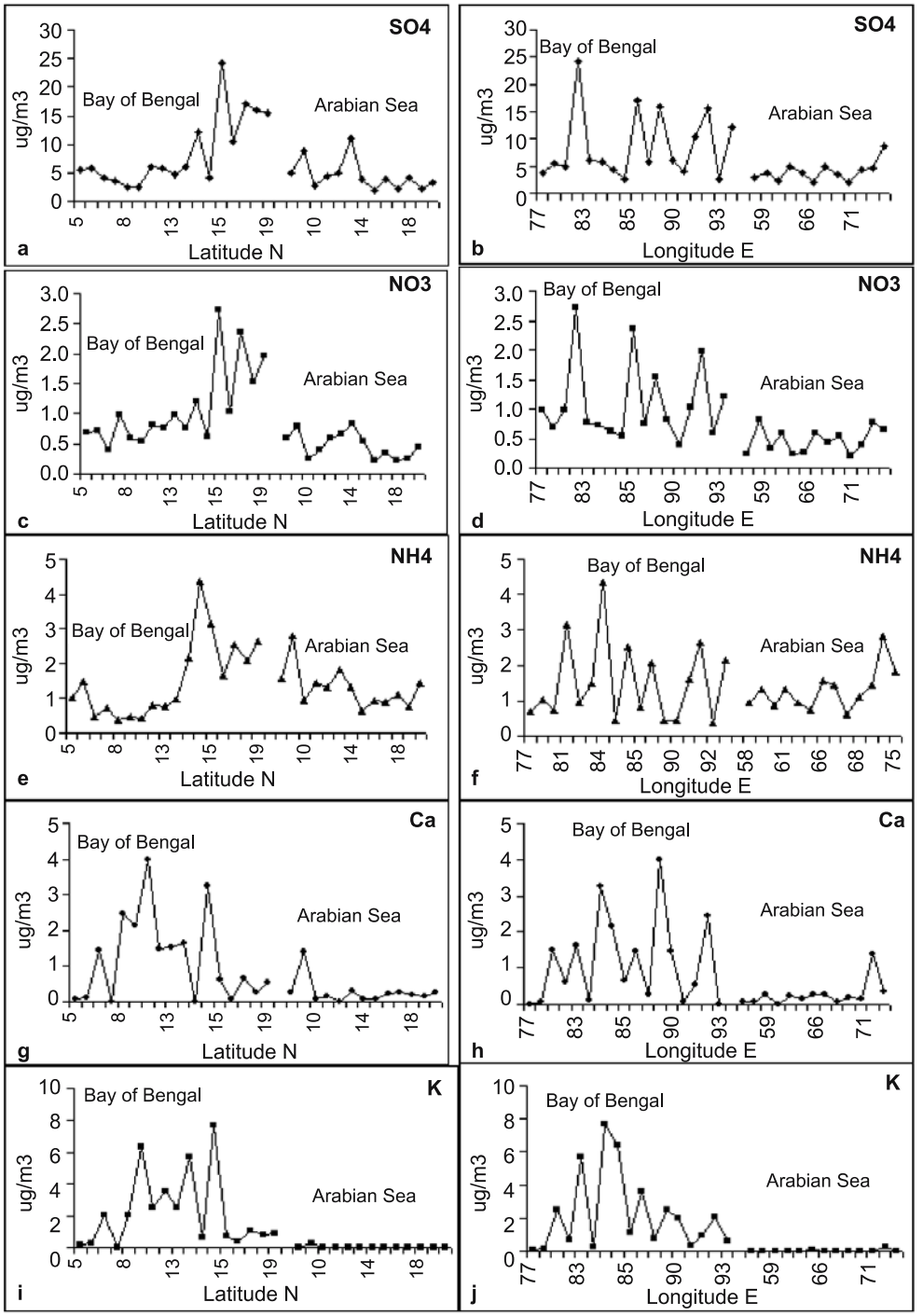

Figure 2. Latitudinal and longitudinal variations of major ions over Bay of Bengal (BoB) and Arabian Sea (AS).

3.5 *Airmass trajectory analysis*

In order to explain the chemistry and sources of pollutants, airmass trajectories were calculated for all the samples on a daily basis (12:00 h) for different ship positions (latitudes and longitudes). Trajectories were calculated using CGER METEX programme (http://cgermetex.nies.jp/metex/manual01.html) for past 120 h at 500 m height. Figure 5 is the back trajectory analysis of airmasses during the campaign period. We have chosen nss SO_4^{2-} concentrations to correlate the origin of airmasses over BoB and AS as it is a good indicator of long range transport of pollution due to oxidation of SO_2 to SO_4^{2-}. The following four classes of air trajectories were obtained over BoB and AS:

Table 4. *Per cent non-sea-salt (nss) fraction of PM_{10} aerosols over Bay of Bengal and Arabian Sea.*

	Bay of Bengal	Arabian Sea
nss SO_4^{2-}	92.8	96.9
nss Ca^{2+}	84.3	90.7
nss K^+	49.2	55.2
nss Cl^-	0.0	42.9
nss Mg^{2+}	0.0	0.0

