

Atmospheric deposition of phosphorus to the Northern Indian Ocean

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Air–sea deposition of phosphorus is an important external source of this macronutrient for ocean primary productivity. Although global budgets have shown that atmospheric input of phosphorus is comparable to its riverine supply, studies on regional scale are rather limited. The present article summarizes sources and atmospheric pathway of phosphorus to the ocean surface as a case study of the Northern Indian Ocean. Water-soluble aerosol phosphorus (P_{Inorg}) concentrations are significantly high over the Bay of Bengal (BoB) compared to those over the Arabian Sea (ARS). Relative increase in the supply of aerosol-P over BoB is attributed to acid-processing of mineral dust during long-range atmospheric transport in addition to its supply from anthropogenic sources. Our estimate of P-deposition to the Northern Indian Ocean ($\sim 1.2 \text{ Gmol-P year}^{-1}$) is comparable to its atmospheric deposition in other oceanic regions such as the North Atlantic and is also consistent with the model-based projections for the Northern Indian Ocean. These results highlight the importance of atmospheric source in influencing the biogeochemical cycle of phosphorus in the Northern Indian Ocean.

Keywords: Air–sea deposition, aerosol-P, atmospheric deposition, Northern Indian Ocean.

Introduction

THE long-range atmospheric transport of phosphorus (P), a limiting nutrient for marine phytoplankton productivity¹, and its air–sea deposition act as an additional source of P to surface waters^{2–5}. Several studies have focused on the marine biogeochemical budget of P by considering the fluvial input as an external source of new nutrients to the euphotic zone, a well-ventilated zone for primary production¹. Nevertheless, contribution from anthropogenic sources through atmospheric pathway is grossly under-represented in the supply of P to surface ocean. Recent studies have shown that aeolian supply of P is of comparable magnitude to its input via rivers^{6,7}. In order to better constrain the marine budget of P, these studies have addressed the need to assess the atmospheric supply of P, particularly to the oceanic regions located downwind of the pollution sources^{7–11}.

The Northern Indian Ocean has been reported to be heavily influenced by polluted continental air masses from south and south-east Asia during winter (December–February) and spring inter monsoon (March–May) season^{12–16}. An evidence for the supply of P resulting from large-scale application of fertilizers, biomass burning emissions in the Indo-Gangetic Plain (IGP) and chemical processing of mineral dust originating from deserts is an integral part of these recent studies¹⁷. This observation is further supported by air–mass back trajectories (AMBTs) and chemical composition of airborne particulate matter sampled in IGP outflow to the Bay of Bengal^{18,19}.

Soils in the IGP and Brahmaputra valley together cover an area of ~ 71 million hectares (as per the FAO record) and are characterized by fine alluvium. Furthermore, fluvial bed load sediments are depleted of nitrogen, phosphorus and organic matter. Therefore, extensive use of fertilizers has been applied for agricultural purposes in the IGP to meet growing food requirement and sustainability. Until recently, the geochemical budget of marine phosphorus has not considered the contribution from atmosphere^{6,7}. Earlier studies have invoked mineral dust deposition in the ocean surface as a dominant source of soluble inorganic phosphorus to the marine environment. However, recent studies have highlighted the contribution of anthropogenic sources such as its contribution from biomass burning emissions and fertilizers. Owing to large-scale burning of post-harvest agricultural waste in the IGP during October–November (Paddy residue) and April–May (wheat residue), contribution of aerosol-P is quite significant and could serve as a potential atmospheric source²⁰. The present article presents the current state of understanding with respect to air–sea deposition of P from observations and model studies for the Northern Indian Ocean. The large spatio-temporal variability associated with N/P molar ratio is an important issue in the present-day context of increase in their anthropogenic sources.

