# The Influence of Atmospheric Deposition of Pollutant Elements on Cross – Domain Causal Relationships at Three Tropical Freshwater Lakes of India

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**Abstract:** This long - term study, conducted at three fresh water lakes of India, was designed to investigate (i) atmospheric deposition of major nutrient elements, (ii) microbial biomass ($C_{mic}$) and activity at land – water interface and their relation to lake ecosystem functioning as influenced by catchment inputs and, (iii) the modifying influence of atmospheric deposition on trans – surface causal relationships.

The results showed significant between site differences ($p < 0.001$) in the atmospheric deposition of $NO_3^-$, $NH_4^+$, $PO_4^{3-}$, $SO_4^{2-}$, $Ca^{2+}$ and $Mg^{2+}$ with consistently rising input across time. The interface of lake Baghdara, characterized by woodland catchment, was found to be rich in phenolics and supported low $C_{mic}$ and activity. Alkaline phosphatase in humus declined by 13.60 to 25.30 % overtime but $C_{mic}$, SIR and FDAase at land – water interface increased significantly in response to atmospheric deposition. Nutrient deposition, although increased primary productivity, had significant ($p < 0.001$) modifying influence on $C_{mic}$ and lake productivity relationships with a time – lag of 3 years. The study demonstrated that, if the present trends of atmospheric deposition of pollutant elements are continued, it will modify the microbial processes at land – water interface and productivity of lakes and will alter the cross – domain causal relationships of fresh water tropical lakes in long – run.

**Keywords :** Atmospheric deposition, Lake, Microbial Biomass, Productivity

## 1. Introduction

Wide acceptance of the role of nutrients in water quality deterioration has led the control of external nutrient loading as one of the most widely used approach in lake ecosystem restoration. Since the nutrient enrichment of surface water bodies has been considered as a local phenomenon around point sources and urban areas, minimizing the surface – nutrient inputs has been used as an effective tool to combat eutrophication. However, recent studies have indicated that the solute chemistry of surface waters depends on chemical constituents imported through weathering, surface discharge and run – off as well as on pollutant aerosols added into the system through atmospheric deposition[1,2,3]. More importantly, during recent years, anthropogenic activities have dramatically increased the atmospheric deposition of pollutant aerosols in many parts of the world including India[3, 4, 5]

Air – driven deposition can contaminate surface water bodies even those situated away from the source of emission. This may lead restoration strategies, based on reduction of surface nutrient loading, to a failure. The impact of long range atmospheric deposition of pollutant aerosols on terrestrial environment is well documented, impact of air – driven pollutant aerosols on lentic systems have received attention only recently[2,3,6]. Scientific investigations during last two decades have indicated that the water bodies, even those situated away from direct human interference, are receiving increasingly high amount of pollutant aerosols through atmospheric deposition. For instance, even the high elevation lakes of Sierra, Nevada have evidenced nutrient enrichment through atmospheric sources[1]. Studies conducted in our laboratory have also indicated enrichment of surface water bodies with air – borne nutrients[6] and / or toxic metals[7] depending upon the emitting sources. Atmospheric deposition alters ecosystem attributes either directly through nutrient supply or indirectly through catchment modifications or altered interface interactions[3]. For instance, air driven nutrients delivered directly onto to the water surface coupled with light and temperature optima promote proliferation of phytoplankton leading the system towards eutrophy[2]. Nutrient poor ecosystems with historically low rates of mineralization are more vulnerable to changes from such nutrient inputs.

Although the evidences are scanty, atmospheric nutrient inputs may alter microbial biomass and mineralization of nutrients at land – water interface. Microbial biomass, the principal component of decomposer system regulating nutrient availability to other components of the ecosystem, is regulated by climate across broad ecosystem types but at finer scale it depends, to a large extent, on litter quality[8], hydrology and anthropogenic perturbations[9]. The land – water interface provides suitable habitat for microbial colonization. In an woodland lake, we observed that the auto – and allochthonous organic matter were the major determinant of microbial biomass and activity at land – water interface which, in turn, regulate productivity of the lake[10]. Continuous long – term atmospheric deposition of pollutant aerosols could alter nutrient mineralization at land – water interface causing long term impacts on other site of the surface interface. However, there is a dearth of studies explicitly addressing the potential synergisms between land – water interface, terrestrial catchment and atmospheric nutrient inputs, especially for tropical lakes of India [3]. The present study was an effort to investigate the influence of long – term atmospheric deposition of nutrient elements on cross – domain causal relationships at three fresh water lakes of India having varied catchment features.

