

Influence of atmospheric dry deposition of inorganic nutrients on phytoplankton biomass in the coastal Bay of Bengal

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Abstract

The aerosols from continents contain relatively higher amounts of inorganic nutrients than those of marine origin and can make a notable contribution to the coastal biological productivity. To test this hypothesis, the composition of aerosols over the city of Visakhapatnam (central east coast of India) were studied when continental flow was dominant and its impact on phytoplankton biomass was estimated through microcosm experiments between September 2013 and November 2014. Higher nitrate (NO_3^-) and ammonium (NH_4^+) concentrations were observed in the aerosols collected in January while higher phosphate (PO_4^{3-}) was observed in September. Simultaneous observations of aerosols over the city and neighboring coastal waters revealed that the concentrations of nitrate in ambient aerosols ranged from 0.56 to 1.89 $\mu\text{g m}^{-3}$ and 0.09 to 0.86 $\mu\text{g m}^{-3}$, respectively. Our results suggest that 52-89% of city's aerosols borne nitrogen deposited over waters within 10 km from the coastline. Microcosm experiments were conducted by spiking the surface water samples, collected from the coastal Bay of Bengal (BoB), with the dust borne nutrients. Upon spiking, dissolved inorganic nitrogen ($\text{NO}_3^- + \text{NH}_4^+$) increased from 0.3 to 11.7 $\mu\text{mol L}^{-1}$ and the N:P ratio increased from 2 to 97. This led to enhanced phytoplankton biomass (1.5 to 4 times) upon spiking. The increase in phytoplankton biomass was linearly related to dissolved N: P ratios in water as aerosol deposition increased the N: P ratios in the microcosms, leading to phytoplankton growth. Though aerosols did not contribute to bioavailable silicate, our microcosm experiments showed linear relationships between ambient silicate phytoplankton biomass, and concentrations of Fucoxanthin (a marker pigment for diatoms). This indicates that the availability of silicate in coastal waters facilitated dominant diatom growth in the presence of higher N: P ratios due to aerosol deposition. The deposition of soluble aerosol nitrogen appears to support ~3 to 33% of the biological production in the coastal waters off Visakhapatnam with higher contribution in winter (~33%) than in summer (10%). This study suggests that atmospheric deposition of nutrients enhances phytoplankton biomass in waters along the central east coast of India during the winter monsoon period, in particular, supporting the hypothesis stated above.

1. Introduction

Several studies have highlighted the significance of the atmospheric pathway for transport of nutrients from continents to oceans (Delany et al., 1967; Duce et al., 1991; Prospero et al., 1996) and its role in ocean biogeochemical cycles (Jickells et al., 2005; Paerl and Whittall, 1999) at regional and global scales (Paerl, 1985; Spokes et al., 2000; Duce et al., 2008). Atmospheric deposition is one of the potential sources of nutrients to coastal waters and may be important in regulating the phytoplankton biomass (Paerl, 1985; Jickells, 1998; Duce et al., 2008; Mahowald et al., 2011; Boyd et al., 2010). It is estimated that the magnitudes of inputs of dissolved inorganic nitrogen (DIN) through river flux and atmospheric deposition to oceans are almost equal (Duce et al., 1991; Jickells, 1998; Boyd et al., 2010). Srinivas et al. (2011a) made a comparative study between atmospheric dry-deposition of total soluble nitrogen (2 to $167 \mu\text{mol-N m}^{-2} \text{d}^{-1}$) and river supplies ($214 \mu\text{mol-N m}^{-2} \text{d}^{-1}$; Kumar et al., 1996) and suggested that these two sources are of comparable magnitudes, by considering the upper limit of nitrogen deposition under the increasing anthropogenic scenario. Srinivas and Sarin (2013) presented the first data set on atmospheric inputs of N, P and Fe to the Northern Indian Ocean. Based on observations in the marine atmospheric boundary layer (MABL) during periods of continental outflow (January – April), the dry-deposition fluxes were estimated to be $2 - 167$, $0.5 - 4.8$ and $0.02 - 1.2 \mu\text{mol m}^{-2} \text{d}^{-1}$ for nitrogen, phosphorus and iron respectively over the Bay of Bengal; significantly higher than that over the Arabian Sea ($0.2-18.6$; $0.3 - 0.9$ and $0.001 - 0.015 \mu\text{mol m}^{-2} \text{d}^{-1}$ respectively). Based on these values, Srinivas and Sarin (2013) suggested that atmospheric anthropogenic sources can account for about 13% of the carbon fixation in the Bay of Bengal. It has been estimated that ~20 and ~30% of the annual mean new production is supported by atmospheric nitrogen inputs in the South China Sea (Kim et al., 2014) and eastern Atlantic surface waters (Knap et al., 1986; Owens et al., 1992, Michaels et al., 1993; Spokes et al., 2000), respectively. Wet deposition of DIN is found to account for 10 to 20% of the new production in the western Mediterranean Sea (Loye-Pilot et al., 1993). Atmospheric deposition of reactive nitrogen (N), in the forms of HNO_3 and NH_3 , may also change surface seawater alkalinity and pH, and consequently inorganic carbon species composition that particularly affects coastal ecosystems (Doney et al., 2007; Hagens et al., 2013; Sarma et al., 2015).

The Bay of Bengal (BoB) or the northeastern Indian Ocean is a tropical basin, which is bordered by land to the north, east and west but open to the Indian Ocean in the south. This region comes under the influence of seasonally reversing monsoon winds (Prasanna Kumar et al., 2002). The airflows from South Asia and South-East Asia, from January to March, transport

pollutants to the boundary layer over BoB (Srinivas et al., 2011a). The BoB receives huge amounts of freshwater from the Himalayan rivers (Ganges and Brahmaputra) and several peninsular rivers (Mahanadi, Godavari etc.) leading to stratification in the upper water column. This serves to reduce the supply of nutrients from deeper waters through vertical mixing (Madhupratap et al., 2003). Hence, the supply of nutrients from atmospheric deposition may be significant for biological production in the Bay. Enhanced chlorophyll associated with atmospheric dust was reported in the BoB using remote sensing data (Patra et al., 2007). Seasonally variable winds blow from north-east to south-west (continental air) during winter (December-February) and spring (March-April) resulting in the transport of pollutants in the downwind direction (Srinivas et al., 2011a). During the same period, nutrients in surface waters along the western coastal BoB are low due to weak vertical mixing (Ramaiah et al., 2010; Sarma et al., 2013). Gauns et al. (2005) showed that primary productivity in the coastal waters of the Arabian Sea and Bay of Bengal are comparable during winter and intermonsoons (both spring and fall) periods despite weak vertical mixing in the latter than in the former basin. Srinivas et al. (2011a) noted that the Bay receives more nutrients from atmospheric deposition compared to the Arabian Sea and estimated that 1.1 and 0.03 PgC y^{-1} of the primary production is supported by aeolian transported nutrients, respectively, assuming that the dust flying over the marine atmosphere gets deposited into the surface waters and all nutrients were utilized by phytoplankton. While increasing ambient concentrations of N and P in surface waters, the deposition of air-borne nutrients may also modify N: P ratios in the water and this could alter the phytoplankton community composition. We hypothesize that a large concentration of anthropogenic aerosols, originating from the coastal city of Visakhapatnam, gets deposited over the proximate coastal waters where it may be responsible for enhancing phytoplankton production. Our objective here is to test this hypothesis by conducting experiments in the city and adjoining coastal BoB with focused observations when wind blew from land.

