

Atmospheric outflow of nutrients to the Bay of Bengal: Impact of anthropogenic sources

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Abstract

The air-sea deposition of nutrients (N, P and Fe) to the oceanic regions located downwind of pollution sources in south and south-east Asia is gaining considerable attention in the present-day scenario of climate change. We report here a case study on the atmospheric outflow of nutrients from the Indo-Gangetic Plain (IGP) to the Bay of Bengal (BoB). Air mass back trajectories suggest conspicuous downwind transport of chemical constituents from the IGP to the BoB during the late NE-monsoon (January-April); thus, representing BoB as one of the unique oceanic regions influenced by anthropogenic sources over a short span of 3 - 4 months. During the course of this study (November'09 – March'10), nutrient (NO_3^- , NH_4^+ , N_{Org} , PO_4^{3-} and Fe_{ws}) concentrations in the atmospheric outflow show pronounced temporal variability. The inorganic nitrogen (NH_4^+ -N: ~ 90 % of N_{Inorg}) dominates the total soluble nitrogen (N_{Tot}). Although the contribution of organic nitrogen is not significant, the mass ratio of $\text{N}_{\text{Org}}/\text{N}_{\text{Tot}}$ in the outflow varied from 0.07 to 0.40. The abundances of P_{Inorg} and Fe_{ws} varied from 0.4 to 4.8 nmol m^{-3} and 0.2 to 0.6 nmol m^{-3} , respectively. The high abundance of K^+ and significant (P-value < 0.05) correlation with P_{Inorg} and Fe_{ws} , suggests their significant contribution from biomass burning emissions (BBEs). The characteristic mass ratios of nutrients ($\text{N}_{\text{Inorg}}/\text{N}_{\text{Tot}}$: 0.92 ± 0.13 , $\text{N}_{\text{Org}}/\text{N}_{\text{Tot}}$: 0.21 ± 0.11 , and $\text{P}_{\text{Inorg}}/\text{nss-Ca}^{2+}$: 0.35 ± 0.23) in the IGP-outflow show striking similarity with those reported over the BoB. These results have implications to further increase in the atmospheric deposition of nutrients and their impact on biogeochemistry of surface Bay of Bengal.

1. Introduction

In the present-day scenario of growing anthropogenic activities, atmospheric transport and deposition of nutrients (N, P and Fe) to the oceanic regions located downwind of pollution sources are gaining considerable attention due to their potential impact on biogeochemistry of ocean surface (Duce, 1986; 1991; 2008; Jickells et al., 2005; Mahowald et al., 2011; Prospero et al., 2009). A significant increase in the eutrophication caused by atmospheric deposition of reactive nitrogen (Paerl et al., 2002; Paerl and Whitall, 1999) further emphasizes the need to assess the impact of air-sea deposition of nutrients to oceanic regions. The increasingly growing use of fertilizers, emissions from biomass burning and fossil-fuel combustion, and animal live stock are the potential sources of this macro nutrient. Until recently, studies on the atmospheric deposition of nitrogen have mainly focused on the soluble inorganic species (Duce et al., 2008 and references therein). However, studies on the atmospheric deposition of organic nitrogen and its bioavailability in sea water are rather sparse (Cornell, 2011; Lesworth et al., 2010; Mace et al., 2003; Neff et al., 2002; Russell et al., 1998; Spokes et al., 2000; Violaki et al., 2010), making it difficult to assess its global significance in the reactive nitrogen cycle.

In addition to the deposition of N, atmospheric source of phosphorus (P) has been also suggested to significantly influence the biogeochemical cycle of nutrients in the ocean surface (Mahowald et al., 2008; Paytan et al., 2009). Owing to lower abundance of P in the atmosphere, earlier studies have suggested its minor role on ocean biogeochemistry, largely based on small contribution from mineral dust (Froelich et al., 1982; Smil, 2000). However, recent estimates have addressed to the concept of mobilization of P from other sources such as mining of phosphate rocks, increase in use of fertilizers (Benitez-Nelson, 2000; Mahowald et al., 2008; Paytan and McLaughlin, 2007), biomass burning emissions (Mahowald et al., 2005; Migon and Sandroni, 1999) and fossil-fuel combustion (Anderson et al., 2010). It is suggested that atmospheric supply of N is far more higher compared to input of soluble inorganic phosphorus. Under these conditions, the atmospheric deposition (high N and low P) can perturb the Redfield ratio (N: P = 16:1) in the surface waters (Duce et al., 2008; Jickells, 2006). In this context, P-limitation is documented in the Mediterranean Sea (Herut et al., 2002; Markaki et al., 2010; Migon et al., 2001) and open ocean waters of Atlantic (Ammerman et al., 2003; Mills et al., 2004; Sanudo-Wilhelmy et al., 2001). These studies emphasize the need to assess the atmospheric sources of phosphorus with respect to present-day increase in contribution from anthropogenic sources.

In contrast to N and P, much of the open ocean waters are limited by the availability of iron (Boyd et al., 2010; Coale et al., 1996; Martin et al., 1990). The significance of mineral dust as a dominant source of this micro nutrient to the surface ocean is well recognized (Baker and Croot, 2010; Boyd and Ellwood, 2010; Jickells et al., 2005; Mahowald et al., 2009; Schulz et al., 2012). However, recent studies have emphasized the importance of anthropogenic combustion sources (fossil-fuel and bio-mass burning) as a potential source of soluble iron to the seawater (Chuang et al., 2005; Guieu et al., 2005; Kumar et al., 2010; Luo et al., 2008; Schroth et al., 2009; Sholkovitz et al., 2009; Sholkovitz et al., 2012; Srinivas et al., 2011a). These observations further emphasize the need to assess the atmospheric input of nutrients on a regional scale.

The widespread dispersal of airborne pollutants, from south and south-east Asia, over the pelagic Indian Ocean has gained much attention in recent years (Lamarque et al., 2010; Lawrence and Lelieveld, 2010; Lelieveld et al., 2001; Quinn and Bates, 2005; Ramanathan et al., 2001). Furthermore, modelling studies are projecting enhanced deposition of reactive nitrogen to the surface waters from pollution sources in south Asia (Duce et al., 2008; Galloway et al., 2004). However, there is no clear consensus on the source specific contribution of P and Fe (i.e., anthropogenic vis-à-vis natural) to the Northern Indian Ocean (Kumar et al., 2010; Srinivas and Sarin, 2012; Srinivas et al., 2011a). It is, thus, relevant to assess the long-range transport of nutrients in the continental outflow and their subsequent deposition to the downwind oceanic regions.

During the late NE-monsoon (January-April), atmospheric outflow from the Indo-Gangetic Plain (IGP) is a conspicuous feature and dominant source of chemical constituents to the marine atmospheric boundary layer (MABL) of the Bay of Bengal (Kumar et al., 2010; Sarin et al., 2010; Srinivas and Sarin, 2012). These studies have essentially characterized the chemical composition of particulate matter (PM_{2.5} and PM₁₀), and have documented the abundances of nutrients (N, P and Fe) within the MABL of Bay of Bengal (Srinivas and Sarin, 2013a). These studies have emphasized dominant role of anthropogenic sources (biomass burning emissions, BBEs) in supplying the soluble P and Fe to the Bay of Bengal (Kumar et al., 2010; Srinivas and Sarin, 2012). In addition, dominance of inorganic nitrogen (mainly as NH₄⁺-N; > 90 % of N_{Inorg}) to total aerosol water-soluble nitrogen is noteworthy over the Bay of Bengal associated with the outflow from the Indo-Gangetic Plain (Srinivas et al., 2011b). This observation is further supported by long-term observation of aerosols (2001 -2009) from the Bay of Bengal (Srinivas et al., 2011b). However, it is relevant to state that these studies have relied mainly on air-mass back trajectory analyses to assess the transport of nutrients and chemical species from the Indo-Gangetic Plain. Therefore, the study reported in this manuscript aims to investigate the temporal variability in the atmospheric outflow of anthropogenic nutrients from the Indo-Gangetic Plain to the Bay of Bengal; and to establish the diagnostic ratios of nutrients vis-à-vis those reported over the Bay of Bengal.

2. Methodology

In this study, fine mode (PM_{2.5}, N = 46) aerosols were collected during November'09 – March'10 from a downwind site (Kharagpur: 22.3 °N, 87.3 °E) in the Indo-Gangetic Plain (IGP). A high-volume air sampler (HVS, Thermo Anderson) was setup on the roof top of geology department in the campus of IIT Kharagpur (Fig.1). The flow rate of air sampler was periodically calibrated and ascertained as ~ 1.07 m³ min⁻¹. All samples were collected on pre-combusted (~ 450°C) tisuquartz filters (PALLFLEX[®]TM) for a total time period of 20 – 22 hrs; filters were sealed in plastic Zip-lock bags and stored at – 19°C until the time of analysis. Prior to the assessment of gravimetric mass, samples were equilibrated for ~24 hrs under a clean laminar flow bench (Class – 1000) and PM_{2.5} concentration was ascertained from the tare weight of the filters. Subsequently, aerosol samples were analyzed for water-soluble nutrients using analytical protocols described in our earlier publications (Srinivas and Sarin, 2013a and references therein).