## 2. Material and methods

The study was conducted during 1999 – 2005 at three lake sites, Fatehsagar ($24^{0}35^{/}$ N and $73^{0}37^{/}$ E); Pichhola ($24^{0}34^{/}$ N and $73^{0}37^{/}$ E) and Baghdara ($24^{0}31^{/}$ N and $73^{0}50^{/}$ E) located in Udaipur district of southern Rajasthan, India (Tab. 1). The climate is tropical with 760 mm annual rain fall. The soil is alfisol with pH above neutral. The details of climate and catchment characteristics are described in Pandey and Pandey[11].

Atmospheric deposition monitoring was carried out using bulk samplers made up of a 5L high density polyethylene bottle connected to a Teflon funnel and devised with PVC needles on top to avoid bird nesting. Analyses of phosphate, nitrate, ammonium and sulphate in bulk samples were executed spectrophotometrically and calcium and magnesium in atmospheric samples were analyzed by a Perkin – Elmer (Model 2130) atomic absorption spectrophotometer.

Humus samples collected from land – water interfaces were analyzed for microbial biomass and activity. Microbial biomass – C in humus was determined using chloroform fumigation extraction procedure[12]. An extraction efficiency coefficient of 0.38 was used to convert soluble C into microbial biomass – C ($C_{mic}$). Basal respiration was measured following Wardle[13]. The substrate induced respiration (SIR) was determined by amending 20 g of sample (moisture content, 55 %) with 120 mg of glucose at the beginning of the incubation[14]. $CO_2$ inhibition by streptomycin sulphate and actidione relative to SIR was used as relative measure of active bacterial and fungal biomass, respectively for computing bacterial to fungal ratio[13]. Microbial metabolic quotient was calculated as the ratio of BR to SIR. Alkaline phosphatase activity was determined as

**Tab.1 General characteristics of the lakes**

| Lake | Size ($km^2$) | Depth (m) | Characteristics |
|---|---|---|---|
| Fatehsagar | 4.0 | 13.4 | Situated at NW margin of the city; cemented pavement, receives atmospheric deposition from urban sources and run – off from rocky terrain characterized by scattered growth of woody perennials. |
| Pichhola | 7.0 | 10.5 | Situated at SW margin of the city, water chemistry is regulated by atmospheric deposition, catchment flushing through agricultural and urban run – off. |
| Baghdara | 1.8 | 8.5 | Situated about 20 km SE of the city in the midst of a tropical forest. Receives nutrients from phosphate fertilizer factories and woodland catchment. |

described in Tabatabai and Bremner[15]. For fluorescein diacetate hydrolytic activity (FDAase), the humus suspension was filtered and incubated with fluorescein diacetate at $24^0$ C. Fluorescein thus formed was determined at 490 nm as described in Schnurer and Rosswall[16].

Humus samples were further analyzed for total – N and total phenolics. Total – N in humus and bottom sediment was measured through Kjeldahl analysis and total phenolics using Folin – Ciocalteau reagent method[17]. Organic – C and total – P in bottom sediment was measured as described by Jackson[18]. Lake primary productivity was measured using Light and Dark bottle method[19], and chlorophyll as described by Maiti [20]. For phytoplankton density, the method described in Pandey and Pandey[11] was followed.

Significant effects were assessed using analysis of variance (ANOVA). Temporal variability were expressed in terms of coefficinent of variation (CV) and correlation analyses were used as a test for linearity.