2. Material and methods

2.1. Study area and atmospheric aerosol collection

Visakhapatnam is an active port city in the state of Andhra Pradesh and located among the Eastern Ghats (mountain range), central east coast of India. It is the second largest city in the state covering an area of ~ 550 km² and is primarily an industrial city. The anthropogenic sources to aerosols in and around this city mainly include ore transport through an open belt system, burning of coal by the National Thermal Power Corporation (NTPC) and fossil fuel burning mainly by vehicles whereas fertilizer usage in agricultural activities in the suburbs contributes to organic and

inorganic nitrogen and inorganic phosphate.

Aerosol sampling was done in the Visakhapatnam city (17.69° N; 83.22° E) at the top of our laboratory building (20 m above mean sea level) located about 200 m from the coastline (Fig. 1). Atmospheric aerosols (referred as total suspended particulates,(TSP) collected for a duration of ~24 hours) were sampled on pre-combusted quartz fiber filters (395.5 cm², PALLFLEX) using a High Volume Sampler (Envirotech 430) at a flow rate of 1.2 m³ min⁻¹ in September (6/09/2013), January (9/01/2014), March (26/03/2014) and November (25/11/2014). Simultaneous collection of PM₁₀ aerosol samples ($\geq 10 \mu\text{m}$; referred as Respirable Suspended Particulate matter collected for ~6 hour duration) was done at the laboratory building and adjoining coastal waters in December 2013 and March 2014. The coastal water sampling site was ~10 km from the coastline. The aerosols were sampled using a Respirable dust sampler APM 460BL (Envirotech, India) at ~ 0.9-1.4 m³ min⁻¹ flow rate on board a trawler (Fig. 1). To avoid contamination from the boat's exhaust, the trawler was anchored (with its engine shut off) at the location where other boat activities were minimal. The sampling was carried out only when the air flow from land was dominant as we planned to examine the differences in magnitude and the composition of aerosols collected over the city and coastal waters and also to estimate the fraction of aerosols getting deposited over the coastal waters.

2.2 Microcosm experiments

The microcosm experiments were conducted using one-day integrated aerosol samples in September 2013, and January, March and November 2014 (Table 1). On the same days of aerosol collection, quadruplicates of 20 L of surface seawater were collected in transparent Nalgene containers at ~5 km from the coast and filtered through 200 μm mesh to remove larger zooplankton. The aerosol samples were treated for 30 minutes with 250 mL Milli-Q water using ultrasonic agitation. The extracted solution was filtered through a 0.7 μm GF/F filter to remove black carbon. The filtered extract was then divided into 3 fractions of 100 mL, 100 mL and 50 mL. The 100 mL fractions were used to spike duplicates of surface seawater collected in 20 L Nalgene bottles and the 50 mL fraction was used to measure the initial concentrations of water soluble nutrients in the aerosol sample. The initial concentrations of nutrients in the surface seawater samples were measured before they were spiked with aerosol-borne nutrients. Two of the 20 L Nalgene bottles were processed as controls (without any nutrients additions). These microcosm experiments were conducted in duplicate under natural light conditions in a water bath with swirling of bottles at regular intervals to avoid settling of plankton.

2.3 Measurements of nutrients and pigments

Sub-sampling of seawater from the incubated microcosm bottles was carried out on alternate days (September) or daily (January, March and November) for one week to examine progressive changes in the concentrations of nutrients and phytoplankton pigments. The concentrations of nutrients in seawater were measured using standard colorimetric techniques (Grashoff et al., 1992). Nitrate was reduced to nitrite using cadmium-mercury amalgam and complexed with sulphanilamide and N (1-naphthyl) Ethylene diamine to form azodye. The absorbance of this dye was measured at 543 nm. Ammonium in water was made to react with hypochlorite under alkaline conditions to form monochloramine, which in the presence of phenol-nitroprusside yielded indophenol blue complex. The absorbance of the complex was measured at 630 nm. Inorganic phosphate in water was allowed to react with acidified ammonium molybdate to yield phosphomolybdate complex and the same was reduced using ascorbic acid in presence of antimonyl ions leading to the formation of phosphomolybdenum blue complex. The absorbance of complex was measured at 882 nm. Dissolved silicate in samples was treated with ammonium molybdate under acidic conditions to form yellow silicomolybdic acid, which was reduced to silicomolybdenum blue complex by ascorbic acid. Its absorbance was measured at 810 nm. The analytical precision for ammonium, nitrate, phosphate and silicate was ± 0.02 , ± 0.02 , ± 0.01 and $\pm 0.02 \mu\text{mol L}^{-1}$, respectively.

Nutrients in aerosols were measured using Ion Chromatography (850 Metrohm Professional, Switzerland). Each filter containing total aerosols collected was treated for 30 minutes with 50 mL of Milli-Q water using an ultrasonicator to extract nutrients. The water extract was transferred into a clean Teflon tube. Nutrients (ammonium, nitrate and phosphate) were analyzed using a cation exchange column (Metrosep C4-150/4.0) and an anion exchange column (Metrosep A Supp 5 250/4.0) and measured by conductivity. Samples were introduced into the chromatographic column through a 20 μL loop. The eluent used for anions was a mixture of Na_2CO_3 (3.2 mM), NaHCO_3 (1 mM) and H_2SO_4 (100 mM) at a flow rate of 0.7 mL min^{-1} whereas the eluent for cations was HNO_3 (2 mM) at a flow rate of 0.9 mL min^{-1} . The detailed Ion Chromatography procedure is discussed by Kumar et al. (2010). A similar procedure was followed for blank filter analysis and corrections with respect to filter and instrumental blanks were applied to the samples. Calibration was done by a set of standards containing either anions or cations of variable concentrations. The overall precision of these measurements is $<10\%$.

For pigment analysis, 500-1000 mL coastal water samples were filtered onto 25 mm $0.7 \mu\text{m}$ GF/F filters using a vacuum pump (Rocker 600 from Tarsons, India) at $<200 \text{ mb}$ vacuum. The filtration was done under low light conditions to avoid photo-degradation of the pigments.