We have defined the water-soluble inorganic nitrogen (N_{Inorg}) as the sum of mass concentrations of NH₄⁺ and NO₃⁻. In order to assess the contribution of N_{Inorg}, a 1/8th portion of the filter was extracted with 50 ml of Milli-Q water (Millipore Co., Specific resistivity > 18.2 MΩ-cm) and analyzed for

NO_3^- and NH_4^+ on Dionex- 500 Ion chromatograph equipped with suppressed conductivity detector. Likewise, another aliquot of filter (i.e., 1/8th portion) was extracted in 30 ml of Milli-Q and filtered through pre-combusted GF/F filters (Whatmann[®]TM, pore size: 0.45 μm). These extracts were subsequently analyzed for water-soluble organic carbon (WSOC) and total soluble nitrogen (N_{Tot}) on a TOC-TN analyzer (Model: Shimadzu, TOC-5000a). The water-soluble organic nitrogen (N_{Org}) was, then, estimated by subtracting the contribution of N_{Inorg} from N_{Tot} . Along with aerosol samples, filter blanks were also extracted and analyzed for WSOC and N_{Tot} . The procedural blank contributes to ~ 1.6 and 0.2 % of minimum and maximum signal of N_{Tot} measured on TN analyzer. Likewise, the contribution of blank varies from 14 to 2 % of min-max signal measured during the analysis of WSOC. The analytical accuracy of the measurements of N_{Inorg} and N_{Tot} has been ascertained by analyzing the Merck standard solutions (Srinivas and Sarin, 2013a; Srinivas et al., 2011b).

An aliquot of water extract was filtered through 0.45 μm Millipore filter (Millipore Co.) and analyzed for water soluble inorganic phosphorus ($\text{P}_{\text{Inorg}} = \text{PO}_4^{3-}$) using 2 m long liquid-core waveguide capillary cell (LWCC from World Precision Instruments, WPI Inc.) coupled to a USB-4000 spectrophotometer (from Ocean Optics Inc.). The absorption of phospho-molybdenum blue complex was measured at a wavelength of 710 nm (Srinivas and Sarin, 2012) with respect to calibration standards made from suitable dilution of stock solution of 2 mM KH_2PO_4 . The analytical accuracy of the measurement (i.e., $\leq 5\%$) was ascertained from the independent analysis of 1000 ppm Merck commercial standard solution. For further details on analytical measurements reference is made to our earlier publication (Srinivas and Sarin, 2012).

For water soluble iron (Fe_{ws}), 8 – 10 filter punches (ca. 2.0 cm dia) were extracted with Milli-Q water and filtered through 0.45 μm Millipore filters. Subsequently, extracts were acidified with 2N distilled HNO_3 to a pH of 2 and analyzed for soluble iron on GF-AAS (Perkin Elmer model, HGA-800). For analysis, calibration standard were prepared by suitable dilution of commercial Merck[®] 23 element standard. To check the accuracy, certified USGS rock standard (W-1 basalt) is digested and measured for the Fe on GF-AAS; results are in agreement within 6 %.

Based on replicate analyses of samples and standards, the overall analytical precision is better than 10 % for all measurements. All samples were suitably corrected for procedural filter blanks. The method detection limits (MDL), defined as the three times the standard deviation of procedural blanks are normalized with the average volume of filtered air (~ 1400 m^3). The MDL of NH_4^+ , NO_3^- , N_{Tot} , PO_4^{3-} and Fe_{ws} were 0.10, 0.62, 2.45, 0.02 and 0.01 nmol m^{-3} respectively. For detailed analytical protocol reference is made to our earlier publications (Srinivas and Sarin, 2012; Srinivas et al., 2011a; Srinivas and Sarin, 2013a; Srinivas and Sarin, 2013b; Srinivas et al., 2011b).

2.2. Error & Statistical analysis

Due to the differences in analytical uncertainties associated with the measurements of N_{Inorg} and N_{Tot} (external precision of 5 %, based on the replicate analyses of samples and standards), error propagation has been made for each sample similar to the approach suggested in our earlier paper (Srinivas et al., 2011b). Only those samples were retained where the error (or standard deviation) is less than the abundance of organic nitrogen ($\text{N}_{\text{Org}} > \sigma_{\text{N-Org}}$). Based on this criterion, a total of 18 samples were considered for assessing the abundances of N_{Org} and $\text{N}_{\text{Org}}/\text{N}_{\text{Tot}}$ during the study period.

In addition, statistical tests were performed to ascertain the significant differences in the atmospheric abundances of nutrients and their molar ratios between the two sampling periods (viz., December'08 – January'09, collected from the MABL of Bay of Bengal and those from the Indo-Gangetic Plain during November'09 – March'10).

3. Results and Discussions

3.1. Air-mass back trajectory analysis

Back trajectory analyses provide important information on the source regions of pollutants. In this study, 7-day isentropic back trajectories (i.e., at an arrival heights of 100, 500 and 1000 m) were computed using hybrid single particle lagrangian integrated trajectory model (HYSPLIT, version 4.0; (Draxler et al., 1999)) from the NOAA air resources laboratory (using GDAS database). This analysis has clearly demonstrated the advective downwind transport of pollutants from the upwind regions of the Indo-Gangetic Plain. The air mass back trajectory cluster at an arrival height of 500 m, during all the sampling days, is presented in Fig.2. Similar to our observation, based on the air mass trajectory analysis, Kumar et al. (2010) had also documented the impact of anthropogenic sources on the MABL of Bay of Bengal during December'08 – January'09. Thus, our sampling site (Fig.1) is relevant in order to study the atmospheric outflow from the IGP to the Bay of Bengal.

3.2. Temporal variability

During the continental outflow, the mass concentration of fine mode aerosols (PM_{2.5}) show pronounced temporal variability (range: 23.1 – 177.2 µg m⁻³) and, also, reflected in the individual nutrient species (Fig.3). A brief statistical description about the nutrient mass concentrations from the IGP-outflow is presented in Table 1. In addition, a comparison is also made for the atmospheric abundances of nutrients and their diagnostic mass ratios between the IGP-outflow (this study, November'09-March'10) and the Bay of Bengal (December'08 – January'09; See Table 1). Similar to the PM_{2.5} mass, concentrations of nutrients are high in early winter (December – January) and decreases during rest of the sampling days (i.e., February – March). Relatively high mass concentration of nutrients in the IGP-outflow during early winter months could be explained by the shallow boundary layer height (~ 500 m) and strong NE-monsoonal winds. These results are similar to our earlier observations from the Bay of Bengal during the two research cruises conducted in December'08-January'09 and March-April'06; relatively high concentrations of nutrients occurring in the winter compared to that in spring-intermonsoon (Srinivas and Sarin, 2013a). However, seasonal difference in data from two cruises was explained in terms of variability in the source strength of atmospheric outflow from the Indo-Gangetic Plain (also supported by the AMBTs).

Our observations from the IGP-outflow (this study) reinforce the pronounced seasonal pattern of nutrient abundances documented from the Bay of Bengal (Srinivas and Sarin, 2013a). Furthermore, it is relevant to state that the chemical composition of water soluble nutrients (N, P and Fe) and chemical species (nss-K⁺, nss-Ca²⁺ and nss-SO₄²⁻) in aerosols from a continental sampling site provide crucial information regarding their emission from source region, subsequent transport and transformation pathways to the MABL during the study period (November'09 - March'10). Therefore, we compared the chemical composition of aerosols from the sampling site with that

collected over the BoB during December'08 – January'09 (PM_{2.5}, PM₁₀) and March – April'06 (TSP).

3.2.1. Inorganic and Organic Nitrogen (N_{Inorg} & N_{Org})

The mass concentration of NH_4^+ dominates (Av: 93 ± 7%) the inorganic nitrogen (N_{Inorg}) and varies from 109 to 559 nmol m⁻³ (Av : 290 ± 108 nmol m⁻³) in the IGP-outflow. Likewise, NO_3^- abundance ranges from 0.5 to 183.6 nmol m⁻³ (Av : 11.6 ± 12.5 nmol m⁻³) during November'09 – March'10 and its fractional contribution in N_{Inorg} averages ~ 7 ± 8 %. However, relatively high concentration of NO_3^- (range: 19 – 183.6 nmol m⁻³) and, therefore, its percentage contribution (range: 6 to 24 %) towards water-soluble inorganic nitrogen has been observed in early winter samples (21st November – 5th December'09). During the later sampling days, NO_3^-/N_{Inorg} show little variability. Overall, the water-soluble inorganic nitrogen (i.e., $N_{Inorg} = NH_4^+ + NO_3^-$) accounts for ~ 90 % of N_{Tot} (N_{Inorg}/N_{Tot} : 0.92 ± 0.13). This observation suggests the predominance of N_{Inorg} in the IGP-outflow. These results were consistent with our earlier findings in the MABL (Srinivas et al., 2011b), where the fractional contribution of N_{Inorg} (i.e., mainly as NH_4^+ -N, 98 % of N_{Inorg}) to N_{Tot} averages ~ 88 ± 13 % during December'08 – January'09. Fig.4 depicts the significant linear correlations observed between NH_4^+ , N_{Inorg} and N_{Tot} in fine mode aerosols collected from the IGP-outflow and over the Bay of Bengal.

During the study period, the abundance of N_{Org} ($N_{Tot} - N_{Inorg}$) varied from 18 to 234 (Av: 96 ± 63) nmol m⁻³. However, the molar mass ratio of N_{Org}/N_{Tot} during November'09 – March'10 averages ~ 0.25 ± 0.10 (range: 0.12 to 0.40). Similar to the contribution of N_{Inorg}/N_{Tot} , the fractional abundance ratio of N_{Org} to N_{Tot} in fine mode aerosols also shows well agreement between the IGP-outflow (this study) and the Bay of Bengal (Srinivas et al., 2011b). Significant contribution of N_{Org}/N_{Tot} is only observed for winter samples (December – February, N = 13). Despite the similar transport pattern of AMBTs (Fig.2), it remains uncertain to assess the source region of organic nitrogen and its enhanced concentration during sampling days in winter. For rest of the sampling days, the abundance of N_{Inorg} is comparable with the N_{Tot} within the spread of the data and therefore, indicates no significant contribution of N_{Org} . A comparison of atmospheric abundances of N_{Org} and their relative contribution from the sampling site in the Indo-Gangetic Plain with that documented for other locations is presented in Table 2. From this table, it is implicit that the mass fraction of N_{Org} is of comparable levels with that documented from the South China Sea, Yellow Sea (Shi et al., 2010), East China Sea (Nakamura et al., 2006), Gulf of aqaba (Chen et al., 2007), Eastern Mediterranean (Mace et al., 2003) and North Atlantic (Lesworth et al., 2010). It is also noteworthy that a similar mass fraction of N_{Org} in N_{Tot} is reported over the broadleaf forest areas in Japan (Miyazaki et al., 2014). Our mean value of N_{Org}/N_{Tot} is also consistent with that document for the typical urban regions such as northern California (Zhang et al., 2002).

