Atmospheric pathways of phosphorous to the Bay of Bengal: contribution from anthropogenic sources and mineral dust

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ABSTRACT

The continental outflow from the Indo-Gangetic Plain and Southeast Asia, during the late NE monsoon (January–March), dominates the transport of chemical constituents to the marine atmospheric boundary layer (MABL) of the Bay of Bengal (BoB). During the rest of the year, prevailing wind regimes and meteorological conditions do not favour the atmospheric transport of continental products. Here we report on the spatio-temporal variability of inorganic phosphorous ($P_{Inorg} = PO_4^{3-}$) in the MABL and its dry-deposition flux to the surface BoB. On the basis of the abundance of P_{Inorg} in $PM_{2.5}$ ($0.1–0.8\ nmol\ m^{-3}$) and PM_{10} ($0.3–2.8\ nmol\ m^{-3}$), we document its dominant occurrence in the coarse mode ($D_a \geq 2.5\ \mu m$). The analytical data also provide evidence for the chemical processing of mineral dust by acidic species and mobilisation of P_{Inorg} during the long-range atmospheric transport. However, significantly high P_{Inorg}/non-sea-salt Ca^{2+} ratios over the BoB suggest dominant contribution from anthropogenic sources (fertilisers and biomass burning emissions). P_{Inorg} concentration over the Arabian Sea is about 4 to 5 times lower and is primarily associated with the mineral dust from desert regions. The dry-deposition flux of P_{Inorg} to the BoB varies by one order of magnitude ($0.5–5.0\ \mu mol\ P\ m^{-2}\ d^{-1}$; Av: $0.02\ Tg\ P\ yr^{-1}$). These results have implications to the air-sea deposition of phosphorous over oceanic regions downwind of the pollution sources and impact on the biogeochemistry of surface waters.

Keywords: aerosol phosphorus, size-distribution, chemical processing, mineral dust, anthropogenic sources, Bay of Bengal, Arabian Sea

1. Introduction

Phosphorous is one of the limiting nutrients for the primary productivity in both terrestrial and marine ecosystems (Chester, 2002; Fang, 2004). It has long been suggested that the marine primary productivity over the geological time scale was limited by the availability of phosphorous (Tyrrell, 1999). Recent observations also suggest that productivity in some of the oceanic regions (Mediterranean Sea and Sargasso Sea) and in marine basins, fed by large quantity of freshwater, is limited by the availability of phosphorous (Paytan and McLaughlin, 2007 and references therein). The role of phosphorus limitation in nitrogen fixation has also been emphasised for open ocean waters of the Atlantic and Pacific (Sanudo-Wilhelmy et al., 2001; Mills et al., 2004; Moutin et al., 2008). In this context,

atmospheric mineral dust is considered as a major source of phosphorous to the ocean surface (Herut et al., 1999a; Ridame and Guieu, 2002; Krom et al., 2004; Anderson et al., 2010; Nenes et al., 2011).

However, the present-day increase in the supply of phosphorous from anthropogenic sources would lead to significant changes in the marine biogeochemical cycle of nutrients (Schlesinger, 1997; Mahowald et al., 2008). It has been argued that the global atmospheric cycle of phosphorous is significantly perturbed as a result of the extensive use of fertilisers and mining of phosphate rocks (Jahnke, 2000). The emissions from biomass burning has also been suggested to be a significant source of phosphorous (Mahowald et al., 2005, 2008; Anderson et al., 2010 and references therein). Recent studies have documented that the magnitude of atmospheric input of phosphorous may attain comparable levels with respect to its riverine supply (Compton et al., 2000; Paytan and McLaughlin, 2007). The uptake of anthropogenic species (non-sea-salt (nss)-SO_4^{2-}, NO_3^-) by

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Ca-rich mineral aerosols during the long-range transport has been documented in the literature (Wolf, 1984; Rastogi and Sarin, 2006; Saliba and Chamseddine, 2012). The chemical uptake of acidic species enhances the solubility of Ca-rich minerals such as carbonates and apatite $(Ca_3(PO_4)_2)$, and hence mineral dust is considered as a significant source of P_{Inorg} from ambient aerosols.

Therefore, atmospheric transport and air-sea deposition of phosphorous demand further investigation in the regional context. This article presents the first data set on the concentration of water-soluble inorganic phosphorus (P_{Inorg}) in ambient aerosols, its sources, size distribution and dry-deposition towards the Bay of Bengal (BoB).

2. Experimental methods

2.1. Study region and cruise tracks

The Bay of Bengal (BoB) and the Arabian Sea (ARS) are the two limbs of the northern Indian Ocean. The prevailing meteorological conditions during the late NE monsoon (January–March) favour the advective transport of pollutants from the Indo-Gangetic Plain and Southeast Asia to the BoB. In this context, earlier studies have highlighted the impact of continental outflow on the marine atmospheric boundary layer (MABL) of the northern Indian Ocean (Lelieveld et al., 2001; Ramanathan et al., 2001; Kumar et al., 2010).

Cruise tracks undertaken in the BoB, on board ORV Sagar Kanya, during March–April 2006 (Cruise: SK-223A) and January 2009 (SK-254) are shown in Fig. 1. During March–April 2006, prevailing winds over the BoB were initially north-easterly and changed to south-westerly in the later part of the cruise (a transition period), whereas the winds were predominantly north-easterly during the cruise in January 2009 (Fig. 1). A detailed description of meteorological conditions during all cruises has been reported in earlier publications (Alappattu et al., 2008; Kumar et al., 2010; Subrahamanyam et al., 2011). For comparison, chemical data on aerosol samples collected from the ARS (Fig. 1) have been used to characterise the contribution of inorganic phosphorous associated with the mineral dust from the desert regions. Although wet deposition is considered as a major source of nutrients to the ocean, wet precipitation events over ARS and BoB seldom occur during the late NE monsoon (January–March). We have, thus, addressed the dry-deposition fluxes of phosphorous to the surface waters of these two oceanic regions.

2.2. Sample collection and analysis

As a part of the Indian National Programme on 'Integrated Campaign of Aerosols and Trace Gases Radiation Budget (ICARB)', aerosol samples from MABL of the BoB and the ARS were collected using high-volume samplers (flow rate: $1.1\,m^3\,min^{-1}$). During 19th March–12th April 2006, PM_{10} samples ($n = 23$) were collected from the BoB. In a later cruise undertaken from 27th December 2008 to 28th January 2009, two size fractions of aerosols, $PM_{2.5}$ ($n = 31$) and PM_{10} ($n = 33$), were collected over the bay region. In addition, PM_{10} samples ($n = 19$) were collected from the ARS during 18th April–11th May 2006. All samples were collected using PALLFLEX®™ tissuquartz filters and were analysed for water-soluble ionic species (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^- and NO_3^-), crustal elements (Al, Ca, Fe and Mg) and carbonaceous species (EC and OC). The relevant details and analytical procedures are described in earlier publications (Kumar et al., 2008, 2010). The nss components of SO_4^{2-}, K^+ and Ca^{2+} were assessed by correcting their measured concentrations for the contribution from sea salts (Keene et al., 1986), where nss-$SO_4^{2-} = SO_4^{2-} - 0.253 \times Na^+$; nss-$K^+ = K^+ - 0.037 \times Na^+$; nss-$Ca^{2+} = Ca^{2+} - 0.038 \times Na^+$, and 0.253, 0.037 and 0.038 are the ratios of SO_4^{2-}, K^+ and Ca^{2+} to Na^+ in seawater, respectively.

For the analysis of P_{Inorg} (PO_4^{3-}) (this study), one quarter of the sample filter was extracted with Milli-Q water (specific resistivity $\leq 18.2\,M\Omega\,cm$). The water extract was filtered through $0.45\,\mu m$ (25 mm in diameter) Millipore filter and concentration of P_{Inorg} was measured using a 2-m-long liquid core waveguide capillary cell (LWCC, WPI Model) connected to USB-4000 spectrophotometer (Ocean Optics). Reagents and preparation of standard solutions for the determination of P_{Inorg} are similar to those described by (Li and Hansell, 2008). The absorption of the phosphate–molybdenum blue complex was measured at a wavelength of 710 nm (Zhang and Chi, 2002). The overall reproducibility in the measurement of P_{Inorg} is within 10%, assessed based on the repeat analysis of a commercial standard (Merck) and repeat checks of sample extracts. The detection limits, reported as three times the standard deviation of procedural blanks and normalised to average volume of air filtered ($\sim 1400\,m^3$), for Na, Ca, Ca^{2+}, K^+, NO_3^-, SO_4^{2-} and P_{Inorg} are 0.35, 0.38, 0.08, 0.17, 0.18, 0.31 and $0.013\,nmol\,m^{-3}$, respectively.