Immediately after filtration, the filters were folded and wrapped in a vial and stored at -20°C until analysis. The extraction was performed by soaking the filters overnight at -20°C in 5 mL of 90%:10% acetone:water (HPLC grade from Merck chemicals) in amber coloured bottles to prevent photo-decomposition. A tissue homogenizer (Ultra Turrax, Germany) was used to break the cells keeping the bottles on an ice filled tray followed by cold centrifugation at -10°C (Eppendorf, Germany). The extracts were centrifuged for 30 minutes at 500g (where $g=9.81\text{m/s}^2$). The supernatant was mixed manually with a buffer made of 28 mM aqueous Tetra Butyl Ammonium Acetate (AR Grade, Fluka) at pH 6.5 and methanol (GC Assay 99.7% pure, Merck) at 90:10 ratio. About 400 μL of extract was injected into a High Pressure Liquid Chromatograph (Agilent 1200 series) at a pressure of 200b (maximum) with a solvent flow rate of 1.1 mL min^{-1} (Thomas and Heukelem; 2001). The peaks of marker pigments were identified using high purity standards (DHI Lab Products, Denmark). The precision of the pigment analyses was $\pm 1\%$. A total of 16 pigments (Pyridine, Fucoxanthin, Zeaxanthin, 19'-but-fucoxanthin, HF-19, 19Hex, Violaxanthin, Astaxanthin, Diadinoxanthin, Alloxanthine, Diatoxanthin, Lutein, Chlorophyll a, Chlorophyll b, Pheophytin, B-carotene) can be detected from this methodology using CHEMTAX software.

2.4 Air Mass Back trajectories

Air mass back trajectories (AMBTs) have been used to trace the possible source regions of aerosols to the sampling site. The back trajectories were computed using the GDAS database of the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory and the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT program, version- 4.0, <http://ready.arl.noaa.gov/HYSPLIT.php>) using arrival heights of 100, 500 and 1000 m (Draxler, 2002). The AMBTs were calculated for 7-day periods preceding our aerosol collection.

3. Results

3.1 Winds and nutrient composition of aerosols

The air mass back trajectories suggested that the source of the aerosols was central India (terrestrial) during September whereas it was from north-eastern parts of peninsular India during January and November (Fig. 2). In contrast, the aerosols appeared to have a marine source in March. The TSP concentrations in air over Visakhapatnam were higher during January ($188\mu\text{g m}^{-3}$), followed by September ($121\mu\text{g m}^{-3}$), November and March ($76\text{-}80\mu\text{g m}^{-3}$). While anthropogenic aerosols might have contributed to higher TSP amounts in winter, aerosols transported long distance by high altitude winds from the west might have possibly contributed to

those in September (Prospero et al., 2002). The concentrations of soluble nitrate in the aerosols (Table 1) was higher during January ($1.17 \mu\text{mol m}^{-3}$) followed by September and November ($\sim 0.65 \mu\text{mol m}^{-3}$) and March ($0.12 \mu\text{mol m}^{-3}$). Higher concentrations of ammonium were found during January ($0.5 \mu\text{mol m}^{-3}$) followed by November ($0.1 \mu\text{mol m}^{-3}$). Measurable phosphate was observed during September ($0.13 \mu\text{mol m}^{-3}$). It was close to the detection limit during November 14 ($0.02 \mu\text{mol m}^{-3}$) and below detection limits during other periods (Table 1). The study region received aerosols from the Indian subcontinent (September and January) and from marine regions (March), respectively, but a mixture of both continental and marine in November (Figure 2). Relatively higher concentrations of nitrate and ammonium were observed in the continental dust and mixed dust than marine dust. Measurable phosphate in September seems to have been associated with long distance transport.

3.2 Aerosol and Nutrient deposition in the coastal Bay of Bengal

Simultaneous collection of aerosols over the city and coastal water during March and December, when winds blew from the land to the sea, facilitated quantification of nutrient deposition between the city and the coastal sampling site. Relatively higher TSP concentrations were observed both over land and coastal waters during March (averages and standard deviation of 73 ± 14 and $63 \pm 5 \mu\text{g m}^{-3}$, respectively) than in December (averages and standard deviation of 55 ± 17 and $31 \pm 13 \mu\text{g m}^{-3}$, respectively) (Fig 3). We estimated the depositional fluxes of transported atmospheric dust over the coastal waters by assuming that the aerosol concentration differences between land and sea stations resulted from some aerosol deposition to the coastal waters as the air masses moved offshore. Based on the observed differences between TSP in air over the city (only 200m inland) and over the coastal waters ($\sim 10\text{km}$ offshore), the amount of TSP deposited over the coastal waters (within 10 km from the coast) was estimated to be between 15 and 45%. Consequently, proportional amounts of nutrients might have been deposited in coastal waters. For instance, the concentration of nitrate in the TSP over land varied between 0.56 and $1.89 \mu\text{g m}^{-3}$ whereas it was 0.09 to $0.86 \mu\text{g m}^{-3}$ over coastal waters indicating that 0.29 to 1.61 (0.79 ± 0.52) $\mu\text{g m}^{-3}$ of nitrate might have been deposited in the coastal waters. This suggests that 52-89% of the air-borne nitrate is deposited in coastal waters (Figure 3), which might significantly influence biological production and plankton composition in the coastal waters.

3.3 Response of phytoplankton to dust supplied nutrients in the coastal Bay of Bengal

Marginally higher concentrations of nitrate were observed in the surface waters of the coastal Bay of Bengal (Table 2) during January 2014 ($1.94 \mu\text{mol L}^{-1}$) than in other sampled periods (0.70 to 1.79

$\mu\text{mol L}^{-1}$) whereas ammonium was much higher during September 2013 ($1.62 \mu\text{mol L}^{-1}$) than in the other periods (0.12 to $0.3 \mu\text{mol L}^{-1}$). The concentrations of phosphate were higher during March 2014 ($0.56 \mu\text{mol L}^{-1}$) than in other periods (0.23 to $0.43 \mu\text{mol L}^{-1}$). Lower dissolved silicate concentrations were observed during January and March (1.5 and $0.86 \mu\text{mol L}^{-1}$, respectively) than in September and November (5.49 and $3.05 \mu\text{mol L}^{-1}$, respectively). All nutrients in surface waters are relatively enriched in September. The N:P ratios during the entire study period were significantly lower (2.21 to 9.13) than the theoretical Redfield ratio of 16 . The low N:P ratio in surface waters suggests that addition of inorganic nitrogen may increase phytoplankton biomass since Sarma et al. (2013) observed that nitrate is a limiting nutrient for biological production in the coastal Bay of Bengal.

The growth of phytoplankton in the ocean depends on the availability of macro and micronutrients. Macronutrients nitrogen (N) and phosphorus (P) are required by cells in higher quantities and at relatively similar ratios across different taxa (Redfield et al., 1963).

