

# Aerosol iron solubility in a semi-arid region: temporal trend and impact of anthropogenic sources

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## ABSTRACT

A systematic 1-yr chemical data for the fine mode (PM<sub>2.5</sub>) air-borne particulate matter, collected from a high-altitude site (Mt. Abu, 22.7°N, 74.6°E, 1680 m asl) in a semi-arid region of western India, reveal characteristic temporal variability in the abundance of mineral dust and fractional solubility of aerosol Fe (referred as water-soluble Fe). A notable feature of the data is seen from an inverse relationship between aerosol Fe (range: 50–915 ng m<sup>-3</sup>) and fractional Fe solubility (range: 0.06–16.1%). A significant increase in the solubility of iron, during wintertime, is also marked by a uniform decrease in the mass fraction of mineral dust. These observations suggest that the advective transport of pollutants and Fe derived from combustion sources is one of the possible causes for the enhanced solubility of iron over a semi-arid region in western India. The relative dominance of anthropogenic sources, during wintertime, is also supported by a two to threefold increase in NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup> as well as enrichment of heavy metals in PM<sub>2.5</sub> (ascertained from Pb/Al and Cd/Al ratios).

## 1. Introduction

The atmospheric transport and deposition of mineral dust is considered as one of the major sources by which iron is supplied to the ocean surface (Duce and Tindale, 1991; Fung et al., 2000; Jickells et al., 2005; Mahowald et al., 2005, 2009). Also, iron is hypothesized as a limiting nutrient in controlling the phytoplankton production in the high-nutrient-low-chlorophyll (HNLC) regions (Martin et al., 1991). These are recognized as potential regions in modulating the CO<sub>2</sub> uptake due to the increase in phytoplankton productivity and thus, affecting the global carbon cycle and climate (Meskhidze et al., 2003; Jickell et al., 2005; Solmon et al., 2009). On average, dust aerosols contain 3.5% of iron mainly existing in the form of aluminosilicate (Duce and Tindale, 1991; Zhu et al., 1997). The aerosol Fe associated with mineral dust remains insoluble at pH conditions typical of sea water (Stumm and Morgan, 1981; Zhu et al., 1997). However, a finite fraction of the aerosol Fe [referred as dissolved iron], is used by phytoplanktons. Several field based studies, incorporating different analytical techniques, have documented large variability (0.01–80%) in the water-soluble fraction of aerosol iron and continues to remain a major source of uncertainty in assessing the impact of atmospheric supply of iron to the marine ecosystem and the biogeochemical cycle (Chen and Siefert,

2004; Hand et al., 2004; Mahowald et al., 2005; Baker et al., 2006).

One of the proposed mechanisms for the enhanced solubility of iron is the chemical processing of aerosol particles, during long-range transport, mediated by the presence of acidic components (e.g. SO<sub>2</sub> and its oxidation products) and sunlight (Zhuang et al., 1992; Johansen et al., 2000; Meskhidze et al., 2003). On the contrary, some of the field experiments have indicated that the soluble iron can be enriched as a result of direct emission from fossil-fuel combustion and biomass burning (Chuang et al., 2005; Sedwick et al., 2007; Sholkovitz et al., 2009). The mixing state of atmospheric aerosols is also considered as one of the important factors affecting the fractional solubility of aerosol Fe (Baker and Croot, 2008). In the source regions, aerosol particles are represented as a mixture of both externally and/or internally mixed state, which acquire internally mixed state during long-range transport. The condensation of gaseous species (on pre-existing particles) as well as in-cloud processing of aerosols is responsible for the increase in internal mixing state, which is populated in the fine mode. Baker and Jickells (2006), have argued that the increase in aerosol iron solubility is influenced by the decrease in particle size (i.e. increase in surface area to volume ratio) during atmospheric transport rather than differences in the composition of aerosols. On the contrary, Buck et al. (2008a) have demonstrated that there is no such obvious correlation and that aerosol composition from the source region is a dominant factor in controlling the iron solubility.

It is, thus, imperative to investigate and understand various sources and factors affecting the fractional solubility of aerosol

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iron at regional scale in order to further constrain its role in the biogeochemical models (Mahowald et al., 2009). In this context, chemical data from high-dust semi-arid regions of south-Asia are lacking in the literature. With this motivation, fine mode ( $PM_{2.5}$ ) aerosol samples were collected from a high-altitude site located in a semi-arid region of western-India, an important source region for the atmospheric supply of dust to the Arabian Sea. Sampling from a high-altitude site is considered to be advantageous in order to study the long-range transport of mineral dust across western India (Kumar and Sarin, 2009). In this manuscript we report on the atmospheric abundance of mineral dust and fractional solubility of iron in the fine mode aerosols ( $PM_{2.5}$ ). We have also evaluated the temporal trend and impact of anthropogenic sources affecting the water-soluble fraction of iron.

