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Article

India's Riverine Nitrogen Runoff Strongly Impacted by Monsoon Variability

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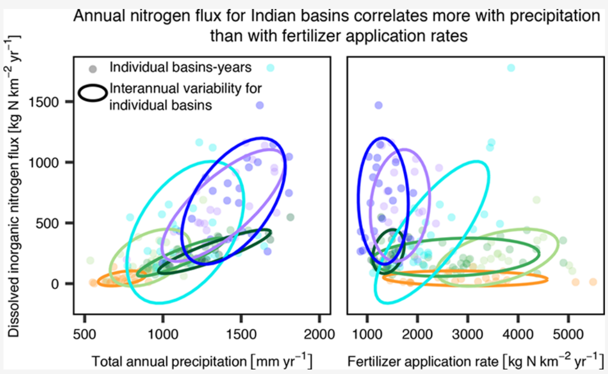
ABSTRACT: Agricultural intensification in India has increased nitrogen pollution, leading to water quality impairments. The fate of reactive nitrogen applied to the land is largely unknown, however. Long-term records of riverine nitrogen fluxes are nonexistent and drivers of variability remain unexamined, limiting the development of nitrogen management strategies. Here, we leverage dissolved inorganic nitrogen (DIN) and discharge data to characterize the seasonal, annual, and regional variability of DIN fluxes and their drivers for seven major river basins from 1981 to 2014. We find large seasonal and interannual variability in nitrogen runoff, with 68% to 94% of DIN fluxes occurring in June through October and with the coefficient of variation across years ranging from 44% to 93% for individual basins. This variability is primarily explained by variability in precipitation, with year- and basin-specific annual precipitation explaining 52% of the combined regional and interannual variability. We find little correlation with rising fertilizer application rates in five of the seven basins, implying that agricultural intensification has thus far primarily impacted groundwater and atmospheric emissions rather than riverine runoff. These findings suggest that riverine nitrogen runoff in India is highly sensitive to projected future increases in precipitation and intensification of the seasonal monsoon, while the impact of projected continued land use intensification is highly uncertain.



KEYWORDS: *agricultural intensification, Indian monsoon, dissolved inorganic nitrogen loading, climate variability*

1. INTRODUCTION

Nitrogen fertilizer application rates in India rose by over an order of magnitude from 1970 to 2015.[1,2] While this has significantly increased agricultural productivity, a large fraction of the applied fertilizer is lost to the environment as reactive nitrogen. The fate of this reactive nitrogen in India is largely unknown.[3] Globally, more than one-fifth of total reactive nitrogen resulting from anthropogenic activity is transported to aquatic ecosystems,[4] increasing rates of eutrophication. In India, eutrophication has resulted in coastal phytoplankton blooms and hypoxia.[5−9] With India poised to become the world's most populous country by 2027[10] and with its continuing high rate of economic development,[11] fertilizer use rates are expected to rise massively under most development scenarios over the 21st century.[12,13] Concurrently, the projected increase in annual and extreme precipitation over India[14−17] may lead to a higher proportion of reactive nitrogen making its way to aquatic ecosystems.

It is therefore critical to understand nutrient runoff from India's major river basins, as well as drivers of its spatial, seasonal, and interannual variability, in order to characterize the fate of reactive nitrogen, assess water quality impacts, and design more effective management strategies in the face of change. Nitrogen flux from river basins in India is not routinely monitored, however. DIN flux was measured in July and August of 2011 for several peninsular rivers of India.[18] Nitrate concentrations measured in August 2001[19] and extreme nitrate concentrations measured in 1998[20] were also combined with observations of discharge to estimate DIN flux for those additional two years in some rivers.[21] In addition, model-based estimates are available for some basins for the year 2000.[22] DIN flux estimates from these studies differ substantially (Table S1) and provide little information about seasonal and interannual variability in fluxes. Climate,[23] fertilizer inputs,[1,24] and water storage capacities[25] vary widely across river basins in India, likely further contributing to spatial and temporal variability in nitrogen fluxes.