Due to the similarity in the origin of AMBTs from the sampling site in IGP with that documented over the Bay of Bengal, two-tailed unpaired t-test (at a significance level, $\alpha = 0.05$) has been used to ascertain the significant similarities/differences of the atmospheric abundances of water-soluble nitrogen species and molar ratios between them (Table 1). From this table, it is noteworthy that there were considerable differences in the water-soluble nitrogen species in terms their atmospheric abundances between the IGP-outflow and Bay of Bengal. Relatively high concentrations were observed in the IGP-outflow compared to that reported from the MABL, suggesting the decrease in

source contribution with distance. However, no statistically significant differences are observed between the IGP-outflow and Bay of Bengal for molar ratios (viz., $\text{NO}_3^-/\text{N}_{\text{Inorg}}$, $\text{NH}_4^+/\text{N}_{\text{Inorg}}$, $\text{N}_{\text{Inorg}}/\text{N}_{\text{Tot}}$ and $\text{N}_{\text{Org}}/\text{N}_{\text{Tot}}$). This observation highlights their common source (i.e., mixed contribution from BBEs, animal-livestock, fertilizers and vehicular emissions) and long-range atmospheric transport to the MABL.

3.2.2. Water-soluble Inorganic Phosphorus (P_{Inorg})

The mass concentration of water-soluble inorganic phosphorus (P_{Inorg}) in the IGP-outflow varies from 0.4 to 4.8 nmol m^{-3} ($\text{Av} : 1.2 \pm 0.9 \text{ nmol m}^{-3}$; Fig.3) and are comparable with those reported over the Bay of Bengal during December'08-January'09 (Table.1). It is noteworthy that mass concentrations of P_{Inorg} from the IGP-outflow are consistent with that documented in BBEs from the Sahel region (western Africa) during DABMEX (Dust and Biomass burning experiment) by Paris et al., (2010). The atmospheric abundances of water-soluble phosphorus in the downwind oceanic regions influenced by BBEs are somewhat lower than that reported for source regions, for example, the IGP-outflow sampled over the Bay of Bengal (Srinivas and Sarin, 2012), Gulf of Aqaba (Chen et al., 2007) and the East Asian outflow sampled over the South China Sea (Hsu et al., 2010b). However, our concentrations of P_{Inorg} in the IGP-outflow are considerably higher than that reported over the North Atlantic (Baker et al., 2007) and the Arabian Sea (Srinivas and Sarin, 2012) where the deposition of mineral dust is a major source of nutrients (Table 3). This observation further signifies the overwhelming source contribution of anthropogenic BBEs as a potential source of P to the surface ocean.

In this study, we have assessed the relative dominance of P_{Inorg} from anthropogenic sources over the mineral dust based on the molar mass ratios of $P_{\text{Inorg}}/\text{nss-Ca}^{2+}$ in the IGP-outflow. In a previous study conducted over the Bay of Bengal, we have used a similar approach where the abundance of nss-Ca^{2+} is used as a proxy for mineral dust (Srinivas and Sarin, 2012). The $P_{\text{Inorg}}/\text{nss-Ca}^{2+}$ varied from 0.06 to 1.01 ($\text{Av} : 0.35 \pm 0.23$) in the IGP-outflow during November'09 – March'10. These results were somewhat similar to those observed over the Bay of Bengal during December'08 – January'09 ($\text{Av} : 0.30 \pm 0.29$) and March – April'06 ($\text{Av} : 0.09 \pm 0.05$) and relatively higher than that documented over the Arabian Sea in April-May'06 ($\text{Av} : 0.03 \pm 0.02$; Srinivas and Sarin, 2012). It is noteworthy that no statistically significant difference is found between the IGP-outflow (this study) and the Bay of Bengal (winter cruise, December'08-January'09) with regard to their molar mass ratios of $P_{\text{Inorg}}/\text{nss-Ca}^{2+}$ (See Table 1).

It has been suggested that non-sea salt potassium (nss-K^+) in the fine mode aerosols can be used a tracer for identifying the impact of BBEs (Andreae, 1983; Echalar et al., 1995; Gaudichett et al., 1995; Reche et al., 2012). In this study, significantly high concentrations of nss-K^+ were observed in the atmospheric outflow from IGP. Therefore, we have used $\text{nss-K}^+/\text{nss-Ca}^{2+}$ similar to $P_{\text{Inorg}}/\text{nss-Ca}^{2+}$ ratios and compared it with that documented from the MABL during December'08 – January'09. The molar mass ratio of $\text{nss-K}^+/\text{nss-Ca}^{2+}$ varied from 5.2 to 27.5 ($\text{Av} : 13.8 \pm 11.3$) in the IGP-outflow. Similar ratios were observed in the fine mode aerosols collected from the MABL of Bay of Bengal ($\text{Av} : 9.1 \pm 3.6$) during December'08 – January'09 (Srinivas and Sarin, 2012). It is noteworthy that no statistically significant differences in the molar ratios ($\text{nss-K}^+/\text{nss-Ca}^{2+}$ and

$P_{\text{Inorg}}/\text{nss-Ca}^{2+}$) are evident in the IGP-outflow from the sampling site and over the Bay of Bengal ($P > 0.05$, See Table 1).

In an earlier study, based on significantly higher abundances of P_{Inorg} and molar ratios ($P_{\text{Inorg}}/\text{nss-Ca}^{2+}$ and $\text{nss-K}^+/\text{nss-Ca}^{2+}$) over the Bay of Bengal during winter months (December'08 – January'09), we have assessed the dominant role of phosphorus contribution from BBEs (Srinivas and Sarin, 2012). Therefore, our observations with respect to atmospheric abundances of high P_{Inorg} , nss-K^+ , $P_{\text{Inorg}}/\text{nss-Ca}^{2+}$ and $\text{nss-K}^+/\text{nss-Ca}^{2+}$ in the IGP-outflow reaffirms the argument that BBEs, indeed, could be a potential source of this nutrient. In this context, Mahowald et al., (2005) had reported a low ratio of 0.0029 ± 0.0001 for P/BC from BBEs (BC and EC are synonymously used in the literature). In this study, the $P_{\text{Inorg}}/\text{EC}$ averages $\sim 0.009 \pm 0.005$ for IGP-outflow (November'09 – March'10) and 0.008 ± 0.004 for the Bay of Bengal (December'08 – January'09). These results are somewhat comparable to that reported for the BBEs (Mahowald et al., 2009 and references therein).

As there are no statistically significant differences in the molar mass ratios, $P_{\text{Inorg}}/\text{nss-Ca}^{2+}$ and $\text{nss-K}^+/\text{nss-Ca}^{2+}$, between the IGP-outflow and over the BoB, we have combined the two data sets to assess their common sources. A linear relationship is noteworthy between P_{Inorg} and nss-K^+ ($r = 0.67$; $P\text{-value} < 0.05$; slope = 0.016; Fig.5a). Likewise, there exists a significant linear relation between molar concentrations of P_{Inorg} and nss-Ca^{2+} ($r = 0.6$; $P\text{-value} < 0.05$; slope = 0.25; Fig.5b) for fine mode aerosols collected from the IGP-outflow and the Bay of Bengal. It is relevant to state here that the slope of the regression reported in this study is significantly higher than that reported for PM_{10} (slope = 0.07; 3 fold higher; January'09) and TSP (slope = 0.03; 8 fold higher; March-April'06) sampled over the Bay of Bengal and an order of magnitude higher than that reported over the Arabian Sea (slope = 0.0044; (Srinivas and Sarin, 2012)). Relatively high slope of $P_{\text{Inorg}}/\text{nss-Ca}^{2+}$ in the IGP-outflow might be indicative of anthropogenic source contribution of this macro nutrient than that from the crustal dust. Also, a moderately significant correlation is found between the nss-K^+ and P_{Inorg} ($r = 0.67$; $N = 72$; $P\text{-value} < 0.05$; 3 outliers from the data were excluded from the regression which are beyond two standard deviations of the mean ratio of $\text{nss-K}^+/P_{\text{Inorg}}$). Based on these significant correlations, we infer the dominant contribution of P_{Inorg} from BBEs.

