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Impact of anthropogenic sources on aerosol iron solubility over the Bay of Bengal and the Arabian Sea

Bikkina Srinivas · M. M. Sarin · Ashwini Kumar

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Abstract Ambient aerosols collected from the marine atmospheric boundary layer of the Bay of Bengal and the Arabian Sea have been studied to assess the fractional solubility of aerosol iron, defined as Fe_{ws} (%) = $Fe_{ws}/Fe_{Tot} \times 100$; where Fe_{Tot} is total aerosol iron and Fe_{ws} is water soluble iron. The mass concentration of Fe_{Tot} over the two oceanic regions is not significantly different. However, the fractional solubility is 1–2 orders of magnitude higher over the Bay of Bengal (1.4–24%) compared to that over the Arabian Sea (0.02–0.4%). The spatio-temporal variability in Fe_{ws} (%) is attributed to differences in the nature of the mineral dust over the two oceanic regions. The Arabian Sea receives coarse dust from desert regions; whereas transport of alluvial dust from the Indo-Gangetic Plain is a dominant source to the Bay of Bengal. The poor fractional solubility (<1%) of Fe from mineral dust, hitherto overestimated in the literature, is documented for the Arabian Sea. A significant linear relationship (P-value < 0.001) between Fe_{ws} (%), Fe_{Tot} and nss-SO_4^{2-} over the Bay of Bengal provides evidence for the chemical processing of mineral dust. Furthermore, the role of anthropogenic sources (biomass burning and fossil-fuel combustion) in enhancing the Fe_{ws} (%) is discernible from the chemical composition of fine mode ($PM_{2.5}$) aerosols over the Bay of Bengal. The potential impact of these Fe-dust depositions on phytoplankton carbon fixation and surface ocean biogeochemistry is discussed.

Keywords Arabian Sea · Bay of Bengal · Aerosols · Aerosol iron solubility · Mineral dust

Introduction

Atmospheric deposition of mineral dust, considered as a major source of Fe to the open ocean, has potential to enhance primary productivity; which in turn influences the atmospheric CO_2 budget and, hence, climate (Duce and Tindale 1991; Jickells et al. 2005; Mahowald et al. 2009). The atmospheric supply of Fe also has an impact on surface ocean biogeochemistry and on the production of DMS (Cooper et al. 1996; Jickells and Spokes 2001; Turner et al. 1996; Zhuang et al. 1992).

A number of studies carried out in high nutrient-low chlorophyll regions (Subarctic Pacific, Equatorial Pacific, and Southern ocean) have highlighted the role of atmospheric deposition of mineral dust in mediating glacial-interglacial CO_2 changes and have emphasized the need to understand the atmospheric transport and deposition of soluble Fe to open ocean (Boyd et al. 2000; Coale et al. 1996; Jickells et al. 2005; Mahowald et al. 2009; Martin 1990; Martin and Fitzwater 1988). In this context, modeling approaches have suggested the transport of mineral dust from semi-arid and arid regions of the world as the dominant source of soluble

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iron to the open ocean (Luo et al. 2005; Mahowald et al. 2005). Studies on the solubility of mineral dust suggest that, in general, the fractional solubility of Fe is very poor, less than ~1% (Jickells and Spokes 2001). Recently, aerosols collected over the remote North Pacific Ocean show enhanced Fe-solubility compared to that from the source regions of mineral dust (Solmon et al. 2009). This variability has been explained in terms of uptake of acidic anthropogenic species by mineral aerosols during long-range transport. However, particle-size and concentration of mineral dust, mineralogy, presence of organic compounds and solar irradiation can also play a role in Fe-solubility (Zhuang et al. 1992). More recently, the importance of combustion sources (biomass burning and fossil-fuel combustion) has been suggested as a potential source of soluble iron (Chuang et al. 2005; Guieu et al. 2005; Kumar and Sarin 2010; Kumar et al. 2010; Luo et al. 2008; Schroth et al. 2009). Although meteoritic debris (cosmic dust) and volcanic ash have been considered as a significant contributor of Fe to seawater, these estimates are not well constrained due to their episodic fallout nature. These observations stress the need for regional scale studies on the control for iron solubility in ambient aerosols.

This manuscript addresses the issue of fractional solubility of aerosol-Fe over the Bay of Bengal and the Arabian Sea. Based on the optical and chemical properties of aerosols, it has been documented that the marine atmospheric boundary layer (MABL) of the northern Indian Ocean is significantly influenced by the continental outflow from the south and south-east Asia during winter and spring seasons (Kumar et al. 2008a; Kumar et al. 2008b; Lelieveld et al. 2001; Ramanathan et al. 2001; Sudheer and Sarin 2008). Therefore, these two oceanic regions provide an ideal location to study the impact of continental outflow on fractional solubility of aerosol-Fe from mineral dust entering the MABL.