We have estimated P_{Dust} by multiplying P/Al ratio of 0.008 [as the revised composition of Upper Continental Crust (UCC) (McLennan, 2001); $P = 700$ ppm; $Al = 8.04\%$] with the Al concentration in aerosol samples (this study). In this calculation, it is implicit that Al is solely derived from the Continental Crust. A ratio of 0.012 for P/Al has been suggested for Saharan dust by Guieu et al. (2002). Similarly, Herut et al. (1999a) had suggested a ratio of about 0.011 for the Saharan aerosols. However, there are no data available for dust composition from the Thar Desert. We have used the P/Al ratio of UCC by
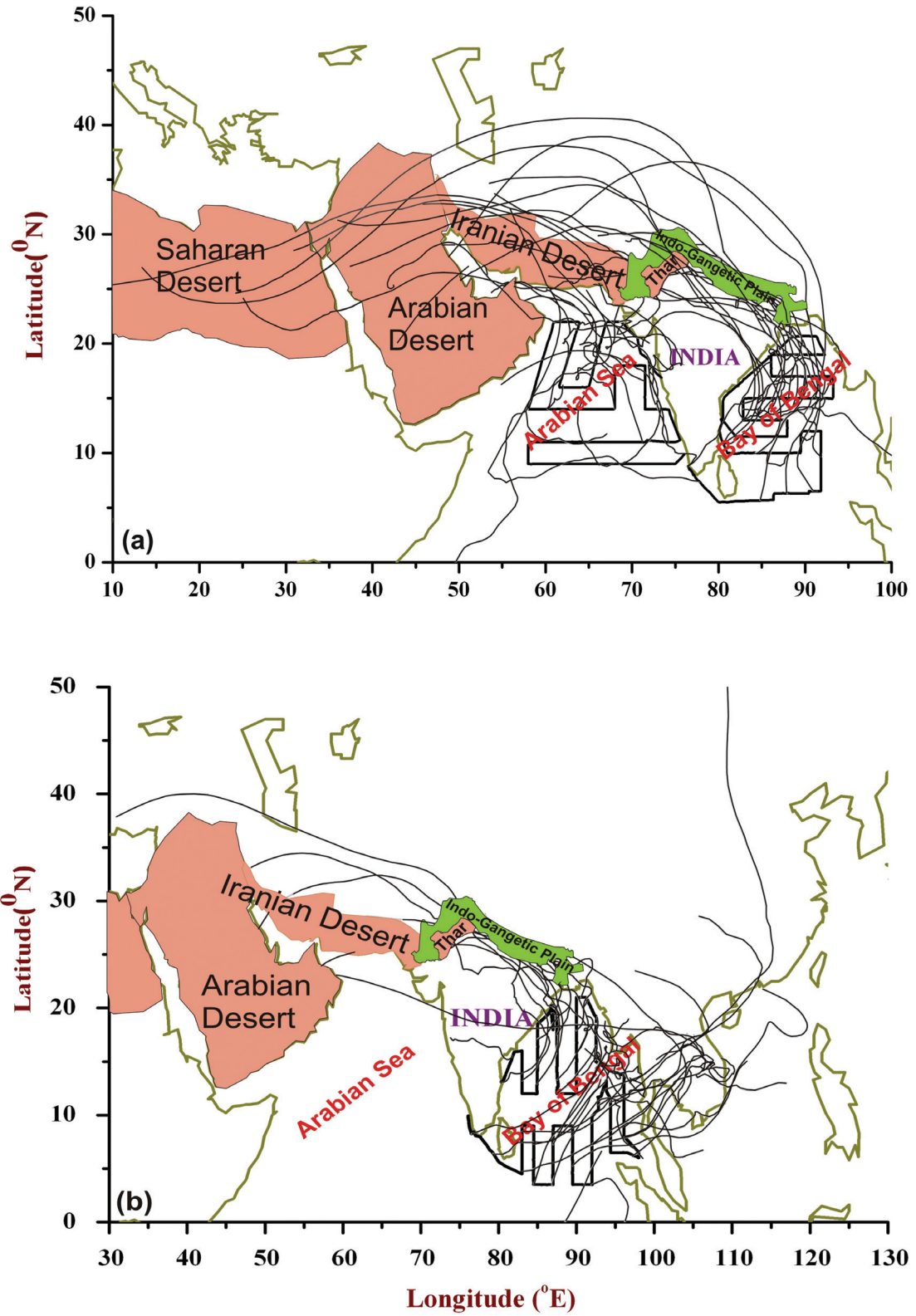
Fig. 1. Cruise tracks undertaken on board ORV Sagar Kanya: (a) Bay of Bengal (BoB; SK-223A: March–April 2006) and Arabian Sea (ARS: SK-223B: April–May 2006); (b) BoB (SK-254: January 2009); 7-d air mass back trajectories (AMBTs) are computed at an arrival height of 1000 m.

Taylor and McLennan (1985), subsequently revised by McLennan (2001).

2.3. Statistical analyses

A summary of the chemical data (mean with $\pm 1\sigma$) obtained from cruises in the BoB (March–April 2006 and January 2009) and the ARS (April–May 2006) is presented in Tables 1 and 2. A comparison between the abundances of P_{Inorg} and chemical constituents is presented in terms of their statistical significance. Linear regression analysis of the measured chemical constituents is also performed to infer about the provenance of aerosol phosphorus. Furthermore, ANOVA analysis (including post-hoc tests) has been carried out to evaluate the seasonal variability in the molar ratios of nss-K^+/nss-Ca^{2+}, nss-SO_4^{2-}/nss-Ca^{2+} and P_{Inorg}/nss-Ca^{2+} for the data (Table 2) obtained from all cruises (March–April 2006, April–May 2006 and January 2009).

3. Results and discussions

3.1. Air mass back trajectory (AMBT) analyses

The isentropic AMBTs were computed (for 7 d) using the HYSPLIT Model (Version-4; Draxler, 2002) by reanalysis of NCEP data set (GDAS, global data set from 2006 to present, http://ready.arl.noaa.gov/hysplit-bin/trajasrc.pl).

Table 1. Chemical composition (mean $\pm 1\sigma$) of aerosols (PM_{10}) from the Bay of Bengal (BoB) and the Arabian Sea (ARS)

	BoB		ARS
Parameter	January 2009[a]	March–April 2006[b]	April–May 2006[b]
P_{Inorg}^a	1.3 ± 0.5	1.1 ± 0.3	0.3 ± 0.1
Na^+	75 ± 35	37 ± 27	83 ± 106
NH_4^+	171 ± 182	50 ± 35	BDL
K^+	17 ± 7	6 ± 3	3 ± 2
Mg^{2+}	9 ± 6	7 ± 3	8 ± 11
Ca^{2+}	6 ± 3	17 ± 9	21 ± 14
Cl^-	9 ± 10	0.4 ± 0.3	62 ± 110
NO_3^-	14 ± 10	1.2 ± 1.4	8 ± 7
SO_4^{2-}	123 ± 79	62 ± 26	35 ± 12
Al	41 ± 35	34 ± 20	32 ± 17
Ca	8 ± 4	18 ± 9	22 ± 15
Fe	8 ± 8	11 ± 6	8 ± 5
Mg	13 ± 5	12 ± 6	20 ± 11
OC	542 ± 434	157 ± 132	18 ± 13
EC	159 ± 119	33 ± 13	7 ± 3

All concentrations are expressed in nmol m^{-3}.
[a]This study.
[b]Kumar et al. (2008).

Air mass back trajectories computed at an arrival height of 1000 m (and above) suggest that the origin of air masses during January 2009 cruise is mainly from the Indo-Gangetic Plain and Southeast Asia (Fig. 1). In contrast, air masses originate from the desert regions of Sahara, Arab, Oman, Somali and Thar, as observed during cruises undertaken from March to May 2006 (Fig. 1). The advective transport of mineral dust from the Middle East, Arabian Peninsula, Oman, Iran, Thar and Somalia to the MABL of ARS has been well recognised (Middleton, 1986; Tindale and Pease, 1999; Prospero et al., 2002). A detailed description on the transport pathways of aerosols to the ARS and BoB, during January to April, has been reported in earlier publications (Siefert et al., 1999; Dey et al., 2004; Kumar et al., 2008; Badarinath et al., 2010; Srinivas et al., 2011). However, a brief discussion is presented on the transport pathways of the mineral dust to these oceanic regions (BoB and ARS).

