

Contribution of point sources and non-point sources to nutrient and carbon loads and their influence on the trophic status of the Ganga River at Varanasi, India

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Abstract To determine the possible contributions of point and non-point sources to carbon and nutrient loading in the Ganga River, we analyzed N, P, and organic carbon (OC) in the atmospheric deposits, surface runoff, and in the river along a 37-km stretch from 2013 to 2015. We also assessed the trophic status of the river as influenced by such sources of nutrient input. Although the river N, P, and productivity showed a declining trend with increasing discharge, runoff DOC and dissolved reactive phosphorus (DRP) increased by 88.05 and 122.7% between the Adpr and Rjht sites, indicating contributions from atmospheric deposition (AD) coupled with land use where agriculture appeared to be the major contributor. Point source input led to increased river concentrations of NO_3^- , NH_4^+ , DRP, and DOC by 10.5, 115.9, 115.2, and 67.3%, respectively. Increases in N, P, and productivity along the gradient were significantly negatively correlated with river discharge ($p < 0.001$), while river DOC and dissolved silica showed positive relationships. The results revealed large differences in point and non-point sources of carbon and nutrient input into the Ganga River, although these variations were strongly influenced by the seasonality in surface runoff and river discharge. Despite these

variations, N and P concentrations were sufficient to enhance phytoplankton growth along the study stretch. Allochthonous input together with enhanced autotrophy would accelerate heterotrophic growth, degrading the river more rapidly in the near future. This study suggests the need for large-scale inter-regional time series data on the point and non-point source partitioning and associated food web dynamics of this major river system.

Keywords Atmospheric deposition · DOC · Ganga River · Surface runoff · Point source · Trophic status

Introduction

Nitrogen (N) and phosphorus (P) are among the most important nutrients that drive the eutrophication of rivers and streams. Addressing the spatial connectivity of source inputs of these nutrients to rivers is important to ensure the maintenance of patterns and processes essential to sustain the system (Merseburger et al. 2011). In seasonally dry tropical regions, spatial connectivity is constrained, together with anthropogenic drivers, by marked temporal changes in the wet-dry seasons (Pandey et al. 2014). Seasonal precipitation and river flow, for instance, are the major factors affecting river nutrient concentrations (Chen and Hong 2011; Pandey et al. 2014). Nutrient exports from watersheds are highly variable and most often depend on the nature of human modifications; regional studies will thus become more important than global-scale analyses (Alvarez-Cobelas et al. 2008). Worldwide, approximately one half the

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population live in cities, increasing from 29% in 1950 to 47% in 2000 and expected to reach 81% by 2030 (Simms 2008), with massive impacts on river ecosystems (Tong and Chen 2002; Fu et al. 2009; Ternus et al. 2011; Kebede et al. 2014). Furthermore, land use change has emerged as one of the most important drivers modifying the proximal factors controlling stream metabolism. Nutrient input from agricultural and urban sources in the form of non-point sources (NPSs) such as fertilizer application and fossil fuels or point sources (PSs) such as sewage and industrial release is continuously degrading the water quality of rivers and streams (Bernot et al. 2010). Some of the rivers and streams receive 10 to 90% of their average annual flow from wastewater treatment plants (Mouri et al. 2011). However, in a previous study of 35 rivers, Caraco and Cole (1999) observed that the average watershed load of nitrates was over 20-fold higher than the point source loading. Approximately 3–20% of the P (Caraco 1995) and 18% of the N (Carpenter et al. 1998) applied to croplands are exported to surface waters. The global anthropogenic N load to freshwater systems from croplands, including both leaching and runoff, is estimated at 24.4 million t N year⁻¹, and from point sources, it is approximately 8.2 million t N year⁻¹ (Mekonnen and Hoekstra 2015).

Although they can be controlled, problems associated with point sources persist as a major issue for developing countries, including India, due to poor management efforts. NPS-driven changes are more widespread and difficult to control due to their extensive and uncertain nature, complexity and diversity of landscape features, variability over time, and seasonal and climate-dependent regulation (Liu and Diamond 2005; Bowes et al. 2005; Mouri et al. 2011). The amount of source input depends on land use heterogeneity; agricultural watersheds, for instance, contribute much higher inputs through surface runoff and leaching than forested watersheds, and even higher than urban watersheds (Zhu et al. 2008). The latter, however, are typically more complex in terms of the numbers and types of sources and pollutants (Lundy and Wade 2013) and are the third most important cause of lake deterioration in the USA (US EPA 1990). Earlier studies have shown that in more than 90% of the major watersheds of the USA, over one half of the N was added through NPS (Puckett 1994). In northeastern US watersheds, approximately 31% of the anthropogenic N is being added through atmospheric deposition (AD) and approximately 24% by food and

feed sources (Zhu et al. 2008). Approximately 90% of NH_x in the atmosphere comes from agricultural sources, with major contributions from animal wastes followed by the volatilization of fertilizers; due to its short atmospheric life, it may be deposited in the same region (Boyer et al. 2002). Excessive inputs of N and P through sewage, AD, and surface runoff, especially in human-disturbed areas, often lead to Si limitation (Pandey et al. 2016b). Studies in Victoria Harbor have shown that sewage inputs with high N and P may contribute to Si limitation even in up to 94% of the cases (Xu et al. 2008). In addition, agricultural activities, an important non-point source contributing to N and P, reduce Si release to surface waters (Beman et al. 2005; Hagen et al. 2010). Such sources also contribute a high input of C to these systems, resulting in a shift in the proportion of autotrophs and heterotrophs (Dodds and Cole 2007). The availability of two distinct sources of C (autochthonous and allochthonous) allows the uncoupling of primary and secondary production with implications for food web dynamics (Van den Meersche et al. 2009; Lau et al. 2009). A number of studies assessed the contributions of point and non-point sources of pollution in temperate (Jeunesse et al. 2002; Pieterse et al. 2003; Bowes et al. 2005, 2008; Merseburger et al. 2005; Munafo et al. 2005) and tropical rivers (Chen et al. 2013; Wua and Chen 2013). In India, such studies are very scarce (Jain et al. 1998), and for the Ganga River, no data on this issue are available thus far, despite it being the second most populated river basin in the world.

The Ganga River basin, an economic and ecological support system of India, has surface water and groundwater potential of approximately 525.02 and 170.0 km³, respectively (Misra 2011). India contributes 674 billion m³ year⁻¹ to the global gray water footprint (GWF) related to N loads accounting for only 5% of the global GWF. Interestingly, however, of the 674 billion m³ year⁻¹, the Ganges Basin alone accounts for 480 m³ year⁻¹ (over 71%) of the GWF, indicating its immense contribution to water pollution related to anthropogenic N (Mekonnen and Hoekstra 2015). Model studies show that the Ganges Basin is the second largest dissolved inorganic nitrogen (DIN) exporter in the world (Dumont et al. 2005). The Ganga River is experiencing massive changes in its water quality driven mainly by anthropogenic factors, including intensive agriculture, urbanization, and industrial release (Misra 2011; Pandey et al. 2014, 2016a). However, no

systematic data are available that evaluate the point and non-point sources of nutrient input and dispersal in this river system despite the fact that such information is important for implementing integrated river basin management strategies.

In view of consistently rising anthropogenic pressure and strong seasonality in hydrology, such annually resolved records would provide valuable insight into the spatiotemporal variability in carbon and nutrients reaching the river and the links therein with the river water quality and trophic status. The objectives of this study were to (1) analyze the contributions of point and non-point sources of carbon and nutrients to the Ganga River, (2) identify the downstream and trans-river trends in water quality as influenced by source inputs and river flow, and (3) examine the influence of these inputs on the trophic status of the river. Here, we present data on the percentage changes in N, P, and organic carbon (OC) inputs from a major point source and non-point sources, and the induced changes in the water quality, productivity, and trophic status of the Ganga River. We hypothesized that variable inputs of N and P from point and non-point sources coupled with river flow affect the distribution of nutrients with associated shifts in the trophic status of the Ganga River at Varanasi. To address this hypothesis, we tested the carbon, nutrients, and trophic status in relation to atmospheric deposition, surface runoff input, and a major point source along a 37-km stretch of the Ganga River upstream and downstream from the Varanasi urban core.