Current understanding: case study from Northern Indian Ocean

Size–distribution and provenance of aerosol-P

A comparison of mass concentration of soluble fraction of aerosol-P in two size fractions ($\text{PM}_{2.5}$ and PM_{10} refer

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to particles having an aerodynamic diameter is equal to or <2.5 and $10\ \mu\text{m}$) collected from the marine atmospheric boundary layer of the Bay of Bengal, reveals the coarse mode (Figure 1)²¹. A similar size-distribution (coarse mode) of crustal elements (Al, Fe, Ca and Mg) and water-soluble non-sea-salt (nss) Ca^{2+} (derived from uptake of acidic species on mineral dust during transport); together with significant linear correlation of nss- Ca^{2+} and inorganic P (P_{Inorg}), suggest significance of mineral dust as a source of aerosol-P. Furthermore, statistically significant correlation is observed between mass concentration of mineral dust and P_{Inorg} in ambient aerosols collected over the Arabian Sea (April–May 2006) and the Bay of Bengal (March–May 2006 and January 2009) (Figure 2). During this period, the isentropic air–mass back trajectories exhibit their origin from desert sources; and therefore, the observed relation signifies the source-specific contribution of phosphorus from desert dust. This argument is further supported by the close match between crustal elemental ratios of aerosol collected during March–May 2006 over the Northern India Ocean and that reported from the Thar Desert (Table 1). Although it originated from the desert region, mass concentration of aerosol-P from the Arabian Sea is significantly lower than that observed over the Bay of Bengal. Despite its proximity to desert region compared to Bay of Bengal, the low concentration over Arabian Sea can be explained by differences in the fractional solubility of mineral dust over the two basins.

Recently, Srinivas *et al.*²² had demonstrated significant differences in the solubility of mineral dust transported to

both the oceanic basins of the Northern Indian Ocean. The assessed aerosol iron solubility over the Bay of Bengal is relatively high (Av: 7%) compared to poor fractional solubility over Arabian Sea (no more than 1%; Av: 0.4%)²². The reason for the difference is attributed to acid-processing of mineral dust in addition to source-specific contribution as reflected in the low solubility of Arabian Sea²¹. Based on a similar analogy, atmospheric processing of mineral dust during long-range transport from desert region could explain the observed differences in the atmospheric abundances of water-soluble inorganic phosphorus over the Northern Indian Ocean. A significant (P -value < 0.05) linear relationship between the nss- Ca^{2+} and P_{Inorg} (ref. 21), hints at the acid-processing of mineral dust over the Bay of Bengal than that over the Arabian Sea. Based on these arguments, we suggest that mineral dust transport during spring–intermonsoon could serve as a source of soluble P over the Northern Indian Ocean. However, as mentioned earlier, the influence of polluted continental air masses from south and south-east Asia on the atmospheric boundary layer of the Bay of Bengal is considerably high compared with the Arabian Sea during December–April^{13,20,21}. Therefore, we suggest that significant contribution of aerosol-P is derived from an anthropogenic source, viz. biomass burning emissions and fertilizers in addition to that from acid-processing of mineral dust over Bay of Bengal.

Seasonal variability – change in source type and strength

There are no significant differences in the atmospheric mass concentration of soluble P over the Bay of Bengal between winter (January 2009) and spring cruises (March–April 2006). However, notable differences are observed in the chemical composition of airborne particulates sampled over the Bay region during the above two seasons. The winter cruise was characterized by the dominance of anthropogenic constituents, mainly carbonaceous and water-soluble inorganic species ($\sim 70\%$) (ref. 14); whereas mineral dust concentration accounts for $\sim 50\%$ of TSP mass in the spring cruise¹³. In addition, it is documented that fine dust from the alluvial soils of IGP is the source of P in the Bay of Bengal during winter cruise (January 2009) compared to that for spring–intermonsoon (March–April 2006) during which dust is transported from the Thar desert²¹.