Methodology

Cruise tracks and meteorological conditions

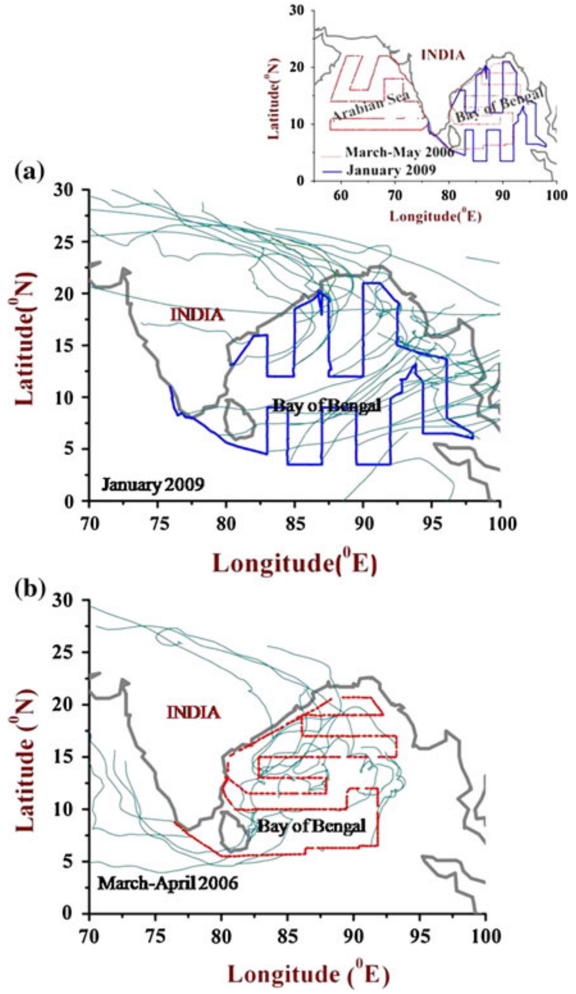

Fig. 1 Aerosol sampling tracks and air mass back trajectory cluster at an arrival height of 500 m over the Bay of Bengal during (**a**) January 2009, (**b**) March–April 2006. *Inset* also shows cruise track conducted in the Arabian Sea (April–May 2006)

Aerosol samples (Fig. 1) were collected from the MABL of the Bay of Bengal and the Arabian Sea during three research cruises (shown as inset in Fig. 1, March–April 2006, April–May 2006 and January 2009) conducted onboard ORV Sagar Kanya as a part of the national programme on integrated campaign of aerosols and trace gases radiation budget (ICARB). The meteorological parameters, measured during January 2009, show that prevailing winds were predominantly from north-easterly with an average speed of about 5.5 ± 2.5 (range = 0.3–12.4) m s^{-1}. The relative humidity (RH %) varied from 41 to 99% (avg = 73 ± 10%). During March–April 2006 and April–May 2006, the winds were initially north-easterly and later changed to south-westerly. Details on meteorological parameters have been described in earlier studies (Alappattu 2007; Kumar et al. 2008a, b; Sudheer and Sarin 2008).

Aerosol sampling and analysis

A total of 42 samples of TSP (19 from Arabian Sea and 23 from Bay of Bengal) were collected between 18 April and 11 May 2006, 19 March and 12 April 2006, respectively. During a subsequent cruise, two size fractions (31 samples of $PM_{2.5}$ and 33 of PM_{10}^{-}) were collected from the Bay of Bengal between 27 December 2008 and 28 January 2009. All aerosol samples were collected on PALLFLEX®™ tissuquartz filters using high volume samplers (Thermo Anderson Inc.,), operated at a flow-rate of 1.13 m^3 min^{-1}. After collection, filters were sealed in zip-lock bags and stored at −19°C until analysis. The mass concentrations of $PM_{2.5}$ and PM_{10} were ascertained gravimetrically based on the filter weights before and after the sampling period. Prior to taking their weight, the filters were equilibrated to room temperature under a clean laminar flow bench (Class-100) for ~24 h and the balance was maintained at constant relative humidity (36 ± 6%) and temperature of 20 ± 3°C. Aerosol samples ($PM_{2.5}$, PM_{10}) were analyzed for soluble iron (Fe_{ws}) and total aerosol iron (Fe_{Tot}). In addition, water soluble inorganic constituents (Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}), crustal elements (Al, Ca, Mg), heavy metals (Cd, Pb) and carbonaceous aerosols (EC, OC) were measured.

For the determination of total aerosol iron (Fe_{Tot}), 8–10 circular filter punches (ca. 2.0 cm dia) were placed in Teflon vials and digested (using 0.5 ml HF and 4.0 ml HNO_3; 4.0 ml Milli-Q water) in a microwave digestion system (pressure ~100 bar). After digestion, 20.0 ml of Milli-Q water was added to the Teflon vials and the solutions were stored in precleaned 30 ml poly-propylene bottles. Subsequently, analysis for Fe, Al, Ca and Mg were performed on ICP-AES (Horiba, Jobin–Yvon model), Cd and Pb on GF-AAS (Perkin Elmer, AAnalyst 100 coupled to HGA-800). The calibration standards were prepared from commercial MERCK®™ (23 elements) standard solution. Based on the repeat measurements of samples and standards, the overall analytical uncertainty in the measurement of Fe_{Tot} was better than 5%.

Analytical protocol for soluble iron (Fe_{ws})

Water soluble iron (Fe_{ws}) was extracted with 10 ml of Milli-Q water (specific resistivity ≥18.2 MΩ-cm) from 8 to 10 circular filter punches (ca. 2.0 cm dia), preceding an analytical step of ultra-sonication for about 10 min. Subsequently, water extracts were filtered through 0.45 µm PTFE filter cartridges and acidified to pH ~2 with distilled HNO_3 and analyzed for Fe_{ws} on GF-AAS. The choice of Milli-Q water as a standard medium for the extraction of aerosol-Fe has evolved by consensus in order to avoid variations among the different laboratories and in the use of buffer solutions of varying pH. Likewise, use of ambient seawater has inherent difficulty in reproducing in situ conditions due to varying degree of organic ligands and pH. The use of Milli-Q water for extraction of aerosol-Fe is consistent with earlier studies reported in the literature (Buck et al. 2010; Kumar et al. 2010; Sedwick et al. 2007). It is also important to mention that the concept of aerosol-Fe solubility is applicable only for remote oceanic regions where the chemical composition of wet-precipitation events approaches detection limits and is comparable to de-ionized water. Due to the relatively low pH of de-ionized water (~5.6), aerosol iron solubility may be somewhat higher than in alkaline sea water (pH ~8.1). In this study, detection limits for Al, Fe, $Fe_{ws,}$ Cd, and Pb, defined as three times the standard deviation of filter blanks ($n = 7$) and normalized to average volume of air filtered (~1,400 m^3), are 80, 11, 0.15, 0.035, and 0.13 ng m^{-3}, respectively. The overall precision based on repeated measurements of samples and standards is better than 10%. Variability in the blank concentration of respective constituent account for less than 5% of the minimum concentration measured during the study. The estimated analytical uncertainties in our measurements of Fe_{ws} (±10%) and Fe_{Tot} (±5%) yield an overall relative uncertainty of about 11% on the assessment of fractional solubility of aerosol iron ($Fe_{ws}\% = Fe_{ws}/Fe_{Tot} \times 100$; Fe_{ws} = water soluble Fe; Fe_{Tot} = total aerosol-Fe).