Materials and methods

Study area

This 3-year study (March 2013 to February 2016) was conducted at three sampling stations on the Ganga River covering approximately 37 km representing upstream and downstream locations from the Varanasi urban core (25° 18' N; 83° 1' E) in the middle of the eastern Indo-Gangetic plains 76.19 m above msl (Fig. 1a). The Ganga River basin (1,086,000 km²) is the fourth largest trans-boundary river basin in the world covering approximately 26.2% of the total geographical area of India. This largest river basin in India is bounded by the Himalayas in the north, the Arawali Range in the west, the Vindhyas in the south, and the Brahmaputra ridges in the east. The river originates in the western

Himalayas, flows southeast through the plains of north India and Bangladesh, and finally empties into the Bay of Bengal. A number of tributaries, including the Kali, Ramganga, Yamuna, Gomati, Ghaghara, Gandak, and Koshi rivers, join the river downstream. The climate of the basin is tropical monsoonal to humid subtropical. The year shows distinct seasonality: a humid monsoon season (July–October) with relative humidity reaching close to saturation, a cold winter (November–February) with minimum temperatures sometimes below 4 °C, and a hot dry summer (March–June) when temperatures sometimes exceed 46 °C. A southwest monsoon brings most of the rainfall with over 80% occurring during the monsoon season. The river is fed by the Himalayan snow from April to June and by rain-driven runoff from July to October with recurrent floods. The average monsoonal discharge is more than 10 times the average dry season discharge. The rainfall fluctuates between 780 mm in the upper part through 1040 mm in middle stretch, 1820 mm in the lower delta region, and reaches 2500 mm in the northeast. The soils of the region are highly fertile eutric cambisols and shallow luvisols.

The sampling stations differ in their land use, anthropogenic disturbances, and sources of nutrient input (Table 1). Each station represents three sites with three subsites at each for AD, surface runoff, and river water sampling. Three additional subsites, an urban side, mid-stream, and an offside, were selected for river water sampling. Samples of AD, surface runoff, and river water all were collected in triplicate. Samples for AD were collected biweekly, river water at monthly intervals, and surface runoff on an event basis. For point source input characterization, sampling was performed at the Assi drain outlet (zero distance) downstream up to the Raj Ghat site through Ravidas Ghat (Rvht), Assi Ghat (Asht), Bhadaini Ghat (Bhht) to Rajghat (Rjht), which are situated at approximately 100 m, 1.2 km, 1.7 km, and 7 km, respectively, from the drain outlet (Fig. 1b). The Assi drain, previously a rivulet, joins the Ganga River south of the Varanasi city boundary, now turned into a drain that adds industrial and domestic waste into the river.

Measurements

To establish cross-domain linkages, we collected data representing changes in all three domains. The chosen determinants were atmospheric deposition of nutrients, surface runoff chemistry, river water chemistry,

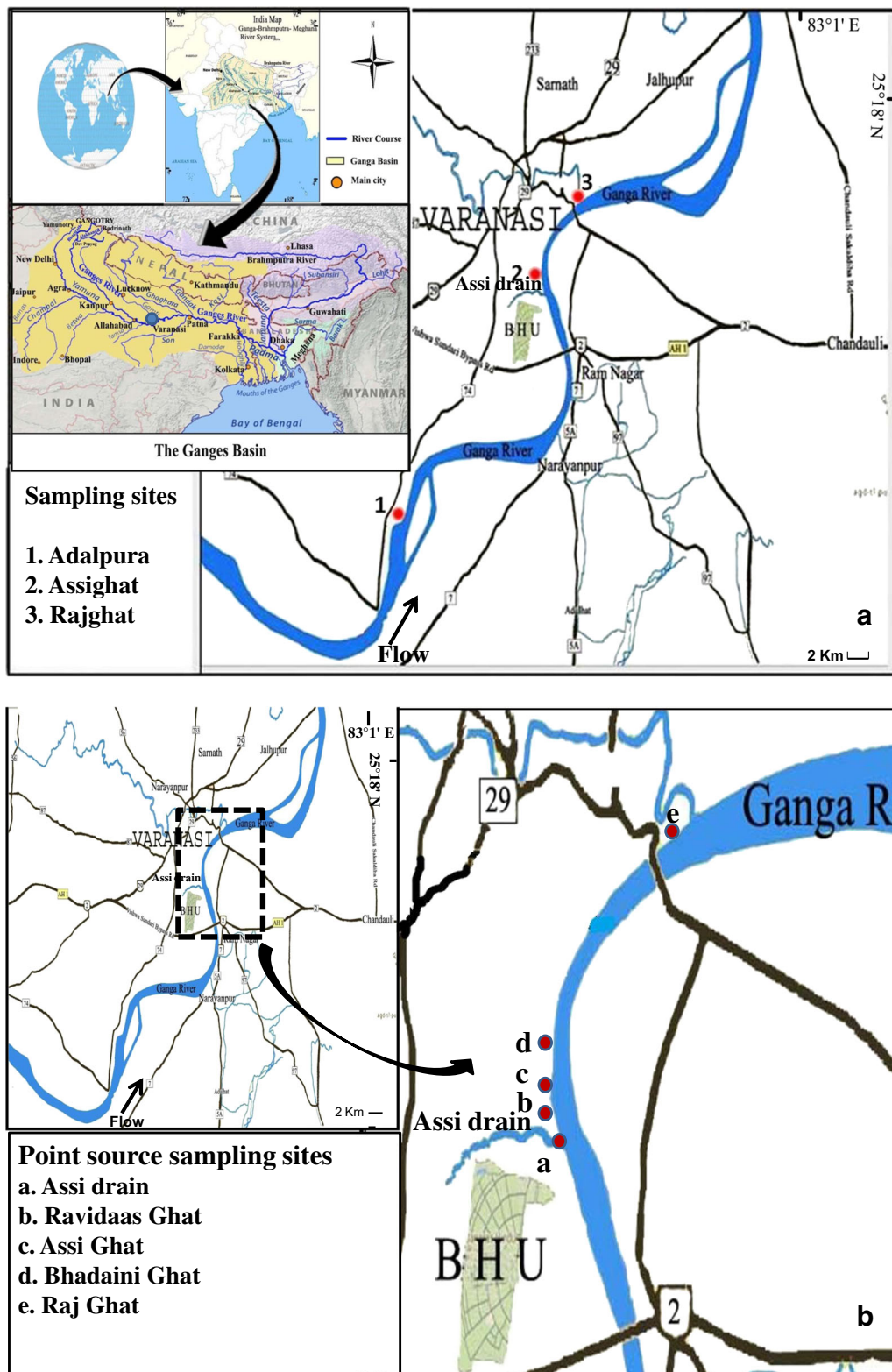


Fig. 1 Map showing the locations of **a** main sampling sites 1, 2, and 3 and point-to-downstream sampling sites **a–e**. **b** An expansion of downstream Assi drain area marked in the box of the map

Table 1 Description of sampling sites and sources of input

Site	Abbreviation	Subwatershed area (km ²)	Population (Census 2011)	River stretch up to next site (km)	Residential category	Major source
Adalpura	Adpr	84	~ 28,370	30	Rural	AD, runoff flux, land use
Assighat	Asht	72	~ 1,72,440	7	Urban	AD, runoff flux, Assi drain
Rajghat	Rjht	90	~ 2,39,500	–	Dense urban core	AD, runoff flux, sewage outlets, industrial outlets, bathing, and washing

productivity variables, and point source-linked downstream variations.

Atmospheric deposition

Atmospheric deposition samples were collected from all the subsites of each sampling station. Samples were collected in triplicates at biweekly intervals. Samples of atmospheric deposits were collected using a bulk sampler made up of a 5-L high-density polyethylene bottle connected to a Teflon funnel. The samplers, devised with PVC needles on the top to prevent bird nesting, were maintained at a height of 2 m, and samples were collected biweekly. Thymol was used as a biocide in the collection buckets to avoid changes in nutrient concentration. The funnels were rinsed with distilled water to collect the deposition from the funnel walls. The NO₃⁻, NH₄⁺, and PO₄³⁻ in the bulk samples were spectrophotometrically analyzed following brucin sulfanilic acid method (Voghel 1971), phenate method (Park et al. 2009), and molybdate stannous chloride method (APHA 1998), respectively. The OC was measured following the KMnO₄ digestion method (Michel 1984). To calculate the pollution load, concentrations thus obtained were converted to rate of deposition considering the time and mouth area of the bulk sampler. Values were then converted to Kg per hectare and subsequently on subwatershed scale following Pandey et al. (2014) for Adalpura (84 km²), Assighat (72 km²), and Rajghat (90 km²).

Runoff chemistry

For the collection of surface runoff samples, sites were selected primarily according to land usage such as rural, urban, agricultural, woodland, etc., and the strategy was to collect stormwater representing the total washout effect featuring a particular site at a single outlet from

the area marked with artificial barriers. From each such area, marked for the purpose, three outlets were installed for collecting the runoff. Runoff samples were collected manually during rain events in triplicate from each site on an event basis and initiated from the first flush using presterilized plastic bottles. Surface runoff samples were collected from each discharge outlet located at about 5–20-m distance from the river bank. Samples were collected initiating from the first flush, and considering the duration of the runoff; the sampling intervals were kept 10 to 30 min. NO₃⁻ in the runoff was quantified using the brucin sulfanilic acid method (Voghel 1971), NH₄⁺ following the phenate method (Park et al. 2009), and total dissolved nitrogen (TDN) using high-temperature persulfate digestion. Dissolved reactive phosphorus (DRP, orthophosphate) was measured with the ammonium molybdate stannous chloride method (APHA 1998) and DOC by the KMnO₄ digestion procedure. Dissolved organic nitrogen (DON) was calculated as TDN minus DIN (NO₃⁻ and NH₄⁺) (Perakis and Hedin 2002). For quantifying pollution load, runoff volume was determined considering the flow velocity, measured using a flow meter, and the cross-sectional area of discharge outlet. The values were then converted to pollution load on subwatershed scale by multiplying the discharge volume with mean concentration of nutrients.