We have also estimated the relative contribution of anthropogenic vis-à-vis to natural source (i.e., biomass burning vis-à-vis mineral dust) to P_{Inorg} in the IGP-outflow. This approach similar to that reported in our earlier publication of (Srinivas and Sarin, 2012). Briefly, we have subtracted the contribution of mineral dust (P_{Dust}) from the measured abundances of P_{Inorg} . The resultant fraction for each sample is estimated for the sampling days and referred as “anthropogenic component (P_{Anth}). The contribution of phosphorus from mineral dust fraction is assessed based on the measured Al concentration (range: 9 – 53 nmol m^{-3} ; on average mineral dust account $\sim 10\%$ of $\text{PM}_{2.5}$ mass) and using a mass ratio of P/Al from the upper crust (~ 0.0087 ; (McLennan, 2001; Taylor and McLennan, 1985)). Although, the applicability of this ratio depends on the geographical location, it is somewhat consistent with that documented for the source soils of IGP and desert dust (Srinivas and Sarin, 2012). Using the similar approach, Mahowald et al, (2005) have estimated the relative contribution of BBEs and land use changes ($\sim 23\%$ of total phosphorus) in supplying this macro nutrient to the tropical Amazonian rain forest. This estimation of P_{Anth} can be represented by the following mathematical expression.

$$P_{\text{Anth}} = P_{\text{Inorg}} - P_{\text{Dust}}$$

$$P_{\text{Anth}} = P_{\text{Inorg}} - [(P/Al)_{\text{Dust}} * Al_{\text{meas}}]$$

In this study, the fractional contribution of P_{Anth} toward P_{Inorg} varies from 50 to 94 % (Av: 75 ± 12 %). This is similar to that observed for the Bay of Bengal during December'08 – January'09, where the $P_{\text{Anth}}/P_{\text{Inorg}}$ averages $\sim 80 \pm 14$ % (range: 35 – 100 %). These observations suggest the predominance of anthropogenic combustion sources from the IGP-outflow in contributing to the aerosol-P. The overwhelming contribution of anthropogenic phosphorus in the IGP-outflow than that derived from the mineral dust, is of comparable level with that reported from the Bay of Bengal (76 ± 18 % in January'09; 77 ± 10 % in March-April'06) and is somewhat higher than that documented for the Arabian Sea (~ 30 %; (Srinivas and Sarin, 2012)).

3.2.3. Water-soluble Iron (Fe)

In the atmospheric outflow from IGP sampled during November'09 – March'10, water-soluble iron (Fe_{ws}) mass concentration exhibits pronounced temporal variability (Fig.3) and varied from 0.2 to 0.6 nmol m^{-3} . A comparison of atmospheric abundances of soluble Fe in the IGP-outflow with other observations reported in the literature is presented in Table 4. It is noteworthy that Fe_{ws} concentrations from the IGP are consistent (See Table 4) with that reported from the urban regions such as Atlanta (Georgia, US; (Rastogi et al., 2009)), Dearborn (Michigan, USA; (Oakes et al., 2010)) and Sahel (west Africa) region (Paris et al., 2010); also consistent with the oceanic regions such as Bay of Bengal (Srinivas et al., 2011a) and Gulf of Aqaba (Chen et al., 2007). The impact of BBEs from these regions have documented in the literature. However, the soluble iron abundances from this study are slightly higher than that observed in the Asian outflow (Chuang et al., 2005), Barbados (Western Equatorial Atlantic, (Trapp et al., 2010)) and oceanic regions such as East China Sea (Hsu et al., 2010b) and North Pacific (Buck et al., 2013).

In addition, the concentrations reported in this study are far higher than that observed over the Arabian Sea (Srinivas et al., 2011a) and the North Atlantic (Chen and Siefert, 2004; Siefert et al., 1999). Based on these observations, it is implicit that atmospheric abundances of soluble Fe show geographical variability (Mahowald et al., 2009). The relatively high concentration of Fe_{ws} in the source region samples is mainly due to their predominant contribution from biomass/bio-fuel burning emissions, whereas the slightly lower concentrations reported in the Asian outflow and over the South China Sea is due to atmospheric processing of mineral dust and slight contribution from anthropogenic sources (Chuang et al., 2005; Zhang et al., 2013). Rather lower concentrations from the North Atlantic and Arabian Sea is due to less anthropogenic influence and the sole contribution from mineral dust (Srinivas et al., 2011a; Trapp et al., 2010).

Preliminary analyses show a significant (P-value < 0.05 ; Fig.6) correlation of $nss-K^+$ with soluble Fe (i.e., Fe_{ws}) in the IGP-outflow and the Bay of Bengal, hints the impact of BBEs. Earlier study by Guieu et al., (2005) has suggested the potential bioavailability of soluble Fe derived from the BBEs to the open ocean regions. Similar to our observations, Paris et al., (2010) have also reported significant linear trends between Fe_{ws} and soluble K^+ in biomass burning aerosols collected from the North Africa (Saheli region) during a field campaign, AMMA-DABEX (Analysis Multidisciplinary

of African Monsoon-Dust and Biomass). Likewise, another study by Oakes et al., (2010) have documented the strong correlation between water-soluble iron and nss-K^+ in $\text{PM}_{2.5}$ samples collected from the Atlanta during forest fires.

Based on the AMBTs over the IGP (Fig.2) together with MODIS (Moderate resolution Imaging Spectro radiometer) fire counts (supplementary material, Fig.S1) for the sampling days, we suggest the significant influence of biomass/bio-fuel burning emissions over the IGP during the study period. In addition, a close match of mass ratio of non-sea-salt potassium to soot carbon (i.e., $\text{nss-K}^+/\text{EC}$) from the study region (0.47 ± 0.12) is consistent with the reported literature range for the BBEs (Ram and Sarin, 2010; Srinivas and Sarin, 2014b) and with that documented for the IGP-outflow over the Bay of Bengal (Srinivas et al., 2011a). All these observation strongly suggest the significant impact of BBEs in the atmospheric outflow to the Bay of Bengal. In addition, no significant differences ($P\text{-value} > 0.05$; Table 2) were observed for the mass ratio of $\text{nss-K}^+/\text{Fe}_{\text{ws}}$ between the IGP-outflow (this study) and the Bay of Bengal (Kumar et al., 2010), reaffirms the impact of biomass/bio-fuel burning emissions from the Indo-Gangetic Plain on the MABL of Bay of Bengal.

Another striking feature of the data pertains to a linear relationship between atmospheric soluble iron (Fe_{ws}) and WSOC in the fine mode aerosols collected from the IGP-outflow and over the Bay of Bengal. A similar relationship between soluble Fe and WSOC has been also documented by Hsu et al., (2010a) for bulk aerosols collected from East-china Sea. The scatter plot between these two variables in $\text{PM}_{2.5}$ samples is shown in Fig.7. It is noteworthy that the slope of the regression between the source region (i.e., Indo-Gangetic Plain) and downwind oceanic region (i.e., Bay of Bengal) has not been significantly different. This observation suggests their common source signature of both species. More recent study by Chang-Graham et al., (2011) have identified the metal-bound water-soluble organic matter (or organometallic species) in the ambient aerosols collected from the BBEs and highlights its potential source contribution. Therefore, the prevailing linear relationship between Fe_{ws} and WSOC could be due to contribution the from BBEs in the IGP. Three outliers have been omitted from the regression for the $\text{PM}_{2.5}$ sampled over the Bay of Bengal whose mass ratio between these two parameters are more than two standard deviations of the data. However, the inclusion of outliers has not made significant variation in slope of the regression ($r = 0.52$; slope = 0.11). It is also possible that these samples have significant contribution from sources other than biomass burning emissions, (for example from mineral dust).

The temporal trend between WSOC and Fe_{ws} for the IGP-outflow and the Bay of Bengal is provided in the supplementary information (Fig.S2). For the $\text{PM}_{2.5}$ fraction sampled over Bay of Bengal, WSOC shows a good correlation and similar temporal trend with that of nss-K^+ over the Bay of Bengal (Srinivas and Sarin, 2014a). Covariability in the temporal trend among WSOC, nss-K^+ and Fe_{ws} together with their inter-relationship (Table 5) further suggest their common source from BBEs. However, it is relevant to state that a linear relationship could also result from similar transport pattern of the chemical species. A comparison of mass ratios of $\text{nss-K}^+/\text{EC}$, $\text{nss-K}^+/\text{OC}$, OC/EC and WSOC/OC in aerosols collected from a downwind site in the IGP with those reported over the Bay of Bengal is consistent with that documented for BBEs (Srinivas and Sarin, 2014b). Based on these observations, we suggest that the enhanced concentration of soluble iron could also be attributed to contribution from BBEs.

In a recent study by Paris et al., (2011) has shown the significant linear relation between water-soluble iron and oxalic acid in the fine mode aerosols sampled from the BBEs. In this regard, a positive linear correlation between low molecular weight diacids (in which oxalic acid is the predominant diacid) and WSOC has been shown by Miyazaki et al., (2009) in aerosols collected from the upwind region of IGP. Furthermore, their study have also documented the high concentrations of nss-K⁺ for the entire sampling period and is attributed to biomass/ bio-fuel burning emissions (Miyazaki et al., 2009). Based on these observations, we infer significant contribution from BBEs in the IGP-outflow to the Bay of Bengal.

4. Linear regression analysis and diagnostic ratios of nutrients

Table 5 presents the Pearson's correlation coefficient of the multiple linear regression analysis performed between the nutrient mass concentrations (NH₄⁺, NO₃⁻, P_{Inorg} and Fe_{ws}) and other anthropogenic species viz., nss-K⁺, water-soluble organic carbon (WSOC), nss-SO₄²⁻ and elemental carbon (EC). Significant correlations were observed among WSOC, Fe_{ws}, nss-K⁺ and NH₄⁺, suggest their emissions from the common source, agricultural crop-waste residue burning and its subsequent downwind long-range atmospheric transport to the sampling site. Similarly, a significant moderate and strong correlation of NH₄⁺ with NO₃⁻ and nss-SO₄²⁻, suggests their formation of NH₄NO₃, NH₄HSO₄ and (NH₄)₂SO₄ in the IGP-outflow.

In order to ascertain the sources of nutrients in the atmospheric outflow from the Indo-Gangetic Plain, a comparison of diagnostic ratios (NH₄⁺/N_{Inorg}, N_{Inorg}/N_{Tot}, N_{Org}/N_{Tot}, P_{Inorg}/nss-Ca²⁺, nss-K⁺/nss-Ca²⁺ and Fe_{ws}/nss-Ca²⁺) has been attempted with those over Bay of Bengal (Fig.8). A noteworthy feature of the chemical composition data is evident from the relative dominance of N_{Inorg} (as NH₄⁺-N) in N_{Tot} in the IGP-outflow and over BoB. In addition, the contribution of N_{Org} in the outflow from IGP is no more than 40 % and is similar to that reported for coastal regions in the Bay of Bengal (Srinivas et al., 2011a). A close similarities in the diagnostic mass ratios of P_{Inorg}/nss-Ca²⁺, P_{Inorg}/EC, nss-K⁺/Ca²⁺ and nss-K⁺/Fe_{ws} serve as finger printing of the outflow from Indo-Gangetic Plain in the MABL.