Supporting parameters

Water soluble ionic constituents were analyzed on a Dionex-500 Ion Chromatography system equipped with suppressed conductivity detection. Carbonaceous species (EC, OC) were measured using a Sunset EC-OC analyzer based on NIOSH protocol (Birch and Cary 1996). The fractions of non-sea-salt-sulfate (nss-SO_4^{2-}) and K^+ (nss-K^+) were obtained by subtracting the contribution of sea salt from the measured SO_4^{2-}

and K^+ concentrations (nss-SO_4^{2-} = SO_4^{2-} −0.253 × Na^+; nss-K^+ = K^+ −0.037 × Na^+; where SO_4^{2-}, Na^+, and K^+ are the measured concentrations in aerosol samples; 0.253 and 0.037 are SO_4^{2-}/Na^+, K^+/Na^+ ratio in sea-salt; (Keene et al. 1986)). For detailed analytical protocols, reference is made to (Kumar et al. 2010; Rengarajan et al. 2007). Although, $PM_{2.5}$ samples were collected during January 2009 and analyzed for Fe_{ws} and Fe_{Tot}, we only used PM_{10} samples for comparison of the aerosol-Fe data during March–May 2006 over the Bay of Bengal and the Arabian Sea.

Results and discussion

Air mass back trajectory analysis

Air mass back trajectory (AMBT) analyses are useful for identifying the source regions that contribute to aerosol chemical composition over sampling sites. During January 2009, 7-days AMBTs were computed for the arrival height at 100, 500, and 1000 m from the National Oceanic and Atmospheric Administration (NOAA) GDAS database using the hybrid single-particle lagrangian integrated trajectories (HYSPLIT) program (Draxler 2002). The analysis of AMBTs reveals that the northern part of the Bay of Bengal (N-BoB) was influenced by the continental outflow from the Indo-Gangetic Plain (between 27 December 2008 and 10 January 2009, here after referred as IGP-outflow) and that the south Bay of Bengal (S-BoB) was influenced by the outflow from south-east Asia (between 11 and 28 January 2009, here after referred as SEA-outflow). A typical air mass back trajectory cluster at arrival height of 500 m over the Bay of Bengal is presented in Fig. 1a. Aerosol samples collected from the Bay of Bengal during 19–31 March 2006 (except 30 March 2006) are influenced by the IGP-outflow. During 1–12 April 2006, AMBTs over the Bay of Bengal show localized pattern (i.e., recirculating within the MABL; Fig. 1b). Over the Arabian Sea, AMBTs show origin from desert dust regions (Kumar et al. 2008a).

Aerosol iron solubility

The mass concentration of total aerosol iron (Fe_{Tot}) in the MABL of the Bay of Bengal and the Arabian Sea during the study period (Table 1) overlaps within the spread of the data (Bay of Bengal: 31–1,651 ng m^{-3}; Arabian Sea: 92–1,010 ng m^{-3}). However, the fractional solubility of aerosol iron (Fe_{ws} %) is significantly different over the Arabian Sea (0.02–0.4%) and the Bay of Bengal (1.4–24.0%). Table 1 presents the concentrations of total aerosol-Fe (Fe_{Tot}) and fractional solubility of aerosol iron (Fe_{ws} %) for the two study regions. The significant difference in Fe_{ws} (%) between the two oceanic regions is attributed to differences in the chemical characteristics of mineral dust. The Arabian Sea receives coarse dust from the surrounding desert regions (Saharan and Arab Deserts on the western side, Thar Desert on the eastern side), whereas transport of fine alluvial dust from the Indo-Gangetic Plain is a dominant source to the Bay of Bengal.

The mineralogical composition of soils adjoining the Thar Desert, situated in the northwestern part of India, suggests that quartz is the most abundant mineral followed by K-feldspar, mica, calcite, chlorite, and plagioclase (Yadav and Rajamani 2004). In addition, some heavy minerals (granite, amphibole, titanite and zircon) have also been identified. In contrast, the Indo-Gangetic Plain is characterized by alluvial soils (in the flood plains of major rivers), mostly granitic in nature with 84–85% quartz, 8–11% of feldspars, and 4–7% of mica (Tripathi et al. 2007). Despite the mineralogical differences, the physical size of the mineral dust is responsible for the higher Fe_{ws} (%) over the Bay of Bengal. Based on the chemical characterization of aerosols, earlier studies (Kumar et al. 2008b; Lelieveld et al. 2001; Sudheer and Sarin 2008) have highlighted the impact of continental outflow on the MABL of the Bay of Bengal during late NE-monsoon (January–April). In addition, during long-range transport, atmospheric processing of mineral dust by anthropogenic acidic species (HNO_3 and H_2SO_4) can enhance the fractional solubility of aerosol iron (Kumar et al. 2010; Meskhidze et al. 2003; Solmon et al. 2009). The enhanced fractional solubility of aerosol-Fe over the Bay of Bengal, as compared to that over the Arabian Sea, can also be attributed to the chemical processing of fine alluvial dust. In PM_{10}, Fe_{Tot} and Fe_{ws} show pronounced and similar temporal variability, suggesting a common source. The scatter plot of mass concentrations of Fe_{Tot} and Fe_{ws} in $PM_{2.5}$ against PM_{10} suggests the coarse mode nature of Fe_{Tot} and fine mode

Table 1 Aerosol iron and its fractional solubility over the Arabian Sea and the Bay of Bengal