Though the methodology we adopted for pigment analysis can potentially identify sixteen pigments our results indicates that only seven (Chl a, Fuco, Zea, Din, 10 Hex, B-carotenoid and Chl c2) could be detected in our samples. However, the three pigments (19 Hex, B- carotenoid and Chl c2) were very close to detection limits and hence we prefer to report only four pigments (Chl a, Fuco, Zea and Din) (Table 3). Addition of aerosol extracts enhanced the concentrations of nutrients in the coastal waters used in the microcosm experiments. The increases in nitrate were higher during September ($13.67 \mu\text{mol L}^{-1}$) followed by January ($3.93 \mu\text{mol L}^{-1}$), November ($2.4 \mu\text{mol L}^{-1}$) and March ($0.46 \mu\text{mol L}^{-1}$). The increases in phosphate were notable only during November ($1.05 \mu\text{mol L}^{-1}$) and September ($0.77 \mu\text{mol L}^{-1}$) (Table 4). The increases of ammonium were similar to those of nitrate (higher during September ($1.94 \mu\text{mol L}^{-1}$) followed by January ($0.93 \mu\text{mol L}^{-1}$)). Relatively lower increases in nutrients were observed during March when the aerosols appeared to have a marine source (Figure 2). As a result of adding varied amounts of inorganic nitrogen and phosphate to surface waters, the N:P ratio increased from 7.93 to 20.2 during September , 9.13 to 97.2 during January and from 3.5 to 7 during March but the change was not significant in November (Tables 2 and 4).

The concentrations of nutrients decreased with time in spiked microcosms, presumably due to biological consumption (Fig 4). In some control microcosms, however, random increases in nutrients have been found. Even though large sized grazers ($>200 \mu\text{m}$) were removed by filtration, bacteria and microzooplankton might have passed through the filter and remained in the coastal

water subsamples used for the microcosm experiment. Hence, the changes in nutrients concentration rather reflect net variations in microcosm waters. In spite of such notable nutrient releases in control microcosms, majority of spiked microcosms exhibited gradual decreases in nutrients presumably due to enhanced biological uptake. The uptake of nitrogenous nutrients is more striking in September and January (Table 2; Fig. 4). The least removal of nutrients during March is consistent with low supplies of nutrients through aerosols (Table 1).

The relative contributions of the four marker pigments used in this study to the total pigment concentrations in coastal waters indicated that Bacillariophyceae (Fucoxanthin) was the dominant group during the entire study period (62-70%) followed by Cyanophyceae (Zeaxanthin) (6-20%) and Dinophyceae (Diadinoxanthin) (5-23%) (Table 5). Less variability in the concentrations of Fucoxanthin ($0.23\text{-}0.29\ \mu\text{g L}^{-1}$), Zeaxanthin ($0.02\text{-}0.06\ \mu\text{g L}^{-1}$), Diadinoxanthin ($0.03\ \text{to}\ 0.08\ \mu\text{g L}^{-1}$) and phytoplankton biomass (as Chl-a) ($0.61\ \text{to}\ 0.81\ \mu\text{g L}^{-1}$) was observed in the control microcosms during the entire study period, except in November when higher concentrations of Fucoxanthin ($0.66\ \mu\text{g L}^{-1}$) and Diadinoxanthin ($0.24\ \mu\text{g L}^{-1}$) were observed (Table 5). After the spiking of aerosol nutrients into seawater, Fucoxanthin increased significantly during September (88%; $t\text{-stat}=-1.4$; $p<0.01$), November (58%; $t\text{-stat}=-2.1$; $p<0.05$), January (174%; $t\text{-stat}=-3.2$; $p<0.05$) and March (165%; $t\text{-stat}=-1.9$; $p<0.05$) (Fig. 5). Similar increases in Zeaxanthin and Chlorophyll-a were found to be statistically significant during all experimental periods (Table 5).

4. Discussion

4.1 Response of phytoplankton to atmospheric deposition of nutrients

The amounts of nutrients in aerosols are related to their sources and sizes of the particles (Church and Jickells, 2004) modulated by atmospheric processes. The composition of aerosols (Table 1) during the study period revealed higher nutrient concentrations during January (nitrate and ammonium) followed by September (nitrate and phosphate) and November (nitrate, ammonium and phosphate) when aerosols were transported from terrestrial sources, as shown by the AMBTs (Fig. 2). This indicates the export of anthropogenic pollutants from the adjacent terrain, a known hub for some major industrial and mining activities. Agricultural industry in this region uses nitrogen and phosphorus fertilizers such as diammonium phosphate, urea etc. On the other hand, lower concentrations of nutrients were observed in aerosols during March (Table 1) when aerosols were contributed by marine sources (Fig. 2). However, several investigators reported that the coastal Bay of Bengal receives anthropogenic aerosols during spring (Srinivas and Sarin, 2014; Srinivas et al., 2011a,b and Kumar et al., 2008; Sarin et al., 2010). The difference might have arisen from

temporal variations in the direction of the winds.

The strong salinity stratification in the BoB reduces upward mixing of subsurface waters (Prasanna Kumar et al., 2002; Madhupratap et al., 2003) so that phytoplankton are severely limited by nutrient availability (Gauns et al., 2005). The present microcosm experimental results showed an increase in phytoplankton biomass by 1.5 to 4 times due to supply of nutrients from aerosols confirming that the biological productivity in the coastal Bay waters was limited by nutrient availability. The increase in phytoplankton biomass was linearly related to the inorganic nitrate concentrations ($r^2=0.68$; $p<0.001$) and N:P ratios ($r^2=0.85$; $p<0.001$) that were measured after the aerosol spike was added. No significant relationships were found with phosphate ($r^2=0.05$, $p>0.05$) and ammonium ($r^2=0.006$, $p>0.05$). Fucoxanthin displayed a linear relationship with nitrate ($r^2=0.95$; $p<0.001$) and the N:P ratios ($r^2=0.55$; $p<0.001$) suggesting that nitrate addition promoted diatom biomass. The concentrations of silicate in incubated waters showed a linear relationship with phytoplankton biomass ($r^2=0.38$; $p<0.05$) and Fucoxanthin ($r^2=0.85$; $p<0.001$) indicating that the addition of nitrate by aerosols facilitated the proliferation of diatoms using ambient silicate. The coastal waters of the Bay of Bengal contain relatively higher concentrations of silicate through river discharge (Sarma et al., 2013). Our experiments also revealed that the nutrients added through aerosol extracts were consumed in the microcosm waters within 1 - 2 days further demonstrating that the low ambient nutrient concentrations (nitrate in particular) are limiting the biological productivity in these waters (Fig. 4). The present results are consistent with those from fertilization experiments, using nutrient rich deep Bay waters, conducted in the coastal BoB (Sarma et al., 2013).