River water chemistry

River water samples were collected at monthly intervals in triplicate from each subsite, namely urban, mid, and offside, from directly below the surface (15–25-cm depth) in acid-rinsed 5-L plastic containers from all the sampling stations. Following similar protocol, water samples were collected also from Assi drain mouth, and downstream for point source impact analysis. pH, TDS, conductivity, and salinity were measured on site

using a multiparameter tester (Oakton 35425-10, USA). Biological oxygen demand (BOD) and dissolved oxygen (DO) was quantified following standard methods (APHA 1998). Nitrate was estimated using the brucin sulfanilic acid method, NH_4^+ following the phenate method (Park et al. 2009), DRP following the ammonium molybdate stannous chloride method, and DOC by KMnO_4 digestion. Dissolved silica (DSi) was quantified following the molybdate blue method (Diatloff and Rengel 2001).

Biological variables

Chlorophyll a (Chl a) was measured following an acetone extraction procedure (Maiti 2001) and gross primary productivity (GPP) by the light-and-dark bottle method (APHA 1998). The trophic state index (TSI) was estimated using the Carlson index based on Chl a and phosphorus (Carlson 1977):

$$\text{TSI}(\text{Chl a}) = 10 \frac{6 - (2.04 - 0.68 \times \ln \text{Chl a})}{\ln 2}$$

$$\text{TSI}(\text{DRP}) = 10 \left(6 - \frac{\ln \frac{48}{\text{DRP}}}{\ln 2} \right)$$

Statistical analysis

The range of variability was assessed by the mean values and standard deviations (SD). Correlation and regression analyses were used to test for significant relationships between the variables. Principal component analysis (PCA) was used to ordinate the sampling locations and environmental variables. SPSS software (version 16) was used for statistical analysis.

Results and discussion

Although a number of trends emerged, the major focus of this study was to evaluate (1) source partitioning with special reference to N, P, and OC and (2) trophic status with respect to changing river water chemistry.

Non-point source partitioning

Distinct spatial and temporal variations in AD of nutrients and OC were recorded along the study gradient, with values being lowest at Adpr and highest at Rjht. AD- NO_3^- , NH_4^+ , PO_4^{3-} , and OC ranged from 4.07 to 10.7, 1.9 to 3.9, 0.3 to 1.2, and 4.4 to 10.6 $\text{kg ha}^{-1} \text{ season}^{-1}$, respectively (Fig. 2). Compared to Adpr, AD- NO_3^- , NH_4^+ , PO_4^{3-} , and OC at Asht increased by 87.6, 40.7, 121.2, and 86.8% and at Rjht by 163.4, 101.2, 299.2, and 141.03%, respectively (Fig. 3). The PO_4^{3-} showed the highest percentage increases in AD input. The AD was lowest in 2013 and increased consistently over time, attaining maxima in 2015. Seasonally, deposition was lowest in the monsoon season and highest in winter. NPSs are diffuse in nature; AD, for instance, is added directly onto water surfaces and indirectly through AD-coupled lateral flow (surface runoff). Such inputs, being widespread and difficult to control, have received worldwide attention. Over two decades, the Ganges Basin underwent large changes in land use patterns with increases in built-up land and agricultural areas and large declines in forest cover, scrubland, and water bodies. This densely populated river basin with approximately 73.44% agricultural land, large urban-industrial activities, and westerly wind influences receives approximately 3.32 Tg of reactive N (Nr) and 173.2 Gg of P annually through atmospheric deposition (Pandey et al. 2016a). The subwatershed scale calculations in this study show that, each year, Rjht subwatershed receives 396.37 t DIN, 33.3 t DRP, and 287.29 t OC as AD input. Through these subwatersheds, the river receives 116.1 t DIN, 21.15 t DRP, and 1421.55 t DOC through surface runoff. These values are 1.8- to 4.3-fold higher than those received at upstream site Adpr. Based on the available data, for instance, at subwatershed 2, when all sources compared, atmospheric deposition alone contributes about 14.01% of DIN, 9.9% of DRP, and 3.73% OC in total pollution load reaching to the river in the study region. Since the land use at subwatershed 2 consists mainly of agricultural lands upstream and urban areas downstream, these observations suggest that, as a non-point source, AD input, where agricultural and urban emissions assume predominance, can be a prominent source of pollution load to the Ganga River in Varanasi region. The study further showed a marked increase in AD input between 2013 and 2015. A sharp increase in AD input attaining maxima in 2015 could be linked to the fastened

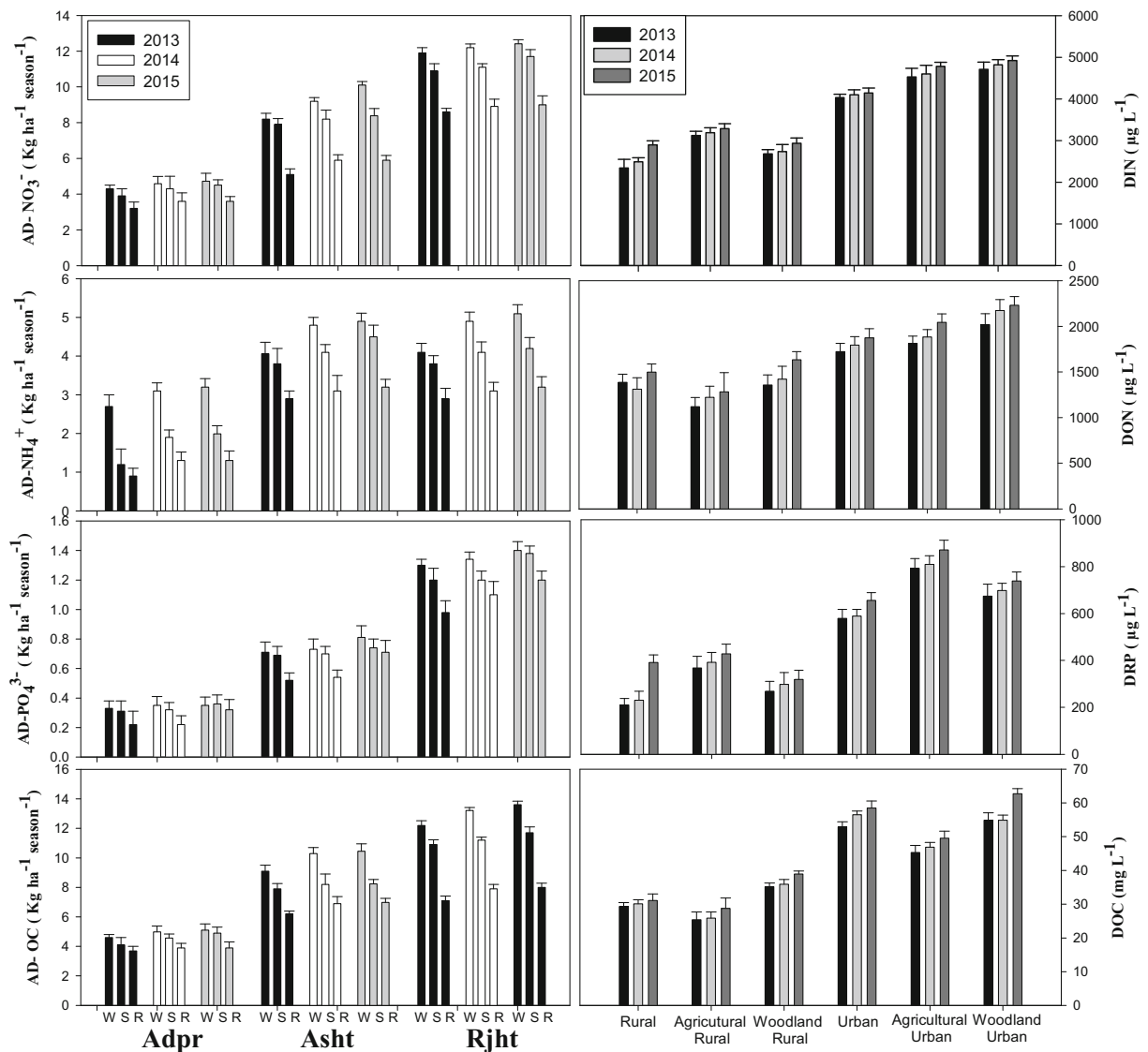


Fig. 2 Atmospheric deposition and concentration of N, P, and OC in surface runoff flux from Adpr to Rjht site. *W* winter, *S* summer, *R* rainy season. Values are mean ($n = 12$ for AD and $n = 36$ for surface runoff) \pm 1SD

developmental activities the Varanasi region witnessed during this period. After the change of the government in India in May 2014, Varanasi region experienced unprecedented economic boom, marked by increased developmental activities, tourist flux, and urban expansion coupled with mobilization of resources and infrastructure. These changes could lead marked increase in atmospheric deposition in 2015, compared to 2013.