Arabian Sea			Bay of Bengal					
TSP ($N = 19$, April–May 2006)			TSP ($N = 23$, March–April 2006)			PM_{10} ($N = 33$, January 2009)		
Sample id	Fe_{Tot}[a]	Fe_{WS} (%)	Sample id	Fe_{Tot}[a]	Fe_{WS} (%)	Sample id	Fe_{Tot}[a]	Fe_{WS} (%)
AS-501	1,010	0.05	AS-402	807	5.3	AS-802	909	7.6
AS-503	976	0.02	AS-403	1,116	4.3	AS-804	667	5.8
AS-504	648	0.08	AS-405	799	6.2	AS-806	430	6.8
AS-506	315	0.04	AS-406	938	4.3	AS-808	773	4.3
AS-507	364	0.11	AS-408	1,010	6.6	AS-810	1,317	6.0
AS-508	381	0.03	AS-409	1,221	4.2	AS-812	1,651	3.6
AS-510	434	0.09	AS-410	946	7.0	AS-814	1,649	3.3
AS-511	450	0.02	AS-412	706	3.2	AS-816	493	2.5
AS-512	953	0.04	AS-413	546	3.8	AS-818	198	4.7
AS-513	452	0.22	AS-414	600	3.8	AS-820	127	9.4
AS-517	232	0.04	AS-416	421	12.0	AS-822	254	6.9
AS-518	92	0.43	AS-417	420	7.2	AS-824	1,270	6.2
AS-520	327	0.08	AS-418	272	6.8	AS-826	898	7.3
AS-521	298	0.12	AS-420	242	5.6	AS-828	461	1.4
AS-522	255	0.03	AS-421	245	13.6	AS-830	213	5.2
AS-524	153	0.03	AS-422	668	2.9	AS-832	339	7.5
AS-525	296	0.19	AS-424	184	5.2	AS-834	382	14.6
AS-527	432	0.07	AS-425	150	11.1	AS-836	355	8.6
AS-528	372	0.02	AS-426	309	13.1	AS-838	329	5.7
			AS-428	294	3.5	AS-840	306	12.7
			AS-429	527	2.6	AS-842	260	9.3
			AS-430	547	2.3	AS-844	254	7.1
			AS-431	595	2.6	AS-846	230	10.4
						AS-848	31	3.2
						AS-850	95	23.9
						AS-852	210	11.9
						AS-854	196	3.5
						AS-856	220	2.7
						AS-858	169	6.9
						AS-860	96	6.6
						AS-862	113	3.1
						AS-863	159	5.2
						AS-864	91	3.5

[a] Concentration (ng m^{-3})

Fe_{ws}(%) fractional solubility of total aerosol-Fe (Fe_{Tot}) defined as $Fe_{ws}/Fe_{Tot} \times 100$ and Fe_{ws} is water soluble Fe

existence of Fe_{ws} (Fig. 2). Furthermore, both Fe_{ws} and Fe_{Tot} show spatial variability with systematically higher concentrations over N-BoB than over S-BoB. The relatively high concentrations of Fe_{Tot} and Fe_{ws} are consistent with the high aerosol loading over N-BoB due to the proximity of the continental sources.

The relatively low fractional solubility of aerosol iron (Fe_{ws} %) over the Arabian Sea is consistent with earlier studies (Siefert et al. 1999). In the coarse mode (>3 μm) aerosols over the Arabian Sea, Siefert et al. (1999) have shown that the contribution of Fe(II) to total aerosol iron (Fe_{Tot}) is no more than 2% during

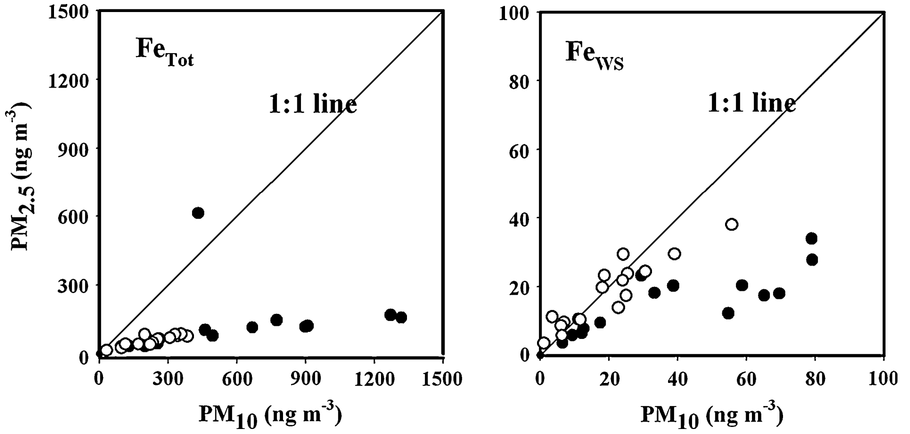

Fig. 2 Scatter plots for aerosol iron (Fe_{Tot}) and water soluble iron (Fe_{ws}) indicating their relative distribution in $PM_{2.5}$ and PM_{10} over the Bay of Bengal (January 2009). *Close* and *open circles* represent samples collected from N-BoB and S-BoB, respectively

spring-intermonsoon (May 1995). The low iron solubility over the Arabian Sea, as documented in the present study, can be explained by a lack of atmospheric processing of coarser desert dust (quartz as the abundant mineral) with anthropogenic acidic species. Furthermore, acid mobilization of iron from mineral aerosols is not favoured due to relatively high abundance of $CaCO_3$. It has been reported that $CaCO_3$ is the major neutralizing constituent over the Arabian Sea (Kumar et al. 2008a, b). This would constrain the availability of acidic species (H_2SO_4) to mobilize iron from mineral dust. Total aerosol iron (Fe_{Tot}) and water soluble iron (Fe_{ws}) over the Arabian Sea and the Bay of Bengal are plotted on a logarithmic scale with 0.01, 1, 5, 10, 25, and 50% solubility (Fig. 3). This plot shows that the solubility over the Arabian Sea and the Bay of Bengal is in the range of 0.01–1 and 1–50%, respectively.