It has been reported that the high-altitude transport of mineral dust from the Middle East, Arabia, Iran and Thar Deserts to the ARS is a conspicuous feature during the summer months (March–May). However, at lower altitude, the Findlater jet counters the atmospheric transport from the desert areas to the ARS (Tindale and Pease, 1999). The periodicity and frequency of dust storms significantly increase during Spring and Summer months (Middleton, 1989). This observation is further corroborated by the total ozone mapping spectrometer absorbing aerosol index (TOMS AAI) data (based on the analysis of a 13-year data, from 1980 to 1992), suggesting that dust storm activity starts during March–May from the Arabian and Iranian Deserts and its magnitude peaks during June–July (Prospero et al., 2002). Likewise, the dust storm activity from the Thar region begins in the early Spring inter-monsoon (March–April) and increases in strength during May–June as inferred from the intermonsoon TOMS Frequency of Occurrence and AAI (Prospero et al., 2002). Herman et al. (1997) have highlighted the application of satellite mapping of large dust events using TOMS of the NIMBUS-7 Satellite. A recent study has also emphasised the high-altitude transport (between 3 and 5 km) of dust storm originating from the Persian Gulf and ARS region and travel towards the Indian subcontinent (Badarinath et al., 2010). On the basis of LIDAR studies, an evidence for high-altitude dust transport (~ 3–3.5 km), during March–April 2006, has been reported by Raj et al. (2008). Likewise, high aerosol loading over the ARS observed during INDOEX field experiment was explained by invoking high-altitude transport of mineral dust from desert (viz., Arabia, Thar, Iran, Somalia) regions (Lelieveld et al., 2001; Ramanathan et al., 2001).

Table 2. Phosphorous concentration and diagnostic ratios in aerosols (PM_{10}) from the Bay of Bengal (BoB) and the Arabian Sea (ARS)

Parameter	BoB		ARS
	January 2009	March–April 2006[a]	April–May 2006[a]
P_{Inorg}	1.3 ± 0.5	1.1 ± 0.3	0.3 ± 0.1
P_{Dust}^{b}	0.3 ± 0.3	0.3 ± 0.2	0.2 ± 0.1
P_{Anth}^{c}	1.0 ± 0.5	0.8 ± 0.2	0.1 ± 0.1
P_{Dust}/P_{Inorg}	0.2 ± 0.2	0.2 ± 0.1	0.7 ± 0.3
P_{Anth}/P_{Inorg}^{d}	0.8 ± 0.2	0.8 ± 0.1	0.3 ± 0.3
$^{d}P_{Inorg}/nss\text{-}Ca^{2+}$	0.54 ± 0.54	0.09 ± 0.05	0.03 ± 0.02
$^{d}nss\text{-}K^{+}/nss\text{-}Ca^{2+}$	5.0 ± 3.9	0.4 ± 0.1	0.07 ± 0.08
$^{d}nss\text{-}SO_4^{2-}/nss\text{-}Ca^{2+}$	35.1 ± 19.0	4.3 ± 1.7	2.1 ± 1.1
$^{d}nss\text{-}Ca^{2+}/Ca^{2+}$	0.66 ± 0.26	0.95 ± 0.04	0.89 ± 0.12
$^{d}Ca^{2+}/Ca$	0.76 ± 0.16	0.92 ± 0.05	0.95 ± 0.14
$^{e}NH_4^{+}/SO_4^{2-}$	0.55 ± 0.34	0.40 ± 0.20	–
$^{e}(NH_4^{+} + Ca^{2+})/SO_4^{2-}$	0.60 ± 0.33	0.67 ± 0.20	0.59 ± 0.23
$^{e}(Na^{+} + NH_4^{+} + Ca^{2+})/SO_4^{2-}$	1.07 ± 0.17	0.97 ± 0.06	1.8 ± 1.2

P_{Inorg}, P_{Dust} and P_{Anth} are in nmol m^{-3} (this study).
[a]Kumar et al. (2008).
[b]P_{Dust} is estimated by multiplying the P/Al ratio in the Upper Continental Crust (~ 0.008; McLennan, 2001) and measured Al concentration in the ambient aerosols (i.e., $P_{dust} = 0.008 \times Al_{meas}$).
[c]Anthropogenic fraction of water-soluble inorganic phosphorous ($P_{Anth} = P_{Inorg} - P_{Dust}$).
[d]Molar ratio.
[e]Equivalent ratio.

3.2. P_{Inorg} concentration over BoB

The mass concentration of water-soluble inorganic phosphorous ($P_{Inorg} = PO_4^{3-}$) in PM_{10} averages about 1.1 ± 0.3 nmol m^{-3} during March–April 2006 (Table 1). During sampling in January 2009, mean P_{Inorg} concentration centres on 0.3 ± 0.2 nmol m^{-3} and 1.3 ± 0.5 nmol m^{-3} in $PM_{2.5}$ and PM_{10}, respectively. A comparison of the seasonal data (Average with $\pm 1\sigma$) suggests that P_{Inorg} concentrations in PM_{10} are not significantly different (P-value > 0.05) during March–April 2006 and January 2009 (Table 1). Despite this similarity in the abundance pattern, evidence for different sources of P_{Inorg} is discernible from the analytical data. The abundance of water-soluble calcium (Ca^{2+}) and P_{Inorg} in two size fractions ($PM_{2.5}$, PM_{10}), collected during January 2009, suggests their dominant occurrence in the coarse mode. The mass ratio of $PM_{2.5}$ to PM_{10} for Ca^{2+} and P_{Inorg} is computed for each sample and then averaged for all samples. The corresponding ratio centres on 0.31 ± 0.18 and 0.25 ± 0.14 for Ca^{2+} and P_{Inorg}, respectively, indicating their dominant contribution from mineral dust.

The contribution of P_{Inorg} from mineral dust has been investigated based on the chemical data presented in Tables 1 and 2. The mass concentration of SO_4^{2-} (with nss-SO_4^{2-}/$SO_4^{2-} \geq 90\%$) accounts for ~ 67 and 60% of ΣWSIC in March–April 2006 and January 2009, respectively (where WSIC refers to water-soluble inorganic constituents and taken as sum of concentrations of all cations and anions).

During these two sampling campaigns in the BoB, the equivalent ratio of NH_4^{+}/SO_4^{2-} and $(NH_4^{+} + Ca^{2+})/SO_4^{2-}$ is far less than one (Table 2), suggesting chemical uptake of excess acid by mineral dust. This is also evident from the significant (P-value < 0.05) linear correlation between nss-Ca^{2+} and nss-SO_4^{2-} (Fig. 2a; regression statistics are presented in Table 3) and among aerosol Ca and nss-Ca^{2+} (Fig. 2b; see Table 3). The dominant contribution of nss-SO_4^{2-} in the MABL of BoB is clearly evident from the steep slope of its linear regression lines with nss-Ca^{2+} for data from January 2009 ($m = 23$; $R^2 = 0.75$; $P < 0.05$) and March–April 2006 ($m = 2.3$; $R^2 = 0.62$; $P < 0.05$; Fig. 2a; see Table 3). This is in sharp contrast to the lower slope of linear regression for data from the ARS ($m = 0.83$; $R^2 = 0.67$; Fig. 2a; see Table 3). It is noteworthy that the solubility of aerosol Ca (defined as nss-Ca^{2+}/Ca) in March–April 2006 is relatively high compared with that in January 2009 (Tables 2 and 3). Another notable observation relates to significantly high (P value < 0.05; t statistic $= 5.89$) Ca^{2+} concentration (range: 4–36 nmol m^{-3}; Av: 17 ± 9 nmol m^{-3}) during March–April 2006 compared to January 2009 (range: 0.4–13 nmol m^{-3}; Av: 6 ± 3 nmol m^{-3}). Furthermore, on average, mineral dust concentration (assessed based on measured Al concentration and assuming Al content of 8.0% in the mineral dust) accounts for ~ 33 and 50% of the aerosol mass in January 2009 and March–April 2006, respectively. The relatively high solubility (Av: $92 \pm 5\%$) of aerosol Ca together with high abundance of mineral dust (Av: 49% of particulate
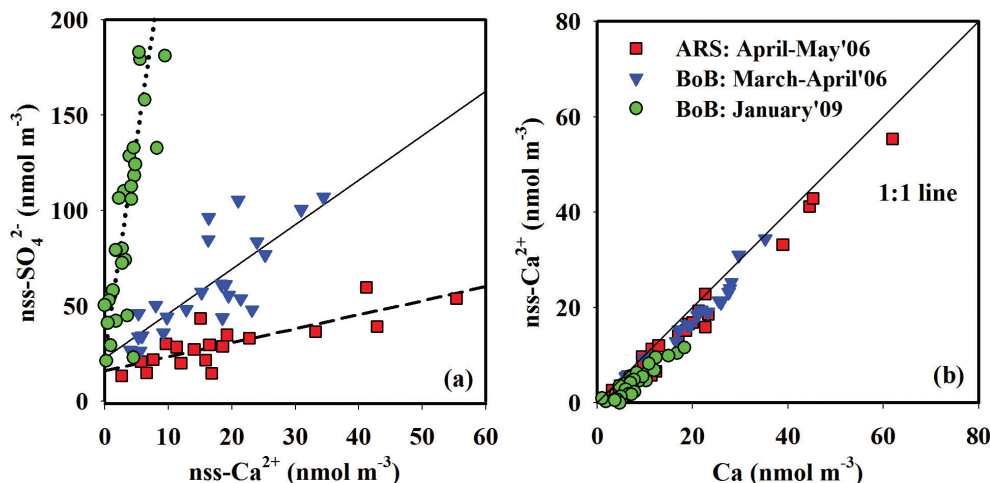
Fig. 2. The scatter plots between (a) nss-Ca^{2+} and nss-SO$_4^{2-}$ and (b) aerosol Ca and nss-Ca^{2+} suggest a reactive uptake of acidic species (H$_2$SO$_4$) by the mineral dust during long-range atmospheric transport. The dominant contribution of nss-SO$_4^{2-}$ in the MABL of Bay of Bengal (BoB) is clearly evident from the steep slope of the linear regression line compared to that in the Arabian Sea (ARS). Data for BoB (March–April 2006) and ARS (April–May 2006) are adopted from Kumar et al. (2008).