The concentration of P_{Inorg} during winter cruise is derived from the combined contribution of alluvial dust, which is characterized by low Ca/Al and $\text{Ca}^{2+}/\Sigma\text{WSIC}$, and from anthropogenic sources (fertilizers and biomass burning emissions) (Figure 3). In contrast, dust transport from the Thar region (Rajasthan, India) is a source of soluble P over the MABL of Bay of Bengal during

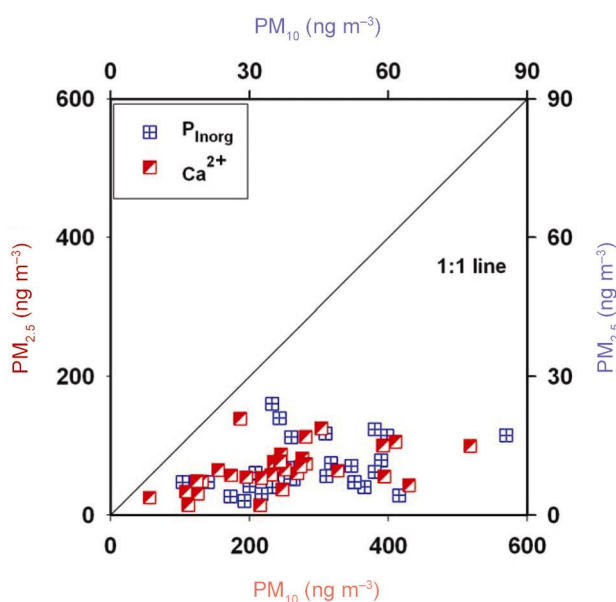


Figure 1. Scatter plot for atmospheric mass concentrations of water-soluble calcium (Ca^{2+}) and phosphorus (P_{Inorg}) in $\text{PM}_{2.5}$ and PM_{10} (axes on left and bottom refer to Ca^{2+} ; top and right axes correspond to P_{Inorg}) sampled over the Bay of Bengal during January 2009 suggests their size-distribution in coarse mode²¹.

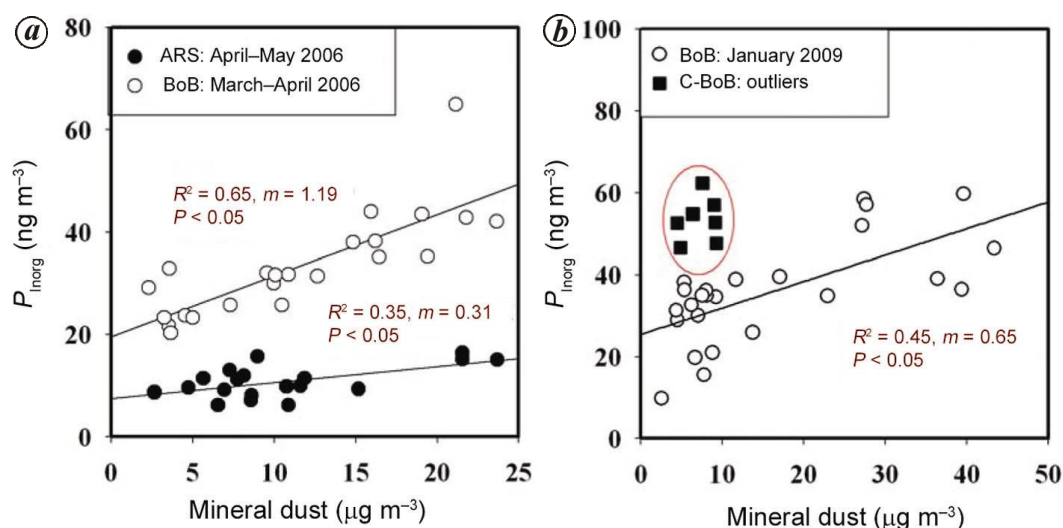


Figure 2. Scatter plot for mass concentrations of mineral dust and aerosol soluble phosphorus (P_{Inorg}) over the Arabian Sea (ARS) and Bay of Bengal (BoB). Outliers (shown as rectangles) are characterized by high contribution from anthropogenic sources ($P_{\text{anth}}/P_{\text{Inorg}} > 0.85$).