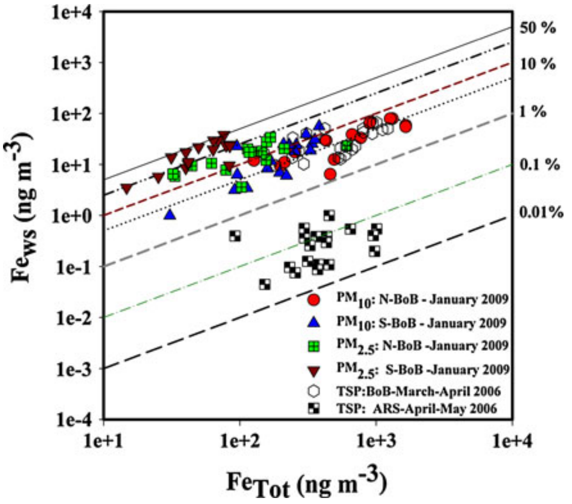

Fig. 3 Total aerosol-Fe (Fe_{Tot}) and water soluble Fe (Fe_{ws}), over the Arabian Sea (*ARS*) and the Bay of Bengal (*BoB*), on logarithmic scale with 0.01, 0.1, 1, 5, 10, 25 and 50 percent solubility lines. It is clearly evident that the solubility over the Arabian Sea is in the range of 0.01 – 1 and 1 – 50% over the Bay of Bengal

Evidence for chemical processing of mineral dust

The fractional solubility of aerosol iron (Fe_{ws} %) in PM_{10}, during January 2009, varied from 1.4 to 9.4% and 2.7 to 23.9% over N-BoB and S-BoB, respectively. In addition, our results show a linear relationship between Fe_{ws} (%) and Fe_{Tot} ($R^2 = 0.40$; $n = 17$; P-value $= 0.006$) and nss-SO_4^{2-} ($R^2 = 0.53$; $n = 17$; P-value $= 0.001$; Fig. 4) over S-BoB, suggesting the role of acid uptake by mineral aerosols in enhancing the fractional solubility of aerosol iron. Note that one outlier (aerosol sample collected on 20 January 2009) was excluded from this analysis. In contrast, we found no significant correlation for the chemical data from N-BoB (Fig. 4). The linear relationship over S-BoB can therefore be explained in terms of mixing state of aerosols. Although direct information on the mixing state of aerosols over the Bay of Bengal is not available from our field campaign, it has been suggested that the extent of internal mixing is enhanced with the aging of aerosol population (Baker and Croot 2010). The aging of aerosols can be invoked for the air masses associated with the outflow from south-east Asia (Fig. 1a). This hypothesis is further supported by the average equivalent ratio of (NH_4^+ + Ca^{2+})/nss-SO_4^{2-}, which is about 0.86 ± 0.23 for N-BoB and 0.43 ± 0.26 for S-BoB, suggesting

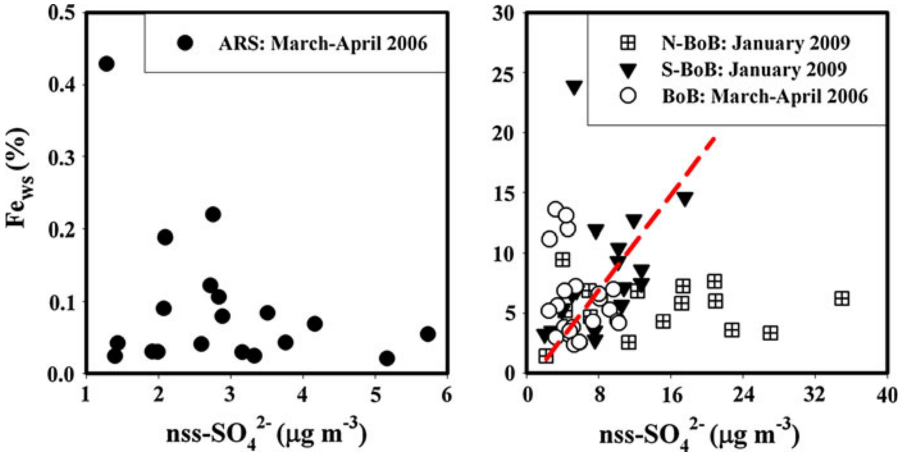

Fig. 4 Scatter plot of fractional solubility of aerosol iron (Fe_{ws} %) versus $nss-SO_4^{2-}$ from the north and south Bay of Bengal (*N-BoB*, *S-BoB*; PM_{10}) and the Arabian Sea (*ARS*, TSP samples)

relatively high acidity over S-BoB. Likewise, a significant linear relation between total acidity ($TA = nss-SO_4^{2-} + NO_3^{-}$, in ng m^{-3}) and Fe_{ws} % ($R^2 = 0.40$; $n = 17$; P-value < 0.001) over S-BoB provides further support to the hypothesis that acid processing of mineral dust had enhanced the fractional solubility of aerosol iron.

It is noteworthy that the mass concentration of mineral dust was relatively high over N-BoB owing to close proximity of continental sources. The contribution of Fe from anthropogenic combustion sources is not expected to be significant in PM_{10} mass fraction. This is also evident from the data that PM_{10} contains 65% of $PM_{2.5}$ mass. Therefore, in order to investigate the fractional solubility of aerosol iron, we have analyzed soluble iron in $PM_{2.5}$ samples. In $PM_{2.5}$, the fractional solubility of aerosol iron (Fe_{ws} %) varied from 3.5 to 49.7% over the Bay of Bengal. It is noteworthy that the solubility in the fine ($PM_{2.5}$) mode approaches 50%, emphasizing the dominant role of combustion sources in enhancing the fractional solubility of aerosol-Fe. Earlier studies have suggested that the interrelationships of soluble Fe, $nss-K^+$, OC, EC, and anthropogenic constituents in fine mode aerosols provide direct evidence for their common source signature in the continental outflow (Chuang et al. 2005; Kumar et al. 2010; Paris et al. 2010).