## 2. Sampling site and experiments

The sampling site was located at Gurushikhar—a remote mountain peak at the southern end of Aravali range (hereafter re-

ferred as Mt. Abu,  $22.7^{\circ}N$ ,  $74.6^{\circ}E$ , 1680 m asl) in western India. The proximity of the site to the great Thar Desert ( $\sim 300$  km) provides an ideal opportunity to study the atmospheric transport of mineral aerosols and temporal variability over western India (Fig. 1). The annual rainfall at Mt. Abu averages around 600–700 mm and all of it occurs during the period of southwest monsoon (June–September). Under the influence of the monsoonal winds, substantial amount of moisture and sea salts are transported inland to the sampling site. In contrast, northeast winds during wintertime favour long-range transport of anthropogenic chemical constituents from northern India (Kumar and Sarin, 2009). The northern part of India (Indo-gangetic Plain), located upwind of the sampling site (Fig. 1), is densely populated area with diverse types of industries (textiles, sports, leather goods and power plants). In the urban regions, aerosol emissions from combustion of fossil-fuels (viz. coal, petrol and diesel oil used in electrical power generation, oil refining, iron and steel production, domestic heating and transportation) are dominant. The use of leaded gasoline and inefficient two-stroke engines are still in use in the densely populated regions of northern India.

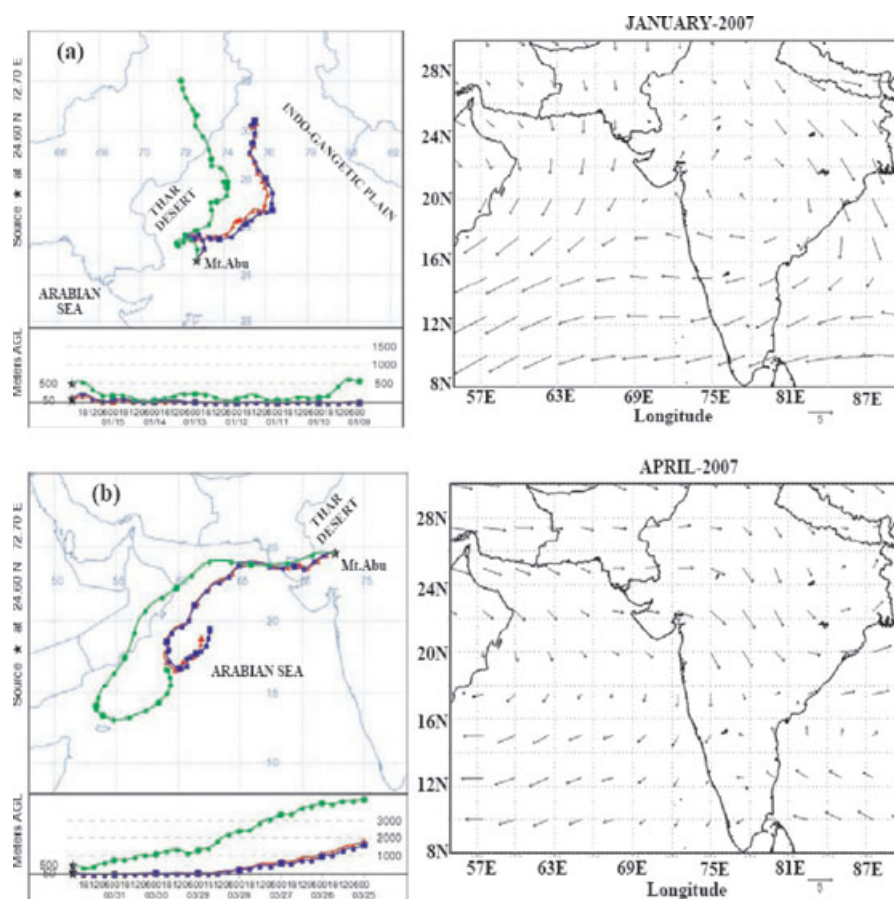


Fig. 1. Map showing sampling site (Mt. Abu) and important source regions influencing the transport of aerosols. Typical 7-d air mass back trajectories (using HYSPLIT model; <http://www.arl.noaa.gov/HYSPLIT.php>) at the study site during (a) winter: 16 January 2007 and (b) summer: 01 April 2007. Also shown are the monthly average wind regimes during January (representing the winter season) and April (representative of summer season).