Here, we quantify DIN fluxes for seven major river basins in peninsular India (Figure 1A, Tables S2 and S3) for the period

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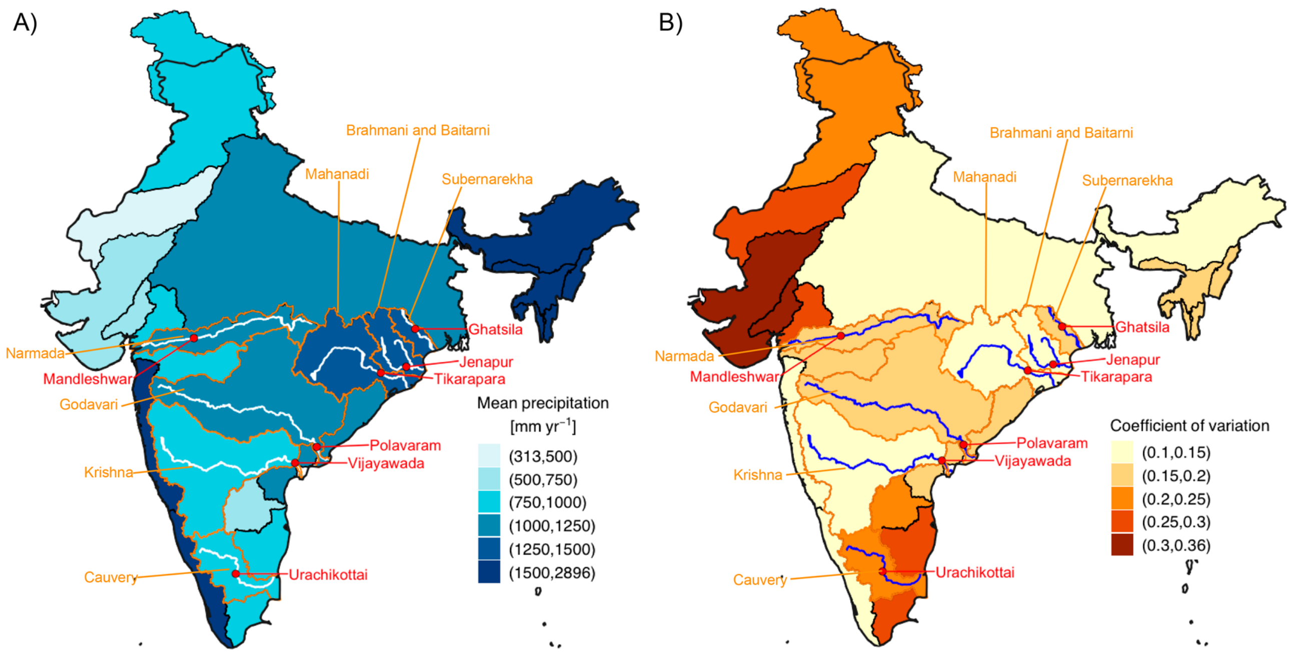

Figure 1. Locations of water quality stations used in this study. Water quality stations used in this study are denoted with red dots and text. The blue polygons represent the Central Water Commission (CWC) basins, with orange borders and text denoting the seven basins used in this study. (A) The shade of blue represents the average annual precipitation. (B) The shade of orange represents the coefficient of variation (i.e., the ratio of the standard deviation to the mean) of total annual precipitation in each CWC basin over the period 1980−2015. The major rivers falling within each of these seven basins are outlined in white (A) and in blue (B).

1981−2014, including its seasonal and interannual variability, by leveraging a recently developed database of water quality and discharge measurements by the India Water Resources Information System (https://www.indiawris.gov.in/wris/). We then examine the drivers of spatial variability in flux across basins as well as primary drivers of the interannual variability in flux within specific basins. We further explore the impact of increases in fertilizer and other nitrogen inputs on riverine nitrogen runoff in India and the ways in which seasonality and year-to-year variability in the monsoon manifests itself in riverine nitrogen fluxes.

2. MATERIALS AND METHODS

2.1. Quantifying DIN Flux. Estimates of DIN flux were obtained by applying the Weighted Regressions on Time, Discharge and Season (WRTDS) method[26] to sporadic DIN concentration measurements and daily discharge measurements. This method estimates long-term variability of water quality parameters through weighted regressions of concentrations on time, discharge, and seasons. Daily nonflow-normalized DIN loads [kg N] are then obtained by multiplying WRTDS-derived daily mean concentrations by observed daily discharge, and monthly or annually averaged DIN flux (Q_{DIN}) [kg N km^{-2}] are estimated by summing daily loads and dividing by the upstream catchment area.