Evidence for biomass burning source

The $nss-K^+$ is used as a diagnostic tracer of biomass burning, its concentration is reported to be high in biomass derived aerosols. Evidence for this is constrained in the emission of K^+ and EC, OC during biomass burning emissions (Andreae 1983). Over the Bay of Bengal, during January 2009, $nss-K^+$ ranged from 0.1 to 1.3 (Av = 0.60 ± 0.30) µg m^{-3} and accounts for ~3% of the total water soluble ion species (ΣWSIS) in PM_{10} fraction. Likewise, $nss-K^+$ concentration varied from 0.1 to 0.4 µg m^{-3} in bulk aerosols collected during March–April 2006. In contrast, the contribution of $nss-K^+$ to ΣWSIS is insignificant over the Arabian Sea during April–May 2006. In addition to mineral dust, biomass burning is also identified as a potential source of soluble iron to surface sea water (Guieu et al. 2005; Paris et al. 2010). In this study, the significant linear correlations between $nss-K^+$ and OC during March–April 2006 ($R^2 = 0.82$; $n = 23$; P-value < 0.01) and January 2009 ($R^2 = 0.85$; P-value < 0.001 for $n = 33$) over the Bay of Bengal suggest a significant contribution of biomass burning. A similar correlation between Fe_{ws} and OC ($R^2 = 0.63$; P-value < 0.001; $n = 33$) and $nss-K^+$ ($R^2 = 0.61$; P-value < 0.001; $n = 33$) during January 2009 further supports biomass burning emissions as a possible source of soluble iron. A similar evidence for contribution from biomass burning emissions is seen in the data for March–April 2006.

In $PM_{2.5}$ and PM_{10} size fractions, the average $nss-K^+/Fe_{Tot}$ ratio is 6.8 ± 2.5 and 2.0 ± 1.3, respectively. These ratios are significantly high compared to those reported for the upper continental crust (0.49; Wedepohl 1995) and for other oceanic regions (Mediterranean Sea: 0.63 ± 0.12; Guieu et al. 2005). The biomass burning emissions from agricultural crop waste (rice and wheat straw), associated with the outflow from Indo-Gangetic Plain, are responsible for the high $nss-K^+$ concentration and thus high $nss-K^+/Fe_{Tot}$.

Furthermore, rice straw contains nearly 65% of soluble K^+, thus providing evidence for bio-mass burning as a potential source (Pal 2009). In contrast, nss-K^+/Fe_{Tot} in samples collected during March–April 2006 is relatively low (average: 0.39 ± 0.11) compared to the ones measured in January 2009. The presence of high mineral dust during March–April 2006 would also lead to low ratio of nss-K^+/Fe_{Tot}. A comparison of the data from cruises in March–April 2006 and January 2009 suggest that samples from the latter cruise are characterized by high concentrations of nss-K^+, nss-SO_4^{2-}, Fe_{ws}, and OC. The impact of biomass burning emissions is dominant during wintertime in the Indo-Gangetic Plain and hence long-range transport of chemical constituents is important over the Bay of Bengal. The high Fe_{ws} (%) in $PM_{2.5}$ over the Bay of Bengal is attributed to fine dust with high surface area to volume ratio which is conducive for the uptake of acidic species. In addition, the contribution from biomass burning sources is discernible based on linear relation among nss-K^+, OC, and Fe_{ws}.

Although the magnitude of Fe_{ws} from combustion sources (biomass burning and fossil-fuel combustion) is not significant compared to that from mineral aerosols, the enhanced fractional solubility of aerosol iron (Fe_{ws} %) is an important issue for the global models. The spatial variability in the fractional solubility of aerosol iron over the Bay of Bengal and the Arabian Sea is presented in Fig. 5. These contours are generated using Ocean Data View software (Schlitzer 2002) and are representative for the entire sampling period. This plot shows relatively low solubility of iron over the eastern Arabian Sea compared to that over western Arabian Sea. A comparison of the fractional solubility of aerosol iron from different oceanic and continental regions is presented in Table 2. The fractional solubility of aerosol iron over the Bay of Bengal is relatively high

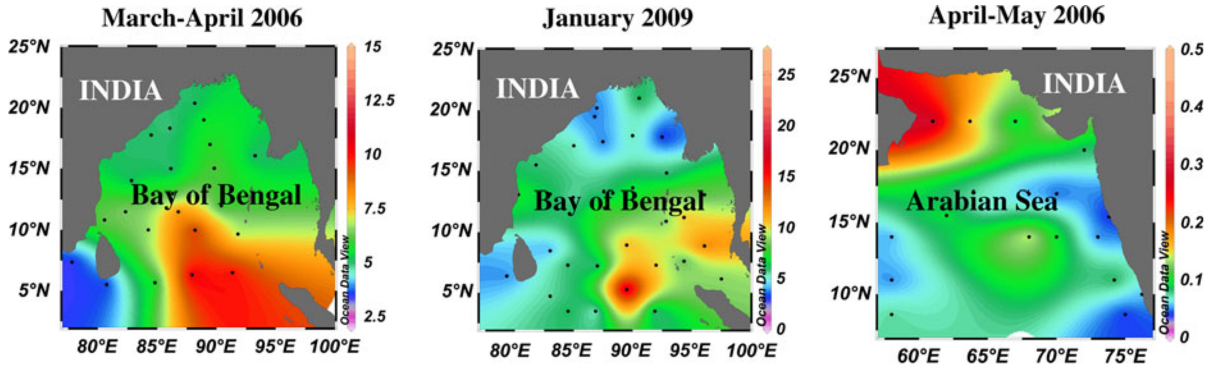

Fig. 5 Spatial variability of fractional solubility of aerosol iron (Fe_{ws} %) over the Bay of Bengal and the Arabian Sea