## 3. Results and discussion

The atmosphere – water transfer may play a crucial role in regulating water chemistry directly and / or indirectly through catchment modifications. Role of such cross – domain relationships becomes more important in areas characterized by potential anthropogenic perturbations where pollutant aerosols become the abundant constituent of the atmosphere. For instance, nitrate concentrations in Norweian lakes doubled over less than a decade due to air driven deposition [21]. For establishing significant ecosystem level trends and causal relationships, long – term records are necessary because individual portions of such records considered separately may indicate increases or decreases or no change in a chosen variable. In this long – term study, although air – borne nutrient input varied significantly ($p < 0.001$) with time and site (Tab. 2), the region witnessed sizably high atmospheric deposition of nutrient ions even higher than those observed in some other industrial regions of India[22]. For instance, lake Fatehsagar, receives 29.87 Kg $ha^{-1}$ $y^{-1}$ of N and Baghdara receives 1.96 Kg $ha^{-1}$ $y^{-1}$ of P through aerial route (Tab. 2).

Microbial biomass ($C_{mic}$) and activity at land – water interface increased significantly overtime and the increases were maximum for Baghdara site (Tab. 3). The humus enzyme FDAase indicated a similar trend but alkaline phosphatase declined significantly overtime (Tab. 4). Lake primary productivity (Tab. 5) and the levels of C, N and P in sediment (Fig. 1) also showed synchrony with nutrient deposition. For initial years, low $C_{mic}$ and activity at Baghdara site (Tab. 2) could be due to high phenolics (Fig. 2) and low nutrient content[23] of tree foliage. On the other hand, herbaceous residues (high N, low phenolics) at Fatehsagar could support relatively high $C_{mic}$ and activity[24]. Effects of herbaceous residues mixed with agricultural inputs were also evident for Pichhola site. Time –lag correlation analysis indicated that the changes in $C_{mic}$ and activity were well associated with the deposition of N and P with a time lag of 2 to 3 years (Fig. 3). The time – lag relationship was more evident for Baghdara site characterized by woodland catchment, where after a lag of three years, $C_{mic}$ and activities increased consistently overtime indicating the influence of atmospheric deposition. Substrate induced respiration (SIR), a relative measure of active microbial biomass, and metabolic quotient ($qCO_2$) also showed significant effects ($p < 0.001$) of atmospheric deposition (Tab. 3). Furthermore, larger proportion of bacterial biomass at Fatehsagar and Pichhola could increase $qCO_2$ for these sites. Enhanced bacterial to fungal ratio over time at Baghdara site indicate improved substrate quality due to atmospheric deposition of nutrients[23]. The declining values of correlation coefficient between $C_{mic}$ and phenolics across time (data not shown) indicate that the deposition of nutrients could lower the negative influence of phenolics on $C_{mic}$ probably through improved quality of humus at land – water interface.

Low $C_{mic}$ and activity coinciding with high concentrations of phenolics suggest that the latter could reduce microbial biomass available to decompose substrate[25]. Presence of lignin on leaves and twigs of woody perennials further delay microbial colonization[8]. The asynchrony between $C_{mic}$ and phenolics slowly altered after a time – lag of 2 – 3 years due probably to continued deposition of nutrient elements. Substrates that resist decomposition stabilize microbial biomass dynamics[24}. This effect although appeared during initial years, especially for Baghdara, was found to have altered after the lag period, as evidenced through increased temporal variability over time.

**Tab. 2 Atmospheric deposition of nutrient ions (Kg ha$^{-1}$ y$^{-1}$) at three lake sites. Values are mean (n = 36) ± 1SE. Data comparison are made between first (1999) and last (2005) year of study**

| Year / Lake | $PO_4^{3-}$ | $NO_3^{-}$ | $NH_4^{+}$ | $SO_4^{2-}$ | $Ca^{2+}$ | $Mg^{2+}$ |
|---|---|---|---|---|---|---|
| 1999 | | | | | | |
| Fatehsagar | 0.99 | 20.05 | 6.20 | 29.18 | 21.76 | 7.12 |
| | ± 0.08 | ± 1.55 | ±0.58 | ±2.66 | ± 1.84 | 0.67 |
| Pichhola | 0.94 | 19.25 | 5.78 | 28.00 | 20.30 | 6.87 |
| | ± 0.07 | ± 1.32 | 0.51 | ± 2.46 | ± 1.70 | ± 0.61 |
| Baghdara | 1.31 | 6.80 | 1.96 | 6.05 | 6.35 | 1.64 |
| | ± 0.27 | ± 0.44 | ± 0.13 | ± 0.46 | ± 0.39 | ± 0.12 |
| 2005 | | | | | | |
| Fatehsagar | 1.41 | 29.87 | 12.15 | 34.00 | 31.50 | 13.10 |
| | ± 0.16 | ± 2.75 | ± 1.30 | ± 3.26 | ± 2.67 | ± 1.05 |
| Pichhola | 1.31 | 28.00 | 11.65 | 31.57 | 29.00 | 11.87 |
| | ± 0.12 | ± 2.66 | ± 1.31 | ± 3.05 | ± 2.46 | ± 1.00 |
| Baghdara | 1.96 | 10.72 | 3.05 | 8.20 | 8.70 | 2.46 |
| | ± 0.15 | ± 0.68 | ± 0.23 | ± 0.70 | ± 0.69 | ± 0.21 |
| ANOVA | | | | | | |
| Time (t) | ** | ** | ** | * | * | * |
| Site (s) | ** | ** | ** | ** | * | * |
| t×s | ** | ** | * | * | ns | ns |