The Ganga River experiences a long low flow period, whereas the high flushing of nutrients and OC through surface runoff is limited to the short monsoon season. PS seemed more effective at regulating N and P during

low flow because of less dilution. NPS such as AD and leaching constantly contribute to river nutrients, while surface runoff is a seasonal phenomenon and is significant only during the monsoon when the hydrological loading of the river is high, leading to a high input of DSi and DOC to the river. In the study area, between Adpr and Rjht, a marked difference in the land use pattern and nature of anthropogenic disturbances result in a clear gradient of nutrient sources. The stretch from Adpr to the upstream Varanasi urban core features agricultural rural land use contributing principally through AD, leaching, and surface runoff. The river between

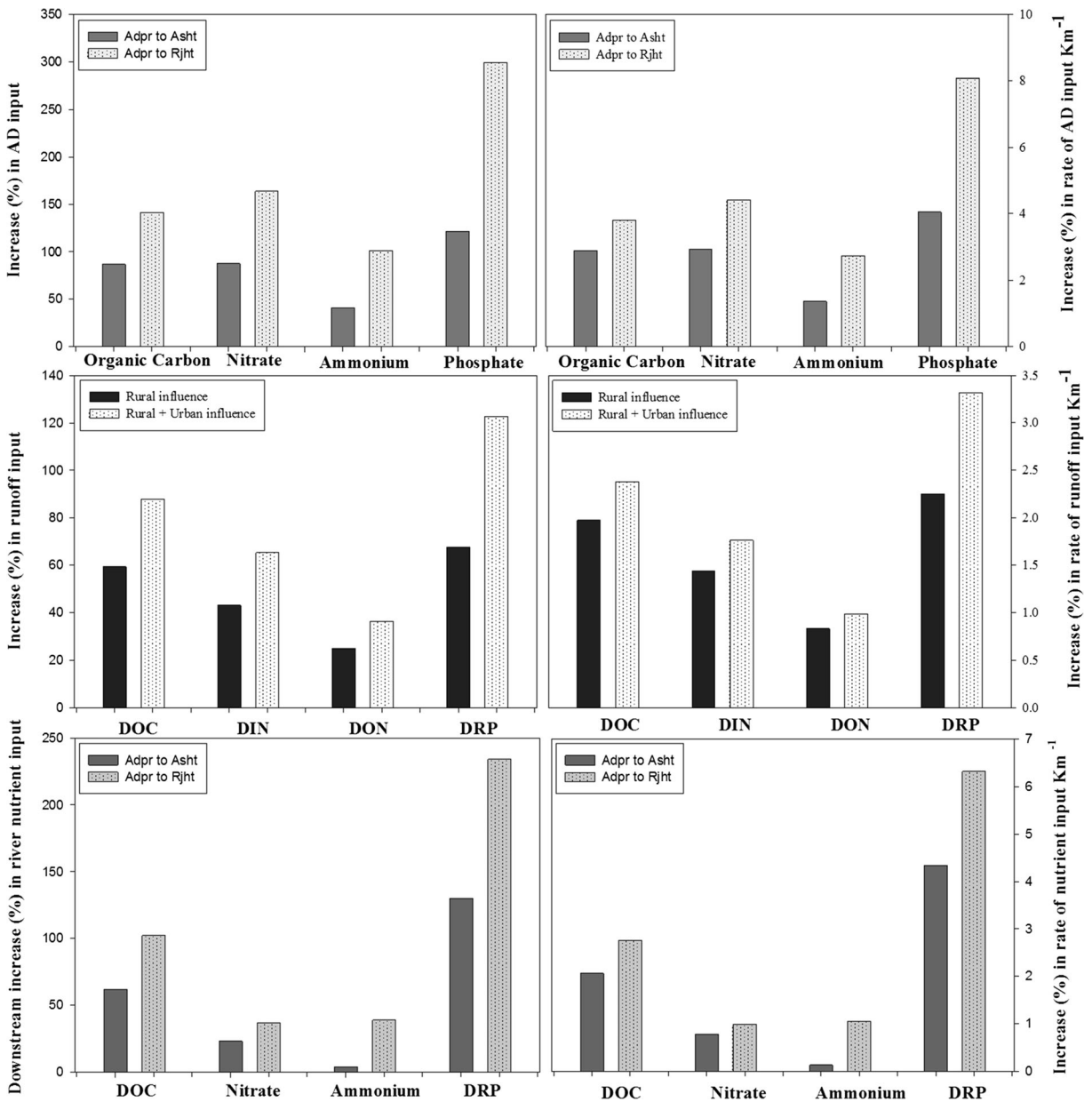


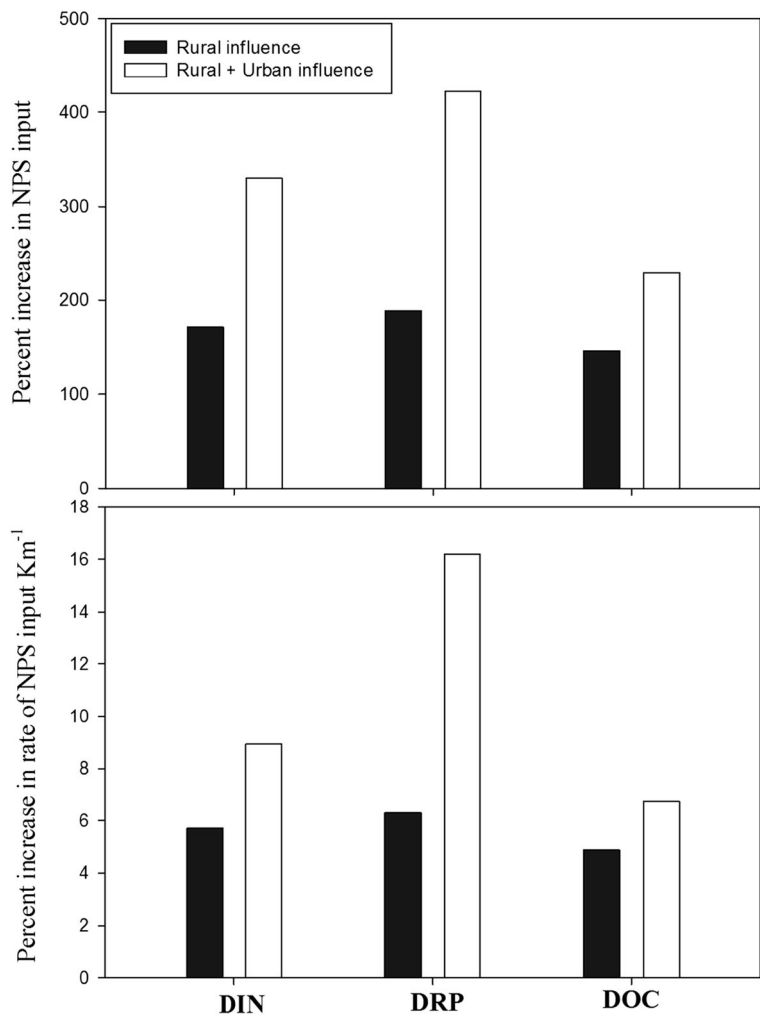
Fig. 3 Percent increase in total input and rate of increase (km^{-1}) relating non-point source and concentration of carbon and nutrient in the river from Adpr to Rjht

Asht and Rjht receives extensive high loads of carbon and nutrients from a number of sewage and wastewater outlets, creating a mixed effect of AD, surface runoff, and PS discharge.

Concentrations of nitrogen, phosphorus, and DOC in the surface runoff increased over time and varied in an almost similar order, being highest at Rjht and lowest at Adpr (Fig. 2). Compared to Adpr, DIN, DON, DRP, and DOC in the runoff increased by 43.02, 29.99, 67.5, and

59.29% at Asht and by 65.25, 36.3, 122.7, and 88.05% at Rjht. Total NPS increases from Adpr to Asht, and Rjht was highest for DRP followed by DIN and DOC (Fig. 4). DIN was the dominant N component being exported through the surface runoff. DIN and DON comprise 66–70 and 30–34% of the runoff N export. The DOC concentration in the runoff ranged from 3.1 to 5.8 mg L^{-1} . Our study showed distinct variations in river nutrients, with changes in AD, land use, and point

Fig. 4 Percent increases in total NPS input and rate of NPS input of DIN, DRP, and DOC in Ganga River



sources. Within-site variations in river N, P, C, and other water quality variables were associated with hydrological flow and subcatchment characteristics. Furthermore, nutrient concentrations showed strong seasonality and a rising trend over time. Earlier studies have shown that seasons play an important role linking river water quality with land use characteristics (Xiao et al. 2016). In addition, agricultural and livestock sources contribute high inputs of N and P to receiving waters (Carpenter et al. 1998; Aneja et al. 2012). Changes in surface runoff chemistry clearly indicated this NPS effect. Concentrations of NH_4^+ and PO_4^{3-} were higher in runoff from agricultural and pasture lands. Sites downstream from Asht to Rjht representing urban land usage, high AD input, together with household and urban-industrial release showed high concentrations of these nutrients. NO_3^- appeared to be the dominant component of N

export through surface runoff. This is a common trend in polluted watersheds except in areas where denitrification and the removal of dissolved inorganic nutrients by plants prevail and DON predominates (Vink et al. 2007). Nitrate concentrations in rivers are relatively less influenced by point sources and more by diffused inputs primarily from agriculture (Neal et al. 2006; Zhang et al. 2015). Correlations presented in this study show that concentrations of nutrients in the surface runoff and in the river followed an almost synchronous trend to the changes in land use. Agriculture-dominated areas added high amounts of PO_4^{3-} , NO_3^- , and OC through AD-coupled surface runoff, and the river concentrations showed a strong synchrony.

