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Modeling and forecasting riverine dissolved inorganic nitrogen export using anthropogenic nitrogen inputs, hydroclimate, and land-use change

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SUMMARY

A quantitative understanding of riverine nitrogen (N) export in response to human activities and climate change is critical for developing effective watershed N pollution control measures. This study quantified net anthropogenic N inputs (NANI) and riverine dissolved inorganic N (DIN = NO_3–N + NH_4–N + NO_2–N) export for the upper Jiaojiang River catchment in eastern China over the 1980–2010 time period and examined how NANI, hydroclimate, and land-use practices influenced riverine DIN export. Over the 31-yr study period, riverine DIN yield increased by 1.6-fold, which mainly results from a ~77% increase in NANI and increasing fractional delivery of NANI due to a ~55% increase in developed land area. An empirical model that utilizes an exponential function of NANI and a power function of combining annual water discharge and developed land area percentage could account for 89% of the variation in annual riverine DIN yields in 1980–2010. Applying this model, annual NANI, catchment storage, and natural background sources were estimated to contribute 57%, 22%, and 21%, respectively, of annual riverine DIN exports on average. Forecasting based on a likely future climate change scenario predicted a 19.6% increase in riverine DIN yield by 2030 due to a 4% increase in annual discharge with no changes in NANI and land-use compared to the 2000–2010 baseline condition. Anthropogenic activities have increased both the N inputs available for export and the fractional export of N inputs, while climate change can further enhance riverine N export. An integrated N management strategy that considers the influence of anthropogenic N inputs, land-use and climate change is required to effectively control N inputs to coastal areas.

1. Introduction

Excessive nitrogen (N) in rivers from anthropogenic activities is of increasing concern worldwide as it degrades aquatic ecosystem health, decreases water quality for beneficial uses, and causes eutrophication and hypoxia in many coastal ecosystems (Edwards and Withers, 2008; Howden et al., 2011; Howarth et al., 2012; Chen et al., 2013). The extent of riverine N export is dependent on various factors, such as anthropogenic N inputs (Howarth, 2008), hydroclimate (McIsaac et al., 2001; Han et al., 2009), and land-use/land-management practices (Groffman et al., 2004; Kaushal et al., 2008; Sobota et al., 2009). Therefore, to effectively guide N management for protecting aquatic ecosystem health, models that predict riverine N export must be responsive to changes in these factors.

Net anthropogenic nitrogen input (NANI) is a nitrogen-budgeting approach that sums N contributions from atmospheric deposition, fertilizer application, agricultural biological fixation, and net import/export of N in food, feed and seed to a watershed (Howarth et al., 2012; Han et al., 2014). NANI has been widely recognized as an effective tool to explain among-watershed or among-year variations of riverine N fluxes (McIsaac et al., 2001; Howarth, 2008; Howarth et al., 2012; Han et al., 2009). Although the relationship between NANI and riverine N export is generally strong, such a relationship is additionally influenced by variations in hydroclimate and land-use (Han et al., 2009; Sobota et al., 2009; Howden et al., 2010). It is commonly observed that years with higher precipitation or discharge exported a higher fraction of NANI by rivers, whereas drier years exported a lower fraction of NANI (Howarth, 2008; Han et al., 2009; Schaefer et al., 2009).

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Therefore combining the influence of NANI with either precipitation or discharge can generally explain the variability in mean N flux among watersheds or years with a higher accuracy (Howarth et al., 2006; Han et al., 2009; Swaney et al., 2012). Furthermore, fractional export of NANI by rivers varies with land-use, with higher factional export generally observed in urban or suburban watersheds than in agricultural or forest watersheds, due to increased impervious surfaces and reduced hydrologic residence times that reduce retention capacity during N transport (Groffman et al., 2004; Kaushal et al., 2008; Elmore and Kaushal, 2008). However, based on our review of the literature there is a paucity of studies that simultaneously considered the influence of NANI, hydroclimate, and land-use on riverine N exports. Such an integrated approach is required for watersheds or regions that have experienced extensive changes in land-use/land-cover, climate and/or agricultural practices over time.

A number of studies indicated that 10–40% of multi-year averaged NANI was exported by rivers (Howarth et al., 2012; Swaney et al., 2012), implying that a considerable proportion of NANI was temporally stored in aquifers, soils and/or biomass (∼27%), although a higher proportion was attributed to denitrification (∼57%) (Van Breemen et al., 2002). These stored N sources, which are not addressed by the NANI budget approach (Swaney et al., 2012), have the potential to release N to rivers in following years (Stålnacke et al., 2003). For example, studies have shown that 25–40% of the annual riverine N loss may originate from the mineralization of soil organic N (Booth et al., 2005; Kopáček et al., 2013) and nitrate exported from groundwater could account for 35–40% of the riverine N flux (Iqbal, 2002; Lindsey et al., 2003). In the Mississippi River, annual riverine nitrate flux was determined by NANI to the watershed from the previous 2–9 yr (McIsaac et al., 2001). However, limited knowledge is available on what proportion of riverine N export in a given year or period is derived from the storage of NANI from previous years. Such information is critical to better understand how watershed N sinks and riverine N exports change in response to changes in NANI and land-use, as well as climate change over extended time periods (Swaney et al., 2012).