Significant at $^{*}$ $p < 0.01$; $^{**}$ $p < 0.001$; ns : not significant

**Tab. 3 Microbial biomass and activity in humus collected from land – water interface of three lakes. Data are for 2005 with values in parentheses representing percent increase (+) or decrease (-) over mean (n = 12) values of 1999**

| Lake | $C_{mic}$ (µg C g$^{-1}$) | BR (µg CO$_2$ – C g$^{-1}$ h$^{-1}$) | SIR (µg CO$_2$ – C g$^{-1}$ h$^{-1}$) | Bacterial/ Fungal ratio | Metabolic quotient[#] |
|---|---|---|---|---|---|
| Fatehsagar | | | | | |
| Winter | 1685.00(+ 42) | 3.11(+ 46) | 9.28(+ 43) | 31.05(+ 18) | 0.318(+ 8) |
| Summer | 2236.00(+ 49) | 4.50(+ 49) | 15.00(+ 48) | 40.80(+ 19) | 0.311(+6) |
| Rainy | 1865.00(+ 45) | 3.36(+ 43) | 9.98(+ 43) | 32.70(+15) | 0.328(+6) |
| Pichhola | | | | | |
| Winter | 1750(+28) | 3.30(+30) | 10.56(+28) | 30.5(+16) | 0.335(+12) |
| Summer | 2360(+34) | 4.90(+35) | 16.68(+32) | 40.6(+17) | 0.324(+7) |
| Rainy | 2040(+32) | 3.62(+ 28) | 11.25(+29) | 31.9(+13) | 0.340(0) |
| Baghdara | | | | | |
| Winter | 797.98 (+59) | 1.52 (+64) | 7.10 (+57) | 18.2 (+26) | 0.214 (+5) |
| Summer | 996.50 (+61) | 2.04 (+59) | 10.04 (+63) | 24.4 (+31) | 0.203 (-3) |
| Rainy | 792.50 (+48) | 1.58 (+52) | 7.30 (+52) | 18.9 (+24) | 0.216 (0) |
| ANOVA | | | | | |
| Time (t) | ** | ** | ** | * | * |
| Site (s) | ** | ** | ** | * | * |
| t×s | ** | * | ** | ns | ns |

[#] Ratio of basal respiration to SIR

Significant at * $p < 0.01$; ** $p < 0.001$; ns : not significant

**Tab. 4 Enzyme activity in humus at land – water interface of three lakes. Data are for 2005 with values in parentheses representing percent increase (+) or decrease (-) over mean (n = 12) values of 1999**

| Lake | FDAase | Alkaline phosphatase (AP) | |
|---|---|---|---|
| | (µg fluorescein g$^{-1}$ soil h$^{-1}$) | ((µg p – NP g$^{-1}$ humus h$^{-1}$) | (n mol p – NP mg$^{-1}$C$_{mic}$ h$^{-1}$)$^{\dagger}$ |
| Fatehsagar | | | |
| Winter | 119.8 (+23.0) | 342.6 (-16.4) | 304.5 (-17.6) |
| Summer | 142.7 (+27.7) | 379.5 (-17.8) | 210.3 (-19.8) |
| Rainy | 124.4 (+ 24.6) | 358.0 (-15.3) | 378.8 (-16.2) |
| Pichhola | | | |
| Winter | 124.6 (+18.0) | 243.6 (-14.0) | 123.6(-15.2) |
| Summer | 159.1 (+22.8) | 291.5 (-15.4) | 98.7 (-16.7) |
| Rainy | 128.0 (+21.0) | 254.7 (-13.1) | 161.2 (-13.6) |
| Baghdara | | | |
| Winter | 102.8 (+32.0) | 196.2 (-19.0) | 230.4 (-21.5) |
| Summer | 138.2 (+37.8) | 242.6 (-21.8) | 189.7 (-23.6) |
| Rainy | 110.6 (+33.7) | 214.3 (-18.0) | 300.3 (-18.4) |
| ANOVA | | | |
| Time (t) | ** | ** | ** |
| Site (s) | ** | ** | ** |
| t × s | ** | * | ** |