In the rural areas, biomass burning (agricultural crop waste) and combustion of biofuels is most common. The boundary-layer dynamics, during winter months, play an important role in efficient trapping of the pollutants and their advective transport away from the source region. It is emphasized that the sampling site is reasonably free from local pollution sources.

In the regional context of western India and based on the meteorological parameter, two distinct seasons have been defined (Fig. 1), viz. summer (March–May) characterized by convective mixing and high-dust loading in the atmosphere and winter (October–February) when the advective transport of pollutants from northern India dominates the aerosol composition. Typical monthly average wind pattern for the month of January (representing the winter season) and April (representing the summer season) are shown in Fig. 1. The remainder months (June–September) represent the wet phase (monsoonal rain) with efficient washout of the atmosphere. The concept of spring season is not strictly relevant in this tropical semi-arid region.

Systematic sampling of  $PM_{2.5}$  was carried out over 12 months (January–December, 2007, covering the complete annual seasonal cycle) by operating a Hi-vol sampler (Thermo Andersen) fitted with inlet having 50% cut-off size for  $2.5\ \mu m$  aerodynamic diameter particles. The Hi-vol sampler was equipped with a volume-flow controller to maintain a constant flow rate. A periodic calibration of sampler was performed (every 2–3 weeks) to check on the variations, in the flow rate if any. Typically, the flow rate varied from  $1.10$  to  $1.15\ m^3\ min^{-1}$  with an uncertainty of 5%. The aerosol samples were analysed for a range of chemical constituents, viz. crustal elements (Al, Fe, Ca and Mg) and water-soluble ionic constituents ( $Na^+$ ,  $NH_4^+$ ,  $K^+$ ,  $Mg^{2+}$ ,  $Ca^{2+}$ ,  $Cl^-$ ,  $NO_3^-$ ,  $SO_4^{2-}$  and  $HCO_3^-$ ). The concentrations of crustal elements were analysed using ICP-AES (JobinYvon, ULTIMA-2) by complete digestion of aerosol samples (Kumar and Sarin, 2009). The anions ( $Cl^-$ ,  $NO_3^-$  and  $SO_4^{2-}$ ) and cations ( $Na^+$ ,  $NH_4^+$ ,  $K^+$ ,  $Mg^{2+}$  and  $Ca^{2+}$ ) were analysed on DIONEX<sup>®</sup> Ion-Chromatograph equipped with suppressed conductivity detector following the procedures described in earlier publications from our laboratory (Rastogi and Sarin, 2005; Rengarajan et al., 2007; Kumar et al., 2008).

The water-soluble iron (WS-Fe) was measured on Graphite Furnace-Atomic Absorption Spectrometer (P-E Model AAnalyst 100 coupled to a HGA 800). In the adopted analytical protocol, about  $15\ cm^2$  section of the sample filter (equivalent to 5-circular punches area =  $3.14\ cm^2$ ) was treated with 10 mL Milli-Q water in a 50 mL Savilex Teflon vial followed by an ultrasonic treatment for 15 min. Subsequently, water extract was filtered through  $0.4\ \mu m$  PTFE filter into a low density polyethylene vials and immediately acidified to  $pH < 2$  using double distilled  $HNO_3$ . The selection of Milli-Q water for WS-Fe, instead of seawater, is based on the rationale that Milli-Q water provides a consistent and reproducible leaching solution (Buck et al., 2006; Sedwick et al., 2007). The data on WS-Fe

reported in this manuscript represent relative amount of soluble Fe (also referred as ‘operational solubility’) as compared to absolute solubility. Sedwick et al. (2007), have attempted to estimate ‘effective solubility’ of aerosol iron over the Sargasso Sea from the ‘operational solubility’. In this study, Cd and Pb were also measured in the same aerosol samples in order to assess the impact of anthropogenic sources on the solubility of iron. Towards this, filter punches (ca. area =  $3.14\ cm^2$ ) were treated with HF and  $HNO_3$  in Teflon vials and digested in a microwave oven (ca. 100 bar pressure and ca.  $250\ ^\circ C$ ). The acid extract was made to a final volume of 15 mL and the concentration of Cd and Pb were measured on Heated-Graphite Furnace-Atomic Absorption Spectrophotometer. Several blank filters were also analysed along with the samples and blank levels were found to be less than 5% of the minimum concentration measured for WS-Fe, Cd and Pb.

