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## Warming increases nutrient mobilization and gaseous nitrogen removal from sediments across cascade reservoirs

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## ABSTRACT

Increases in water temperature, as a result of climate change, may influence biogeochemical cycles, sediment-water fluxes and consequently environmental sustainability. Effects of rising temperature on dynamics of nitrate, nitrite, ammonium, dissolved inorganic nitrogen (DIN), dissolved reactive phosphorus (DRP), dissolved organic carbon (DOC) and gaseous nitrogen (N<sub>2</sub> and N<sub>2</sub>O) were examined in a subtropical river (the Jiulong River, southeast China) by microcosm experiments. Slurry sediments and overlying water were collected from three continuous cascade reservoirs, and laboratory incubations were performed at four temperature gradients (5 °C, 15 °C, 25 °C and 35 °C). Results indicated: (1) warming considerably increased sediment ammonium, DIN and DOC fluxes to overlying water; (2) warming increased retention of nitrate, and to a lesser extent, nitrite, corresponding to increases in N<sub>2</sub> and N<sub>2</sub>O emission; (3) DRP was retained but released from Fe/Al-P enriched sediments at high temperature (35 °C) due to enhanced coupled transformation of carbon and nitrogen with oxygen deficiency. Using relationships between sediment fluxes and temperature, a projected 2.3°C-warming in future would increase ammonium flux from sediment by 7.0%–16.8%, while increasing nitrate flux into sediment by 8.9%–28.6%. Moreover, substrates (e.g., grain size, carbon availability) influenced nutrient delivery and cycling across cascade reservoirs. This study highlights that warming would increase bioreactive nutrient (i.e., ammonium and phosphate) mobilization with limited gaseous N removal from sediments, consequently deteriorating water quality and increasing eutrophication with future climate change.

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### 1. Introduction

Since the middle of the last century, the freshwater systems have been seriously disrupted by increasing human disturbance. According to the International Commission on Large Dams (ICOLD) in 2013, more than 68,000 larger dams (over 15 m high) have been constructed worldwide in rivers for hydropower generation, irrigation, and flood control, with around 38,000 located in China. Typically, cascade hydroelectric dams have been built and the whole river becomes a regulated “lake-like” system. Barbosa et al. (1999) put forward the “Cascading Reservoir Continuum Concept” (CRCC), proposing a theoretical framework for addressing ecological processes in such systems. Reservoirs act as sources of

greenhouse gases, N<sub>2</sub>O and CH<sub>4</sub> (Giles, 2006; Guérin et al., 2006; Rosa et al., 2004), and dam construction can alter nutrient biogeochemical and ecological processes (Beusen et al., 2009; Conley et al., 1993; Garnier et al., 2010; Harrison et al., 2012; Hartmann and Moosdorf, 2012; Humborg et al., 2000; Maavara et al., 2014; Teodoru and Wehrli, 2005) due to changes in hydraulic residence time, turbidity and oxygen saturation (Friedl and Wuest, 2002; Klaver et al., 2007; McGinnis et al., 2006; Teodoru and Wehrli, 2005). A number of studies have shown that there have been long-term warming water temperatures in streams and rivers due to interactive effects of global warming and watershed land use change, e.g., urbanization (Kaushal et al., 2010; Webb and Nobilis, 2007). However, little is known about how cascade hydroelectric dams change nutrient transformation in coastal rivers and thereby influence nutrient fluxes from land to the ocean in response to warming (Liu et al., 2015; Ouyang et al., 2010).

Sediments can act “sinks” or “sources” of nutrient elements

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(Havens et al., 2001) by retaining nutrient compounds through abiotic adsorption, biotic uptake (Arango et al., 2008; Bowes et al., 2003; Kaplan et al., 2008) or release back to the water column through organic matter decomposition or reduction (Aguilar and Thibodeaux, 2005; Jensen and Andersen, 1992). There are many factors influencing biogeochemical transformations near the water-sediment interface, including temperature, organic content, redox conditions, grain size, etc. Temperature is an important factor influencing nutrients biogeochemical cycles (Dalsgaard and Thamdrup, 2002; Duan and Kaushal, 2013; Gudasz et al., 2010; House and Denison, 2002; Liikanen et al., 2002). River water temperature varies seasonally and impacts many aspects of river ecosystem (Webb and Nobilis, 2007). High temperatures increase microbial activities in the sediments to promote mineralization of organic nutrients (Cornelissen et al., 1997; Dalsgaard and Thamdrup, 2002), decrease the thickness of the oxidized surface layer and thereby induce release of Fe-bound P (Jensen and Andersen, 1992), and release of  $\text{NH}_4\text{-N}$  as inhibited nitrification (Liikanen et al., 2002). Moreover, mineralization of labile organic matter in response to warming consumes dissolved oxygen, and thereby inhibits nitrifying bacteria but promotes denitrification (Van Luijn et al., 1999). Other factors, including pH and grain size, also influence the transformation of nutrients in sediments (Jin et al., 2006; Zhang et al., 2002).