Table 1. Mass concentration (nmol m^{-3}) of soluble-P in aerosols collected from the Bay of Bengal and Arabian Sea and comparison with other oceanic regions

Region	Period	Av \pm Sd	Reference
Bay of Bengal	January 2009	0.3–2.8 (1.3 ± 0.5)	20
Bay of Bengal	March–April 2006	0.7–2.1 (1.1 ± 0.4)	20
Arabian Sea	April–May 2006	0.2–0.5 (0.4 ± 0.1)	20
South Atlantic Ocean	September–October 2001	0.004–0.18	33
Finokalia, Crete	September 1999–September 2000	0.13	33
Erdemli, Turkey	January–December 1999	0.26	33
Gulf of Aqaba	August 2003–September 2005	0.09–2.0	34
Gulf of Aqaba	August 2003–November 2004	0.5 ± 0.2	35

March–April 2006 (spring–intermonsoon cruise). The chemical composition data (Ca/Al , $\text{Ca}^{2+}/\Sigma\text{WSIC}$ and AMBTs; ref. 21) is consistent with that reported for source region^{23,24}. Also, it can be inferred from the relative decrease in Indo-Gangetic outflow from winter months (December–February) to spring–intermonsoon (March–April) that there is a significant decrease in supply of aerosol-P from anthropogenic sources during a later sampling period. The enhanced Ca^{2+}/Ca with low equivalent mass ratio of $\text{NH}_4^{2+}/\text{SO}_4^{2-}$ and significant linear relation of P_{Inorg} and nss-Ca^{2+} in the MABL of Bay of Bengal during March–April 2006 (ref. 21) could have occurred due to acid-processing of mineral dust as a source of P_{Inorg} during this period (Figure 3).

Chemical composition data for aerosols collected from the Arabian Sea and Bay of Bengal during March–May 2006, and the data from Bay of Bengal during January 2009 exhibit significant linear relation between mineral dust concentration and P_{Inorg} (Figure 2). Although abundance of mineral dust correlates with concentration of P_{Inorg} , contribution of P from anthropogenic sources is also

evident in the Bay of Bengal. In Figure 2b, some of the samples collected from the central Bay of Bengal during January 2009 are characterized by low dust concentration but with high contribution of aerosol-P from anthropogenic sources (as evident through $P_{\text{anth}}/P_{\text{Inorg}}$ ratio which is greater than 0.85). However, significant inter-correlations (P -value < 0.05) among nss-K^+ , carbonaceous species (OC, EC) and high $\text{nss-K}^+/\text{EC}$ (Av: ~ 0.47), suggest the impact of biomass burning emissions on the MABL of Bay of Bengal^{14,22}. The significant linear relationship of nss-K^+ with P_{Inorg} , suggests the contribution of P from biomass burning emissions²⁵. Furthermore, we have estimated the contribution of anthropogenic fraction of aerosol-P to the Bay of Bengal during January 2009 (ref. 21), as follows.

$$P_{\text{anth}} = P_{\text{Inorg}} - P_{\text{dust}}$$

Here, P_{anth} is the contribution of phosphorus from anthropogenic sources; P_{dust} refers to contribution of phosphorus from mineral dust i.e. $(\text{P}/\text{Al})_{\text{UCC}} \times \text{Al}_{\text{meas}} = 0.009 \times \text{Al}_{\text{meas}}$,