5. Conclusions

The fine mode (PM_{2.5}) aerosols collected from a sampling site in the Indo-Gangetic Plain have been used to assess the abundances of nutrients in the atmospheric outflow to the Bay of Bengal. Relatively high concentrations of nutrients (NH₄⁺, NO₃⁻, N_{Org}, N_{Tot}, P_{Inorg}, Fe_{ws}) in the atmospheric outflow from the Indo-Gangetic Plain, compared to those documented over the Bay of Bengal, are attributed to the proximity to the anthropogenic sources. The diagnostic ratios of N_{Inorg}/N_{Tot}, N_{Org}/N_{Tot}, P_{Inorg}/nss-Ca²⁺, nss-K⁺/nss-Ca²⁺ and nss-K⁺/Fe_{ws} in the IGP-outflow are, however, not statistically different from those over the Bay of Bengal. These results have implications to long-term monitoring of atmospheric deposition of nutrients to the oceanic regions (Bay of Bengal) located downwind of the pollution sources.

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Table 1. Statistical description (range, mean $\pm 1\sigma$) of analytical data on atmospheric abundances of nutrients and their molar ratios in the atmospheric outflow from the Indo-Gangetic Plain and over the Bay of Bengal.

Parameter	IGP-outflow (Nov'09-Mar'10)		#Bay of Bengal (Dec'08-Jan'09)		Two-tailed t-test
	Range	Av \pm Sd (X_g)	Range	Av \pm Sd (X_g)	t-Score, df, *P-value
NO ₃ ⁻	0.5 -184	30 \pm 41 (12)	\leq 0.13 - 22	4.2 \pm 5.4 (3)	3.1, 69, < 0.05
NH ₄ ⁺	109 -559	290 \pm 108 (270)	16 -561	178 \pm 139 (129)	4, 75, < 0.05
N _{Inorg}	109 – 703	320 \pm 136 (292)	18 - 565	182 \pm 142 (133)	4.3, 75, < 0.05
^s N _{Org}	18 – 234	96 \pm 63 (79)	7 - 173	50 \pm 41 (37)	2.8, 28, < 0.05
^s N _{Tot}	212 – 676	388 \pm 137 (367)	24 -738	278 \pm 209 (194)	3.2, 75, < 0.05
P _{Inorg}	0.39 - 4.79	1.19 \pm 0.89 (0.99)	\leq 0.01 - 0.78	0.33 \pm 0.17 (0.29)	5.2, 74, < 0.05
Fe _{ws}	0.21 - 0.58	1.06 \pm 0.56 (0.91)	0.06 - 0.68	0.30 \pm 0.16 (0.25)	7.1, 75, < 0.05
nss-K ⁺	18.0 - 98.4	50.9 \pm 15.1 (48.6)	2.1 - 29.1	12.6 \pm 5.8 (11.3)	13.4, 75, < 0.05
nss-Ca ²⁺	1.7 - 9.2	4.0 \pm 1.5 (3.8)	0.1 - 2.9	1.4 \pm 0.7 (1.2)	8.8, 68, < 0.05
NO ₃ ⁻ /N _{Inorg}	0.004 - 0.26	0.07 \pm 0.08 (0.04)	0.001 - 0.19	0.03 \pm 0.04 (0.02)	2.6, 69, < 0.05
NH ₄ ⁺ /N _{Inorg}	0.74 - 1.00	0.93 \pm 0.08 (0.92)	0.81 - 1.00	0.98 \pm 0.04 (0.98)	3.2, 75, < 0.05
N _{Inorg} /N _{Tot}	0.60 - 1.09	0.92 \pm 0.13 (0.96)	0.62 - 1.22	0.88 \pm 0.12 (0.87)	1.4, 75, > 0.05
^s N _{Org} /N _{Tot}	0.12 - 0.40	0.25 \pm 0.10 (0.23)	0.12 - 0.38	0.20 \pm 0.07 (0.19)	0.5, 32, > 0.05
P _{Inorg} /nss-Ca ²⁺	0.06 - 1.01	0.35 \pm 0.23 (0.28)	0.07 - 1.66	0.30 \pm 0.29 (0.23)	0.8, 67, > 0.05
nss-K ⁺ /nss-Ca ²⁺	5.2 - 27.5	13.8 \pm 4.8 (13.1)	2.5 - 23	9.1 \pm 3.6 (8.5)	0.8, 68, > 0.05
nss-K ⁺ /Fe _{ws}	26 – 160	60 \pm 31 (54)	17 - 156	50 \pm 27 (44)	1.5, 75, > 0.05

Note: All concentrations were in nmol m⁻³. # Data obtained from Srinivas et al., (2011a,b) and Srinivas and Sarin, (2012).

^s Only those data were considered whose propagated error in the estimation of N_{Org} is less than its concentration (See Material and Methods).

* denotes the statistical significance of the measured parameter at 95 % C.I. X_g refers to geometric mean.

Table 2. Mass concentration of water-soluble organic nitrogen and its fractional contribution to the total soluble nitrogen from the Indo-Gangetic Plain and other oceanic regions.

Location	N_{Org} (nmol m ⁻³)	N_{Org}/N_{Tot}	Reference
Indo-Gangetic Plain	18 -234	0.25	This study
Bay of Bengal	0 - 202	0.17	Srinivas et al, 2011a
Arabian Sea	2.4 - 12.5	-	Srinivas and Sarin, 2013
Northern. California	3.1 - 57.8	0.20	(Zhang et al., 2002)
Gulf of Aquaba	0.05 - 29	0.11	Chen et al., 2008
Eastern Mediterranean	BDL - 150	0.26	(Mace et al., 2003)
Broad leaf forest, Northern Japan	-	0.2	(Miyazaki et al., 2014)
North Atlantic Ocean	-	0.24	(Lesworth et al., 2010)
Yellow Sea	-	0.17	(Shi et al., 2010)
Eastern Mediterranean	-	0.23	(Violaki et al., 2010)
East China Sea	-	0.24	(Nakamura et al., 2006)
South China Sea	-	0.33	(Shi et al., 2010)
Southeast US	-	0.33	(Lin et al., 2010)
Hawaii	-	0.16	(Carrillo et al., 2002)

Table 3. Atmospheric abundance of soluble phosphorus (P_{Inorg} , in nmol m^{-3}) from different oceanic regions

Geographical Location	Geographical coordinates	Season	Sample type	P_{Inorg}	Reference
Indo-Gangetic Plain	22.3 °N, 87.3 E	W & SIM	PM _{2.5}	0.4 - 4.8	This study
Bay of Bengal	0 - 25 °N, 70 -100 °E	W	PM _{2.5}	0.1 - 0.8	Srinivas and Sarin, 2012
Bay of Bengal	0 - 25 °N, 70 -100 °E	SIM	TSP	0.3 - 2.8	Srinivas and Sarin, 2012
Arabian Sea	0 - 25 °N, 55 - 70 °E	SIM	TSP	0.2 - 0.5	Srinivas and Sarin, 2012
Gulf of Aqaba	24 - 32°N, 120 - 128°E	-	TSP	0.02 - 2.0 (0.4)	Chen et al., 2008
South China Sea	24 - 32°N, 120 - 128°E	-	TSP	0.21 ± 0.20	Hsu et al., 2010
North Atlantic				0.004 - 0.11	Baker et al., 2007
Sahel, West Africa	8 - 14 °N, 2 - 5 °E		-	≤ 3.2 - 5.8	Paris et al., 2010

Here S, F, W and SIM refer to summer, fall, winter and spring-intermonsoon seasons, respectively.

Table 4. Atmospheric abundance of soluble iron (Fe_{ws} , in pmol m^{-3}) from different oceanic regions.

Geographical Location	Geographical coordinates	Season	Sample type	Fe_{ws}	Reference
Indo-Gangetic Plain	22.3 °N, 87.3 E	W & SIM	$\text{PM}_{2.5}$	206 - 2578	This study
Bay of Bengal	0 - 25 °N, 70 -100 °E	W	$\text{PM}_{2.5}$	63 - 678	Kumar et al., 2010
Bay of Bengal	0 - 25 °N, 70 -100 °E	SIM	TSP	170 - 1188	Srinivas et al., 2011
Arabian Sea	0 - 25 °N, 55 - 70 °E	SIM	TSP	0.8 - 17.7	Srinivas et al., 2011
Arabian Sea	0 - 25 °N, 55 - 70 °E	SIM	$\text{PM}_{10-3.0}$	$\leq 7.1 - 84.8$	Siefert et al., 1999
Arabian Sea	0 - 25 °N, 55 - 70 °E	SIM	$\text{PM}_{10-3.0}$	$\leq 1.6 - 6.1$	Siefert et al., 1999
North Atlantic	0 - 25 °N, 55 - 70 °E	S		3.2 - 1815	Buck et al., 2010b
North Atlantic	26 - 30 °N	W	$\text{PM}_{2.5}$	0.9 - 35	Chen et al., 2004
North Atlantic	5 - 26 °N	W	$\text{PM}_{2.5}$	7.9 - 400	Chen et al., 2004
North Atlantic	26 - 30 °N	W	$\text{PM}_{2.5}$	1.1 - 13.2	Chen et al., 2004
North Atlantic	6 - 26 °N	W	$\text{PM}_{2.5}$	3.6 - 28.8	Chen et al., 2004
North Atlantic	0 - 6 °N	W	$\text{PM}_{2.5}$	0.5 - 4.6	Chen et al., 2004
North Pacific	30 °N, 150 °W	-		0.7 - 509	Buck et al., 2013
Atlanta, Georgia, US	33°51'43.56"N, 84°41'16.44"W	-	$\text{PM}_{2.5}$	82 - 6607	Rastogi et al., 2009
Dearborn, Michigan, US	42°19'00N, 083°13'53W	F, SIM, S	$\text{PM}_{2.5}$	82 - 7142	Oakes et al., 2010
Barbados, WE.Atlantic	13.17 °N, 59.43 °W	S	TSP	31 -653	Trapp et al., 2010
Gulf of Aqaba, Eilat, Israel	-	R.Y.	TSP	40 - 1000	Chen et al., 2007
East China Sea	24 - 32°N, 120 - 128°E	-	TSP	375 ± 393	Hsu et al., 2010
Asian outflow	-	-	TSP	571 ± 339	Chuang et al., 2005

Note: Here S, F, W and SIM refer to summer, fall, winter and spring-intermonsoon seasons, respectively.