Previous studies have investigated the effects of temperature on sediment nutrient fluxes and transformation in rivers, lakes or reservoirs (Duan and Kaushal, 2013; Gudasz et al., 2010; Wang et al., 2015; Xue and Lu, 2015), but it is still not well known how warming affects the transformation and fate of nutrients, especially nitrogen. Few investigations have been conducted to explore the effects of temperature on the coupled nutrient biogeochemical cycles across cascade reservoirs. According to the IPCC fifth assessment report (AR5), the projected land surface temperature will increase by  $3\text{ }^\circ\text{C}$ – $5\text{ }^\circ\text{C}$  during 2081–2100 in South Asia (Stocker et al., 2013). Given the nonlinear relationship between stream temperature and air temperature, future water temperatures could be the primary factor impacting aquatic ecosystems (Caissie et al., 2001; Mohseni et al., 1998; Webb et al., 2003). In the context of global warming, we tested three hypotheses: (1) high water temperatures would accelerate sediment transformations and fluxes of ammonium, nitrate, nitrite, dissolved reactive phosphorus (DRP), and dissolved organic carbon (DOC) to streamwater as a result of organic matter decomposition; (2) cascade dam reservoirs are likely to produce more  $\text{N}_2\text{O}$  and  $\text{N}_2$  emission to atmosphere with an increase in temperature; and (3) temperature effects on sediment nutrients mobilization vary across cascade reservoirs due to differences in sediment grain size and organic matter. An improved conceptual understanding of warming potential on fluxes of major bioreactive elements (N, P and C) can contribute to our understanding of drivers of change in water quality and ecosystem functions with cascade dam construction and global climate change.

## 2. Materials and methods

### 2.1. Study area

The Jiulong River is a subtropical river with a drainage area of  $14,741\text{ km}^2$  located in southeastern China and influenced by a monsoon climate. Long-term water temperatures in this area are generally within  $7\text{ }^\circ\text{C}$ – $31\text{ }^\circ\text{C}$ . Two main tributaries (North River and West River) discharge water into Xiamen Bay through the estuary and the total discharge is  $12.4 \times 10^9\text{ m}^3\text{ y}^{-1}$ , of which the North River accounts for nearly two-thirds. Annual precipitation varies from 1400 to 1800 mm, 75% of which occurs between April and

October. Hydrology can be characterized as a shift from medium flow in October to low flow in December. The North River is 272 km long and has a catchment area of  $9562\text{ km}^2$ . The upper North River receives a large amount of human waste when passing through two large cities (Longyan and Zhangzhou), and widespread livestock produce a large amount of animal wastes and extreme nutrient pollution (Chen et al., 2013, 2014b). Six large cascade dams along the main stem have been constructed since the late 1990s. The main soil type on hilly land (accounting for 62% of the total catchment) is lateritic red soil (one of the soils forming in subtropical and tropical regions and rich in  $\text{Fe}_2\text{O}_3/\text{MnO}_2$ ), followed by paddy soil alongside rivers and latosolic red soil on the valley floor.

In this study, temperature gradient incubation experiments were conducted for three cascade dam reservoirs in the North River [upper site: Xiaoqi reservoir (XQ), middle site: Xipi reservoir (XP), lower site: Mianliang reservoir (ML)]. XQ, XP and ML have a mean channel length of 11, 8.5 and 9.9 km, and a water capacity of 19.30, 17.10 and 12.74 million  $\text{m}^3$ , respectively. Average water depths are all less than 16 m and there is no obvious temperature stratification. The largest temperature difference between surface and bottom water was less than  $2\text{ }^\circ\text{C}$  in summer. Recent monitoring records (2012.4–2016.4) at XP station indicate that daily mean water temperature ranged from  $10.4\text{ }^\circ\text{C}$  to  $30.2\text{ }^\circ\text{C}$ .

### 2.2. Sample collection and processing

In subtropical areas with Asian monsoon climate, a large amount of fresh sediment can be settled in reservoirs during the flood season (May–September) with subsequent mineralization in the dry season (October–January). Therefore, sediment samples were comparatively studied on October 18 (soon after flood season) and December 24, 2015, when the rivers were at medium flow and low flow conditions, respectively. Surface (10 cm) sediment samples were collected 1 km upstream of the dam with a Petite Ponar grab sampler from the three cascade dam reservoirs and were well-mixed after three times sampling on both sides of the boat. Fifteen liters of river water from XP were collected for later incubation experiments and 300 mL water samples from three reservoirs were collected for water quality analysis. Water temperature and pH was measured in-situ using a WTW TetraCon<sup>®</sup> 325 probe. All the sediment samples were placed into polyethylene bags, kept cold on ice and then transported to the lab. In the lab, sediments were hand-sieved through a 2-mm sieve, and the fractions  $<2\text{ mm}$  were homogenized and kept cold for incubations. In December, overlying water at each site was sampled. All fresh sediments were subjected to centrifugation (5000 rpm, 10 min), and the extracted water was filtered through 25 mm Whatman GF/F filters in a syringe filter. The filtrates (pore water) were stored in a refrigerator. Sediments were dried in a freeze-drier, homogenized and ground in an agate mortar, and the powder was collected for chemical analyses.

### 2.3. Sediment incubations

The sediments were incubated at 5, 15, 25, and  $35\text{ }^\circ\text{C}$  in temperature control incubators. A total of 200 g wet sediment and 450 mL of unfiltered river water were placed in each incubator. After reaching the predefined temperature over-night, the water was carefully added to the sediment in the 500 mL glass flasks. Meanwhile, blank controls without sediment were prepared in 500 mL flasks. Four sets of flasks (4 samples plus controls for each set) were wrapped with aluminum foil and then placed in four temperature incubators. During the 2-day incubation, water samples for nutrients analysis were collected at 0, 6, 12, 24, and 48 h increments and filtered immediately through 25 mm Whatman GF/F filters in a syringe filter. The filtrates were stored in

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