Table 2 Fractional solubility of aerosol iron for different oceanic regions

Region	Type	Duration	Fe_{Tot}	Fe_{ws}	Fe_{ws} (%)	Reference
Bay of Bengal	$PM_{2.5}$	January 2009	98	16.9	23.4	Present study
Bay of Bengal	PM_{10}	January 2009	459	27.4	6.9	Present study
Bay of Bengal	TSP	March–April 2006	590	31.1	6.0	Present study
Arabian Sea	TSP	April–May 2006	444	0.3	0.1	Present study
North Atlantic Ocean	TSP	July–August 2003	653	–	1.4	Sholkovitz et al. (2009)
North Atlantic Ocean	TSP	April–June 2004	49	–	11.6	Sholkovitz et al. (2009)
North Pacific Ocean	TSP	May–June 2002	95	6.5	9.0	Buck et al. (2006)
$^{\$}$Mt. Abu (India)[a]	$PM_{2.5}$	March–May 2007	431	4.6	1.7	Kumar and Sarin (2010)
Mt. Abu (India)[a]	$PM_{2.5}$	October–November 2007	150	11.0	8.0	Kumar and Sarin (2010)

Fe_{ws} *(%)* fractional solubility of total aerosol-Fe (Fe_{Tot}) and Fe_{ws} is the water soluble iron

[a] A continental site in western India-source region of mineral dust to Arabian Sea

compared to North Atlantic and Pacific Ocean. The atmospheric processing of mineral dust during long-range transport in the presence of high abundance of anthropogenic acidic species and contribution from combustion sources are likely factors governing the high fractional solubility of aerosol-Fe. In contrast, the fractional solubility over the Arabian Sea is relatively low compared to the other oceanic regions.

Mixing model for fractional solubility

In $PM_{2.5}$, the fractional solubility of aerosol iron varied from 3.5 to 21.2 and 11.4 to 49.7% over N-BoB and S-BoB. It is noteworthy that N-BoB is characterized by high aerosol-Fe (Fe_{Tot}) concentration but low fractional solubility (Fe_{ws} %), representing one end member with contribution from mineral dust; Aerosols over S-BoB are characterized by higher Fe_{ws} (%) associated with low concentration of aerosol iron (Fe_{Tot}). The scatter plot (Fig. 2) of Fe_{Tot} between $PM_{2.5}$ and PM_{10} suggest the predominant existence of total aerosol iron in the coarse mode (i.e., contribution from mineral dust). The abundance of mineral dust in PM_{10} can account for 33% of the average particulate mass concentration; whereas its contribution to $PM_{2.5}$ is relatively low. Therefore, anthropogenic sources can contribute significantly to Fe_{Tot} in the fine mode. In this regard, the inverse relationship observed between Fe_{ws} (%) and Fe_{Tot} (Fig. 6) can be explained by the mixing of the two end member (mineral dust and combustion sources).

A similar inverse relationship has been used in the North Atlantic to explain the fractional solubility of aerosol iron (Fe_{ws} %), where advection of Saharan desert dust and air mass from North America contribute significantly to the particulate loading (Sedwick et al. 2007). A similar approach of two end member mixing has been used to explain the fractional solubility of aerosol-Fe from a high altitude site (Mt. Abu) in western India, a source region of mineral dust to the Arabian Sea (Kumar and Sarin 2010). A scatter plot and linear relation between $1/Fe_{Tot}$ and Fe_{ws} % ($r = 0.66$; P-value < 0.001; except for two outliers excluded for low PM_{10} concentration $\leq 6\ \mu g\ m^{-3}$), provide further support for the existence of two end members in the study area (see Fig. 6). The mixing curve is generated by assuming that one end member is characterized by Fe_{Tot} concentration of 1,651 ng m^{-3} with fractional solubility of 3.6% (high Fe_{Tot}, low Fe_{ws} %; representative of dust) and the other end member is characterized by 95 ng m^{-3} with a solubility of 24.0% (low Fe_{Tot}, high Fe_{ws} %, representative of combustion sources).

Deposition flux of Fe_{ws}

The air–sea deposition of soluble iron is estimated from the soluble component (Fe_{ws}) of aerosol-Fe

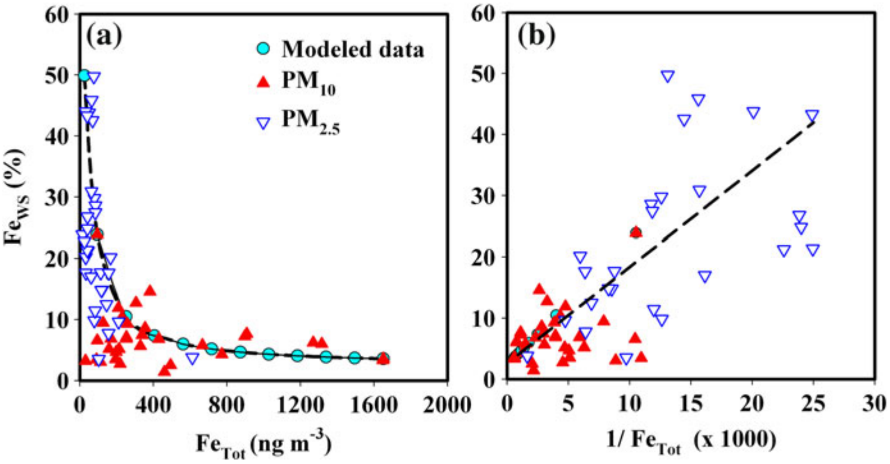

Fig. 6 **a** Fractional solubility of aerosol iron (Fe_{ws} %) in $PM_{2.5}$ and PM_{10} plotted against total aerosol-Fe (Fe_{Tot}). Modeled data points (this study) and hyperbolic relationship (*dash line*) is obtained by a two end member mixing 1 Dust: with high Fe_{Tot} with low Fe_{ws} %. 2 Combustion sources: low Fe_{Tot} with high Fe_{ws} (%). **b** Linear increase of Fe_{ws} % with $1/Fe_{Tot}$, further establishes that fractional solubility of aerosol-Fe can be explained by two end member mixing (mineral dust and anthropogenic combustion sources)