mass) during March–April 2006 is attributed to differences in the source regions of mineral dust and chemical processing during the long-range transport.

On the basis of the above discussion, we argue in favour of the seasonal trend and significant role of chemical processing of mineral dust during March–April 2006 compared to samples collected during January 2009. These differences in the chemical composition of aerosols are attributed to the varying type of mineral dust over the BoB during the two seasons. The AMBTs, during March–April 2006, were from desert regions (Thar Desert) of western India, whereas air masses during January 2009 over the BoB show dominant influence from the Indo-Gangetic Plain. The elemental ratio of Ca/Al in aerosols associated with these air masses averages around 0.81 ± 0.16 for March–April 2006 (Kumar et al., 2008) and 0.35 ± 0.14 for January 2009 (Kumar et al., 2010). The relatively high ratio of Ca/Al is consistent with that reported over the ARS, attributable

to the source of dust from the desert region in western India (Yadav and Rajamani, 2004; Rastogi and Sarin, 2009). In contrast, the lower ratio of Ca/Al over BoB (during January 2009) is largely dictated by the outflow from the Indo-Gangetic Plain (Ca/Al $= 0.23 \pm 0.11$, as measured at downwind site in the Gangetic Plain) (unpublished data; manuscript under preparation). A significant correlation between nss-Ca^{2+} and P$_{Inorg}$ ($R^2 = 0.64$; $n = 23$; P-value < 0.001; Fig. 3a) for the data from March to April 2006 suggests their common source from the mineral dust. The contribution of dust-derived phosphorous has been traced by the linear relation ($P < 0.05$) between nss-Ca^{2+} and P$_{Inorg}$ (Herut et al., 1999a; Markaki et al., 2003). These studies have shown that the air masses originating from the Arabian Peninsula exhibit a linear relation among nss-Ca^{2+} and total inorganic phosphorous (Markaki et al., 2003). Higher abundance of P and Ca in the desert dust (from Thar Desert) has been documented by Yadav and

Table 3. Statistical significance of linear regressions among nss-SO$_4^{2-}$, nss-K$^+$, nss-Ca^{2+} and Ca

| Scatter Plot (Y vs. X)	January 2009	March–April 2006	April–May 2006
		Regression ($Y = mX + b$)	
nss-SO$_4^{2-}$ vs. nss-Ca^{2+}	$m = 22.5$; $b = 24.1$	$m = 2.3$; $b = 22.4$	$m = 0.70$; $b = 15.2$
	$R^2 = 0.75$; $P < 0.05$	$R^2 = 0.64$; $P < 0.05$	$R^2 = 0.60$; $P < 0.05$
nss-K$^+$ vs. nss-Ca^{2+}	$m = 2.24$; $b = 5.90$	$m = 0.26$; $b = 1.28$	$m = 0.014$; $b = 1.07$
	$R^2 = 0.83$; $P < 0.05$	$R^2 = 0.76$; $P < 0.05$	$R^2 = 0.11$; $P > 0.05$
P$_{Inorg}$ vs. nss-Ca^{2+}	$m = 0.07$; $b = 1.00$	$m = 0.03$; $b = 0.59$	$m = 0.0044$; $b = 0.257$
	$R^2 = 0.20$; $P > 0.05$	$R^2 = 0.64$; $P < 0.05$	$R^2 = 0.38$; $P > 0.05$
nss-Ca^{2+} vs. Ca	$m = 0.75$; $b =$	$m = 0.92$; $b = -0.92$	$m = 1.04$; $b = 0.21$
	$R^2 = 0.86$; $P < 0.05$	$R^2 = 0.97$; $P < 0.05$	$R^2 = 0.96$; $P < 0.05$

$m =$ Slope; $b =$ intercept.
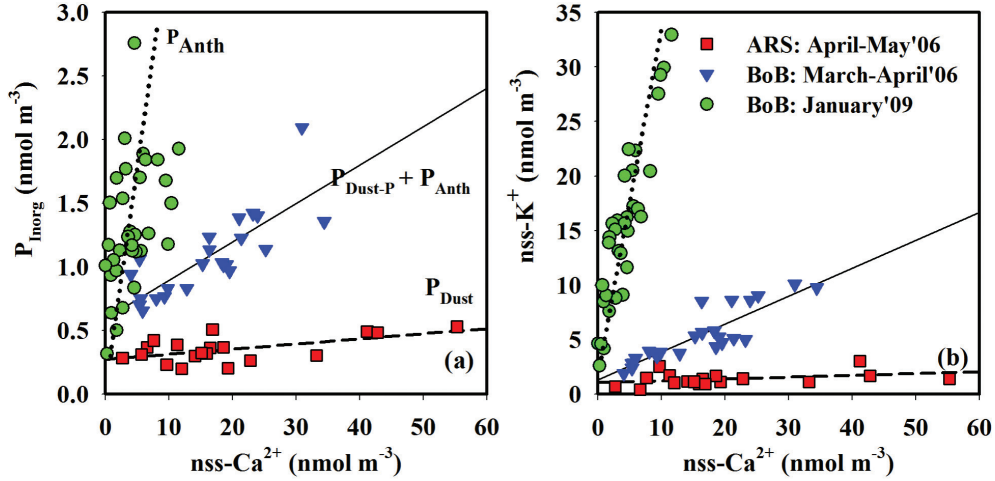
Fig. 3. Scatter plots: (a) data plotted with symbol (▼) provide evidence for the acid processing of mineral dust and, hence, enhanced supply of water-soluble P_{Inorg} during long-range transport to the Bay of Bengal (BoB). Evidence for supply of P_{Inorg} from anthropogenic sources is shown with the dotted line plotted through symbol (●), with steep slope and high P_{Inorg}/nss-Ca^{2+} ratio. For comparison, data from the Arabian Sea (ARS) (■), with high abundance of dust, represent exceptionally lower P_{Inorg}/nss-Ca^{2+} ratios. The scatter plot shown in (b) also exhibits steep slope, with higher abundance of anthropogenic nss-K^+ (relative to nss-Ca^{2+}) during January 2009 compared to March–April 2006 (BoB) and April–May 2006 (ARS).

Rajamani (2004). These desert soils are reported to be rich in Ca which is reflected from the characteristic high elemental ratio (0.73 ± 0.10) of Ca/Al (Yadav and Rajamani, 2004; Rastogi and Sarin, 2006; Kumar and Sarin, 2009). The reactive uptake of acidic species can lead to enhancement in the solubility of Ca-rich minerals such as carbonates, apatite ($Ca_3(PO_4)_2$), and is represented by following equations:

$$Ca_3(PO_4)_2 + H_2SO_4 \rightarrow CaSO_4 + H_3PO_4$$

$$Ca_3(PO_4) + HNO_3 \rightarrow Ca(NO_3)_2 + H_3PO_4$$
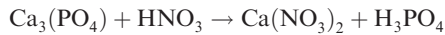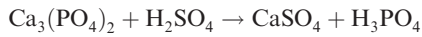
Therefore, the observed linear relation between nss-Ca^{2+} and P_{Inorg} during March–April 2006 (Fig. 3a) over the BoB is attributed to their common source (as evident from the AMBTs originating from the Thar Desert; Fig. 1). In contrast, relatively high concentrations of P_{Inorg} associated with extremely low concentrations of Ca^{2+} during January 2009 (Fig. 3a) suggest the dominance of anthropogenic sources (emissions from biomass burning and use of fertilisers).