In the present study, total NPS contribution was highest for DRP followed by DIN and DOC. The NPS contribution to the total water pollution in China reaches

81% for N and 93% for P (Ongley et al. 2010). In another study conducted in the Jialing River watershed (China), Wu et al. (2012) showed that of the total nitrogen (TN) flushed to the river, land use contributed 67.2% followed by livestock and poultry (30.5%) and rural life (2.2%); livestock and poultry contributed 50% of the TP, followed by land use (48.8%) and rural life (1.2%). In a modeling study using three UK catchments of varying population densities and land use, Bowes et al. (2008) predicted that approximately 75% of the annual TP load was derived from diffuse sources. A study of 86 US rivers showed that although PS continues to contribute > 50% of the N and P reaching US rivers from urban areas, NPS are responsible for over 90% of the N input to over one half of the 86 rivers studied (Carpenter et al. 1998). Among NPS, agriculture is regarded as the principal source of nutrients contributed through enhanced AD, leaching, and surface runoff. Our study showed that large amounts of N, P, and OC are added to the river as AD and surface runoff from Adpr to Asht, which mainly represents an agricultural subwatershed. Earlier studies have shown AD and surface runoff to be important drivers of N, P, and OC input to the Ganga River (Pandey et al. 2013, 2014). In the USA and Europe, only approximately 30% of fertilizer P and 18% of fertilizer N applied to the croplands are removed in the produce, leaving behind approximately 22 kg ha⁻¹ year⁻¹ of surplus P and 174 kg ha⁻¹ year⁻¹ of surplus N, which, in due course, through leaching, surface runoff, or atmospheric deposition reach may the water bodies (Carpenter et al. 1998). Based on soil properties, fertilizer N export from agriculture varies, ranging from 10 to 40% for loam and clay soils, and 25 to 80% for sandy soils (Carpenter et al. 1998). In this respect, the Ganges Basin would show a mixed effect of loam, clay, and sandy soils.

The increased AD and surface runoff inputs at Rjht indicated the effect of intense anthropogenic pressure, including population density, urban land use, and biomass burning. PO₄³⁻ showed the highest percentage increase in AD input followed by NO₃⁻, OC, and NH₄⁺ at Asht and Rjht compared to Adpr. The relatively higher increase at Rjht could partly be attributed to emissions from massive biomass burning. In the burning process, almost 100% of the N and approximately 80% of the P are released into the atmosphere (Singh et al. 1993). This site receives emissions from the burning of more than approximately 25,000 t of dry wood for the cremation of approximately 36,000 dead bodies

annually. Biomass burning accounts for approximately 23% of the P flux in the Amazon Basin through atmospheric transport (Mahowald et al. 2005). Approximately 50% of the global atmospheric sources of P (approximately 1.8 Tg P year⁻¹) are from combustion-related emissions (Wang et al. 2015). Gaseous ammonia is an important precursor pollutant emitted mainly by livestock farms and fertilizer application. Human food consumption, the burning of biomass and fuel, mobile sources, industry, and other activities also contribute to ammonia emission (Sapek 2013). AD-coupled surface runoff chemistry clearly showed the influence of such sources along the study gradient. N and P export coefficients are 3- and 12-fold higher for urban areas than for rural or undisturbed areas (Fluck et al. 1992), as evident also in the high average increase and rate of increase in nutrients per kilometer between the Asht and Rjht sites. At Rjht, for instance, approximately 7 km downstream from Asht, the total NPS-DRP was over 2-fold higher compared to Asht.

Point source partitioning

River water quality showed increasing deterioration downstream the study gradient (Figs. 5 and 6). Concentrations of N, P, and DOC were lowest at Adpr and highest at Rjht and increased consistently over time. Within-site variations in the concentrations of NO₃⁻, NH₄⁺, PO₄³⁻, and DOC were distinct at all sites, with values being highest towards the urban side. With respect to the point source, at zero distance where concentrations of nutrients and DOC were highest, dissolved oxygen was close to zero (Figs. 7 and 8). Nutrients and carbon showed a strong influence of river discharge. Concentrations of NO₃⁻, NH₄⁺, and PO₄³⁻ were high during low flow, while DOC and DSi were high during high flow (Supplementary Fig. 1). DOC and DSi showed significant positive correlations ($p < 0.001$) with river discharge, but other variables were significantly negatively correlated (Supplementary Fig. 2). Compared to Adpr, the concentrations of river NO₃⁻, NH₄⁺, PO₄³⁻, and DOC increased by 23.4, 3.9, 130.2, and 61.9% at Asht and by 36.7, 38.9, 234.0, and 102.1% at Rjht, respectively. The increases, related to NPS, were highest for DRP (Fig. 9). However, after addition from the PS, the increases were highest for NH₄⁺ followed by DOC and DRP. At zero distance, the concentrations of DOC and nutrients were highest and showed a dilution effect downstream to Bhht (Fig. 9). Other water quality

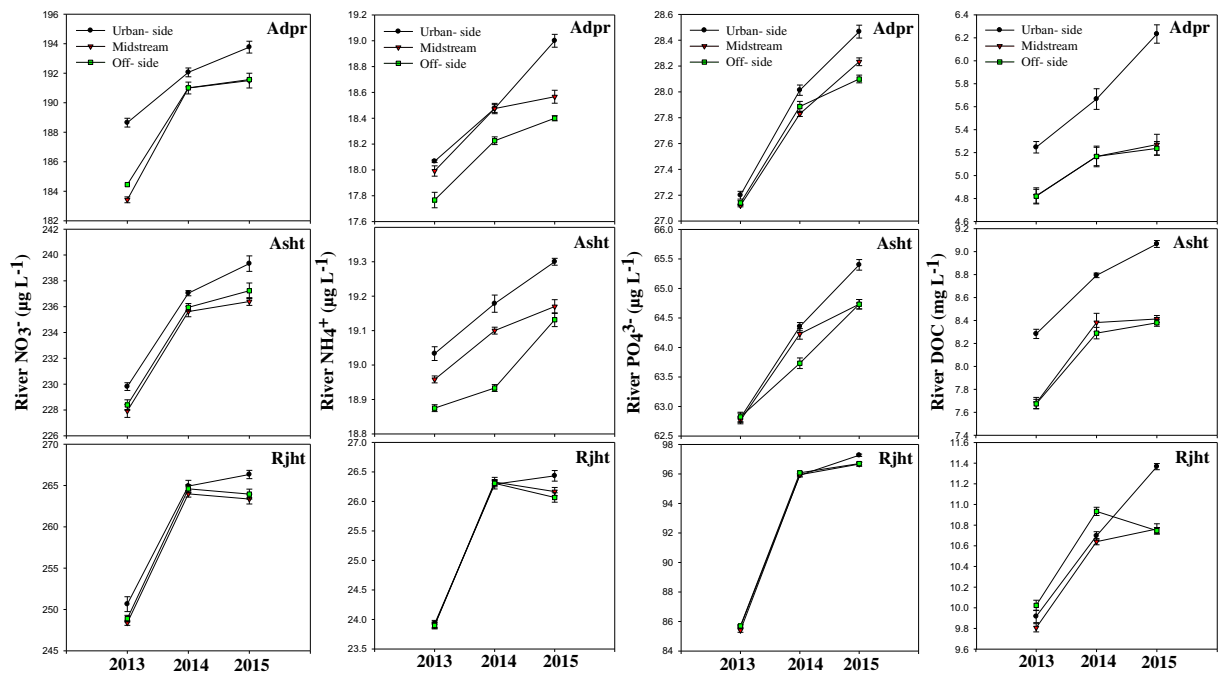


Fig. 5 Between and within site (trans-river) variations in concentrations of N, P, and DOC in the river

parameters showed variable trends with respect to NPS and PS inputs (Table 2). DO and DSi declined downstream. GPP and Chl a also remained highest at Rjht. PCA distinguished the most polluted site (Rjht) from the least polluted site (Adpr) (Fig. 10a). Rvht and Rjht showed similarities in nutrients and DOC as influenced by point sources and cumulative effects downstream (Fig. 10b).