The method is most accurate when continuous daily discharge measurements are available for a minimum of 20 years and when 200 or more concentration measurements are available over that same period.[26] However, reliable flux estimates have been obtained from model application to as few as 60 measurements over a 10-year period.[27] In addition, we eliminated years with fewer than six DIN concentration measurements in order to avoid using years with few observations that may lead to highly uncertain estimates.

2.2. Identifying Dominant Drivers of DIN Flux Variability. We used multiple linear regression to identify the principal drivers of interannual DIN flux variability within individual basins and of both spatial and interannual variability of DIN flux across basins. Candidate predictor variables included variables based on fertilizer application rates, NO_x deposition, precipitation, and temperature (Table S5). We use precipitation rather than runoff or discharge because this provides a more direct link to the variability and change in the physical climate.[28] A statistical model selection approach based on the Bayesian information criterion (BIC)[29] was used to identify a parsimonious subset of predictor variables that explained a large fraction of the observed variability in DIN flux. The BIC approach considers all possible subsets of predictor variables and favors models with a low residual sum of squares while penalizing models with more predictor variables. The (in this case, linear) model with the lowest BIC value optimizes this trade-off. Here we used a BIC difference of two as a threshold for selecting a smaller model with a slightly higher BIC over a larger model with the lowest BIC.[30]

Four candidate variables were used to represent nitrogen inputs to the river basins. These included total nitrogen fertilizer application rate, the sum of fertilizer application rate and NO_x deposition, and the natural log of these two variables. The model selection was set up such that fertilizer application rate and NO_x deposition could each only be selected at most once either alone or in combination.

Forty-two candidate variables were based on precipitation, with variables defined based on climate change indices developed by the Expert Team on Climate Change Detection and Indices (ETCCDI, http://etccdi.pacificclimate.org/list_27_indices.shtml). These variables included total annual precipitation; four variables capturing seasonal precipitation

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during subsets of the months of June, July, August, September, and October; and 37 candidate variables capturing extreme precipitation. The extreme precipitation variables included the number of days with extreme precipitation and the amount of extreme precipitation annually and during the months of June, July, August, September, and October. These months were selected because the majority of rainfall over peninsular India occurs during these monsoon months.[31] The final model was restricted to at most one seasonal and one extreme precipitation variable, and only precipitation variables with positive regression coefficients were considered.

Five candidate variables were based on temperature, including mean annual temperature and four variables based on average seasonal temperature during subsets of the months of June, July, August, and September. The linear model was restricted to selecting at most one temperature variable.

Finally, calendar year was included as a candidate variable to capture any otherwise unexplained long-term trend in DIN fluxes, such as a potential long-term increase in flux not attributable to the other variables in the model.

Spatially and temporally explicit data on wastewater effluent discharged to agricultural lands, agricultural nitrogen fixation and food and feed import are not available for India and could therefore not be included.

2.3. Data Sets Used. We used water quality and discharge observations collected at seven Central Water Commission (CWC) observation stations (Table S2, Figure 1A) released by the India Water Resources Information System (India-WRIS) to quantify upstream DIN fluxes. CWC stations located closest to the mouth of six river basins met the criterion of more than 200 measurements of DIN concentration and more than 20 years of daily discharge measurements. For the seventh basin, the Tikarapara station had only 167 observations of DIN concentrations and 17 years of daily discharge measurements, but these are likely still sufficient for reliable DIN flux estimation.[27] The catchment areas associated with the seven stations were obtained from the USGS Hydro1k data set (https://www.usgs.gov/centers/eros/science/usgs-eros-archive-digital-elevation-hydro1k).

Fertilizer application rates for the seven Indian River basins were estimated based on annual fertilizer application rates at state and national levels. Fertilizer application rate data are available from the Directorate of Economics and Statistics, Department of Agriculture, Cooperation, and Farmers Welfare, Ministry of Agriculture and Farmers Welfare, Government of India (https://eands.dacnet.nic.in/) for 1950−2016 at the national level and for 2004−2016 at the state level. The statewide fertilizer application rate data was converted to basin scale usage based on the fraction of various states falling within the river basins as reported by the India-WRIS Web site (https://www.indiawris.gov.in/wris/) (Table S7). The basin-scale fertilizer application rates for 1980−2003 were estimated based on basin area as a fraction of the national fertilizer application rate. Actual interannual variability in fertilizer usage at the basin scale may be even higher because the state-level estimates may not capture additional interannual variability at the basin scale. There are three main planting seasons in India; seasonal variability in fertilizer application was not considered, however, because data on the timing of fertilizer application is not available.