(Fe_{Tot}) multiplied by the dry-deposition velocity; assuming that the fractional solubility after deposition of mineral dust and/or combustion derived products is comparable with the solubility derived from laboratory studies. This can be expressed by the following equation. The deposition flux (f_d) = $Fe_{ws} \times V_d$; Here V_d = dry-deposition velocity, which is assumed as 1.0 cm s^{-1} in the present study (Jickells and Spokes 2001). The soluble iron flux varied from 8.2 to 57.5 µg m^{-2} day^{-1} over the Bay of Bengal and 0.04 to 0.9 µg m^{-2} day^{-1} over the Arabian Sea for the campaign in March–May 2006. Likewise, during January 2009, the dry-deposition of soluble iron (Fe_{ws}) in PM_{10} aerosols ranged from 0.9 to 68.4 (23.7 ± 19.9) µg m^{-2} day^{-1} over the Bay of Bengal. The fractional solubility of aerosol iron over the Arabian Sea is about 0.1%, suggesting that the availability of soluble iron is two orders of magnitude lower than estimated by earlier studies (Kumar et al. 2008a, b). Although the fractional solubility of aerosol iron over Arabian Sea is low, the amount of dust input to the surface waters from surrounding desert regions compensate for the poor solubility. Our results indicate that regional-scale studies could provide better insight in understanding the relative importance of different sources. The air-sea deposition of soluble iron associated with high fractional solubility of dust and/or combustion derived products can increase the total pool of iron in seawater with a potential stimulating effect on primary production.

Impact on ocean surface biogeochemistry

The surface waters of the Bay of Bengal and eastern Arabian Sea are relatively replete in iron compared to the western Arabian Sea (Wiggert et al. 2006). However, iron limitation has been reported in the surface waters of the central Arabian Sea during spring-intermonsoon (Naqvi et al. 2010; Wiggert et al. 2006). Atmospheric deposition of soluble iron to seawater in this region can be thus important ecologically. By stimulating algal growth, the atmospheric input of iron can also increase dimethylsulfide (DMS) production, as demonstrated during the large scale iron fertilization experiments conducted in the Southern Ocean (Turner et al. 1996).

Earlier studies have documented the dominance of diatoms (80–90%) during winter (December–February) and spring-intermonsoon (March–May) period (Madhu et al. 2006). Diatoms also dominate the algal assemblage (86%) in the Arabian Sea (Sawant and Madhupratap 1996). In the following section, we have estimated the impact of the atmospheric deposition of soluble iron on carbon fixation by diatoms in these waters.

In the Bay of Bengal, aerosol concentration of Fe_{Tot} averages around 460 ng m^{-3}. Assuming a dry-deposition velocity of 0.01 m s^{-1} over an area of 2.2×10^{12} m^2, the atmospheric deposition of total aerosol iron (Fe_{Tot}) is ~0.32 Tg $year^{-1}$. Based on the average fractional solubility of aerosol iron (Fe_{ws} %) taken as ~7%, the deposition of Fe_{ws} to the Bay of Bengal is 0.022 Tg $year^{-1}$. The mean cellular ratio of Fe: C for the diatoms is ~8 µmol/mol (Moore et al. 2002; Okin et al. 2011). Therefore, the amount of carbon fixed by diatoms can be calculated by the following equation.

$$C_{dia}^{Fix} = (Fe_{WS})_{atm} \times \left[\frac{C}{Fe}\right]_{dia}$$

Here C_{dia}^{Fix}, $(Fe_{WS})_{atm}$ and $\left[\frac{C}{Fe}\right]_{dia}$ correspond to the amount of carbon fixed by the diatoms, the magnitude of atmospheric deposition of Fe_{ws} and the cellular uptake ratio of C:Fe by diatoms, respectively. Our data suggest that the deposition of Fe_{ws} can support primary production of ~2.7 Pg-C $year^{-1}$ in the Bay of Bengal. Similarly, the amount of carbon fixed by the diatoms in the Arabian Sea is estimated as ~0.09 Pg-C $year^{-1}$ [Fe_{Tot}: 444 ng m^{-3}; Fe_{ws} (%) = 0.1%; Integrated area of Arabian Sea = 4.93×10^{12} m^2 (Bange et al. 2000)].

Conclusions

The results obtained in this study highlight the role of anthropogenic sources in modifying the atmospheric supply of soluble iron to the surface ocean. The fractional solubility of aerosol iron is 1–2 orders of magnitude higher in the MABL of the Bay of Bengal compared to that in the Arabian Sea. This difference is attributed to the chemical characteristics of the mineral dust reaching the two oceanic regions. The Arabian Sea receives coarse dust from the desert regions; whereas transport of alluvial dust from the Indo-Gangetic Plain is a dominant source to the Bay of Bengal. Furthermore, a linear relationship between Fe_{ws} %, Fe_{Tot}, and nss-SO_4^{2-} provides evidence for the chemical processing of alluvial dust by atmospheric acidic constituents (H_2SO_4, HNO_3) during long-range transport and thus enhance the solubility of aerosol-Fe. In addition, supply of soluble iron from combustion sources (agricultural crop waste, biomass-biofuel burning and fossil–fuel combustion) is demonstrated based on high fractional solubility of aerosol iron over the Bay of Bengal. Our results suggest that dust deposition can significantly increase photosynthetic carbon fixation and potentially DMS production in the central Arabian Sea in spring-intermonsoon time when iron could be limiting for phytoplankton growth.

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