The phytoplankton biomass in coastal waters off Visakhapatnam was dominated by Bacillariophyceae (Bac; 62-70%) followed by Cyanophyceae (Cya; 6-20%) and Dinophyceae (Din; 5-18%) during the present study. However, the Bac/Cya ratio was significantly lower during September (3.8) compared to that in January (8.5) and March (15). The higher ratios in January and March were due to 8-10 fold higher concentrations of Bacillariophyceae as opposed to a decrease in Cyanophyceae. Due to addition of nutrients from the aerosols, phytoplankton biomass increased between 1.5 and 4 times (September>November>March). The contributions of the different phytoplankton groups to the total biomass were also modified with time during the microcosm experiments (Fig 5). For instance, the ratio of Bac/Cya was significantly lower at the end of the experiments than at the beginning during September (by 1.6 ± 0.4), but increased during January (by 3.6 ± 0.3) and November (by 12.3 ± 5) with no change during March (by 0.2 ± 0.4). This suggests that Cyanophyceae responded significantly to the nutrient addition from aerosols

during September whereas Bacillariophyceae responded during January and November. No notable plankton compositional change was found during March when relatively low nitrate ($0.12 \mu\text{mol m}^{-3}$) and no phosphate were added through the aerosols. On the other hand, the Bacillariophyceae to Dinophyceae (Bac/Din) ratio decreased during the entire sampling period by 0.3 to 2.4 except in November when it increased by 0.8 and the net increase in phosphate was high (Table 4). This suggests that Dinophyceae also responded to nutrient addition from aerosols (Fig. 5). Our results indicated that the shifts in phytoplankton composition occurred due to input of nutrients from the aerosols and was also governed by the ambient concentrations (mainly silicate) in the water that was used for the incubations.

4.2 Influence of atmospheric deposition on coastal primary production

The aerosol concentrations of nitrate (0.12 to $1.17 \mu\text{mol m}^{-3}$) and ammonium (0.02 to $0.5 \mu\text{mol m}^{-3}$) varied (Table 1) significantly during the study period (September to November). Using these values, the dry-deposition fluxes of nitrate and ammonium have been calculated using Equation 1:

$$\text{Dry-Deposition flux (f}_{\text{dry}}) = C_{\text{measured}} * V_{\text{dry}} \quad \text{---(1)}$$

Where C_{measured} is the concentration of nutrients (nitrate or ammonium) in the aerosols and V_{dry} is the dry deposition velocity. Duce et al. (1991) suggested dry-deposition velocities for fine ($<2.5 \mu\text{m}$ particles) and coarse mode aerosols ($> 2.5 \mu\text{m}$ particles) to be 0.1 and 1.0 cm sec^{-1} , respectively. In the present study, V_{dry} for NH_4^+ was taken as 0.1 cm sec^{-1} due to its predominant occurrence in the fine mode (Chen et al., 2007; Chen et al., 2006; Srinivas et al., 2011a) while V_{dry} for nitrate was taken as 1.0 cm sec^{-1} . The dry deposition fluxes are associated with an uncertainty by a factor of 3, which is caused due to several factors that include wind speed, terrain roughness, particle composition etc (Srinivas et al 2013). The dry depositional flux of inorganic nitrogen (nitrate and ammonium) varied between 106 ± 31 and $1049 \pm 302 \mu\text{mol-N m}^{-2} \text{ d}^{-1}$ during September to November. In the offshore BoB, Srinivas et al. (2011a) computed a dry depositional flux of inorganic nitrogen to be $2 - 83 \mu\text{mol m}^{-2} \text{ d}^{-1}$; an order of magnitude lower than the present estimate in the coastal waters off Visakhapatnam. To examine its impact on primary production, atmospheric dry deposition of nutrients is converted into equivalent primary production using Redfield stoichiometry (C:N:P=106:16:1). Assuming all the dry deposited nitrogen is completely utilized by phytoplankton, the estimated carbon fixation ranged from 9 to $83 \text{ mgC m}^{-2} \text{ d}^{-1}$. The seasonal primary production in the coastal Visakhapatnam varied between 253 and $434 \text{ mgC m}^{-2} \text{ d}^{-1}$ (Gauns et al., 2005; Kumar and Ramesh, 2005; Prasanna Kumar et al., 2007; Jyothibabu et al., 2008; Ramaiah et al., 2010;

our unpublished data) with relatively higher production during the fall intermonsoon than other seasons. Thus, our study suggests that 3 ± 0.4 to $33\pm 3\%$ of the total primary production is supported by the atmospheric deposition of nutrients to the coastal waters off Visakhapatnam with relatively higher contributions during winter (33%), than in summer (10%) followed by spring (3%). Srinivas et al. (2011a) estimated that $\sim 13\%$ of the primary production in the entire Bay of Bengal is supported by atmospheric deposition of nutrients assuming all particles in the marine atmosphere are deposited over the Bay of Bengal. Sarma et al. (2013) noted that the coastal BoB receives nutrients mainly through extreme atmospheric conditions, such as rainfall, depressions and cyclones during non- monsoon period (October-February), weak upwelling during spring (March-May) and river discharge during summer (June-September). The extreme conditions along the Indian coast are rather short-lived (Maneesha et al., 2011). The west coast of India receives nutrients through vertical mixing due to weak stratification and wind driven mixing supplies significant amount of nutrients to the surface (Prasanna Kumar and Prasad, 1996; de Sousa et al., 1996). Though strong stratification hinders the vertical supply of nutrients during the monsoon period resulting in low primary production on the east coast of India compared to west coast of India (Madhupratap et al., 2003), in contrast, primary production along the east and west coasts of India is almost equal during non-monsoon periods (Gauns et al., 2005). Our present study provides evidence for atmospheric deposition of aerosols being a nutrient source that significantly contributes to phytoplankton biomass and species diversity along the central east coast of India during the non-monsoon period.

5. Summary and Conclusions

The objective of this work was to examine the hypothesis that phytoplankton biomass and primary production in the coastal BoB are supported significantly by the atmospheric deposition of nutrients during the non-monsoon period. The simultaneous collection of aerosol samples over the coastal city of Visakhapatnam and over the adjacent coastal waters (about 10 kms from the coast) suggested that 15-45% of the PM₁₀ particles deposited within 10 km from the coast. On the other hand, the decrease in aerosol nitrate concentrations suggested that 52-89% of the land-derived nitrate was deposited in the coastal waters. A two to four-fold increase in phytoplankton biomass, mainly diatoms, was observed after the addition of aerosol nutrients to the coastal water suggesting nutrient limitation in the coastal BoB. The increases in phytoplankton biomass following the addition of aerosol nutrients displayed linear relationships with concentrations of inorganic nitrogen added and the resultant N: P ratios suggesting that atmospheric nitrogen inputs facilitated phytoplankton growth. Though a bioavailable dissolved silicate contribution from the atmosphere was negligible

due to low dissolution at sea water pH, the ambient concentrations of silicate in the incubated waters showed linear relationships with both phytoplankton biomass and the concentration of Fucoxanthin (Bacillariophyceae/ diatom marker pigment). This suggests that the availability of silicate in the coastal waters selects for the dominant group of phytoplankton upon the atmospheric deposition of nutrients. Based on the nutrient dry deposition rates and the Redfield ratio the contribution of atmospheric inputs to primary productivity is estimated to be 3 to 33% in the coastal BoB. This study suggested that atmospheric deposition of nutrients contributes significantly to phytoplankton productivity and species diversity along the east coast of India during the non-monsoon period. Thus its impact on other components of the ecosystem through changes in plankton biomass and diversity should be evaluated.