### 3. Results and discussion

#### 3.1. Chemical characteristics of fine mode aerosols and temporal variability

The temporal variability in the mass concentration of fine mode ( $PM_{2.5}$ ) aerosols (range:  $5.5$ – $46.1\ \mu g\ m^{-3}$ ), during the two distinct seasons (summer and winter), is presented in Fig. 2a. A large variability is observed during wintertime compared to that in summer season and is attributed to the down-wind (northeast-winds) advective transport of anthropogenic species from emission sources in northern India. The mass fraction of dust (Fig. 2b) has been calculated using Al abundance in the aerosols, and assuming 8.04% of Al as composition of upper continental crust (McLennan, 2001; Rastogi and Sarin, 2006; Kumar and Sarin, 2009). The mass fraction of dust, plotted as a function of Julian days (Fig. 2b), makes significant contribution to the  $PM_{2.5}$  mass during summer time ( $Av = 56.9 \pm 20.6\%$ ) compared to winter months ( $Av = 18.1 \pm 12.5\%$ ), suggesting distinct sources of dust during the two seasons. This is further supported by the air-mass back trajectory analysis depicting the transport of air-masses from the two distinct regions during summer and winter seasons (Figs. 1a and b).

The elemental ratio of Fe/Al has been used to characterize the sources of dust over the study site similar to the approach used by Chiapello et al. (1997). The temporal trend in Fe/Al ratio (Fig. 2c) represents a large variability during winter months compared to the summer season. During the high dust season (summertime), Fe/Al ratios are in the narrow range of 0.55–0.80 (for ca. 85% of the data points), suggesting a characteristic nature of mineral aerosols derived from desert regions (western India). During wintertime, under the influence of prevailing northeasterlies, significantly higher Fe/Al ratios (range: 0.49–1.58; Fig. 2c) in the fine mode ( $PM_{2.5}$ ) are attributable to the widespread pollution sources in the Indo-Gangetic Plain (northern India) (Kumar and Sarin, 2009).

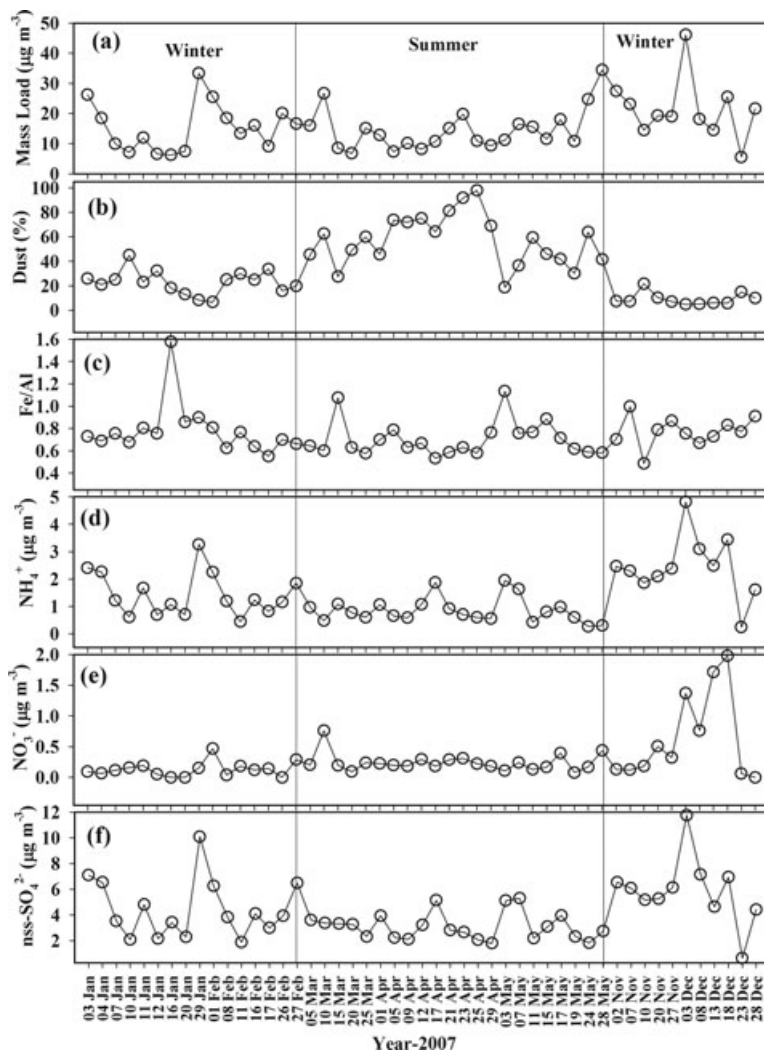


Fig. 2. Temporal variability during winter and summer seasons in 2007 at a high altitude station (Mt. Abu), for (a)  $\text{PM}_{2.5}$  aerosol mass, (b) mass fraction of estimated dust, (c)  $\text{Fe}_A/\text{Al}$  weight ratio, (d) concentrations of  $\text{NH}_4^+$  and (e)  $\text{NO}_3^-$  (f)  $\text{nss-SO}_4^{2-}$ .