3.3. *P_{Inorg} over ARS*

The mean ($\pm 1\sigma$) concentration of P_{Inorg} in aerosol samples collected from the ARS during April–May 2006 (Cruise SK-223B) corresponds to 0.3 ± 0.1 nmol m^{-3}.

A comparison of P_{Inorg} abundances shows significant difference (one-way ANOVA; $F_{(2,72)} = 39.9$; P value < 0.05) between the cruises (March–May 2006 and January 2009). These analyses suggest that concentration of P_{Inorg} over the BoB is significantly high compared to that over the ARS

(April–May 2006). However, concentrations were not significantly different between January 2009 and March–April 2006 (Table 1). Relatively high concentration of P_{Inorg} observed over the BoB is due to the predominance of anthropogenic sources and acid processing of mineral dust (Fig. 3a). A significant difference in the P_{Inorg}/nss-Ca^{2+} ratio in aerosols over the ARS and BoB is noteworthy (Table 2). The P_{Inorg}/nss-Ca^{2+} ratio varied from 0.01 to 0.11 (Av: 0.03 ± 0.02) over the ARS during April–May 2006. In contrast, P_{Inorg}/nss-Ca^{2+} ratio averages around 0.09 ± 0.05 (range: 0.04–0.23) over the BoB during March–April 2006, and the ratio is one order of magnitude higher (Av: 0.54 ± 0.54, range: 0.12–2.4) during January 2009 (Table 2). Relatively high P_{Inorg}/nss-Ca^{2+} ratio over the BoB during January 2009 is attributed to enhanced supply of P_{Inorg} from fertilisers and biomass burning emissions with relatively low Ca^{2+} content. The reactive uptake of acidic species (nss-SO_4^{2-}) by mineral dust is also evident from the mass ratio of water-soluble Ca^{2+} to total aerosol Ca (Fig. 2b) varying from 0.63 to 1.05 (Av: 0.95 ± 0.14), with average nss-Ca^{2+}/Ca^{2+} ratio centring on 0.89 ± 0.12 (Table 2). However, abundance of nss-SO_4^{2-} in the MABL of ARS is significantly lower than that over the BoB (Fig. 2a; one-way anova, $F_{(2, 70)} = 16.8$; P value < 0.05).

3.4. *Anthropogenic source of P_{Inorg} vis-à-vis mineral dust*

A characteristic high abundances of anthropogenic constituents (nss-K^+ and nss-SO_4^{2-}) within the MABL of BoB,

during January 2009, are evident based on relatively high ratios of $nss\text{-}SO_4^{2-}/nss\text{-}Ca^{2+}$ (Fig. 2a) and $nss\text{-}K^+/nss\text{-}Ca^{2+}$ (Fig. 3b). These characteristic ratios in January 2009 over the BoB are relatively high compared to those observed during March–April 2006 and over the ARS (April–May 2006). The difference in the slopes of the regression lines (Figs. 2a and 3b) reflects the seasonal variability in the chemical composition of aerosols associated with the outflow from the Indo-Gangetic Plain (from December to February) and from the desert regions during March–May. Also, a statistical test has been performed in order to ascertain differences in the ratios of $nss\text{-}K^+/nss\text{-}Ca^{2+}$, $nss\text{-}SO_4^{2-}/nss\text{-}Ca^{2+}$ and $P_{Inorg}/nss\text{-}Ca^{2+}$ for the data from January 2009, March–April 2006 (BoB); April–May 2006 (ARS). ANOVA analyses (with post hoc test) suggest that P_{Inorg} concentration and ionic ratios ($nss\text{-}K^+/nss\text{-}Ca^{2+}$, $nss\text{-}SO_4^{2-}/nss\text{-}Ca^{2+}$ and $P_{Inorg}/nss\text{-}Ca^{2+}$) are significantly different ($P < 0.05$) for seasonal data from all cruises (January 2009, March–April 2006 and April–May 2006). A significant difference is evident between $P_{Inorg}/nss\text{-}Ca^{2+}$ for winter (January 2009) and spring intermonsoon (March–April 2006, April–May 2006) cruises [$F(2, 71) = 15.782$, $P\text{-value} < 0.05$]. Likewise, significant difference ($P\text{-value} < 0.05$) is observed for $nss\text{-}K^+/nss\text{-}Ca^{2+}$ [$F(2,71) = 11.56$; $P < 0.05$] and $nss\text{-}SO_4^{2-}/nss\text{-}Ca^{2+}$ [$F(2, 71) = 50.6$; $P < 0.05$] (Table 2, data from January 2009, March–April 2006 and April–May 2006).

The relatively high concentrations of anthropogenic constituents ($nss\text{-}SO_4^{2-}$ & $nss\text{-}K^+$) together with the low abundance of $nss\text{-}Ca^{2+}$ within the MABL of BoB, during the wintertime (January–March), is a characteristic feature of the outflow from the Indo-Gangetic Plain (Kumar et al., 2010; Sarin et al., 2011; Srinivas et al., 2011). The high ratios of $nss\text{-}SO_4^{2-}/nss\text{-}Ca^{2+}$, $nss\text{-}K^+/nss\text{-}Ca^{2+}$ and $P_{Inorg}/nss\text{-}Ca^{2+}$ during January 2009 (Table 2) are attributed to the dominance of pollution sources. In contrast, the relative decrease in the source strength of the continental outflow during the spring intermonsoon (March–April), together with higher abundance of water-soluble Ca^{2+}, is a dominant cause for the low ratios of $nss\text{-}SO_4^{2-}/nss\text{-}Ca^{2+}$, $nss\text{-}K^+/nss\text{-}Ca^{2+}$ and $P_{Inorg}/nss\text{-}Ca^{2+}$ (Figs. 2a, 3a, 3b). The high concentration of Ca^{2+} mainly results from seasonal shift in the source regions of mineral dust during March–April 2006, as evident from the AMBTs which originates from the Thar Desert. The enhanced solubility of aerosol Ca and the linear relation of P_{Inorg} with $nss\text{-}Ca^{2+}$ during March–April 2006 suggest that significant fraction of water-soluble inorganic P is derived from the chemical processing of mineral dust.

The mass concentration of P_{Inorg} in the MABL of BoB is significantly higher than that derived from the mineral dust as its only source. A significant linear relationship of P_{Inorg} and $nss\text{-}K^+$ ($P\text{-value} < 0.05$) suggests fertilisers and/or emissions from biomass burning as their common anthropogenic source. A recent study by Singh and Singh (2008) has shown significant increase in the use of fertilisers (rich in N, P and K) in the Indo-Gangetic Plain during the past few decades. Therefore, atmospheric transport of dust from disturbed soils and agricultural fields could serve as a dominant source of P_{Inorg}. Furthermore, emissions from biomass burning (burning of agricultural crop residue) has been suggested as a significant source of aerosol P, similar to aerosol K^+ (Mahowald et al., 2005, 2008 and references therein; Izquierdo et al., 2012). Thus, a linear correlation between $nss\text{-}K^+$ and P_{Inorg} is attributed to both the emissions from biomass burning and fertilisers. The P_{Inorg}/EC ratio in the fine mode aerosols over the BoB, during January 2009, averages about 0.003 ± 0.002 (also evident from the slope of regression line between EC and P_{Inorg}; $m = 0.001$; $R^2 = 0.54$; figure not shown in this article). A similar ratio of P_{Inorg} to BC (0.0029 ± 0.0001) has been reported for the emissions from biomass burning over the Amazonian region (Mahowald et al., 2008). We, therefore, invoke significant contribution of K^+ and P_{Inorg} from fertilisers associated with the large-scale agricultural activity and biomass burning (burning crop residue) in the Indo-Gangetic Plain.

The phosphorous contribution from dust (P_{Dust}) is subtracted from the total P_{Inorg} in order to obtain the anthropogenic component (P_{Anth}). In Fig. 3a, three distinct relationships among P_{Inorg} and $nss\text{-}Ca^{2+}$ are identified. The line with steep slope (high P_{Inorg} and low $nss\text{-}Ca^{2+}$) is dominated by the anthropogenic phosphorous (P_{Anth}). The line with flat slope (Fig. 3a; low P_{Inorg} and high $nss\text{-}Ca^{2+}$) for the ARS is dominated mainly by P_{Inorg} derived from the mineral dust. The third relationship with an intermediate slope for the data from the BoB (during March–April 2006) is dictated by varying contribution from dust and anthropogenic sources.

