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Research article

Hydrological controls on nitrogen (ammonium versus nitrate) fluxes from river to coast in a subtropical region: Observation and modeling

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A B S T R A C T

Increased anthropogenic nutrient input and losses has caused eutrophication problems in freshwater and coastal ecosystems worldwide. High-frequency observations and modeling of river fluxes in subtropical regions are required to understand nutrient cycling and predict water quality and ecological responses. In 2014, a normal hydrologic year, we carried out daily sampling of the North Jiulong River in southeast China, which drains an agricultural watershed and experiences the Asian monsoon climate. We focused on the distinct characteristics of two important inorganic nitrogen forms (ammonium and nitrate). Our results show contrasting hydrological controls on the seasonal timing and magnitude of ammonium and nitrate concentrations and loads, likely due to differing sources and transport pathways (surface runoff versus baseflow) to the river. Both nitrogen concentrations were enriched in the dry season and diluted in the wet season. Arrival of rains in the pre-wet period in March caused a "first flush" peak event with the highest concentrations of the year, during which ammonium peaked two weeks earlier than nitrate. In contrast, the majority of nitrogen transport occurred during the lower concentrations of the wet season, with seven storms inducing flood events that lasted 24% of the time, contributed 52% of the runoff, and exported 47% of the ammonium and 42% of the nitrate. We found that seasonally piecewise LOADEST models (for pre-wet, wet and post-wet periods) performed better (5–8% error) than a year-round model (12–24% error) in estimating monthly nitrogen loads. However, not all nitrogen dynamics are easily synthesized by this approach, and extreme floods might produce a greater deviation in estimating nitrogen loads. These findings represent important implications for coastal ecology and provide opportunity on improving observation and modeling.

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1. Introduction

Human activities and climate change have caused estuaries and coastal ecosystems worldwide to receive increased riverine nutrient loads (Howarth, 2008; Paerl, 2006; Peñuelas et al., 2013; Strokal et al., 2016; Turner and Rabalais, 1994). Increasing nitrogen (N), principally in the form of inorganic N (nitrate, nitrite, and ammonium) has led to a series of ecological and environmental problems (e.g. water quality deterioration, eutrophication, and harmful algal blooms) that present large threats to socio-economic development and human health (Billen and Garnier, 2007; Davidson et al., 2014; Paerl et al., 2011; Reed et al., 2016). Many studies have demonstrated that wet season and accompanied storm events contribute to the majority of annual nutrient loading to river and coastal water (Adame et al., 2016; De Carlo et al., 2007; Zhang et al., 2009). Aquatic ecosystems are sensitive to the changing nutrient loading and their elemental ratios over seasons and storm events (Chen et al., 2015; Cornelis et al., 2011; Corriveau et al., 2013; Sigleo and Frick, 2007). River N loading to the coastal waters of China have been predicted to increase continually through 2050 (Strokal et al., 2014). Anthropogenic factors (agriculture development, sewage discharge, dam construction, and land use change) within watersheds and climate factors (watershed precipitation and discharge) synergistically affect the magnitude and timing of nutrient loading (Johnson et al., 2007; Statham, 2012). Coastal areas in China are affected by the Asian monsoon climate which is characterized by strong seasonal variations in

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precipitation and frequent storm events. Therefore, observation and modeling of river nutrient fluxes at the seasonal scale has become an important basis for managing watershed nutrients and mitigating coastal eutrophication.

Several models have been developed to simulate river N load. For instance, SWAT (Soil and Water Assessment Tool) estimates nutrient yields in surface runoff and identifies critical source areas in watershed and large river basins (Arnold et al., 1998). WASP (Water Quality Analysis Simulation Program) has been used to estimate the influence of nutrient loads on the water quality of Tampa Bay, U.S.A (Wang et al., 1999). HSPF (Hydrological Simulation Program-Fortran) quantifies N exported annually from the catchment of the Great Barrier Reef, Australia (Hunter and Walton, 2008). These models require detailed, high quality input data which are often difficult to obtain in large catchments. The Load Estimator (LOADEST), a statistical model developed by USGS, makes use of limited flow and water quality datasets to set up a regression formula of corresponding pollutant fluxes, and then estimates their loads at different time scales (Runkel et al., 2004). The LOADEST model is considered to be a useful management tool and has been widely applied to river N load estimations. Johnson et al. (2007) explored nitrate fluxes of Fall Creek watershed in central New York during 1972–2005. Mast, (2013) used LOADEST to analyze temporal trends in stream N over two periods (1970–2010 and 1990–2010). Pellerin et al. (2014) explored the ability of high frequency measurements of nitrate concentration to improve the accuracy of monthly nitrate loads. The LOADEST model has also been optimized by adjusting sampling frequency and obtaining storm samples (Duan et al., 2014; Park and Engel, 2014; Park and Engel, 2015; Toor et al., 2008). In contrast to this abundance of previous work to estimate nitrate loading, modeling efforts on river ammonium loads are scarce. Considering ammonium and nitrate separately, rather than just total dissolved inorganic N is important because they can have different sources, biogeochemical behaviors, and potential ecological effects (Domingues et al., 2011; Dugdale et al., 2007; Tamminen, 1995).