Another line of evidence has been drawn based on the  $\text{Fe}/\text{Al}$  ratio ranging from 0.55 to 0.63 in the weathered soils from northern India, as inferred from the composition of river bank sediments (Sarin et al., 1979). It is, thus, proposed that significant contribution of Fe from mineral dust is unlikely and that the enrichment of  $\text{Fe}/\text{Al}$  ratio in  $\text{PM}_{2.5}$ , during wintertime, may be attributed to anthropogenic sources (fossil-fuel combustion and/or biomass burning). The dominance of biomass burning has been documented based on the  $\text{OC}/\text{EC}$  (organic carbon/elemental carbon) ratios as high as 7–8 in northern India (Rengarajan et al., 2007). The influence of anthropogenic sources on the chemical composition of fine mode aerosols is also well demonstrated by the temporal variability in the chemical constituents (e.g.  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{nss-SO}_4^{2-}$ ) as shown in Figs. 2d–f. The abundances of  $\text{NH}_4^+$  and  $\text{nss-SO}_4^{2-}$  exhibit parallel increase in proportion with the  $\text{PM}_{2.5}$  mass during winter months; re-emphasizing the long-range transport of pollutants from northern India (supported by the air-mass back-trajectories; Fig. 1a). On average,  $\text{NH}_4^+$  and

$\text{nss-SO}_4^{2-}$  account for 10 and 30% of the fine mode ( $\text{PM}_{2.5}$ ) mass during wintertime. It is, however, noteworthy that the concentration of  $\text{NO}_3^-$  is nearly five times less than that of  $\text{SO}_4^{2-}$  (Figs. 2e and f).

### 3.2. Temporal variability of aerosol iron ( $\text{Fe}_A$ ) and water-soluble iron (WS-Fe)

During the summer season, contribution of dust to  $\text{PM}_{2.5}$  mass is relatively high and steadily decreases in winter months (Fig. 2b). Thus, aerosol Fe ( $\text{Fe}_A$ ) concentration also exhibit similar temporal variability (Fig. 3a), with relatively high values occurring during the summer months (range: 161–915  $\text{ng m}^{-3}$ ) compared to the winter season (range: 50–397  $\text{ng m}^{-3}$ ). In contrast, temporal variability of water-soluble Fe fraction (i.e.  $[\text{WS-Fe}/\text{Fe}_A] \times 100$ ) shows an opposite trend over the annual seasonal cycle (Fig. 3b). The percentage of WS-Fe averages around  $8.1 \pm 3.5\%$  (range: 2.5–16.1%) during wintertime and  $1.6 \pm 1.9\%$  (range:

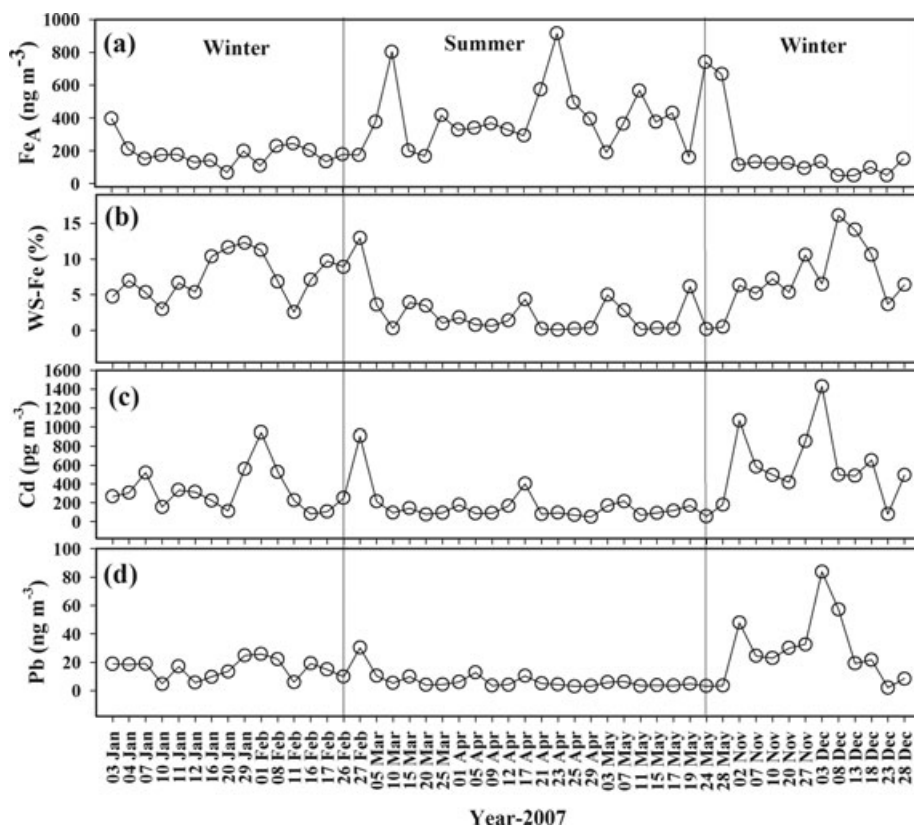


Fig. 3. Temporal variability of (a) total aerosol iron ( $\text{Fe}_A$ ) concentration and (b) water-soluble fraction of iron [WS-Fe (%)], (c) concentration of Cd and (d) Pb, in the aerosol samples collected from Mt. Abu.

0.06–6.1%) during summer months (Fig. 3b). The enhanced fractional solubility of Fe [WS-Fe (%)], during wintertime, is associated with relatively low abundance of dust and  $\text{Fe}_A$ ; whereas lower solubility is a characteristic feature of the high dust season (higher  $\text{Fe}_A$ ). It is noteworthy that the fractional solubility does not show significant correlation with  $\text{nss-SO}_4^{2-}$  (Fig. 4a). This is unlike the process of enhanced dissolution/leaching of aerosol Fe with the increasing acidity (lower pH) suggested by Meskhidze et al. (2003). The abundances of  $\text{nss-SO}_4^{2-}$  and  $\text{NH}_4^+$  (expressed in equivalent units) exhibit significant linear correlation (Fig. 4b;  $r = 0.95$ ), indicating the predominance of  $\text{nss-SO}_4^{2-}$  in the fine mode as ammonium salt.

A wide range of values for water-soluble Fe fraction have been reported during several field experiments from the oceanic regions (Siefert et al., 1999; Johansen and Hoffmann, 2003; Chen and Siefert, 2004; Baker et al., 2006; Buck et al., 2006; Sedwick et al., 2007). Some of them have also reported on the potential link between acid processing during long-range transport and enhancement in the iron solubility. However, these studies do not firmly conclude acid processing as a primary control for iron fractional solubility. More recently, it has been hypothesized that the increase in fractional iron solubility may vary with the mixing state of aerosols as well as deposition of acid and dust particle simultaneously during atmospheric transport (Baker and

Croot, 2008). One of the recent modelling study by Luo et al. (2005), have concluded on the lack of significant correlation between observed sulphate or organic acids and iron solubility. In this study, it is quite reasonable to assume that the long-range transport of fine mode particles sampled from a high-altitude site, during wintertime, are internally mixed. However, based on the observation that the mass fraction of dust significantly decreases during winter months (Fig. 2b), and together with efficient neutralization of  $\text{nss-SO}_4^{2-}$  by  $\text{NH}_4^+$  it can be inferred that chemical processing of aerosols is of minor significance for the increased fractional Fe solubility at this study site. In the regional context, it may also be emphasized that the large buffer capacity of the carbonate dust is relevant during the high-dust season.

### 3.3. Solubility of aerosol iron and role of anthropogenic sources

The relatively low abundance of WS-Fe in aerosol samples collected during the high dust season (summertime), and high solubility of aerosol Fe during the winter months (characterized by lower dust abundance) clearly demonstrates that Fe solubility is not dictated by the abundance of the mineral dust and aerosol Fe. The significantly high  $\text{Fe}_A/\text{Al}$  ratios are characteristic of the