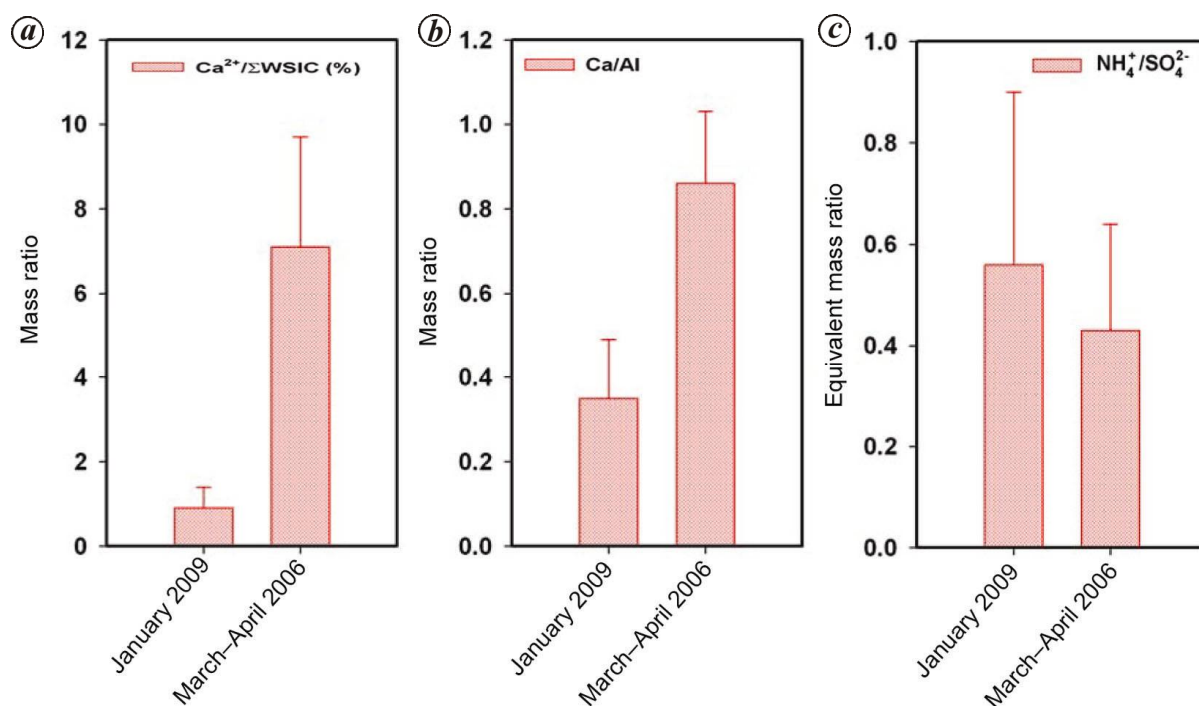


Figure 3. Histogram showing relative differences with regard to (a) fractional contribution of water-soluble Ca^{2+} to total water-soluble inorganic constituents ($\Sigma\text{WSIC}\%$); (b) mass ratio of Ca/Al and (c) equivalent mass ratio of $\text{NH}_4^+/\text{SO}_4^{2-}$, between the two research cruises January 2009 (winter) and March–April 2006 (spring–intermonsoon), conducted during the continental out-flow.

where $(\text{P}/\text{Al})_{\text{UCC}}$ is the abundance ratio of P/Al in upper continental crust²⁶.

This calculation is based on the assumption that Al is derived from atmospheric mineral dust. A supporting argument in its favour is the significant correlation among crustal elements (Al , Ca , Fe and Mg), suggesting that their crustal origin and elemental ratios of Fe/Al and Ca/Al over Bay of Bengal are comparable to the fine alluvium in the IGP²¹. Based on Taylor and McLennan²⁷, P and Al comprise 0.07% and 8.04% of upper continental crust (source of atmospheric mineral dust) and the weight ratio of P/Al is thus considered to be ~ 0.009 . Hence, P_{anth} is assessed by subtracting the contribution of phosphorus from mineral dust (P_{dust}) from the total measured P_{inorg} concentration. We have, therefore, estimated that almost 75% of contribution of aerosol P over the Bay of Bengal during January 2009 is derived from anthropogenic sources, viz. biomass burning emissions and fertilizers from the IGP. In contrast, the contribution of anthropogenic sources to the Arabian Sea is relatively low (<30%) and is dominated by dust input.