Atmospheric deposition was estimated based on global NO_x deposition estimates developed by Lamarque et al.[32] We performed an additional sensitivity analysis using NO_x deposition estimates for 1996−2014 as developed by Geddes and Martin.[33]

Nitrogen input from other sources such as sewage, animal manure, and agricultural nitrogen fixation could not be considered in this study due to a lack of available data.

Water storage capacities for the seven river basins were obtained from the India-WRIS river basin reports (https://www.indiawris.gov.in/wris/#/Basin).

Daily precipitation and temperature data for each basin were obtained from the India Meteorological Department (IMD),[34] which provides gridded precipitation at 0.25° × 0.25° resolution and gridded temperature at 1.0° × 1.0° resolution for India. We performed additional sensitivity tests using daily precipitation data at 0.5° × 0.5° from the Climate Prediction Center (CPC) Global Unified Precipitation data provided by NOAA/OAR/ESRL PSD (https://psl.noaa.gov/data/gridded/data.cpc.globalprecip.html) and using monthly temperature data at 0.5° × 0.5° from the Climatic Research Unit (CRU) data set.[35]

3. RESULTS AND DISCUSSION

3.1. Seasonal and Interannual Variability. We find that substantial DIN fluxes only occur during monsoon months, with 68% (Cauvery basin) to 94% (Godavari basin) of the DIN fluxes occurring in June through October (Figure 2 and Table S2) across the examined basins. This large seasonality is attributable to the fact that over 80% of annual rainfall in India occurs from June to September.[31] In addition, the rainfall that occurs during nonmonsoonal months is largely retained by dams for irrigation purposes.[18] The June to October average DIN flux (average across years and across months) for the seven basins ranges from 8 (Krishna basin) to 111 (Subernarekha basin) kg N km^{-2} $month^{-1}$ (Table S2). The highest average DIN flux occurs in August for six of the basins, while it occurs in October for the Cauvery basin.

For specific years and basins, the month with the highest DIN flux coincides with the month with the highest discharge in 94% of cases, while the month of maximum discharge coincides with the month of highest precipitation in 43% of cases and lags it by one month in 31% of cases due to water capture and release in reservoirs. Taken together, the month of highest DIN flux coincides with the month of highest precipitation in 47% of cases and lags it by one month in 31% of cases. Record high monthly DIN fluxes range from 70 (Krishna basin; October 1998) to 952 (Narmada basin; August 2013) kg N km^{-2} $month^{-1}$.

We also find that annual DIN fluxes exhibit a large interannual variability, with the coefficient of variation ranging from 44% for the Mahanadi basin to 93% for the Krishna basin (Figure 3). The actual interannual variability in DIN flux may be even higher because the Weighted Regressions on Time, Discharge and Season (WRTDS) approach used here to estimate DIN concentrations (and therefore fluxes) at unsampled times underrepresents the observed variability in DIN concentrations in some cases (see the Supporting Information). This observed interannual variability highlights some of the limitations of previous estimates (Table S1, Figure 3). For example, DIN flux estimated by Swaney et al.[21] based on concentration and discharge measurements by Subramanian et al.[20] are unrealistically high due to their focus on only extreme observed concentration values. In addition, the large interannual variability in DIN flux shown here indicates that estimates based on individual years cannot be used to