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Table1. Mean concentrations of nutrients in atmospheric aerosols obtained from samples collected in each month over the city of Visakhapatnam.

Month of sampling	Nutrients ($\mu\text{mol m}^{-3}$)			No. of samples collected
	NO_3^-	PO_4^{3-}	NH_4^+	
September'13	0.66 \pm 3	0.13*	0.03 \pm 0.3	4
January'14	1.17 \pm 5.03	BDL	0.5 \pm 2	3
March'14	0.12 \pm 0.33	BDL	0.02 \pm 0.2	2
November'14	0.65 \pm 2.71	0.02	0.10 \pm 0.44	2

*Concentration found detectable in only one of the number of samples specified in the last column of the -month. BDL=Below Detection Limit($0.01\mu\text{mol m}^{-3}$)

Table2. Variations in the concentrations of nutrients in coastal water samples and their uptake rates obtained from the microcosm incubation experiments.

Months	Nutrients ($\mu\text{mol L}^{-1}$)				N:P	Uptake rates ($\mu\text{mol L}^{-1} \text{d}^{-1}$)			
	NO_3^-	PO_4^{3-}	NH_4^+	Si(OH)_4		NO_3^-	PO_4^{3-}	NH_4^+	Si(OH)_4
September'13	1.79	0.43	1.62	5.49	7.93	3.48	0.27	0.75	0.73
January'14	1.94	0.23	0.16	1.5	9.13	0.65	0.01	0.8	0.01
March'14	1.68	0.56	0.3	0.86	3.53	0.29	0.02	0.03	0.08
November'14	0.7	0.37	0.12	3.05	2.21	0.34	0.07	0.03	0.61

Table 3: Marker pigments along with their symbols used to identify biomass and dominant taxonomic groups of phytoplankton. (Jeffrey and Vesk, 1997) in this study.

Symbol	Pigment name	Marker for
Chl a	Chlorophyll a	Phytoplankton biomass
Fuco	Fucoxanthin	Bacillariophyceae/ Diatoms
Zea	Zeaxanthin	Cyanophyceae
Din	Diadinoxanthin	Dinophyceae

Table 4. Increases in nutrients due to dust dissolution in surface water and the subsequent net increase in pigments in microcosm waters in the incubation experiments during different months. N:P ratios are calculated using total inorganic nitrogen ($\text{NO}_3^- + \text{NH}_4^+$) and phosphate. Pigment symbols are as shown in Table 3.

Month	NO_3^-	PO_4^{3-}	NH_4^+	$\text{Si}(\text{OH})_4$	N:P	Chl-a	Zea	Fuco	Din
	(μmolL^{-1})					$(\mu\text{g L}^{-1})$			
September 6/09/2013	13.67	0.77	1.94	2.13	20.2	0.39	0.08	0.21	0.08
January 9/01/2014	3.93	0.05	0.93	0.05	97.20	1.06	0.04	0.39	0.17
March 26/03/2014	0.46	0.09	0.17	0.11	7.00	0.61	0.02	0.15	0.06
November 25/11/2014	2.4	1.05	0.1	1.05	2.3	1	-0.02	0.38	0.04

Table 5: Statistical evaluations of the increases in pigment concentrations (indicating the rise in plankton populations following nutrient supply through aerosols).

Month	Chlorophyll a				Zeaxanthin				Fucoxanthin				Diadinoxanthin			
	Control	Experiment	T stat value	P value	Control	Experiment	T stat value	P value	Control	Experiment	T stat value	P value	Control	Experiment	T stat value	P value
September'13	0.77	1.17	-1.33	<0.1	0.06	0.14	-2.24	<0.05	0.24	0.45	-1.4	<0.01	0.08	0.16	-1.6	<0.01
January'14	0.81	1.87	-7.3	<0.001	0.03	0.07	-1.8	<0.05	0.23	0.63	-3.2	<0.01	0.03	0.21	-3.64	<0.001
March'14	0.61	1.22	-3.1	<0.01	0.02	0.04	-3.4	<0.01	0.29	0.77	-1.9	<0.05	0.05	0.12	-3.3	<0.01
November'14	0.76	1.56	-2.9	<0.05	0.05	0.05	2.8	<0.05	0.66	1.04	-2.1	<0.05	0.24	0.28	-0.8	<0.1

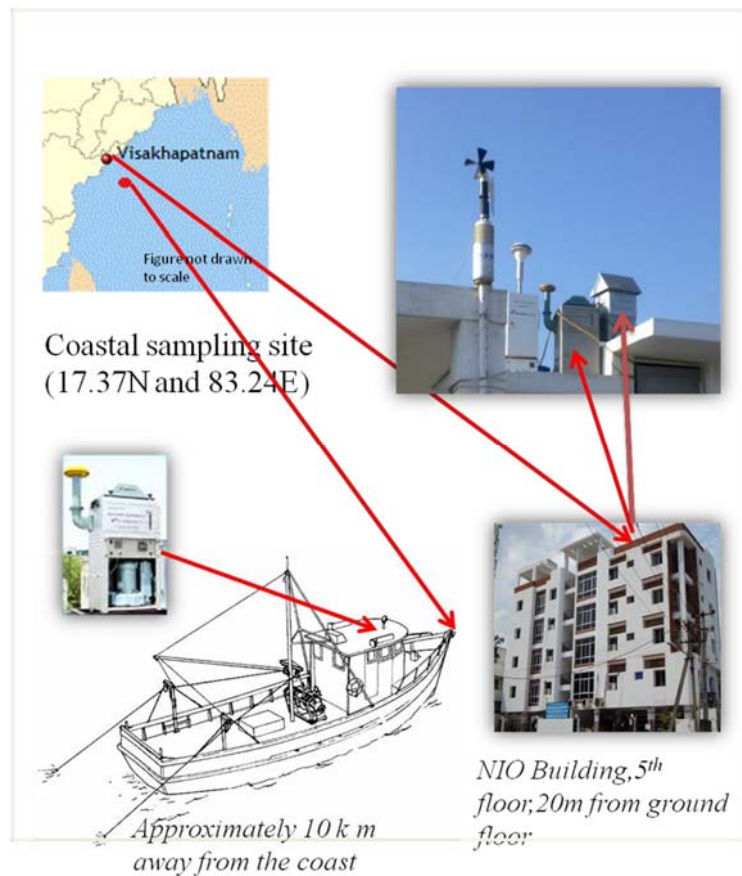


Figure 1. Map showing sampling sites in Visakhapatnam city and the adjacent coastal waters off the eastern coast of India. Locations of high volume air sampler (APM 430; *Envirotech*) used for bulk aerosol sampling in the city and that of water sampling and the Respirable Dust Sampler (APM 460BL; *Envirotech*) in the boat in the coastal Bay of Bengal are shown.