Air–sea deposition

The estimate of dry-deposition flux of atmospheric water-soluble inorganic phosphorus in the Northern Indian Ocean is compared with the available data for other oceanic

Table 2. Atmospheric dry-deposition fluxes of water-soluble inorganic P in the Northern Indian Ocean compared with projected model-based estimates and with other oceanic regions

Region	Gmol-P year ⁻¹	Reference
Bay of Bengal	0.8	20
Arabian Sea	0.4	20
N.Indian Ocean	1.2	20
N.Indian Ocean	1.4	10
*N.Indian Ocean	0.4	7
#N.Indian Ocean	1.2	28
Atlantic Ocean	0.2	29
N.Pacific Ocean	0.7	7
Mediterranean Sea	0.2	7

*Estimated from Mahowald *et al.*⁷, based on projected PO_4^{3-} -deposition of $\sim 13.5 \text{ Gg-P year}^{-1}$ to the Northern Indian Ocean. #Duce *et al.*²⁸ had projected P deposition in the Northern Indian Ocean $110 \text{ Gg P year}^{-1}$ and also estimated solubility of 33%. This gave rise to a soluble flux of $1.2 \text{ Gmol year}^{-1}$.

regions, and also with model-based projections (Table 2). Earlier studies on the Bay of Bengal have demonstrated that atmospheric deposition of this macronutrient in the Northern Indian Ocean is comparable in magnitude to its riverine supply^{20,21}. This observation highlights the importance of atmospheric source of this macronutrient in surface waters of the Northern Indian Ocean and is as important as that of fluvial input to the basins. The estimated total soluble phosphorus deposition to the Northern Indian Ocean is $\sim 1.2 \text{ Gmol-P year}^{-1}$ ($\sim 0.8 \text{ Gmol-P year}^{-1}$ for Bay of Bengal and $\sim 0.5 \text{ Gmol-P year}^{-1}$ for ARS). A

comparison of estimate from this study with model-based projection (~ 0.4 Gmol-P year⁻¹, ref. 7; ~ 1.4 Gmol-P year⁻¹, ref. 10; ~ 1.2 Gmol-P year⁻¹, ref. 28) for the Northern Indian Ocean suggests that in general there is good agreement between the two (Table 2). However, the concentrations reported for the Northern Indian Ocean are relatively high compared to that documented for North Atlantic (~ 0.2 Gmol-P year⁻¹)²⁹ and the Pacific (0.7 Gmol-P year⁻¹)⁷. These observations bring out the importance of anthropogenic sources, particularly in the downwind oceanic regions of pollution sources.

Scope for future research

Although efforts have been made to estimate the deposition fluxes of soluble P in oceans, observations on organic phosphorus are very few. A recent modelling study³⁰ has highlighted the relative importance of organic phosphorus (~ 0.35 Tg-P year⁻¹) and inorganic phosphorus in the global ocean. Although lower by more than a factor of 3, the projected organic phosphorus deposition to the Northern Indian Ocean is ~ 0.2 Gmol-P year⁻¹ (ref. 30). However, large uncertainty is associated with the estimate owing to paucity of data. This necessitates further studies on possible sources of phosphorus, transport and transformation pathways to the marine atmospheric boundary layer. More recently, the significance of atmospheric source to the surface waters was emphasized by providing information on bioavailable phosphorus⁹. In this context, the air–sea deposition of phosphorus is, thus, crucial to assess its impact on the biogeochemical cycle of this nutrient.

Although mineral dust supply dominates the air–sea deposition of P_{Inorg} , in the present-day context of changing climate scenario, contribution from anthropogenic sources (biomass burning emissions and application of fertilizers) overwhelms this natural source in the continental outflow. In addition to the processing of mineral dust during transport, mineralogy can provide crucial information regarding the supply of soluble phosphorus from mineral dust and explain source variability. More recent studies have focused on the oxygen isotopic composition of dissolved phosphorus (as phosphate) and suggested its applicability as a potential tracer for assessing the significance of aeolian phosphorus derived from mineral dust vis-à-vis anthropogenic sources to aquatic bodies^{6,31,32}. The increase in demand to understand the various atmospheric sources and their pathways to the surface ocean from the continental outflows requires long-term assessment of their role in biogeochemistry of the open and coastal waters.

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