We carried out daily sampling in a hydrologic year (2014) to better understand the hydrological processes controlling seasonal and flood-event N fluxes from the main (North) branch of the Jiulong River, which drains an agricultural watershed. We focused on determining the contrasting characteristics and behaviors of two inorganic N forms (ammonium versus nitrate) under the extreme rainfall variations that occur in the Asian monsoon climate. We first present seasonal patterns of nitrate and ammonium concentrations and loads, and then use the LODEAST model approach to determine total fluxes from the river, including developing and evaluating a seasonally piecewise LOADEST model (for the pre-wet, wet, and post-wet periods). Finally, the hydrological controls on river N (ammonium versus nitrate) export are summarized, and perspectives on future observations and modeling of N loads are discussed.

2. Materials and methods

2.1. Description of study site

The Jiulong River watershed is located in southeast China, comprised of the North, West and South Rivers (Fig. 1). The North River is the main branch, with a drainage area of 9570 km^2. Its length to the mouth (Jiangdong) is around 274 km, traversing an elevation change of 325 m. The topography in the North Jiulong River (NJR) is mostly hilly (91% of the catchment area has an altitude greater than 200 m). Land use is comprised of 78% forest (mostly artificial), 16% arable land, 3% urban and residential land, 2% water, and 1% bare and grassland (based on information from the 2007 Landsat Thematic Mapper). Six major dam reservoirs have been constructed in the main branch of the river mainly for hydropower generation and flood control (Wang et al., 2010). The NJR flows through four cities/counties (Longyan, Zhangping, Hua'an and Changtai) and a part of Zhangzhou city, with approximately 1.5 million people in total. Longyan city is located furthest upstream, where animal farming has become the dominant industry since the 1990s. The other counties are downriver and comprised of predominantly agricultural and forest land. The Jiulong River and adjacent estuary have suffered nutrient (N and P) enrichment and eutrophication problems during the past three decades (Chen et al., 2013; Li et al., 2011; Yan et al., 2012).

2.2. Daily sampling and laboratory analysis

Daily surface water samples in 2014 were manually collected at the mouth of the NJR (at the Jiangdong auto monitoring station, operated by Xiamen Environmental Monitoring Center). About 50 mL of water was filtered immediately (0.45 μm membrane) after each sampling, and stored frozen at −20 °C at Jiangdong until monthly delivery to Xiamen University for analysis. The thawed water samples were analyzed by segmented flow automated colorimetry using the manufacturer's standard procedures (San++ analyser, The Netherlands) for ammonium (NH_4–N) and the sum of nitrate and minor nitrite (called NO_3–N here). Dissolved inorganic nitrogen (DIN) was summed from NO_3–N + NO_2–N + NH_4–N. Sampling, preservation and transportation of the water samples to the laboratory were conducted by standard methods for surface waters (American Public Health Association, 2005). The precision for the DIN components was estimated by repeated determinations of 10% of the samples and the relative error was 3–5%. Commercial standard reference materials were used to check the instrument performance.

2.3. Auxiliary data collection and data analysis

Hourly rainfall records from four weather stations within the catchment were obtained from Weather China (http://www.weather.com.cn/). Daily river discharge was obtained from a nearby hydrological station (Punan) and was extrapolated to the sampling site (Jiangdong) using the ratio (1.08) of the catchment area between them (Fig. 1). Baseflow was estimated using an automatic segmentation procedure (BFI (F): Smoothed Minima method). The designation of a flood event was applied only to flow increases that exceeded 2 times the average discharge of 2014. Flood events were defined as starting and ending when discharge was more than 1.2-times the previous baseflow. Daily observed loads (OL) were calculated as products of the concentration and river discharge. The sums of discrete daily loads over various periods (flood events, months, seasons and annual) were used as references for evaluation of model performance.

2.4. LOADEST model calibration and evaluation

LOADEST assists researchers to develop a regression model to estimate constituent loads in rivers and streams. The model is formulated using seven parameters representing river discharge, time-trends, and seasonality (Eq. (1)) (Runkel et al., 2004).

$$\ln(L) = a_0 + a_1 \ln Q + a_2 \ln Q^2 + a_3 \sin(2\Pi \mathrm{dtime}) + a_4 \cos(2\Pi \mathrm{dtime}) + a_5 \mathrm{dtime} + a_6 \mathrm{dtime}^2 \quad (1)$$

where L is load; Q is stream flow or river discharge; dtime is decimal time; a_0 to a_6 are coefficients. The expressions lnQ and dtime are centered over the studied period to avoid multi-collinearity. For

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