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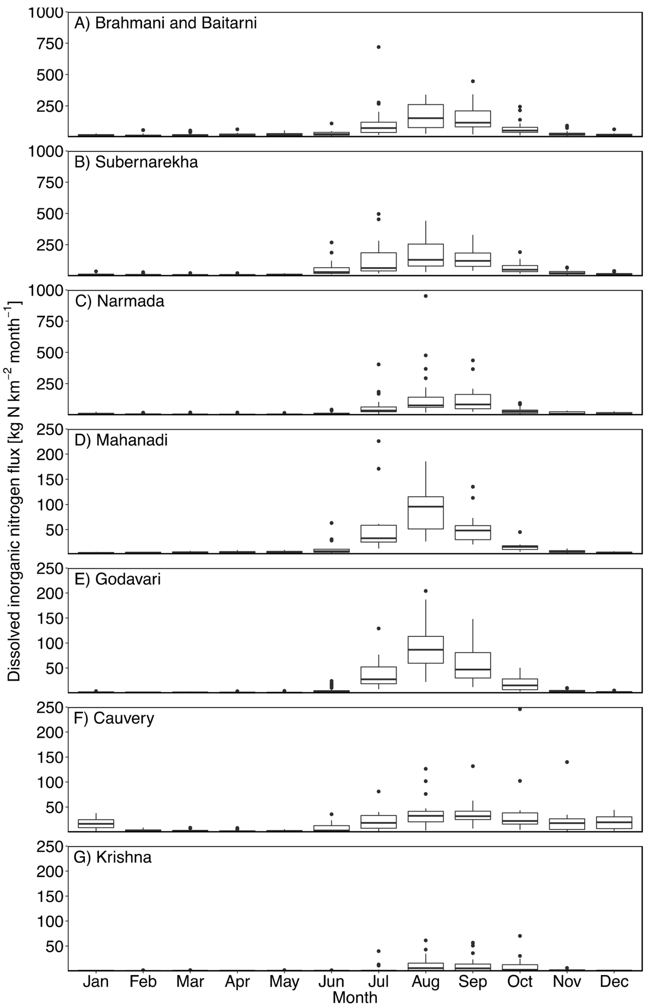

Figure 2. Monthly dissolved inorganic nitrogen fluxes for the seven examined basins for years with sufficient available data (see Figure 3) in the period 1981−2014. Here and in subsequent figures, basins are ordered from largest to smallest mean annual flux (Q_{DIN}). Note that the range of the vertical axes varies between basins. The center line of the box plots represents the median, the lower and upper hinges correspond to the first and third quartiles, the upper and lower whiskers extend from the box hinges to the largest and smallest value within the 1.5·interquartile range (distance between the first and third quartiles), and data beyond the whiskers represents outliers.