Despite large contributions of N and P from non-point sources, point sources still contribute > 50% of these nutrients in high-density areas. In a model estimate, Bowes et al. (2008) showed that between 1970 and 1985, the DRP load from point sources in the Frome River (UK) was 46–62%. The Ganga River receives disproportionately high amounts of N and P from PSs. Municipal sewage discharge and industrial wastewater comprise most PS pollution. There are 36 class I and 14 class II cities discharging approximately 2723.3 million liters per day (MLD) of wastewater into the river along its 2525-km course. The treatment facilities, however, can handle only approximately 44% (1208.8 MLD) of the wastewater. The Ganga River at Varanasi receives 141 MLD of treated sewage and 46.1 MLD of untreated sewage along with 410 MLD of wastewater (CPCB 2013). The Assi drain, located upstream from Asht and flushed by a large section of the city, discharges 66.45

MLD of wastewater (pH 7.46, chemical oxygen demand (COD) 156 mg L⁻¹) with a 4060 kg day⁻¹ BOD load to substantially enhance downstream concentrations of nutrients and organic C, more between Asht and Bhht. The Assi drain adds approximately 535.1 t of DIN and 133.78 t of DRP annually. In this stretch (between Asht and Rjht), atmospheric deposition added directly to water surface contributes approximately 15.23 t of DIN and 1.19 t DRP, while surface runoff adds 289.69 t of DIN and 47.1 t of DRP annually. Assuming a leaching rate of 10 kg ha⁻¹ year⁻¹ N (He et al. 2011), leaching along the agricultural subwatershed of the study stretch contributes approximately 813.5 t of DIN annually. Thus, this single-point source (Assi drain) alone contributes approximately 32% of the total NPS in the study stretch. Sewage input adds substantial amounts of nutrients and OC to the receiving waters (Daniel et al. 2002). Nitrate remains an exception to this pattern possibly due to rapid denitrification in oxygen-deficient sewage. Streams receiving sewage effluent are characterized by low oxygen and high DOC and EC (Daniel et al. 2002). Trans-river variation, an important issue here considered for the first time for the Ganga River, helps to evaluate the influence of point sources and changes regulated by hydrological and biogeochemical processes across the river. In this context,

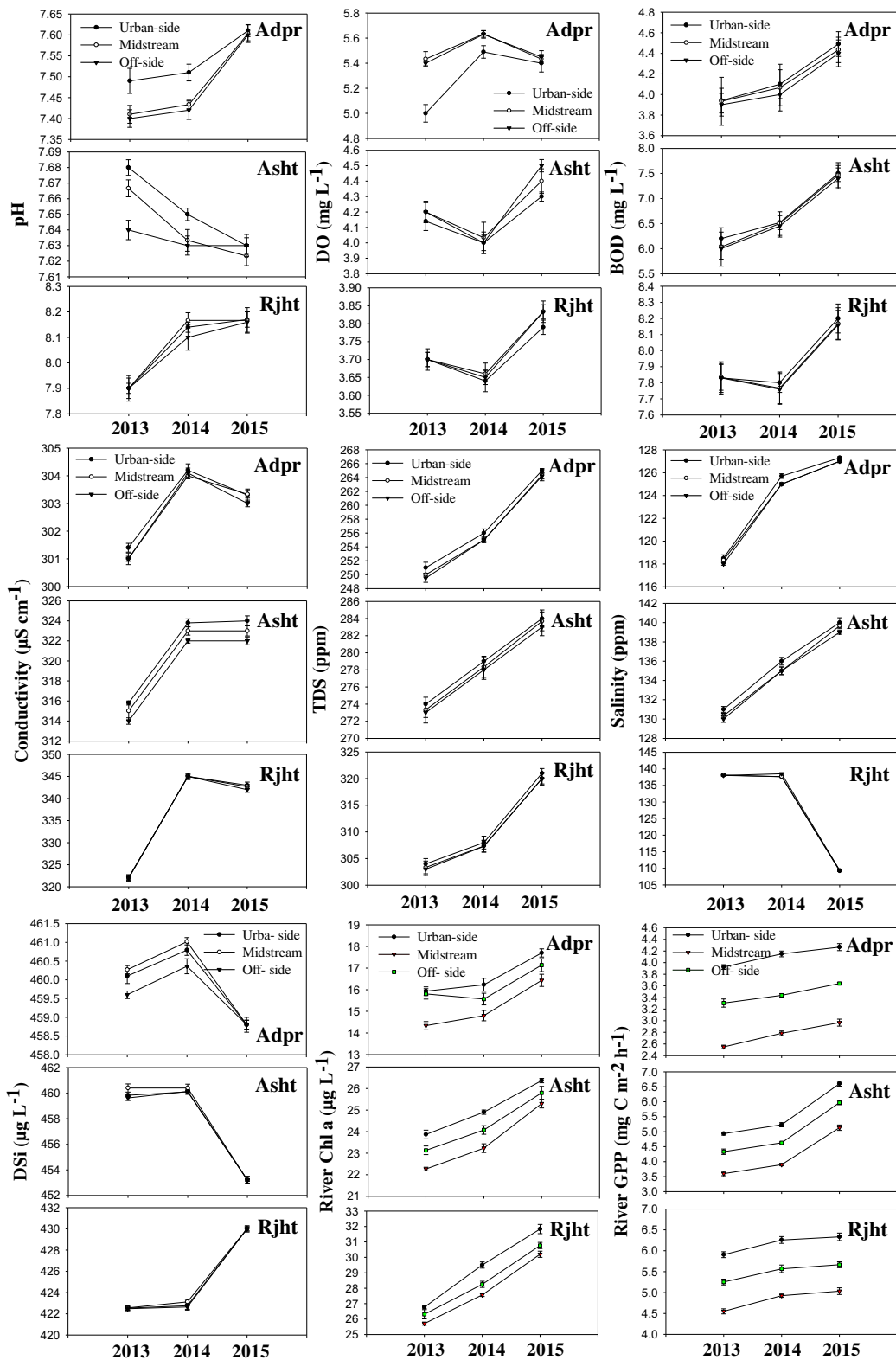


Fig. 6 Between and within site (trans-river) variations in physico-chemical and productivity variables in the river

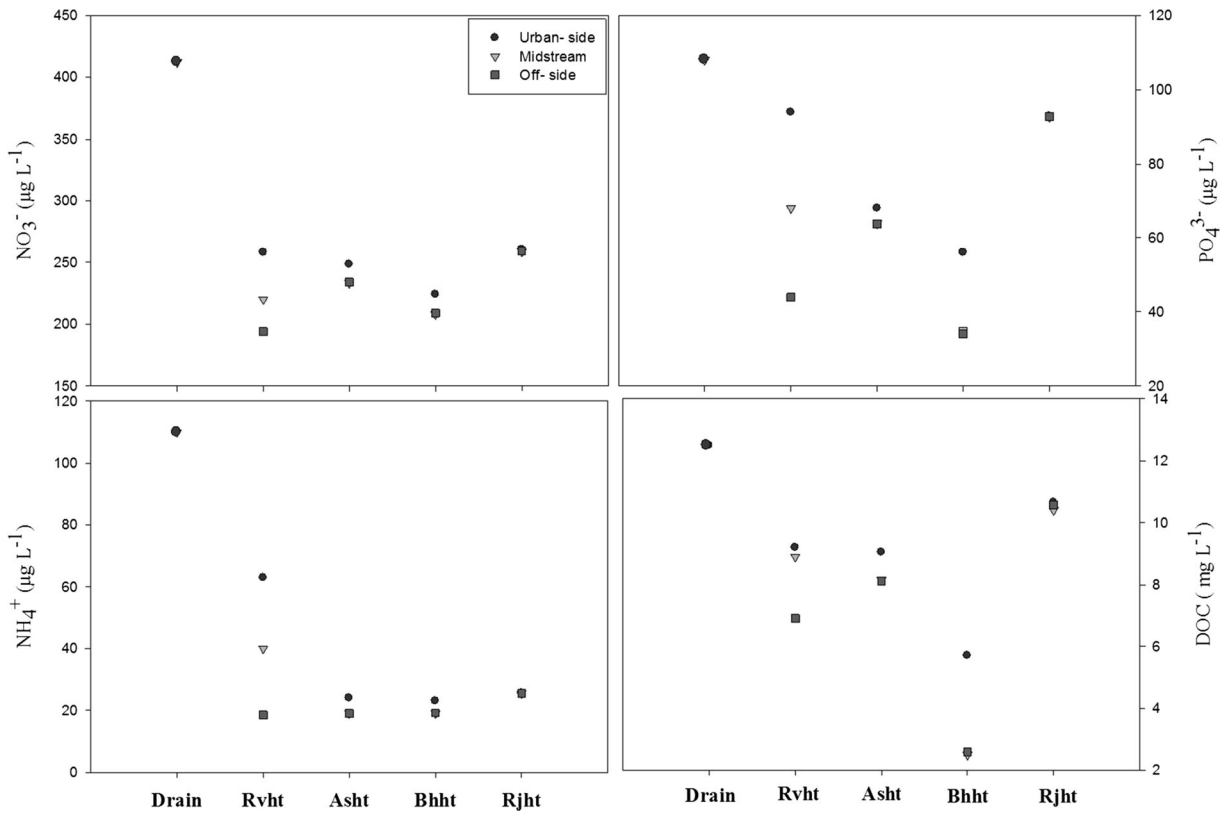


Fig. 7 Point source-to-downstream trends in concentration of N, P, and DOC in the river