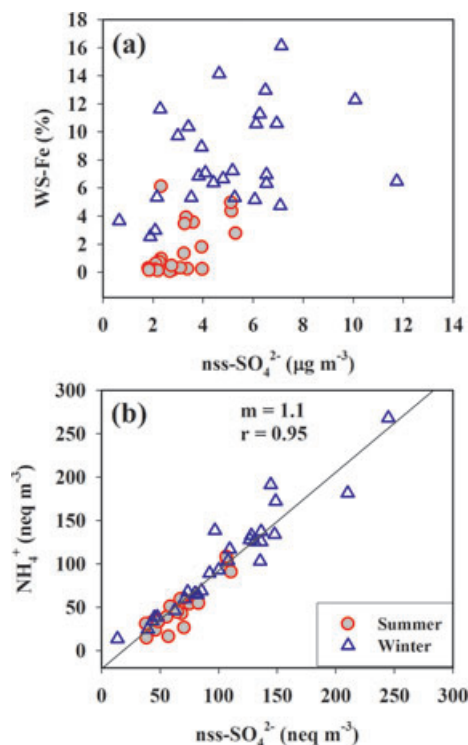


Fig. 4. (a) Large scatter between WS-Fe (%) and nss-SO<sub>4</sub><sup>2-</sup> suggests that chemical processing during aerosol transport is of minor significance for soluble iron fraction at the study site. (b) Linear relationship among abundances (in equivalent units) of nss-SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> in the fine (PM<sub>2.5</sub>) aerosols during summer and winter seasons.

$\text{Fe}_A < 200 \text{ ng m}^{-3}$  (Fig. 5a). Furthermore, we have evaluated the concentrations of Cd and Pb as a function of total aerosol Fe ( $\text{Fe}_A$ ) concentration (Figs. 3c and d); thus documenting a sharp increase in their abundances at low concentration of  $\text{Fe}_A$  (less than  $200 \text{ ng m}^{-3}$ ). It is noteworthy that extremely low  $\text{Fe}_A$  content is characteristic of wintertime when aerosol composition is influenced by the advective transport of pollutants from northern India (Indo-Gangetic Plain) associated with north-easterly winds. The enrichment factors of Cd and Pb (with respect to Al) show exceptionally high values (range: 214–6700 and 85–3650, respectively), during winter months, suggesting dominant impact of anthropogenic sources. The elevated Cd/Al and Pb/Al ratios in the aerosols, compared to the upper continental crust can be used as important indicator of anthropogenic aerosols produced by burning of fossil-fuel (Nriagu and Pacyna, 1988; Arimoto et al., 1995). The plot between WS-Fe (%) and  $\text{Fe}_A$  (Fig. 6), shows a similar enhancement shown by Cd/Al and Pb/Al versus  $\text{Fe}_A$  (Figs. 5b and c), further corroborating that the fractional solubility of Fe is dictated by the contribution from anthropogenic activities (fossil-fuel combustion products) during winter season. In contrast, low solubility of Fe is associated with the high dust conditions and transport from desert and semi-arid region in western India and as far as from Oman

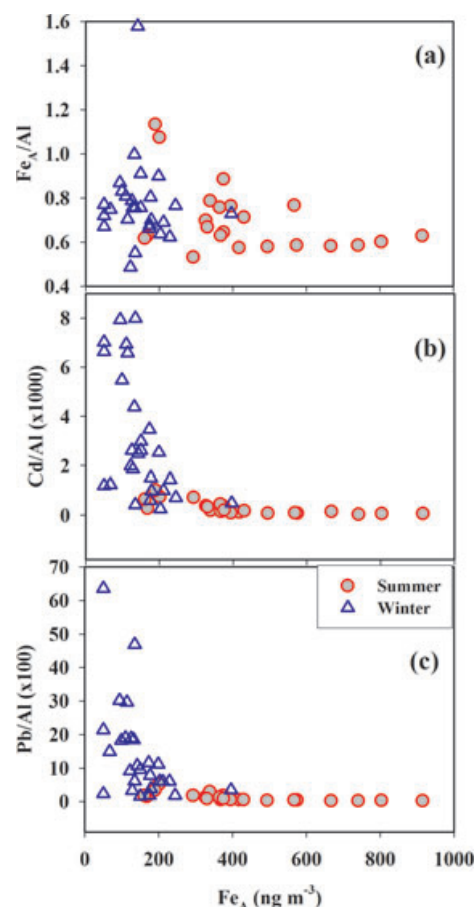


Fig. 5. (a)  $\text{Fe}_A/\text{Al}$ , (b) Cd/Al and (c) Pb/Al ratios are plotted against total aerosol iron concentration. A sharp increase in Cd/Al and Pb/Al ratio at lower  $\text{Fe}_A$  concentration is observed for winter month samples.

desert during summer month. Recently, Buck et al. (2008b), have observed similar type of enrichment of soluble Sb and Pb, over central Atlantic Ocean and suggested the burning of fossil fuel as an important source. Relatively enriched Fe/Al mass ratio has been reported for fuel-oil fly ash and exhaust particle produced by diesel combustion (Desboeufs et al., 2005); where as 77–81% of water-soluble Fe can be contributed by oil combustion products (Schroth et al., 2009). Chuang et al. (2005), has demonstrated significant contribution of particulate soluble Fe concentration from anthropogenic activity rather than processing during long-range transport in the East Asian atmospheric outflow over annual timescales.