Significant at $^{*}$ p < 0.01; $^{**}$ p < 0.001

# Enzyme activity expressed in terms of per unit C$_{mic}$

**Tab. 5 Productivity variables measured at three lakes. Data are for 2005 with values in parentheses representing per cent increase over mean (n = 12) values of 1999**

| Lake | Chlorophyll **a** (mg m$^{-3}$) | Gross primary productivity (g C m$^{-2}$ d$^{-1}$) | Phytoplankton density (cells ml$^{-1}$) |
|---|---|---|---|
| Fatehsagar | | | |
| Winter | 31.09 (46) | 2.46 (48) | 2210 (47) |
| Summer | 36.04 (49) | 3.05 (49) | 2580 (48) |
| Rainy | 27.12 (45) | 1.89 (46) | 1620 (49) |
| Pichhola | | | |
| Winter | 37.86 (34) | 2.82 (36) | 2430 (36) |
| Summer | 42.75 (33) | 3.57 (34) | 2870 (33) |
| Rainy | 33.05 (36) | 2.21 (36) | 1840 (37) |
| Baghdara | | | |
| Winter | 27.68 (57) | 1.84 (54) | 1965 (56) |
| Summer | 34.48 (62) | 2.56 (50) | 2470 (51) |
| Rainy | 24.33 (66) | 1.51 (63) | 1390 (58) |
| ANOVA | | | |
| Time (t) | ** | ** | ** |
| Site (s) | ** | ** | ** |
| t × s | * | * | ** |

Significant at $^{*}$ p < 0.01; $^{**}$ p < 0.001

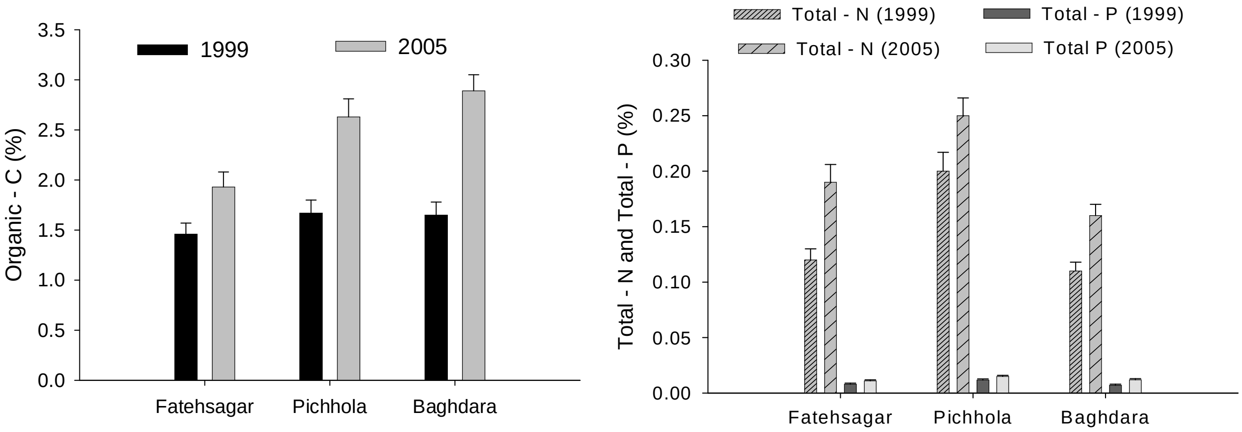

**Fig.1 Chemical characteristics of bottom sediments collected from 10 m reach of three lakes. Between year differences were significant at $p < 0.001$ (Analysis performed using paired t – test)**