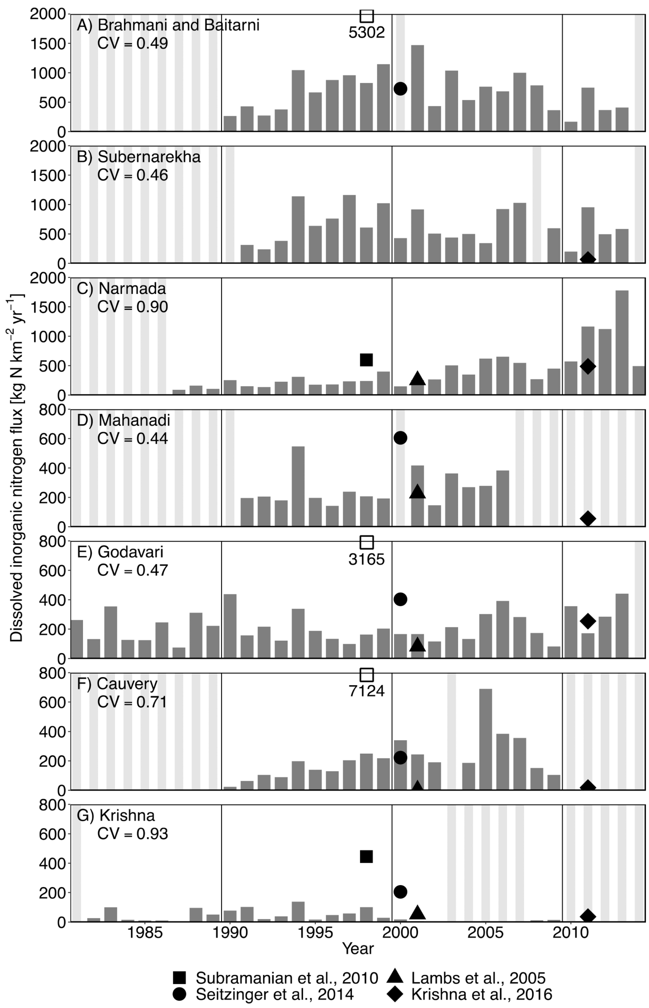

Figure 3. Annual dissolved inorganic nitrogen fluxes (Q_{DIN}) for the seven examined basins for the period 1981−2014. Light gray bars represent years for which Q_{DIN} was not estimated because there were fewer than six available observations of DIN concentration. The coefficient of determination (CV) is a measure of the interannual variability in fluxes and is defined as the ratio of the standard deviation of the annual fluxes to their mean. Available flux estimates from earlier studies (Table S1) are presented as symbols, with empty symbols representing fluxes above the range of the axes, with the value listed under the symbol. Note that the range of the vertical axes varies between basins.

represent long-term average conditions and even less so conditions during other individual years.

3.2. Role of Annual Precipitation. The spatial variability in DIN fluxes is strongly related to variability in annual total precipitation (Figure 4A). Basins with higher precipitation, based on data from the India Meteorological Department (IMD),[34] exhibit larger DIN fluxes. The high average annual DIN fluxes from the Brahmani and Baitarni basin and the Subernarekha basin (679 and 645 kg N km^{-2} yr^{-1}, respectively) correspond to large average annual precipitation in these basins (1429 and 1374 mm yr^{-1}, respectively), while the low average annual DIN fluxes from the Krishna basin (43 kg N km^{-2} yr^{-1}) are consistent with the low average annual precipitation in this basin (753 mm yr $^{-1}$). Overall, average annual precipitation explains 73% of the spatial variability in average annual DIN fluxes across the seven basins (diamonds in Figure 4A).

Year- and basin-specific annual precipitation also explains 52% of the combined spatial and interannual variability across years and basins (dots in Figure 4A, Table S4). Even when considering the superset of possible predictor variables described earlier (Table S5), the best linear model for representing variability across years and across the seven basins consists of annual precipitation combined only with a linear trend with time, which together explain 57% of the observed variability (Table S6). Within specific basins, annual precipitation explains 20% (Krishna basin) to 81% (Godavari basin) of the interannual variability in DIN fluxes. We also conducted a sensitivity analysis using precipitation from the Climate Prediction Center (see Materials and Methods) and found a similar explanatory power of annual precipitation within specific basins (22% to 59%) and combined spatial and interannual variability across basins (57%) (Table S4).

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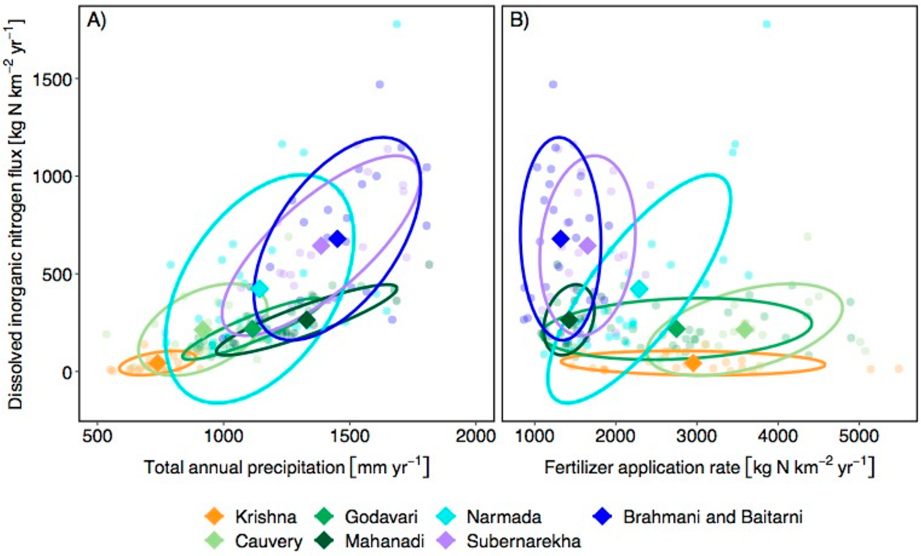

Figure 4. Annual dissolved inorganic nitrogen fluxes (Q_{DIN}) for the seven examined basins presented as a function of (A) total annual precipitation (P_{Annual}) and (B) fertilizer application rate (N_{Fert}) for the corresponding basin and year. Ellipses represent one standard deviation of the variability across years for a given basin, dots mark the annual values for a given basin, and diamonds represent the mean value across years for a given basin.