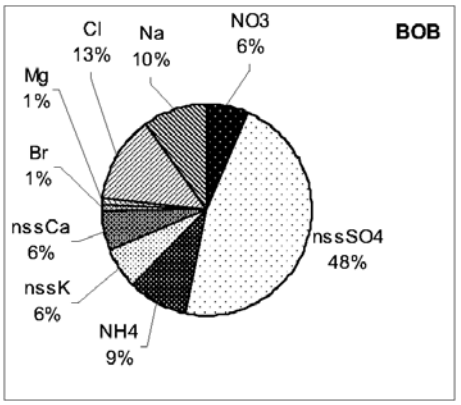

Figure 3. Per cent contribution of water soluble fraction of PM_{10} aerosols over Bay of Bengal.

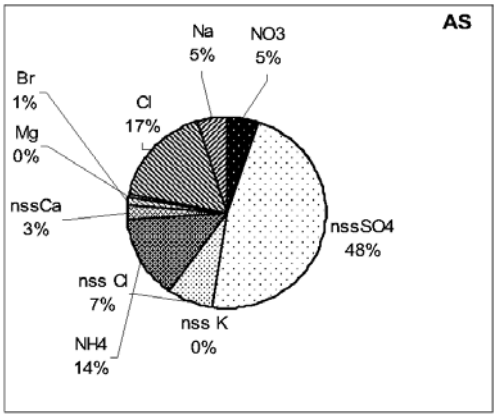

Figure 4. Per cent contribution of water soluble fraction of PM_{10} aerosols over Arabian Sea.

- Air parcel moving through Indian land

  It was primarily dominated over BoB. Six out of 17 times the air moved over BoB from the Indian continent when the concentrations of nss SO_4^{2-} were observed to be the highest.
- Air parcel from northern Arabian Sea

  Some of the days, the wind direction was observed to be coming from north of Arabian Sea close to Middle East and Kucch. These samples showed higher nss SO_4^{2-} concentrations.
- Air parcel from Indian Ocean

  When air parcel was moving from ocean the concentrations of nss SO_4^{2-} were observed to be the lowest both over BoB and AS.
- Air parcel from near African continent

  It was seen on April 29, 2007 that the air was coming from the African continent when nss SO_4^{2-} concentrations were moderately high.

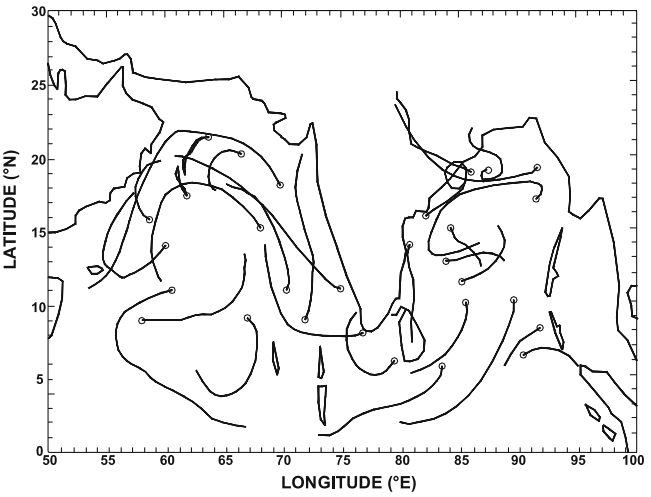

Figure 5. Airmass trajectories over Bay of Bengal and Arabian Sea during ICARB.

3.6 *Chemical composition of TSP and $PM_{2.5}$ at Hyderabad during ICARB*

During ICARB, samples of $PM_{2.5}$ aerosols and Total Suspended Particulate (TSP) were collected at Hyderabad (on the terrace of IICT building). Due to limited availability of samplers, $PM_{2.5}$ aerosol samples were collected during Leg-I while TSP samples were collected during Leg-II for their chemical characterization. $PM_{2.5}$ aerosol samples were collected on PTFE filters using low volume custom made sampler while TSP samples were collected on Whatman 41 filters using high volume sampler (HVS, Envirotech APM 430-411). For $PM_{2.5}$, 1/4 of the filter portion was extracted in 2 ml deionized distilled water using ultrasonic bath for 75 minutes. TSP samples were prepared and analyzed as mentioned in section 2.2.

Table 5 gives the average concentration of chemical components of TSP and $PM_{2.5}$ aerosols at Hyderabad during the ICARB period. $PM_{2.5}$ values have been given for the period of leg-I of ICARB when the ship was moving over BoB while TSP values have been given for the period of leg-II of ICARB when the ship was moving over AS.