Since the 1980s, Chinese rivers, such as the Yangzi River, Yellow River and Zhujiang River, have experienced a significant increase in N concentration and flux associated with rapid economic development, human population expansion, and urbanization (Duan et al., 2000; Xia et al., 2002; Liu et al., 2003; Li et al., 2007; Shen and Liu, 2009). Due to the lack of long-term river N monitoring records, historical N trend analyses have been limited to these major rivers, while little information is available for smaller rivers, which may experience appreciably different N source-sink dynamics. Examining trends in riverine N flux in response to changes in N inputs and land-use is especially important for watersheds in eastern China that have experienced the most rapid economic and society development in China since the 1980s. For these coastal waters along the East China Sea, excessive dissolved inorganic N (i.e., DIN = NO_3–N + NH_4–N + NO_2–N, which is the most bioavailable form of N) from coastal rivers has resulted in serious algal blooms and persistent hypoxia (Duan et al., 2000; Li et al., 2007; Gao and Zhang, 2010). Furthermore, although the relationship between NANI and riverine N export has been widely examined in American and European watersheds, few attempts have been conducted for Chinese watersheds. Accordingly, it is urgent to update and expand the quantitative knowledge of long-term riverine DIN trends in response to changes in NANI and land use over the past several decades for watersheds in eastern China to support the development of efficient coastal N pollution control strategies.

This study investigated a 31-yr record (1980–2010) of riverine DIN exports in response to changes in NANI, hydroclimate, and land use in the upper Jiaojiang catchment (2474 km^2) in eastern China; the third largest river of Zhejiang Province that ultimately flows into the East China Sea. The study is based on detailed data from long-term monitoring of riverine NH_4–N, NO_3–N, and NO_2–N concentrations and discharge, land-use, precipitation, and anthropogenic N sources. The primary objectives of this study were to (i) develop an empirical model of river DIN export that simultaneously incorporates the influences of NANI, hydroclimate, and land use, (ii) use the model to identify individual contribution from NANI, watershed N storage, and background sources to riverine DIN export, and (iii) predict future (2011–2030) trends in riverine DIN export based on scenarios for future changes in NANI, land use and climate. This study will inform development of N management strategies to effectively control N inputs to coastal waters experiencing persistent eutrophication and hypoxia.

2. Materials and methods

2.1. Watershed description

The upper Jiaojiang catchment (120° 13′ 46.065″–121° 0′ 52.464″E and 28° 28′ 10.118″–29° 2′ 22.156″N; elevation ∼15–1000 m) is located in the highly developed Taizhou City area of Zhejiang Province, China (Fig. 1). The Jiaojiang River is the third largest river of Zhejiang Province and flows into Taizhou Estuary and the East China Sea, a coastal area that commonly experiences hypoxia (Duan et al., 2000; Li et al., 2007; Gao and Zhang, 2010). The sampling location (Fig. 1) for this study was 55 km upstream of Taizhou Estuary at an elevation of ∼15 m. The river drains a total area of 2474 km^2 and has an average annual water depth of 5.42 m and discharge of 72.9 $m^3\ s^{-1}$ at the sampling location. The climate is subtropical monsoon having an average annual temperature of 17.4 °C and average annual precipitation of 1400 mm (Fig. 2a). The rainfall mainly occurs in May–September with a typhoon season occurring in July–September. Agricultural land (including paddy field, garden plot, and dryland) averaged ∼12% of total watershed area in 1980–2010, with developed land (including rural and urban residential lands, roads, and mining and industry lands) and woodland, and barren land (including water surface, swamp, rock, and natural reservation land) contributing ∼3%, ∼67%, and ∼18%, respectively. Developed land area was increased by ∼55% since 1980 (Fig. 2b). The economic role of agriculture has been increasingly replaced by industry since the 1990s, resulting in a large reduction in chemical N fertilizer application (∼40%) and cultivated crop area (∼20%) since 2000 (Fig. 2b). Total population within the catchment increased from ∼590,000 in 1980 to ∼740,000 in 2010. Over the 31-yr study period, domestic livestock production (pig, cow, sheep, and rabbit) decreased by ∼25%, poultry production (chicken and duck) increased by 4.8-fold (Fig. 2c), and freshwater aquatic species production (fish and shrimp) increased by 11.7-fold.

2.2. Riverine DIN flux estimate

River water samples were collected once every 4–8 weeks (n = 300 sampling times total) at Baizhiao station from 1980 to 2010 (Fig. 1) by the Taizhou City Environment Protection Bureau. A well-mixed composite water sample (surface and bottom layers at three sites along the cross section) was collected between 8:00 and 9:00 on each sampling date, and nitrogen forms were analyzed by the Taizhou City Environment Protection Bureau. Duplicate samples from the composite sample were acidified with H_2SO_4 in the field (10 ml of concentrated H_2SO_4 per liter) and filtered and analyzed within 4 h of sampling. The concentration of NO_3^- in water sample was determined by the spectrophotometric phenol disulfonic acid method (limit of detection: LOD = 0.01 mg N L^{-1});