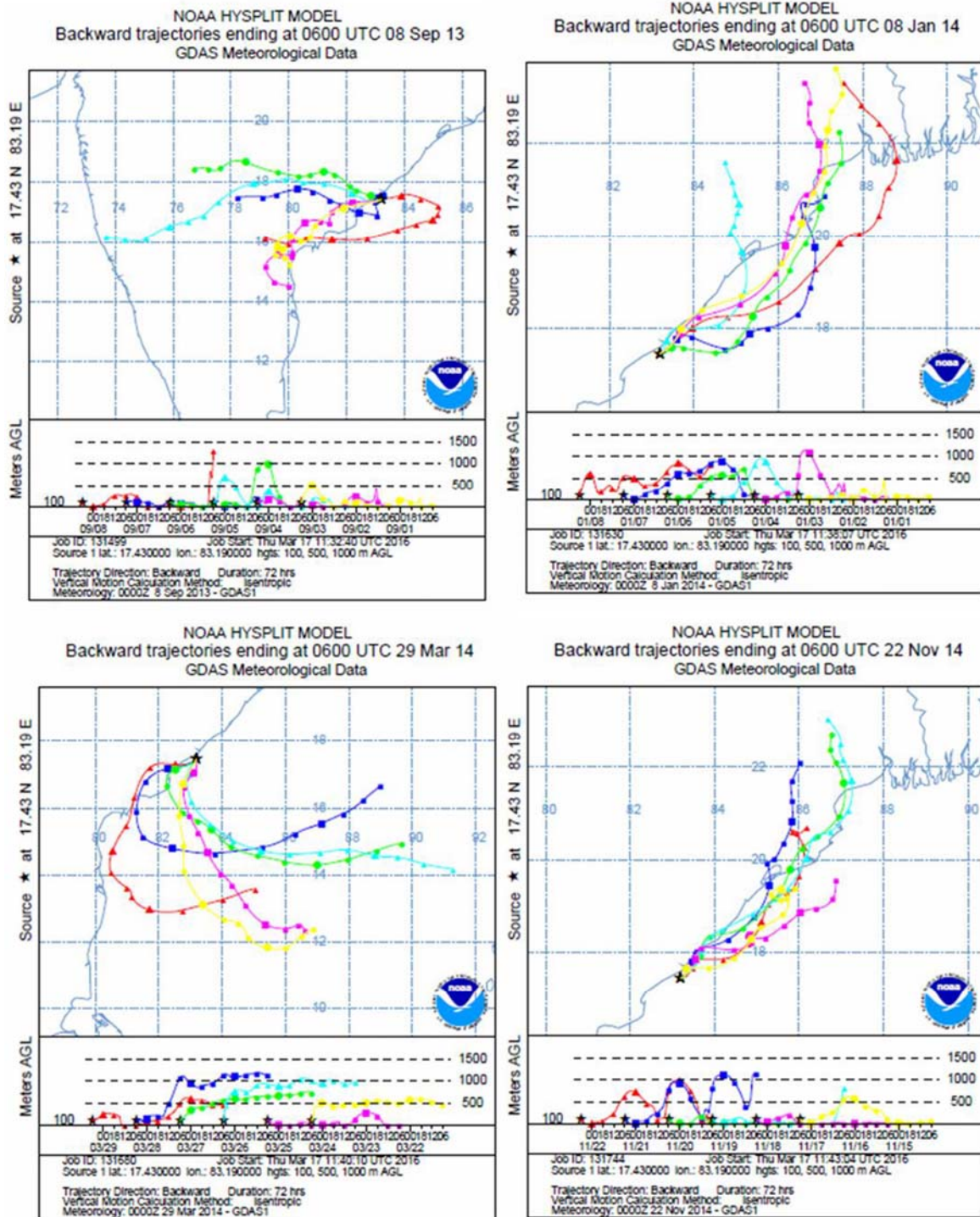


Figure 2. The different colored lines indicate the AMBTs arriving at the sampling location each day during each sampling period.

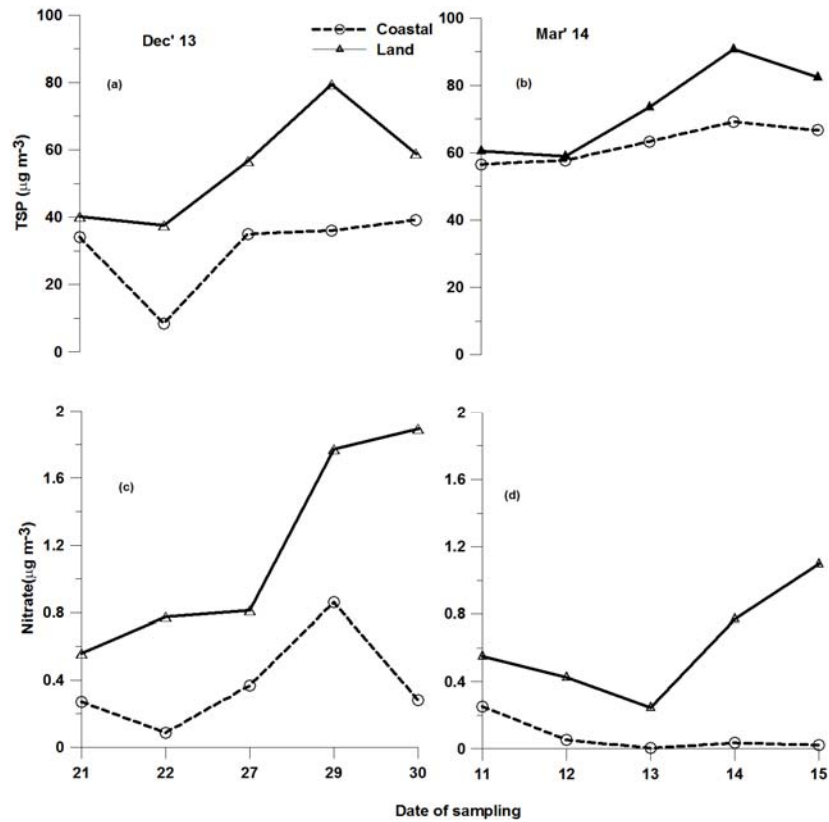


Figure 3: Concentrations of TSP (a, b) and nitrate in aerosols (c, d) during December (a, c) and March (b, d), respectively, over land and coastal regions.