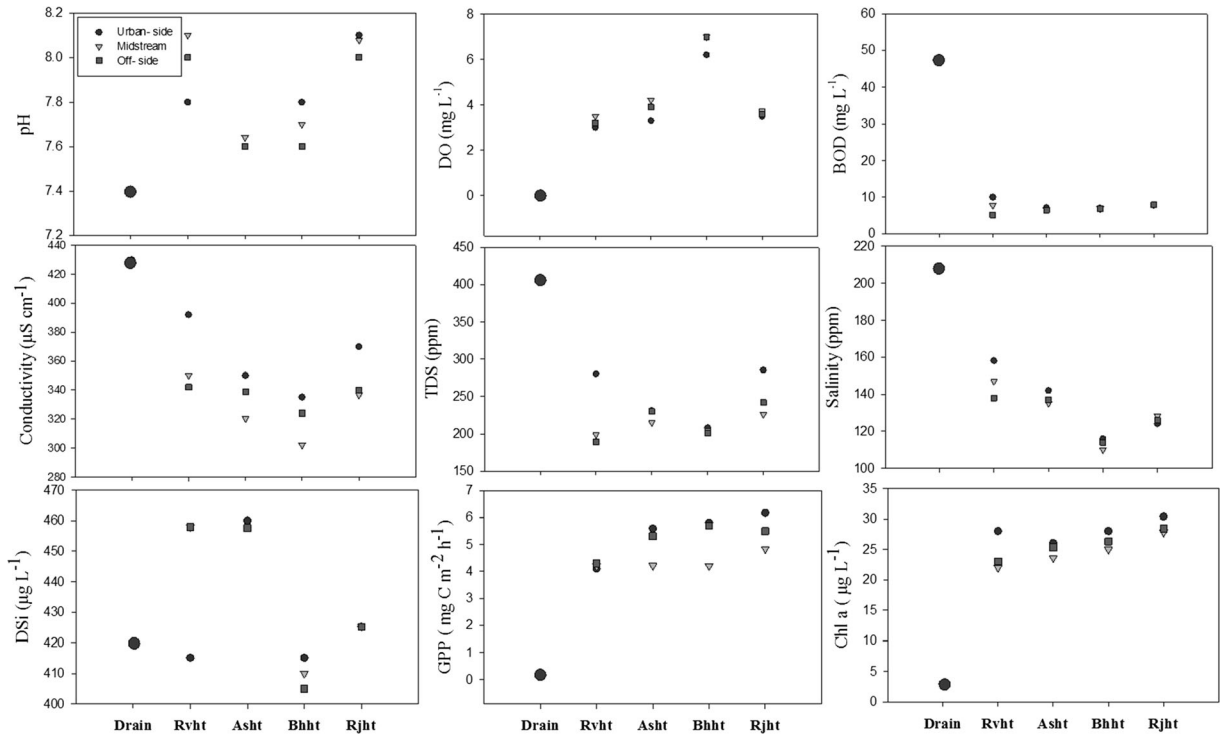
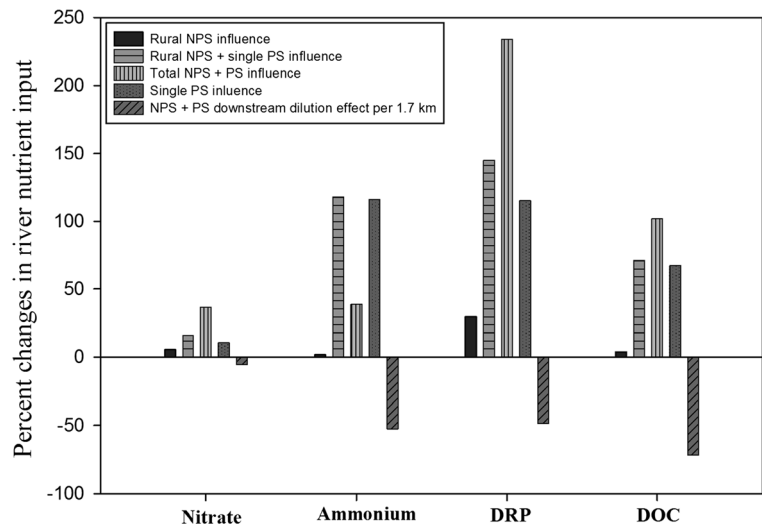


Fig. 8 Trends in physico-chemical and productivity variables from point source to downstream sites in urban side, mid-stream, and off-side

Fig. 9 Source-linked changes in river nutrient input



trajectory variations showed remarkable changes in concentration with flow pattern. During high rain events, for instance, although volume is enhanced, the overall sewage is diluted for nutrients and other constituents. During low flow, from the point source, concentrations of N, P, and C declined downstream but remained higher towards the urban side compared to the off-side and mid-stream.

The synergy between point and non-point source inputs is often disturbed due to the strong hydrological seasonality; consequently, the distribution and seasonal variations in nutrients and other pollutants in the river are altered (Tremblay et al. 2014). Diffuse nutrient inputs from agricultural fields induce greater effects on water chemistry than those flushed through wastewater treatment plants, although N concentration is highly

influenced by PS inputs in non-agricultural streams (Merseburger et al. 2005). As per the US Geological Survey (1994) records, approximately two decades back, PS represented only 5.7% of the total N added to the environment, whereas agricultural NPS accounted for 93.5%, although streams near large cities did account for approximately 77% of the N from PS (Puckett 1994). The river between the Adpr and Rjht sites has a specific feature of meandering, which induces variable effects on the hydrological flow pattern and velocity (Shukla and Raju 2008). The river makes a prominent meander loop with the outside towards Varanasi where the SE flowing Ganga River takes an abrupt swing and flows to the NNW, resulting in the western margin being the down-thrown side and the eastern side the up-thrown block. Unlike other places where cliff

Table 2 Source-linked changes in physico-chemical and productivity variables in the river

Parameters	Before first outlet (NPS) (%)	After first outlet (PS + NPS) (%)	At Rjht (PS + NPS) (%)	Due only to first PS (%) at Rvht	At 1.5 km after first outlet (Rvht mid to Bhht mid) (%)
pH	4.2	8.3	8.0	4.0	- 4.9
Conductivity	3.0	15.6	11.2	12.6	- 13.7
TDS	2.2	16.6	21.0	14.4	1.4
Salinity	1.3	19.1	4.0	17.8	- 0.3
DO	- 14.5	- 36.4	- 32.2	- 21.8	1.0
BOD	20.6	88.2	91.2	67.6	- 11.5
DSi	- 0.4	- 0.4	- 7.6	0.0	- 10.5
GPP	22.2	22.2	59.8	0.0	0.0
Chl a	38.0	38.0	79.0	0.0	13.6

NPS non-point source, PS point source, Rvht Ravidas ghat, Bhht Bhadaini ghat

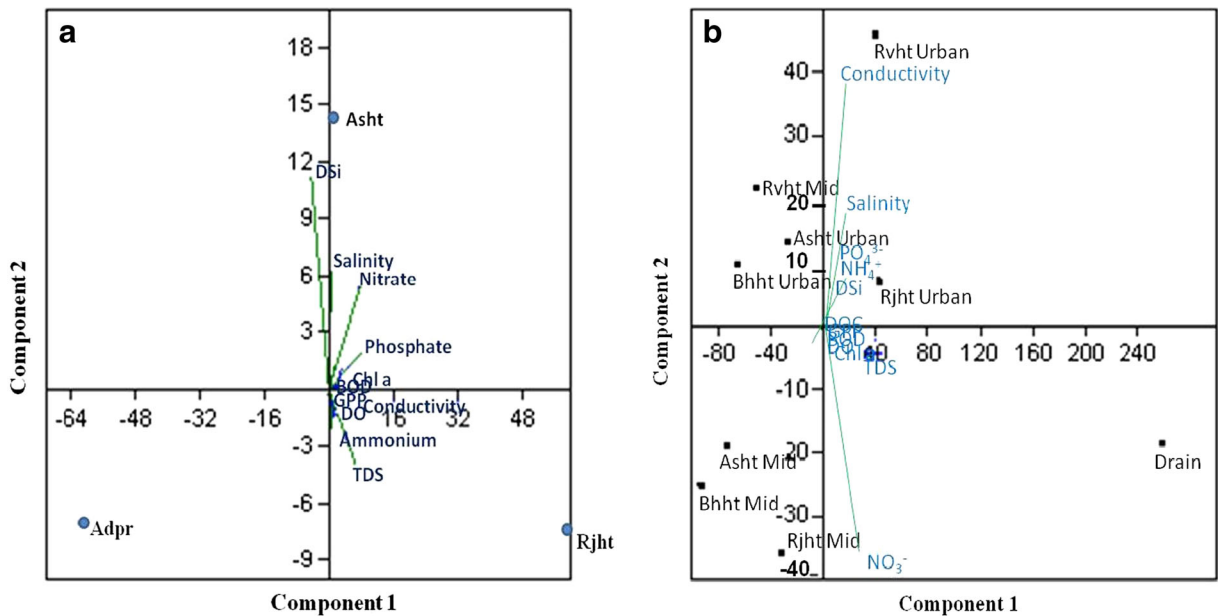


Fig. 10 Principal component analysis (PCA) ordination showing study sites **a** based on nutrient gradient from Adpr downstream and **b** from point sources downstream