Looking beyond the seven basins examined here, we find that precipitation exhibits large interannual variability for all river basins in India (Figure 1B), such that high interannual variability in DIN fluxes is expected across the whole country. This is consistent with research showing large interannual variability but no clear long-term trend in coastal anoxic events in India over the last 50 years,[36,37] which is in contrast to observed decreasing trends in dissolved oxygen concentrations along the United States and European coasts since 1950s.[38]

The high sensitivity of DIN fluxes to annual precipitation suggests that projected climate-changed-driven increases in precipitation in India[14−17,31] will lead to increased riverine nitrogen loading, presenting additional challenges to sustainable management in the region. As has been observed for the Baltic Sea,[39] this could contribute to exacerbated hypoxic or anoxic conditions in Indian waters. Therefore, the impact of future changes in precipitation patterns must be considered in developing resilient management strategies for reducing nutrient pollution in India.

3.3. Role of Fertilizer Use. We find that historical increases in fertilizer application rates over the period considered here have not resulted in higher DIN flux for most river basins in southern India. It is likely, however, that fertilizer application would play a larger role on longer time scales. We find that within specific basins fertilizer application rate is only correlated with annual DIN fluxes for the Narmada and Cauvery basins (Figure 4B). Furthermore, contrary to findings for watersheds in the United States,[40,41] Indian basins with higher average fertilizer application rates (e.g., Krishna and Cauvery basins) actually have lower DIN fluxes relative to basins with lower fertilizer application rate (e.g., Brahmani and Baitarni basin and Subernarekha basin) (Figure 4B). This discrepancy is caused by the large spatial variability in precipitation (Figure 1A, 4A), with basins with the highest fertilizer application rates experiencing some of the lowest precipitation rates, and vice versa.

Prior literature has also alluded to the fact that a large fraction of the nitrogen input in India is lost to the atmosphere as ammonia and nitrous oxide emissions, converted back to nitrogen gas via denitrification, or retained in groundwater as nitrate.[42,43] Our results support these findings, suggesting that agricultural intensification in India is likely impacting groundwater quality and atmospheric emissions more so than riverine fluxes. This hypothesis is also supported by observations of high nitrate concentrations in groundwater in various states across India[44,45] and by increasing ammonia and nitrous oxide emissions that have been attributed to agricultural intensification.[46−49] Quantifying nitrogen losses to groundwater and the atmosphere as well as nitrogen export via agricultural products at a basin scale would make it possible to attribute the increase in fertilizer application to each of these four end points but is beyond the scope of the current study.

Fertilizer application rates in India are projected to increase further over the next several decades across five of the six shared socioeconomic pathways[13] developed in support of the Coupled Model Intercomparison Project Phase 6 (CMIP6) climate simulations,[50] and these continuing increases in fertilizer application rates may change how agricultural intensification impacts riverine DIN fluxes.

3.4. Explanatory Factors for Specific Basins. We find that the degree to which climatic variables (here, precipitation and temperature) and nitrogen inputs (here, fertilizer application and NO_X deposition) explain interannual variability in DIN fluxes varies substantially across the seven examined basins (Figure S1, Table S6). These factors explain much of the variability for the Godavari, Subernarekha, Mahanadi, and Narmada basins (64−91%). For the Godavari basin, annual precipitation and seasonal temperature explain a large fraction of the interannual variability, while for the Subernarekha basin seasonal precipitation for the months of June, July, August, and September (JJAS) alone explains the vast majority of the interannual variability. For the Mahanadi and Narmada basins, seasonal precipitation in JJAS in conjunction with nitrogen inputs explains more than 75% of the interannual variability. Both fertilizer application rates and NO_X deposition have increased over time (Figures S2 and S3), and we found them to be important factors for explaining interannual variability for the Narmada and Mahanadi basins.

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This increasing trend is also reflected in the flow-normalized DIN concentrations and load for these basins (Figure S4).