The “operational solubility” of Fe [WS-Fe (%)] versus total Fe ( $\text{Fe}_A$ ) represents a two component mixing plot (Fig. 6). This is also confirmed by plotting WS-Fe (%) and  $1/\text{Fe}_A$ , yielding a best-fit linear regression line ( $r = 0.82$ ;  $p < 0.001$ ). Based on this observation, a conservative mixing line is generated for the two distinct end members (similar to the approach adopted by Sedwick et al. 2007). One end member is characterized by higher  $\text{Fe}_A$  concentration of  $915 \text{ ng m}^{-3}$  with fractional



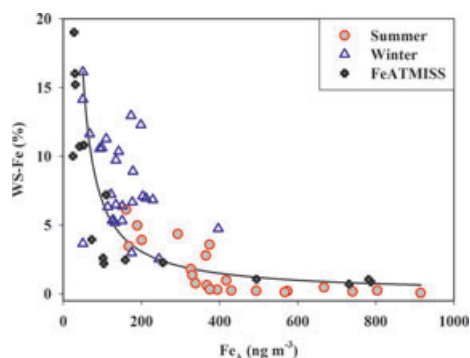


Fig. 6. Relationship between WS-Fe (%) and  $\text{Fe}_A$  for Mt. Abu samples along with the FeATMISS data (Sedwick et al., 2007) from Sargasso Sea. Two of the data points from FeATMISS exceeding  $\text{Fe}_A > 1000 \text{ ng m}^{-3}$  are not shown in the figure.

solubility of 0.06% (during the summertime). The other end member is characterized by lowest  $\text{Fe}_A$  value of  $50 \text{ ng m}^{-3}$  associated with the soluble fraction of 16.1% (representing wintertime data). The modelled curve simulates reasonably well the inverse relationship between WS-Fe (%) and total aerosol Fe, except for the few data points deviating from the mixing line during winter season. The data for the aerosol iron-solubility from Sargasso Sea exhibit similar kind of hyperbolic relationship with the total aerosol iron (Sedwick et al., 2007). For comparison, the FeATMISS (Iron Atmospheric Inputs to the Sargasso Sea; Sedwick et al., 2007) data points collected over time period ranging from days to months have been plotted on the mixing curve representing this study. This comparison suggests that the contribution of aerosol Fe from anthropogenic sources influences the fractional solubility of Fe nearly to the same degree as over Sargasso Sea. Such an approach provides better insight into operational solubility of aerosol Fe and enforcing our conclusion that Fe solubility in the semi-arid region of western India is primarily controlled by composition and chemical nature of aerosols from the source region rather than chemical processing during their long-range transport.

#### 4. Conclusions and implications

The chemical composition of atmospheric particulate matter ( $\text{PM}_{2.5}$ ), studied from high-dust semi-arid region of western India, reveals conspicuous temporal variability in the fractional solubility of aerosol iron dominated by the anthropogenic sources. During wintertime, the advective transport of pollutants (derived from fossil-fuel combustion and biomass burning) from the Indo-Gangetic Plain (northern India), with distinctly high  $\text{Fe}_A/\text{Al}$ ,  $\text{Pb}/\text{Al}$  and  $\text{Cd}/\text{Al}$  ratios, result in enhanced fractional solubility of Fe ( $\text{Av} = 8.1 \pm 3.5\%$ ). The impact of anthropogenic sources is further demonstrated by significant increase in the ambient concentrations of  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$  in fine mode ( $\text{PM}_{2.5}$ ). In contrast, the high-dust season (summertime) asso-

ciated with high abundance of aerosol Fe, is characterized by exceedingly low solubility ( $\text{Av} = 1.6 \pm 1.9\%$ ); suggesting that atmospheric chemical processing is of minor significance.

These results have implications to the atmospheric source of soluble iron associated with large-dust events occurring in the semi-arid regions. In a recent study from the Arabian Sea, the dry-deposition flux of Fe during the period of inter-monsoon (March–April, 2006) was estimated to be  $395 \mu\text{g m}^{-2} \text{ d}^{-1}$  based on the Fe concentration (range:  $90$  to  $1010 \text{ ng m}^{-3}$ ) in ambient aerosols (Kumar et al., 2008). Assuming 10% as the solubility of aerosol Fe (also commonly referred as bio-available Fe in the literature) would lead to over estimation of the supply of dust related soluble iron to the ocean surface. Such regional scale studies are, thus, important to constrain the existing uncertainties in the atmospheric source and delivery of soluble iron for modelling the impact on surface ocean biogeochemistry.

#### 5. Acknowledgment

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