The mass concentrations of P_{Inorg}, P_{Dust} and P_{Anth} and the corresponding ratios of P_{Dust}/P_{Inorg}, P_{Anth}/P_{Inorg} and $P_{Inorg}/nss\text{-}Ca^{2+}$ within the MABL of BoB and ARS are summarised in Table 2. Over the BoB, P_{Anth} contributes significantly, varying from 61 to 94% (Av: $77 \pm 10\%$) during March–April 2006 and from 24 to 96% (Av. $76 \pm 18\%$) during January 2009. It is implicit in this approach that the mobilisation of P_{Inorg} by acid processing of mineral dust is also a part of P_{Anth}. In contrast, P_{Inorg} over the ARS is mainly dominated by desert dust ($P_{Dust} = \sim 70\%$). The spatio-temporal variability of percentage contribution of P_{anth}/P_{Inorg} to the study region is depicted in Fig. 4. From this figure, it is inferred that the contribution of P_{Inorg} from anthropogenic sources (viz., emissions from biomass burning and fertilisers, etc.) dominates over the BoB during the continental outflow.
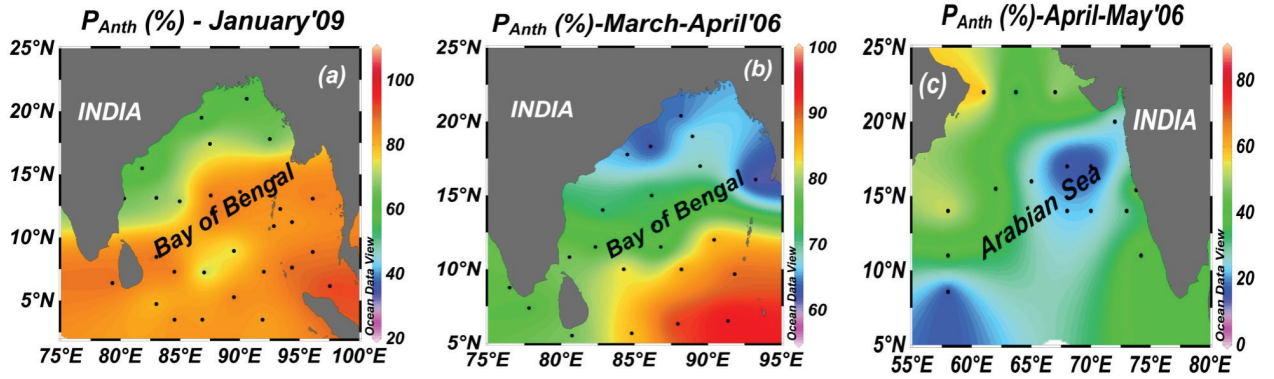
Fig. 4. Spatio-temporal variability in the concentration of anthropogenic water-soluble inorganic phosphorous (P_{anth}) to P_{Inorg} over the Bay of Bengal and the Arabian Sea.

3.5. Atmospheric dry-deposition of P_{Inorg}

The dry-deposition flux is estimated by using the following equation.

$$\text{Dry} - \text{deposition flux, } F_{dry} = V_{dry} \times C_{meas}$$

where V_{dry} and C_{meas} refer to dry-deposition velocity and measured mass concentration, respectively. For the flux computation, we have used a deposition velocity of 2.0 cm s^{-1}. A similar dry-deposition velocity has been used by several studies reported in the literature to estimate deposition fluxes (Duce et al., 1991; Herut et al., 1999b; Markaki et al., 2003; Baker et al., 2006; Chen et al., 2006, 2007). On the basis of experimental data and modelled results, deposition velocity of 0.1 and 2.0 cm s^{-1} has been suggested for fine and coarse mode aerosols by Spokes et al. (2000). Due to the dominant occurrence of P_{Inorg} in PM_{10} (as evident from the mass ratio of $PM_{2.5}/PM_{10}$, Av: 0.25), our experimental data suggest an association of P_{Inorg} in the coarse mode. Therefore, V_d of 2.0 cm s^{-1} is used for estimating the deposition flux in this study.

The dry-deposition flux of P_{Inorg} to the BoB ranges from 1.1 to 3.6 (Av:1.9 ± 0.6) µmol P m^{-2} d^{-1} during March–April 2006 and from 0.5 to 4.8 (Av: 2.3 ± 0.9) µmol P m^{-2} d^{-1} during January 2009 (assuming dry-deposition velocity of P_{Inorg} as 2.0 cm sec^{-1}), whereas to the ARS it varies from 0.3 to 0.9 (Av: 0.6 ± 0.2) µmol P m^{-2} d^{-1} during April–May 2006. These fluxes have been compared with the model-based estimates (Mahowald et al., 2008; Okin et al., 2011). The atmospheric deposition of P_{Inorg} to the BoB (this study) is ~ 0.022 Tg P yr^{-1} (0.024 for January 2009 and 0.019 Tg P yr^{-1} for March–April 2006) and to the ARS is ~ 0.014 Tg P yr^{-1} for April–May 2006 (areas of BoB and ARS are taken as 2.2×10^{12} and 4.93×10^{12} m^2, respectively, and dry-deposition is integrated only for 150 d when continental outflow is dominant). Thus, the total dry-deposition flux of P_{Inorg} (~ 0.036 Tg P yr^{-1}) over these two oceanic regions can

be considered as representative of the annual deposition of P_{Inorg} to the northern Indian Ocean (NIO). A comparison of dry-deposition of P_{Inorg} to the northern Indian Ocean (this study) with the model-based estimates (reported in the literature) is presented in Table 4. We have also compared the P_{Inorg} flux to NIO with that from other oceanic regions. These estimates are of comparable magnitude with the model-based results for the NIO (0.04 Tg P yr^{-1} by Okin et al. (2011) and 0.014 Tg P yr^{-1} by Mahowald et al. (2008)).

3.6. Comparison with riverine supply

The global riverine input of P_{Inorg}, via major rivers, is 0.8–1.4 Tg P yr^{-1} (Compton et al., 2000) and the freshwater influx from the global rivers is about 37.4×10^{12} m^3 yr^{-1} (Martin and Whitfield, 1983; Kumar et al., 1996). It has been reported that Bay receives approximately 1.625×10^{12} m^3 yr^{-1} of freshwater (Sarin et al., 1990; Prasanna Kumar et al., 2004). Thus, the riverine input of P_{Inorg} to the BoB is estimated to be 35–61 Gg P yr^{-1}. Since P_{Inorg} abundance is

Table 4. P_{Inorg} fluxes (Tg yr^{-1}) from the northern Indian Ocean, Atlantic and Pacific

References	N Indian Ocean	N Atlantic	N Pacific
This study	0.036	–	–
Duce et al. (1991)[a]	0.070	0.154	0.336
Prospero, (1996)[a]	0.014	0.154	0.067
Ginoux et al. (2001)[a]	0.097	0.123	0.064
Zender et al. (2003)	0.040	0.190	0.030
Luo et al. (2003)[a]	0.079	0.161	0.025
Tegen et al. (2004)[a]	0.043	0.181	0.039
Jickells et al. (2005)[a]	0.083	0.141	0.050
Mahowald et al. (2008)	0.014	0.028	0.022
Okin et al. (2011)	0.043	0.020	0.026

[a]P_{Inorg} is estimated from the abundance of mineral dust and P concentration in UCC.

relatively high during January 2009 compared to that in March–April 2006 (Spring intermonsoon), it is relevant to compare the atmospheric input with the riverine supply in order to estimate the maximum contribution via air-sea deposition to the BoB. The concentration of P_{Inorg} in MABL of the BoB, during January 2009, ranged from 0.32 to 2.76 nmol m^{-3}. Therefore, dry deposition of P_{Inorg} to the BoB ranges from 0.5 to 4.8 µmol P m^{-2} d^{-1} (5–50 Gg P yr^{-1}). A comparison of these estimates suggests that the atmospheric deposition of P_{Inorg} is of comparable magnitude to its supply via rivers, and that the atmosphere is a significant source of P_{Inorg} to surface waters of the BoB.