Table 5. Multiple linear regression analysis between the nutrient concentrations and other anthropogenic water-soluble species for the fine mode aerosols collected from the IGP-outflow.

Parameter	NH_4^+	nss- K^+	P_{Inorg}	NO_3^-	nss- SO_4^{2-}	EC	WSOC	Fe_{ws}
NH_4^+	1.00							
nss- K^+	0.75	1.00						
P_{Inorg}	0.34	0.33	1.00					
NO_3^-	0.53	0.30	0.49	1.00				
nss- SO_4^{2-}	0.92	0.66	0.19	0.29	1.00			
EC	0.09	0.30	0.41	0.12	0.08	1.00		
WSOC	0.60	0.68	0.41	0.38	0.60	0.56	1.00	
Fe_{ws}	0.61	0.57	0.67	0.66	0.51	0.40	0.69	1.00

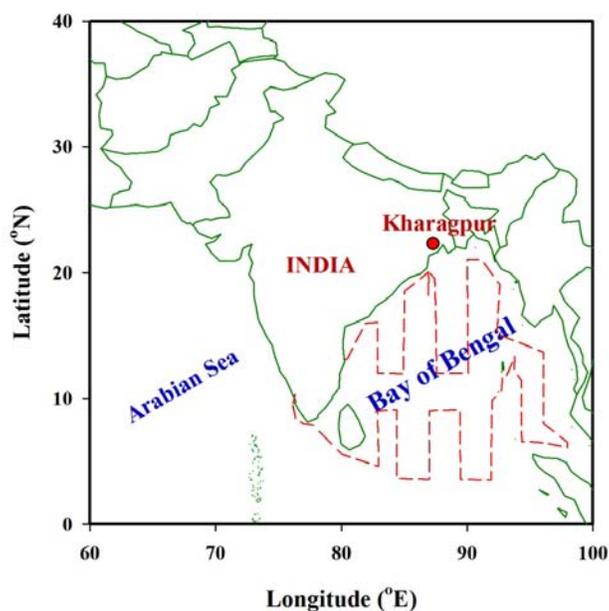


Fig.1. Map showing downwind sampling site at Kharagpur in the Indo-Gangetic Plain. Also shown is a cruise track conducted in the Bay of Bengal (Kumar et al., 2010).

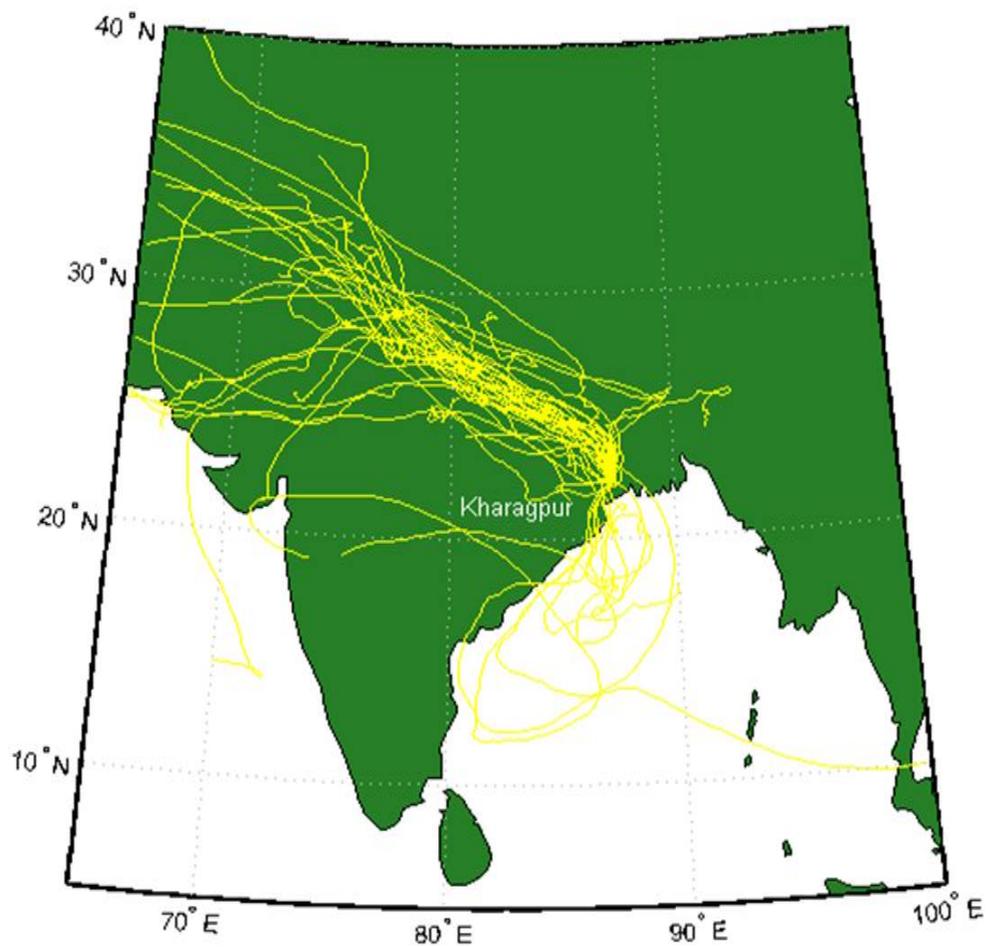


Fig.2. Air mass back trajectory cluster for sampling days (November'09-March'10) at an arrival height of 500 m.

Fig.3. Temporal variability in the concentration of soluble nutrients (NO_3^- , NH_4^+ , P_{Inorg} and Fe_{ws}) in the atmospheric outflow from the Indo-Gangetic Plain.

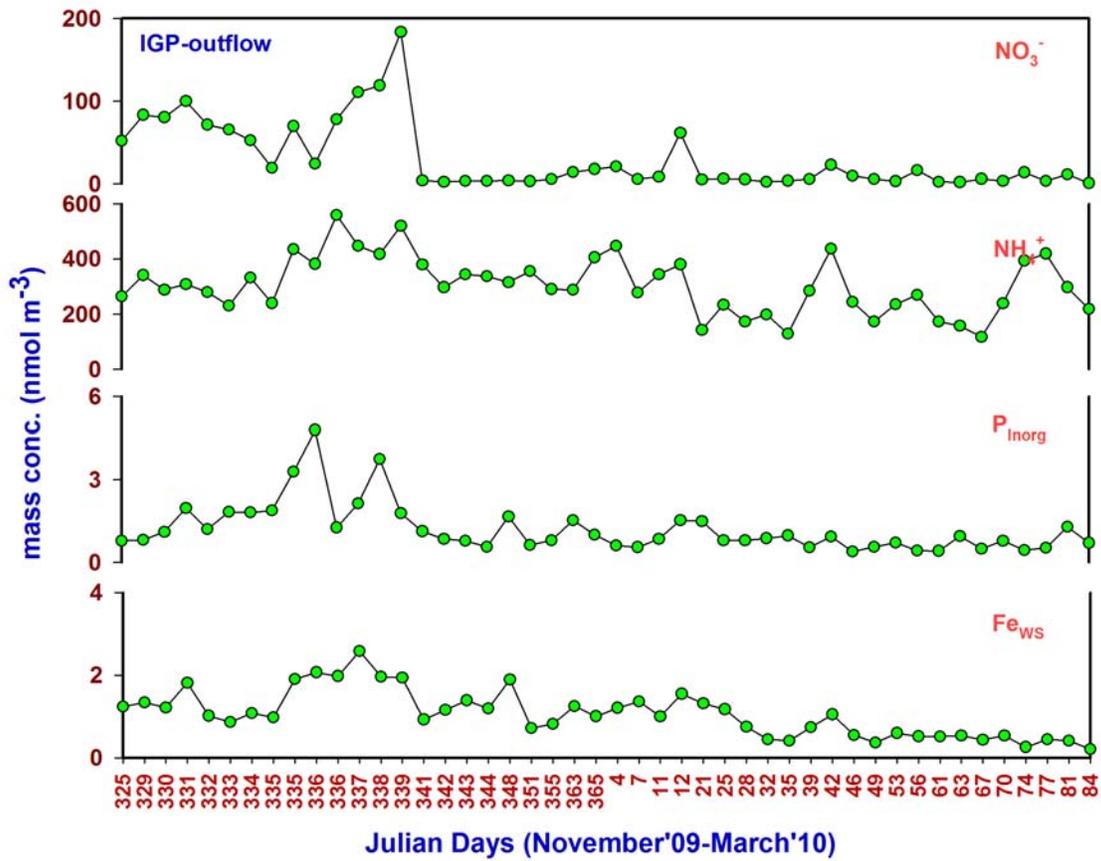


Fig.4. Scatter plot between abundances of NH_4^+ and N_{Inorg} in the (a) IGP-outflow and (b) Bay of Bengal. Likewise scatter plot between N_{Inorg} and N_{Tot} in (a) IGP-outflow and (b) Bay of Bengal, suggests the predominance inorganic nitrogen (i.e., as $\text{NH}_4^+\text{-N}$) towards total soluble nitrogen.