Conversely, the examined climatic factors and nitrogen inputs (Table S5) explain less than half of the interannual variability in DIN fluxes for the Brahmani and Baitarni, Cauvery, and Krishna basins (38−40%) (Figure S1, Table S6). These results imply that, for these three basins, other variables such as population changes, wastewater effluent, agricultural nitrogen fixation, food and feed import, soil characteristics, or water resource management likely explain the remaining interannual variability. In addition to precipitation, seasonal temperature is also found to be important for the Godavari and Krishna basins. The temperature variables are associated with negative drift coefficients, indicating a decrease in DIN fluxes with increasing temperatures. This is likely attributable to increased evapotranspiration leading to lower river discharge as well as an increase in loss of reactive nitrogen via denitrification.[51]

3.5. Study Limitations. Relative to regions such as the United States, where nitrogen runoff has been the subject of long-term monitoring and study, available concentration measurements for the Indian river basins are relatively sparse (see Supporting Information S1). In addition, the lack of available data on related quantities or processes, such as wastewater effluent discharged to agricultural lands, agricultural nitrogen fixation and food and feed import limits the degree to which total anthropogenic nitrogen inputs can be quantified. A full accounting of the fate of applied nitrogen is therefore beyond the scope of the current work. The dominant role of precipitation in explaining the spatial and temporal variability in nitrogen fluxes is clear, however, as is the lack of a large increase in riverine runoff in response to historical increases in fertilizer application.

4. IMPLICATIONS FOR WATER QUALITY MANAGEMENT UNDER CHANGING CLIMATE AND LAND USE

We find that large seasonal and interannual variability exists in DIN fluxes across Indian rivers basins and that a large fraction of this variability can be explained by seasonal or extreme seasonal precipitation. These findings imply that strategies for reducing DIN loading must explicitly consider the role of interannual variability in meteorological conditions. In addition, the strong influence of climatic factors means that water quality management strategies aimed at reducing the occurrence of impairments such as harmful algal blooms and coastal hypoxia must be resilient to climatic variability and change.

Our results suggest that in highly managed river basins in southern India, fertilizer application rates have had only a secondary effect on riverine nitrogen fluxes since the 1980s, although this may change in the future as fertilizer application rates continue to rise. We speculate that historical increases in fertilizer application rates have likely negatively impacted groundwater quality and atmospheric nitrogen emissions more so than riverine fluxes; additional research is needed to substantiate the scale of this impact. The results also point to the differential impact of fertilizer application on DIN fluxes for various river basins and, therefore, reinforce the need for basin-specific management strategies.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c01274.

(S1) Application of WRTDS approach; (S2) sensitivity analyses for individual basins; (Figure S1) contribution of predictor variables to modeled dissolved inorganic nitrogen fluxes (Q_{DIN}) for the seven examined basins; (Figure S2) fertilizer application rates for Indian river basins; (Figure S3) NO_x deposition rates for Indian river basins; (Figure S4) Mann−Kendall trend test on flow-normalized concentration and flow-normalized load for the Narmada and Mahanadi basins; (Figure S5) observed vs WRTDS-estimated natural log of DIN concentration; (Figure S6) comparison of R^2 values between observed vs WRTDS-estimated values; (Table S1) estimates of annual dissolved inorganic nitrogen flux (Q_{DIN}) [kg N km^{-2} yr^{-1}] for major river basins as reported here and in earlier studies; (Table S2) discharge and dissolved inorganic nitrogen (DIN) observations used for quantifying annual fluxes and characteristics of monthly DIN fluxes for each of the seven examined basins; (Table S3) characteristics of the river basins including population, annual rainfall, maximum and minimum temperature, and land use; (Table S4) comparison of R^2 between WRTDS-estimated annual dissolved inorganic nitrogen flux and observed annual discharge and total annual precipitation; (Table S5) predictor variables evaluated; (Table S6) linear model for representing dissolved inorganic nitrogen fluxes in each of the seven basins; (Table S7) percentage of the area of Indian states falling within each of the seven examined basins (PDF)

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Author Contributions

E.S. and A.M.M. designed the study, performed the research, and analyzed the data with input from V.B. and L.R. All authors contributed to the interpretation of results and writing of the manuscript.

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Notes

The authors declare no competing financial interest.

Data and Materials Availability. All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supporting Information. Additional data and code related to this paper were archived in the following GitHub repository https://github.com/evasinha/Sinha_ES-T_2022.

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