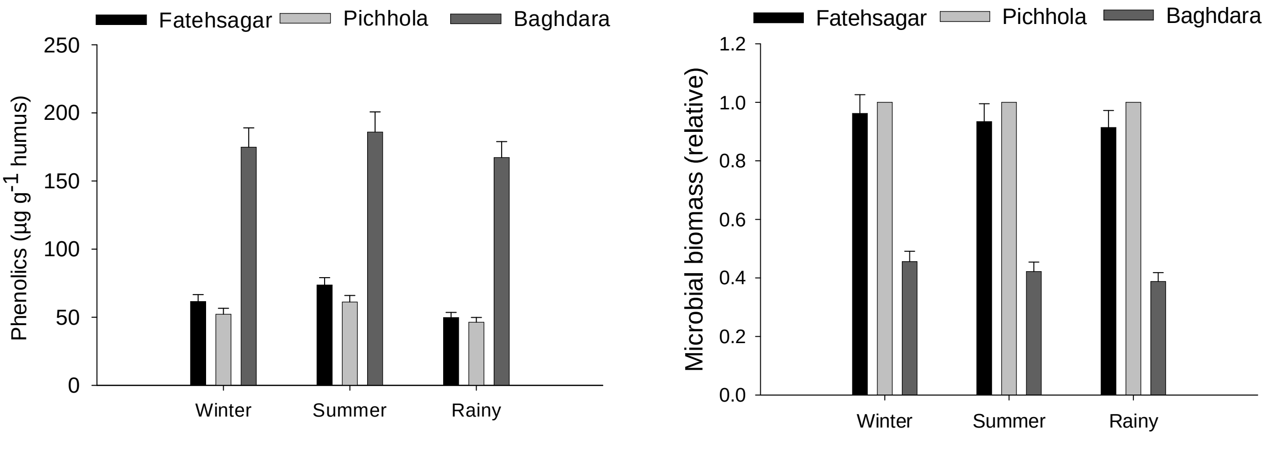

**Fig. 2 Microbial biomass (relative) and phenolics in humus at land – water interface of three lakes. Data (n = 9) are for 2005 (mean ± 1SE). Differences between sites and seasons were significant at $p < 0.01$ and $p < 0.005$ respectively**