4. Conclusion and implications

The spatio-temporal variability in the concentration of water-soluble inorganic phosphorous (P_{Inorg}) has been studied over the BoB and the ARS during the continental outflow (January–April). Our study provides the first field evidence for the acid processing of mineral dust during atmospheric transport and, hence, enhanced solubility of P_{Inorg}. We suggest that high concentrations of sulphate persisting in the continental outflow from the Indo-Gangetic Plain provide most conducive environment for acid mobilisation of phosphorous (and other nutrients) from dust-laden air masses. Nevertheless, contribution from anthropogenic sources (fertilisers and emissions from biomass burning) is overwhelming and account for nearly 75% of P_{Inorg}. In contrast, P_{Inorg} concentrations are 4 to 5 times lower over the ARS and anthropogenic fraction contributes no more than ~30% of P_{Inorg}. The atmospheric dry-deposition of phosphorous (0.4–3.8 Gmol P yr^{-1}) is comparable to its supply via rivers (1.1–2.0 Gmol P yr^{-1}) draining into the BoB. Furthermore, a comparison of atmospheric dry-deposition of P_{Inorg} (0.5–4.8 µmol P m^{-2} d^{-1} during January 2009) with water column productivity (99–566 mg C m^{-2} d^{-1}; Gauns et al., 2005) suggests that Aeolian input can support up to ~10% of in situ primary production. In the present-day scenario of growing anthropogenic activities, these results have implications to significant impact on the biogeochemistry of coastal waters downwind of pollution sources. Our estimate for the dry-deposition of P_{Inorg} to the NIO is of comparable magnitude with the model-based results.

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References

Alappattu, D., Subrahamanyam, D., Kunhikrishnan, P., Ramachandran, R., Somayaji, K. and co-authors. 2008. Spatio-temporal variability of surface-layer turbulent fluxes over the Bay of Bengal and Arabian Sea during the ICARB field experiment. *Boundary-Layer Meteorology* **126**, 297–309.

Anderson, L. D., Faul, K. L. and Paytan, A. 2010. Phosphorus associations in aerosols: what can they tell us about P bioavailability? *Mar. Chem.* **120**, 44–56.

Badarinath, K. V. S., Kharol, S. K., Kaskaoutis, D. G., Sharma, A. R., Ramaswamy, V. and co-authors. 2010. Long-range transport of dust aerosols over the Arabian Sea and Indian region – a case study using satellite data and ground-based measurements. *Global Planet. Change* **72**, 164–181.

Baker, A. R., Jickells, T. D., Witt, M. and Linge, K. L. 2006. Trends in the solubility of iron, aluminium, manganese and phosphorus in aerosol collected over the Atlantic Ocean. *Mar. Chem.* **98**, 43–58.

Chen, Y., Mills, S., Street, J., Golan, D., Post, A. and co-authors. 2007. Estimates of atmospheric dry deposition and associated input of nutrients to Gulf of Aqaba seawater. *J. Geophys. Res.* **112**, D04309.

Chen, Y., Street, J. and Paytan, A. 2006. Comparison between pure-water- and seawater-soluble nutrient concentrations of aerosols from the Gulf of Aqaba. *Mar. Chem.* **101**, 141–152.

Chester, R. 2002. *Marine Geochemistry*. 2nd ed. Blackwell Publishers, New York, 506 pp.

Compton, J., Mallinson, D., Glenn, C. R., Filippelli, G., Follmi, K. and co-authors. 2000. Variations in the global phosphorous cycle. *Marine Authigenesis: From Global to Microbial*, SEPM Special Publication No. 66, ISBN 1-56576-064-6, pp. 21–33.

Dey, S., Tripathi, S. N., Singh, R. P. and Holben, B. N. 2004. Influence of dust storms on the aerosol optical properties over the Indo-Gangetic basin. *J. Geophys. Res.* **109**, D20211–20224, doi:10.1029/2004JD004924.

Draxler, R. R. 2002. *HYSPLIT-4 user's guide*. NOAA Tech Memo, ERL ARL-230, 35.

Duce, R. A., Liss, P. S., Merrill, J. T., Atlas, E. L., Buat-Menard, P. and co-authors. 1991. The atmospheric input of trace species to the world ocean. *Global Biogeochem. Cycles* **5**, 193–259.

Fang, T. H. 2004. Phosphorus speciation and budget of the East China Sea. *Cont. Shelf Res.* **24**, 1285–1299.

Gauns, M., Madhupratap, M., Ramaiah, N., Jyothibabu, R., Fernandes, V. and co-authors. 2005. Comparative accounts of biological productivity characteristics and estimates of carbon fluxes in the Arabian Sea and the Bay of Bengal. *Deep Sea Res. Part II* **52**, 2003–2017.

Ginoux, P., Chin, M., Tegen, I., Prospero, J., Holben, B. and co-authors. 2001. Global simulation of dust in the troposphere: model description and assessment. *J. Geophys. Res.* **106**(D17), 20255–20273.

Guieu, C., Loÿe-Pilot, M. D., Ridame, C. and Thomas, C. 2002. Chemical characterization of the Saharan dust end-member: some biogeochemical implications for the western Mediterranean Sea. *J. Geophys. Res.* **107**, 4258.

Herman, J. R., Bhartia, P. K., Torres, O., Hsu, C., Seftor, C. and co-authors. 1997. Global distribution of UV-absorbing aerosols from Nimbus-7/TOMS data. *J. Geophys. Res.* **102**, 911–922.

Herut, B., Krom, M. D., Pan, G. and Mortimer, R. 1999a. Atmospheric input of nitrogen and phosphorus to the Southeast Mediterranean: sources, fluxes, and possible impact. *Limnol. Oceanogr.* **44**(7), 1683–1692.

Herut, B., Zohary, T., Robarts, R. D. and Kress, N. 1999b. Adsorption of dissolved phosphate onto loess particles in surface and deep Eastern Mediterranean water. *Mar. Chem.* **64**, 253–265.

Izquierdo, R., Benítez-Nelson, C. R., Masqué, P., Castillo, S., Alastuey, A. and co-authors. 2012. Atmospheric phosphorus deposition in a near-coastal rural site in the NE Iberian Peninsula and its role in marine productivity. *Atmos. Environ.* **49**, 361–370.

Jahnke, R. A. 2000. The phosphorus cycle. In: *Earth system science: From biogeochemical cycles to global change* (eds. M. C. Jacobson, R. J. Charlson, H. Rodhe, and G. H. Orians). Academic Press, London, pp. 360–376.

Jickells, T. D., An, Z. S., Andersen, K. K., Baker, A. R., Bergametti, G. and co-authors. 2005. Global iron connections between desert dust, ocean biogeochemistry, and climate. *Science* **308**, 67–71.

Keene, W. C., Pszenny, A. P., Gallloway, J. N. and Hawley, M. E. 1986. Sea salt correction and interpretation of constituent ratios in marine precipitation. *J. Geophys. Res.* **91**, 6647–6658.

Krom, M. D., Herut, B. and Mantoura, R. F. C. 2004. Nutrient budget for the Eastern Mediterranean: implications for phosphorus limitation. *Limnol. Oceanogr.* **49**, 1582.

Kumar, M. D., Naqvi, S. W. A., George, M. D. and Jayakumar, D. A. 1996. A sink for atmospheric carbon dioxide in the northeast Indian Ocean. *J. Geophys. Res.* **101**, 18121–18125.

Kumar, A. and Sarin, M. M. 2009. Mineral aerosols from western India: temporal variability of coarse and fine atmospheric dust and elemental characteristics. *Atmos. Environ.* **43**, 4005–4013.

Kumar, A., Sarin, M. M. and Srinivas, B. 2010. Aerosol iron solubility over Bay of Bengal: role of anthropogenic sources and chemical processing. *Mar. Chem.* **121**, 167–175.

Kumar, A., Sudheer, A. K. and Sarin, M. M. 2008. Chemical characteristics of aerosols in MABL of Bay of Bengal and Arabian Sea during spring inter-monsoon: a comparative study. *J. Earth Syst. Sci.* **117**(S1), 325–332.

Lelieveld, J., Crutzen, P. J., Ramanathan, V., Andreae, M. O., Brenninkmeijer, C. A. M. and co-authors. 2001. The Indian Ocean experiment: widespread air pollution from South and Southeast Asia. *Science* **291**, 1031–1036.

Li, Q. P. and Hansell, D. A. 2008. Intercomparison and coupling of magnesium-induced co-precipitation and long-path liquid-waveguide capillary cell techniques for trace analysis of phosphate in seawater. *Anal. Chim. Acta* **611**, 68–72.

Luo, C., Mahowald, N. and del Corral, J. 2003. Sensitivity study of meteorological parameters on mineral aerosol mobilization, transport and distribution. *J. Geophys. Res.* **108**(D15), 4447, doi:10.1029/2003JD003483.

Mahowald, N. M., Artaxo, P., Baker, A. R., Jickells, T. D., Okin, G. S. and co-authors. 2005. Impacts of biomass burning emissions and land use change on Amazonian atmospheric phosphorus cycling and deposition. *Global Biogeochem. Cycles* **19**, GB4030.

Mahowald, N., Jickells, T. D., Baker, A. R., Artaxo, P., Benitez-Nelson, C. R. and co-authors. 2008. Global distribution of atmospheric phosphorus sources, concentrations and deposition rates, and anthropogenic impacts. *Global Biogeochem. Cycles* **22**, GB4026-4045.