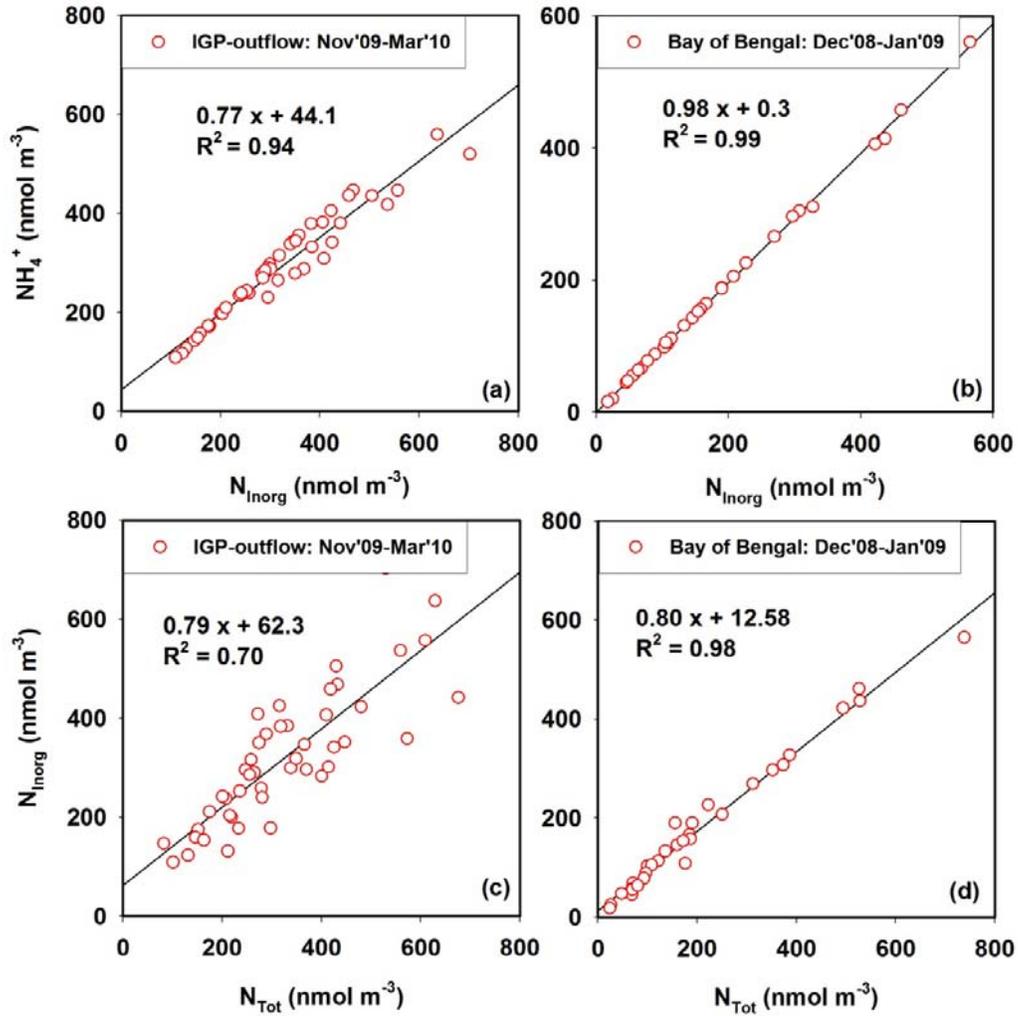


Fig.5. Scatter plot between mass concentration of water-soluble inorganic phosphorus (P_{Inorg}) and (a) non-sea-salt potassium ($nss-K^+$); (b) $nss-Ca^{2+}$ for the fine mode aerosols collected from the IGP-outflow during November'09 – December'10 and from the Bay of Bengal during December'08 – January'09.

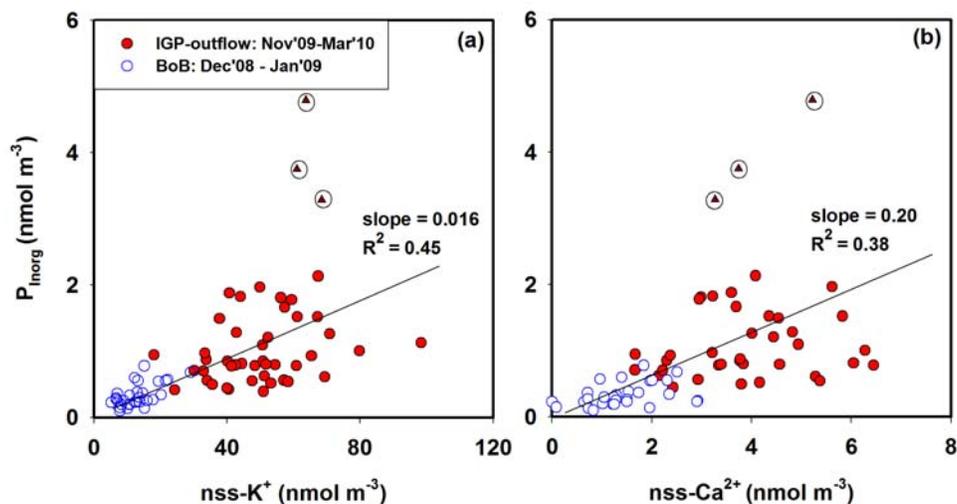


Fig.6. Scatter plot between mass concentrations of water-soluble Fe and non-sea-salt K^+ in aerosols collected from the Indo-Gangetic outflow (this study) and over the Bay of Bengal (January'09; Srinivas and Sarin, 2012).

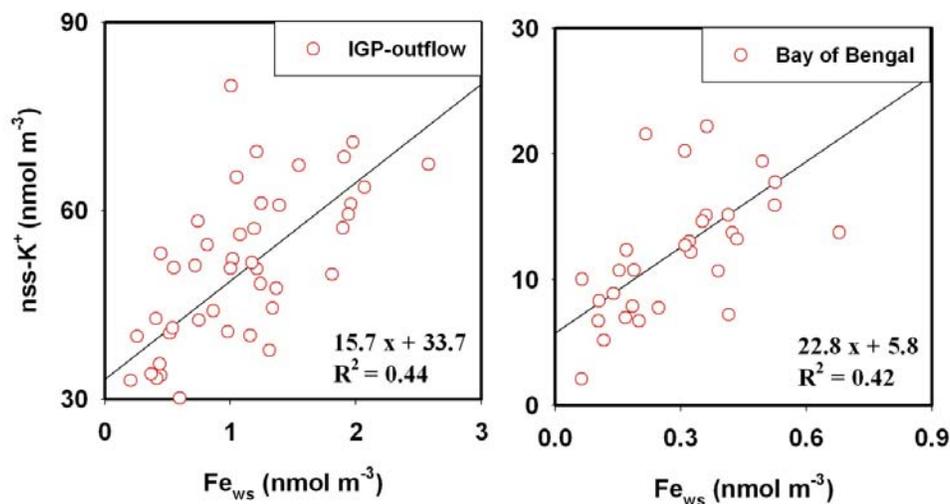


Fig.7. Scatter plot between water-soluble organic carbon (WSOC) and iron (Fe_{ws}) in fine mode aerosols ($PM_{2.5}$) collected from the IGP-outflow (November'09-March'10) and the Bay of Bengal (December'08-January'09). The three outliers show a phase difference between their temporal trends and could have contribution from other sources such as fossil-fuel combustion and mineral dust.

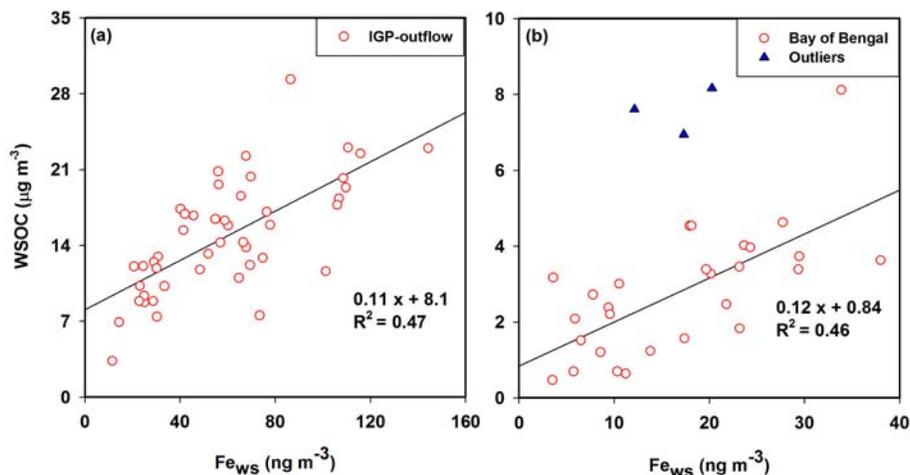
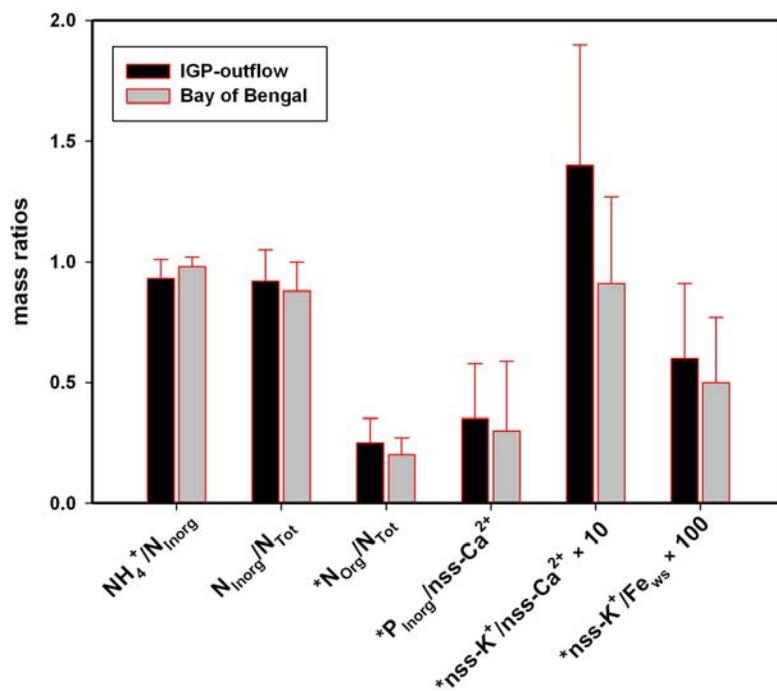


Fig.8. A comparison of diagnostic mass ratios of nutrients in aerosols collected from the Indo-Gangetic Outflow with that from the Bay of Bengal during the continental outflow.