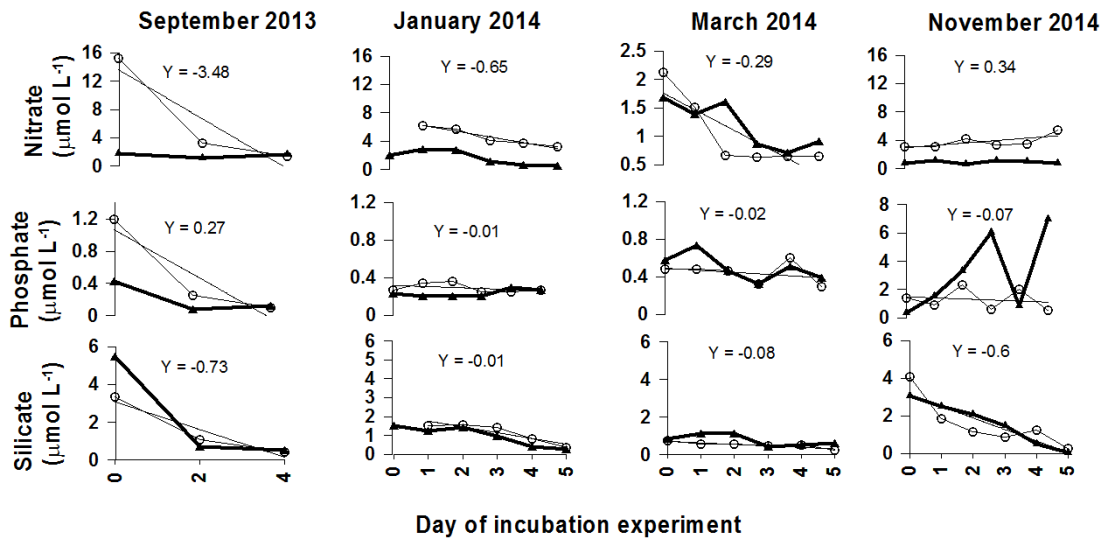


Figure 4: Variations in the concentrations of nutrients in the water during the incubation experiments after spiking with the dust collected during each season. Thick lines with triangles show results of controls (no aerosol additions) while lines with circles are those of the spiked microcosms.

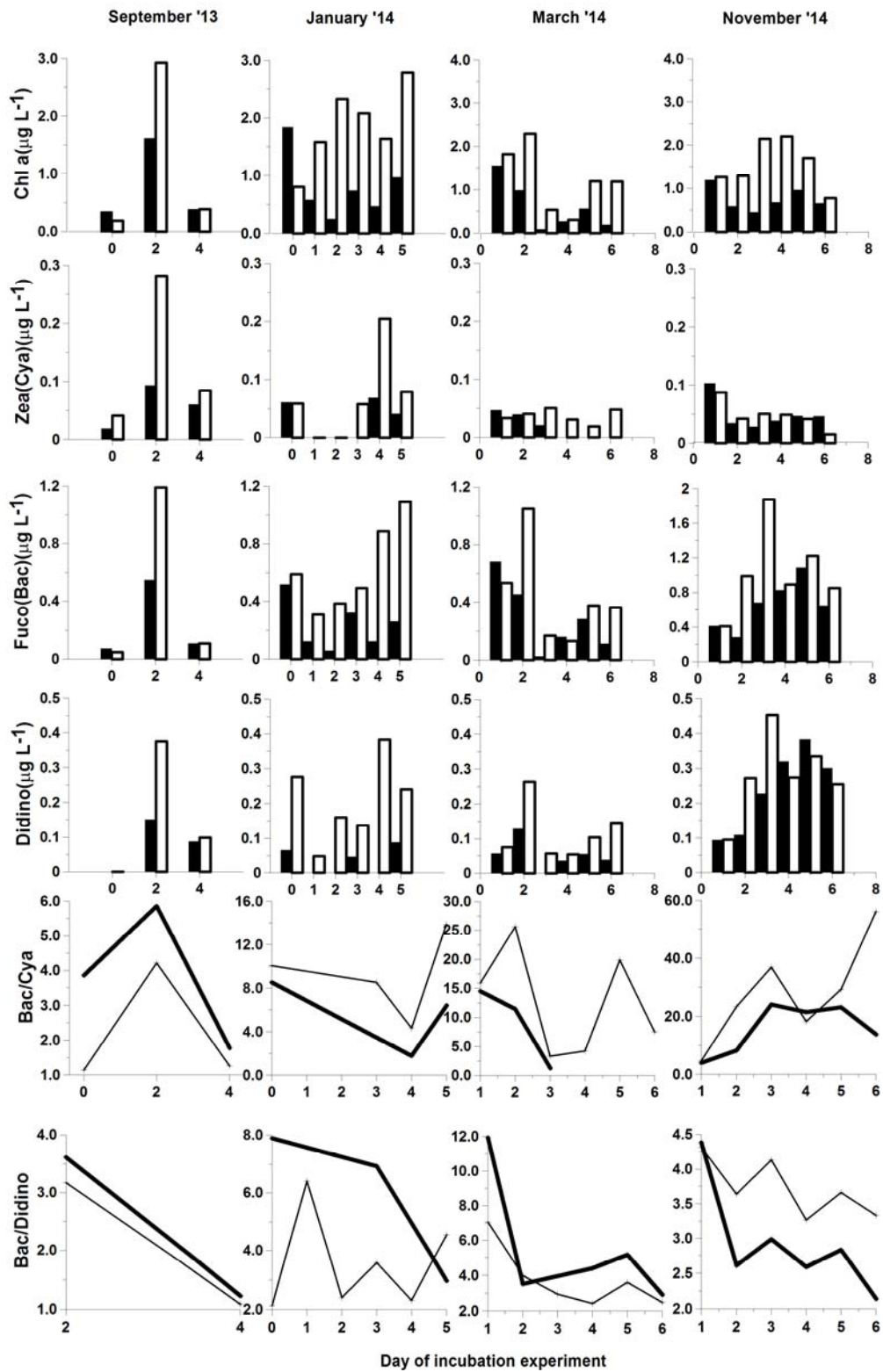


Figure 5: Variations in the concentrations of pigments in water observed during the incubation experiments after spiking with the dust collected for each season. Filled bars/ thick line show results from control experiments while hollow bars/ thin lines show data from the spiked microcosms.