development is observed only on the southern bank, the northern sides are gently sloping near Varanasi, and cliffs are present on both sides of the channel (Shukla and Raju 2008). This effect, together with PS inputs, possibly influenced the well-marked trajectory variations from the Assi drain outlet to Bhht, with declining concentrations of nutrients downstream. The urban side was more enriched in carbon and nutrients than the midstream and off-side, obviously due to the influence of the river flow and direct human disturbances towards the urban side. The high concentrations of DOC, NO_3^- , NH_4^+ , and DRP at Rjht reflect the combined effect of point and non-point sources, although the highest variation in NH_4^+ was possibly associated with a high sewage load. River hydrological features and biogeochemical processes possibly drove the declines in pH, conductivity, salinity, BOD, and DSi downstream up to approximately 1.7 km from the point source outlet. The concentration of DSi decreased in the low flow period possibly due to the cutoff of external sources and enhanced consumption by diatoms (Wang et al. 2016).

Trophic status in relation to changing N and P

The variations in river water chemistry and productivity were large, and we attempted to evaluate the influence of NPS and PS inputs on these variations. Our data

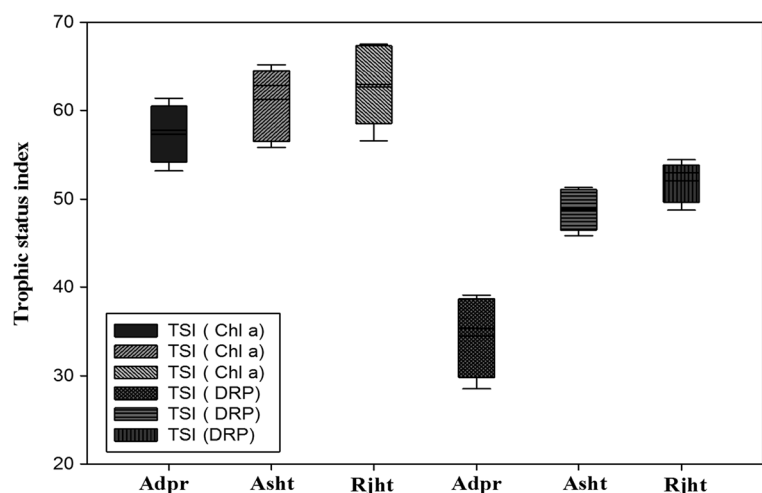
indicated a highly dynamic link and influence of AD, surface runoff, and sewage input on the river’s water chemistry and trophic status. The PS might contribute directly to productivity in the low flow period that persists for a major part of an annual cycle. PS contributions therefore may be expected to supersede such NPS as the surface runoff mainly because of strong seasonal influences. Surface runoff-driven nutrients, added mainly in the rainy season, could not be significantly translated to productivity due to unstable and turbulent water. However, the effect of leaching, such as N from the agricultural subwatershed and AD as an NPS, could be more consistent and uniform in influencing the trophic status of the river. In an extensive study of 54 UK river sites across seven major lowland catchments, Jarvie et al. (2006) observed that point sources of P were a more significant risk for eutrophication, even in rural areas with high agricultural phosphorus loss. Other studies, however, have shown non-point sources such as atmospheric deposition, leaching, and surface runoff to be important drivers of phytoplankton blooms and nutrient limitation in lakes, rivers, and coastal areas (Beman et al. 2005; Bergstrom et al. 2008; Pandey and Pandey 2013). Our earlier studies have shown that AD-N and AD-P account for 11.6–33.3% (for N) and 3.4–10.2% (for P) of the GPP in the Ganga River (Pandey et al. 2014). We found a

consistently rising trend in phytoplankton production (Chl a biomass and GPP) over time at all sites, corresponding closely to the increases in nutrient concentration. The trans-river GPP and Chl a biomass showed a distinct pattern, being highest at the urban side and lowest in mid-stream. The urban side featuring a high concentration of nutrients could be expected to favor phytoplankton development faster than the trans-river sites. NPS between Adpr and Asht contributed to an increase of approximately 22.2% in GPP and 38.0% in Chl a biomass. Compared to Adpr, the Chl a biomass and GPP at Rjht increased by 79.0 and 59.8%, respectively, indicating that the combined effect of NPS and PS induces a stronger influence on phytoplankton production. The TSI of the river, estimated as Carlson's index, ranged from 57.3 to 62.65 and 32.4 to 52.0 based on Chl a and DRP, respectively, and the values were highest at Rjht (Fig. 11). The Carlson's TSI also was highest at Rjht, indicating a combined influence of nutrient flux through different sources. The trophic status, a measure of the food base of an ecosystem, is used to understand food web linkages, biogeochemical processes, and water quality (Dodds and Cole 2007). Carlson's TSI is one of the commonly used tools for characterizing the trophic status or overall health of surface waters. The TSI is a function of six variables—TP, TN, COD, Secchi disk (SD), Chl a, and phytoplankton biomass—and is an important index for eutrophication studies. Based on Carlson's TSI (Chl a) scale, Adpr, Asht, and Rjht fall within the eutrophic range (50–70), but as per the TSI (DRP) scale, Adpr falls in the mesotrophic (< 40) range, while Asht and

Rjht are in the mesotrophic to eutrophic (45–55) range. These observations indicate that despite the source-linked variations, the concentration of nutrients was sufficient to support a reasonably high Chl a biomass along the study stretch. Because the AD input can directly be capitalized by rapidly proliferating phytoplankton, atmospheric deposition could be one possible factor for the increasing phytoplankton biomass with a high TSI scale as reported also by the National Park Service Scientific Investigation Report (Clow and Campbell 2008).

This study further showed that both allochthonous and autochthonous C contributed to the river DOC. In rivers, allochthonous C often exceeds that added from primary production regulated by nutrients. A substantial rise in monsoon season DOC indicates the contribution of NPS allochthonous C, while that during low flow could be closely linked with a PS, as well as atmospheric deposition and leaching-associated autochthonous C build-up. The DOC at Rjht comprises a relatively higher proportion of autochthonous C, indicating the influence of nutrient enrichment on DOC accumulated through autotrophy. Increased allochthonous input and autochthonous C build-up would trigger secondary production or heterotrophy, with associated shifts in food web dynamics as reported for tropical streams (Berggren et al. 2015; Roiha et al. 2016; Yadav and Pandey 2017). The autotrophic and heterotrophic states are used to define rates of photosynthetic and respiratory C fluxes, and the dominance of allochthonous sources allows total respiration to exceed GPP, an effect that has been demonstrated for lakes, rivers, estuaries, and coastal areas

Fig. 11 Trophic state index (TSI) at study sites



(Dodds and Cole 2007), with implications for ecosystem functioning through enhanced CO₂ supply (Wetzel 1995).

Conclusions

The study shows within-site (trans-river) and between-site (spatial gradient) variations in nutrient concentrations as influenced by point sources, land use, and atmospheric deposition along the study stretch of the Ganga River. With respect to DIN and DRP loads reaching to the river, atmospheric deposition showed precedence over the surface runoff. Based on the available data, for instance, at subwatershed 2, when all sources compared, atmospheric deposition alone seemed to have accounted for about 14.01% of DIN, 9.9% of DRP, and 3.73% of OC of the total pollution load reaching to the river in the study region. The study further showed, towards the up-stream urban core, that non-point sources could account for about 38 and 22.2% rise in Chl a and GPP, respectively. The point sources appeared as main contributor enhancing the levels of carbon and nutrients during summer low flow, whereas the surface runoff dominates during monsoon-driven high flow, and atmospheric deposition appeared a consistent source of nutrient supply over the year. Enhanced autotrophy together with large increases in allochthonous C would accelerate heterotrophic growth, degrading the river water quality more rapidly in the near future. Our study suggests the need for regional-scale studies evaluating the contribution of point and non-point sources to support decisions for the sustainable management of the Ganga River.

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