Supplimentary Information

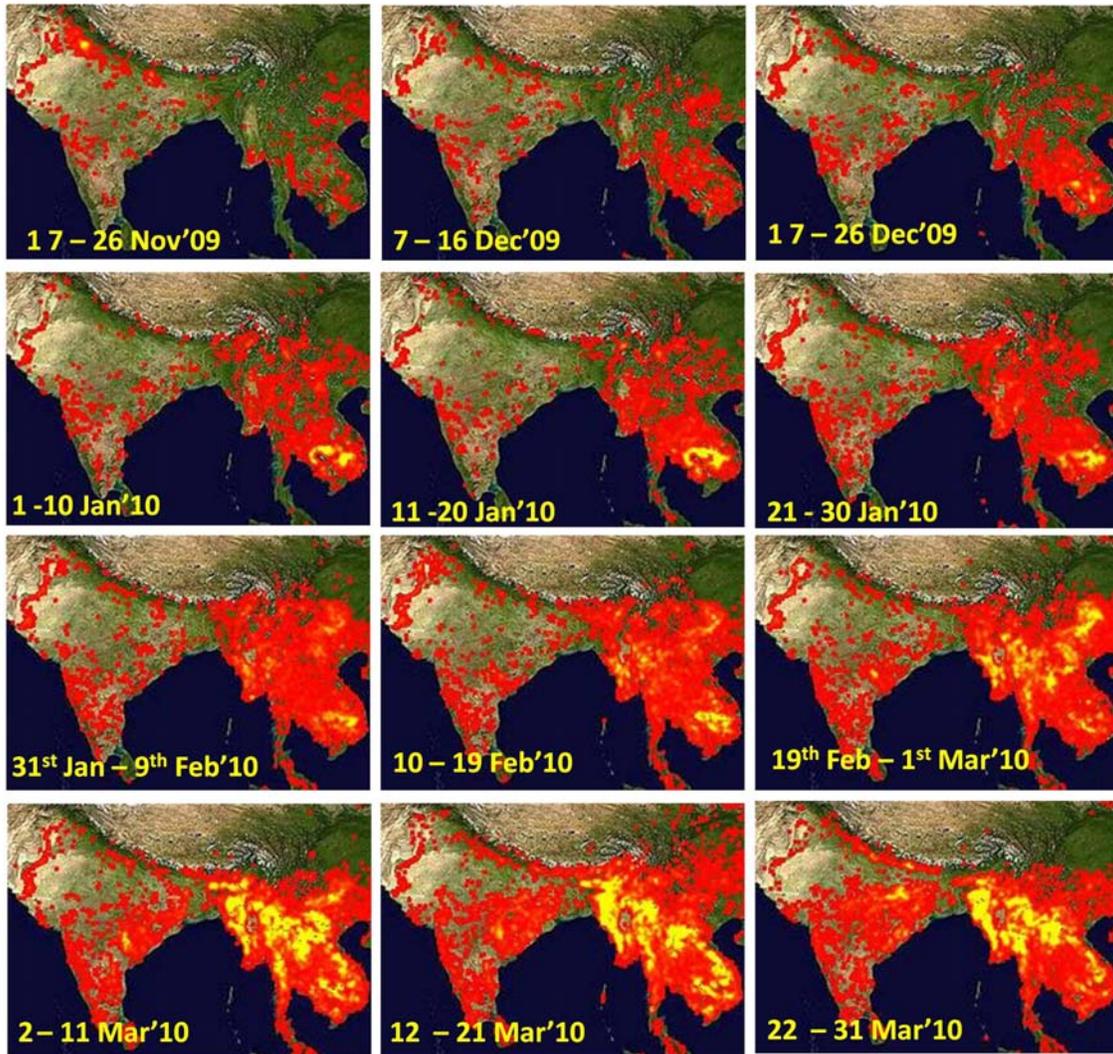


Fig.S1. MODIS fire maps during the sampling days (November'09-March'10). (source: <http://earthdata.nasa.gov/data/near-real-time-data/firms/active-fire-data#tab-content-7>).

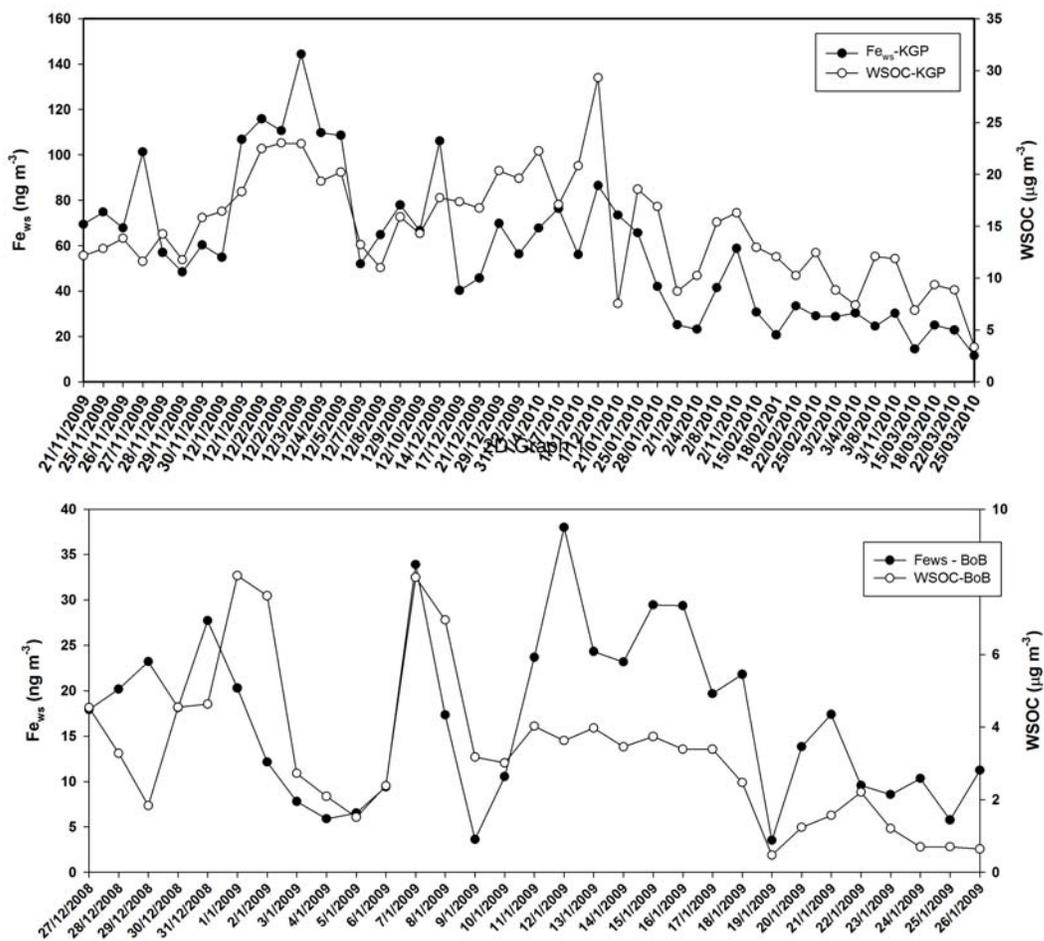


Fig.S2. Covariability in the temporal trend between atmospheric soluble iron (Fews) and water-soluble carbon (WSOC) in fine mode aerosols collected from the IGP-outflow (November'09-March'10) and over the Bay of Bengal (BoB: December'08 – January'09).

Appendix: Table of abbreviations

AMBT: Air mass back trajectory
Al_{meas}: Measured Aluminium Concentration
BBE: Biomass burning emissions
BoB: Bay of Bengal
EC: Elemental carbon
Fe_{ws}: Water-soluble iron
GDAS: Global Data Assimilation System
GF-AAS: Graphite furnace atomic absorption spectrophotometer
IGP: Indo-Gangetic Plain
MABL: Marine atmospheric boundary layer
MDL: Method detection limit
N_{Inorg}: Water-soluble inorganic nitrogen
N_{Org}: Water-soluble organic nitrogen
N_{Tot}: Water-soluble total nitrogen
nss-Ca²⁺: non-sea-salt calcium
nss-K⁺: non-sea-salt potassium
nss-SO₄²⁻: non-sea-salt sulphate
OC: Organic carbon
P_{Inorg}: Water-soluble inorganic phosphorous
P_{Anth}: Water-soluble anthropogenic phosphorous
P_{Dust}: Water-soluble dust derived phosphorous
TSP: Total Suspended Particulate matter
WSOC: Water-soluble Organic Carbon