Markaki, Z., Oikonomou, K., Kocak, M., Kouvarakis, G., Chaniotaki, A. and co-authors. 2003. Atmospheric deposition of inorganic phosphorus in the Levantine Basin, eastern Mediterranean: spatial and temporal variability and its role in seawater productivity. *Limnol. Oceanogr.* **48**, 1557–1568.

Martin, J. M. and Whitefield, M. 1983. The significance of the river input of chemical elements to the ocean. In: *Trace metals in sea water* (eds. C. S. Wong, E. Boyle, K. W. Bruland, J. D. Burton, and E. D. Goldberg). Plenum, New York, pp. 265–296.

McLennan, S. M. 2001. Relationships between the trace element composition of sedimentary rocks and upper continental crust. *Geochem. Geophys. Geosyst.* **2**, ISSN: 1525–2027, article no: 2000GC000109.

Middleton, N. J. 1986. A geography of dust storms in South-West Asia. *J. Climatol.* **6**, 183–196.

Middleton, N. J. 1989. Climatic controls on the frequency, magnitude and distribution of dust storms: examples from India/Pakistan, Mauritania and Mongolia. In: *Paleoclimatology and Paleometeorology: Modern and Past Patterns of Global Atmospheric Transport* (eds. M. Leinen and M. Sarntheined). Kluwer Academic, Norwell, MA, pp. 97–132.

Mills, M. M., Ridame, C., Davey, M., La Roche, J. and Geider, R. J. 2004. Iron and phosphorus co-limit nitrogen fixation in the eastern tropical North Atlantic. *Nature* **429**, 292–294.

Moutin, T., Karl, D. M., Duhamel, S., Rimmelin, P., Raimbault, P. and co-authors. 2008. Phosphate availability and the ultimate control of new nitrogen input by nitrogen fixation in the tropical Pacific Ocean. *Biogeosciences* **5**, 95–109.

Nenes, A., Krom, M. D., Mihalopoulos, N., Van Cappellen, P., Shi, Z. and co-authors. 2011. Atmospheric acidification of mineral aerosols: a source of bioavailable phosphorus for the oceans. *Atmos. Chem. Phys.* **11**, 6265–6272.

Okin, G. S., Baker, A. R., Tegen, I., Mahowald, N. M., Dentener, F. J. and co-authors. 2011. Impacts of atmospheric nutrient deposition on marine productivity: roles of nitrogen, phosphorus, and iron. *Global Biogeochem. Cycles* **25**, GB2022.

Paytan, A. and McLaughlin, K. 2007. The oceanic phosphorous cycle. *Chem. Rev.* **107**, 563–576.

Prasanna Kumar, S., Narvekar, J., Kumar, A., Shaji, C., Anand, P. and co-authors. 2004. Intrusion of the Bay of Bengal water into the Arabian Sea during winter monsoon and associated chemical and biological response. *Geophys. Res. Lett.* **31**, L15304.

Prospero, J. M. 1996. The atmospheric transport of particles to the ocean. In: *Particle Flux in the Ocean* (eds. V. Ittekkot, P.

Schäfer, S. Honjo and P. J. Depetris). John Wiley & Sons Ltd., New York, vol. 57 of SCOPE, pp. 19–52.

Prospero, J. M., Ginoux, P., Torres, O., Nicholson, S. E. and Gill, T. E. 2002. Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total ozone Mapping Spectrometer (TOMS) absorbing aerosol product. *Rev. Geophys.* **40**, 1002, doi:10.1029/2000RG000095.

Raj, P. E., Saha, S. K., Sonbawne, S. M., Deshpande, S. M., Devara, P. C. S. and co-authors. 2008. Lidar observation of aerosol stratification in the lower troposphere over Pune during pre-monsoon season of 2006. *J. Earth Syst. Sci.* **117**, 293–302.

Ramanathan, V., Crutzen, P. J., Lelieveld, J., Mitra, A. P., Althausen, D. and co-authors. 2001. Indian Ocean experiment: an integrated analysis of the climate forcing and effects of the great Indo-Asian haze. *J. Geophys. Res.* **106**, 28371–28398.

Rastogi, N. and Sarin, M. M. 2006. Chemistry of aerosols over a semi-arid region: evidence for acid neutralization by mineral dust. *Geophys. Res. Lett.* **33**, L23815.

Rastogi, N. and Sarin, M. M. 2009. Quantitative chemical composition and characteristics of aerosols over western India: one-year record of temporal variability. *Atmos. Environ.* **43**, 3481–3488.

Ridame, C. and Guieu, C. 2002. Saharan input of phosphate to the oligotrophic water of the open western Mediterranean Sea. *Limnol. Oceanogr.* **47**(3), 856–869.

Saliba, N. A. and Chamseddine, A. 2012. Uptake of acid pollutants by mineral dust and their effect on aerosol solubility. *Atmos. Environ.* **46**, 256–263.

Sanudo-Wilhelmy, S. A., Kustka, A. B., Gobler, C. J., Hutchins, D. A., Yang, M. and co-authors. 2001. Phosphorus limitation of nitrogen fixation by Trichodesmium in the central Atlantic Ocean. *Nature* **411**, 66–69.

Sarin, M. M., Krishnaswami, S., Somayajulu, B. L. K. and Moore, W. S. 1990. Chemistry of uranium, thorium, and radium isotopes in the Ganga–Brahmaputra river system: weathering processes and fluxes to the Bay of Bengal. *Geochim. Cosmochim. Acta,* **54**, 1387–1396.

Sarin, M., Kumar, A., Srinivas, B., Sudheer, A. and Rastogi, N. 2011. Anthropogenic sulphate aerosols and large Cl-deficit in marine atmospheric boundary layer of tropical Bay of Bengal. *J. Atmos. Chem.* **66**, 1–10.

Schlesinger, W. 1997. *Biogeochemistry: An Analysis of Global Change.* 2nd ed. Elsevier, New York, 588 pp.

Siefert, R. L., Johansen, A. M. and Hoffmann, M. R. 1999. Chemical characterization of ambient aerosol collected during the southwest monsoon and intermonsoon seasons over the Arabian Sea: Labile-Fe(II) and other trace metals. *J. Geophys. Res.* **104**, 3511–3526.

Singh, B. and Singh, Y. 2008. Reactive nitrogen in Indian agriculture: inputs, use efficiency and leakages. *Curr. Sci.* **94**, 1382.

Spokes, L. J., Yeatman, S. G., Cornell, S. E. and Jickells, T. D. 2000. Nitrogen deposition to the eastern Atlantic Ocean. The importance of south-easterly flow. *Tellus B* **52**, 37–49.

Srinivas, B., Sarin, M. and Kumar, A. 2011. Impact of anthropogenic sources on aerosol iron solubility over the Bay of Bengal and the Arabian Sea. *Biogeochem.* http://dx.doi.org/10.1007/s10533-011-9680-1. 1–12.

Subrahamanyam, D. B., Kumar, N. V. P. K., Dutt, C. B. S., Anurose, T. J., Kunhikrishnan, P. K. and co-authors. 2011. Characterization of air–sea interaction processes over the Bay of Bengal during the winter phase of ICARB field experiment. *Atmos. Res.* **99**, 97–111.

Taylor, S. R. and McLennan, S. M. 1985. *The Continental Crust: Its Composition and Evolution.* Blackwell, Oxford.

Tegen, I., Werner, M., Harrison, S. P. and Kohfeld, K. E. 2004. Relative importance of climate and land use in determining present and future global soil dust emission. *Geophys. Res. Lett.* **31**, L05105, doi:10.1029/2003GL019216.

Tindale, N. W. and Pease, P. P. 1999. Aerosols over the Arabian Sea: atmospheric transport pathways and concentrations of dust and sea salt. *Deep Sea Res. Part II* **46**, 1577–1595.

Tyrrell, T. 1999. The relative influences of nitrogen and phosphorus on oceanic primary production. *Nature* **400**, 525–531.

Wolf, G. T. 1984. On the nature of nitrate in coarse continental aerosols. *Atmos. Environ.* **18**, 977–981.

Yadav, S. and Rajamani, V. 2004. Geochemistry of aerosols of northwestern part of India adjoining the Thar desert. *Geochim. Cosmochim. Acta* **68**, 1975–1988.

Zender, C. S., Bian, H. and Newman, D. 2003. Mineral dust entrainment and deposition (DEAD) model: description and 1990s dust climatology. *J. Geophys. Res.* **108**, 4416–4435.

Zhang, J.-Z. and Chi, J. 2002. Automated analysis of nanomolar concentrations of phosphate in natural waters with liquid waveguide. *Environ. Sci. Technol.* **36**, 1048–1053.