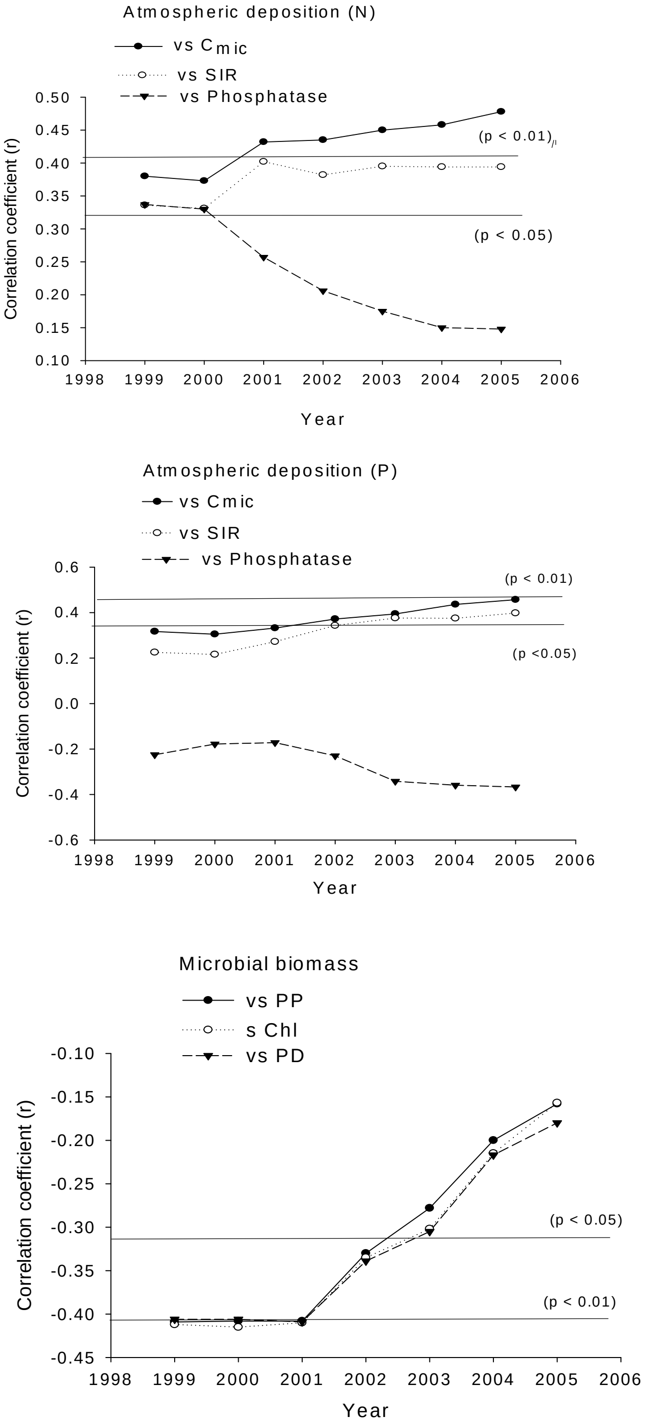

**Fig. 3 Time lag correlation coefficients (n = 36) between cross – domain variables. Values outside the lines marked with P are significantly different at that level of probability.**

Among the humus enzymes, alkaline phosphatase (AP) appeared particularly responsive to nutrient input and declined significantly ($p < 0.001$) with increasing P input across time (Tab. 4). Increased temporal variability in AP over time indicates destabilizing effect of nutrient input. The decreases in AP activity at lake sites were due probably to the atmospherically added P[26]. The AP activity per unit of humus remained lowest at Baghdara (196.2 to 242.6 µg p NP $g^{-1}$ humus $h^{-1}$), but per unit of $C_{mic}$ remained lowest at Pichhola (98.7 to 161.2 n mol p – NP $mg^{-1}$ $C_{mic}$ $h^{-1}$), resulting in high $C_{mic}$ /AP ratio for the latter. High $C_{mic}$ / AP ratio at Pichhola could be due to additional P added through agricultural run – off apart from aerial input. This ratio is an useful measure of phosphate demand by soil organisms and strongly influenced by the quantity of active microorganisms [9]. The ratios observed in the present study are comparable to those reported for pastures and grasslands[9] receiving variable levels of nutrients.

Primary productivity although increased significantly overtime (Tab. 5), the period of low $C_{mic}$ in humus coincided with high GPP, chlorophyll **a** and phytoplankton density in lakes. The phase of declining $C_{mic}$ at land – water interface could increase nutrient release in lake water and consequently the productivity. This relationship however, was slowly altered across the year (after lag period) as the influence of deposition predominated the scene. Time – lag correlation analysis showed that the changes in both, productivity and $C_{mic}$ were clearly associated with air borne nutrients (Fig. 3) indicating that the nutrient input through aerial catchment can affects ecosystem attributes of freshwater lakes. Temporal variability in productivity did not show detectable time – lags due probably to the effect of air driven nutrients – delivered to upper water surface, where light availability promotes rapid uptake and proliferation of phytoplankton[1]. This effect was invariably evident for all the three lakes and was further reflected in terms of increased organic – C and nutrients in bottom sediment. Nutrients, especially P accumulated in sediments release and accelerate productivity during bottom anoxia[10]. Thus, atmospheric – nutrient input could become an important determinant accelerating eutrophy, especially for water bodies situated away from direct human interference.

The data presented in this study indicated significant spatial and temporal variations in the atmospheric deposition of nutrients. The catchment vegetation had important effects on microbial processes at land – water interface which, in turn, affected the lake productivity. Air – driven nutrients significantly raised phytoplankton density, chlorophyll **a** and GPP and altered the microbial processes at land – water interface and the lake productivity relationships with a time – lag of 3 years. The study demonstrated that, if the present trends of atmospheric deposition of pollutant elements are continued, it will significantly modify microbial dynamics at land – water interface and productivity of lakes and will alter the cross – domain causal relationships of fresh water tropical lakes in long – run. The study has relevance in formulating strategies for management and conservation of fresh water tropical lakes.

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