Table 5. *Mean concentration of major ions of $PM_{2.5}$ aerosols (during leg-I of ICARB) and total suspended particulates (TSP, during leg-II of ICARB) at Hyderabad.*

	$PM_{2.5}$ ($\mu g/m^3$)	TSP ($\mu g/m^3$)
Cl^-	0.3	1.0
Br^-	0.04	0.1
NO_3^-	0.5	5.5
SO_4^{2-}	0.8	4.0
Na^+	0.1	0.7
NH_4^+	0.1	2.4
K^+	0.09	0.8
Ca^{2+}	0.8	2.3
Mg^{2+}	0.1	0.3

Table 6. *Major ions in rain water over BoB during ICARB.*

	Concentration (μeq/l)
pH	6.2
EC (μS/cm)	41.7
F^-	1.5
Cl^-	196.4
Br^-	5.0
NO_3^-	32.5
SO_4^{2-}	69.3
Na^+	151.6
NH_4^+	21.0
K^+	6.2
Ca^{2+}	61.7
Mg^{2+}	25.1

Concentration of major chemical components of $PM_{2.5}$ aerosols is given in table 5. It shows that $PM_{2.5}$ aerosols are dominated by SO_4^{2-} followed by $Ca^{2+} > NO_3^- > Cl^- > Na^+ > NH_4^+ > Mg^{2+} > K^+ > Br^-$ (table 5). Fine mode dominance of SO_4^{2-} suggests its formation via gas to particle conversion of SO_2. Surprisingly, Ca^{2+} in $PM_{2.5}$ aerosols is second highest. Normally, Ca^{2+} is reported in coarse mode. Fine mode presence of Ca^{2+} is due to unknown sources. It could be due to various types of industries in the vicinity of site. SO_4^{2-}/NO_3^- ratio in $PM_{2.5}$ was noticed higher (1.66) than TSP indicating that role of NO_3^- is significant over $PM_{2.5}$ size range.

At Hyderabad, the order of concentration of chemical components of TSP was $NO_3^- > SO_4^{2-} > NH_4^+ > Ca^{2+} > Cl^- > Na^+ > K^+ > Mg^{2+} > Br^-$ (table 5). This feature is somewhat different than $PM_{2.5}$ aerosols. At Hyderabad, vehicular sources are dominated over industrial sources in TSP. Being an urban site, a significant amount of oxides of nitrogen is emitted by vehicular transport which gets converted into coarse mode NO_3 aerosols. SO_4^{2-}/NO_3^- ratio in TSP at this site was observed to be 0.72 which is similar to other continental sites but much lower than BoB and AS.

3.7 *Chemical constituents of rainwater over Bay of Bengal during ICARB*

During the campaign, 5 samples of rain water were collected over BoB. The pH, EC and average concentration of chemical components of rainwater are given in table 6. The pH of rainwater was observed from 5.9–6.5 which is lower than the values reported over land sites (Kulshrestha *et al* 2005) indicating more acidity over BoB. Acid rain with very low pH were reported over the Indian Ocean during the INDOEX period (Kulshrestha *et al* 1999). But the pH over BoB is slightly higher than the values reported over the Indian Ocean. The possible reason for slightly higher pH of rain water during ICARB than INDOEX period could be higher Ca^{2+} levels due to closeness to the Indian coast and a different season of collection. Land airmasses carry crustal aerosols close to the coast which neutralize the aerosols resulting in higher pH of rainwater. INDOEX cruises covered larger area over remote ocean during January–March months. Similar to PM_{10} aerosols, SO_4^{2-}/NO_3^- ratio in rainwater have also been noticed higher (ratio = 2.13) which corroborates the fact that over oceanic regions, SO_2 sources predominate as compared to the sources of NO_2. Airmass trajectories also reflect that BoB region has higher influence from airmasses coming from Indian land side which can carry SO_2 over the sea which further gets converted into sulphate aerosols. Calculations show that 73% fraction of SO_4^{2-}, 89% of Ca^{2+} and 19% of Cl^- were contributed by non-marine sources. The major source of non-marine SO_4^{2-} may be transported SO_2 emitted from fossil fuel combustion in the region.

4. Conclusion

The study showed that PM_{10} aerosols were dominated by SO_4^{2-}, NH_4^+ and NO_3^- over both the regions indicating the presence of ammonium sulphate and ammonium nitrate aerosols which play a very important role in radiation budget. Near the coast of India, the concentrations of all the components were noticed higher showing the influence of landmass. Most polluted region over BoB was observed to be 13°–19°N and 70°–90°E while 11°N and 75°E over AS. Although the levels of

components were higher over BoB, per cent non-sea salt fraction was higher over AS. Ratios of SO_4^{2-}/NO_3^- over both the regions were very high when compared to reported values at land sites Allahabad and Kanpur. Of the total SO_4^{2-}, 92.8% was observed as nss SO_4^{2-} over BoB while 96.9% over AS. The nss SO_4^{2-} alone contributed 48% of total water soluble fraction over BoB and AS. Four major air trajectory classes showed that highest nss SO_4^{2-} was observed during airmasses coming from the Indian land side while lowest concentrations were observed when the air was coming from oceanic regions. Moderate concentrations of nss SO_4^{2-} were observed when air was seen moving from Middle East and African continent. pH of rain water over BoB was observed to be acidic as compared to the pH values reported from land stations which may be due to high nss SO_4^{2-} concentration indicating that marine atmosphere contains more free acidity than land atmosphere.

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