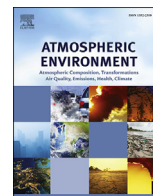




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## Effect of watershed urbanization on N<sub>2</sub>O emissions from the Chongqing metropolitan river network, China



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### HIGHLIGHTS

- Higher N<sub>2</sub>O saturation and fluxes in urban rivers than the remote rural ones.
- The nutrients of the surface water served as effective predictor of saturation and flux of N<sub>2</sub>O.
- Temperature and dilution effect due to consistent rain were responsible for the seasonal changes of N<sub>2</sub>O saturation.
- Urbanization may significantly increase the uncertainty about the current IPCC method for N<sub>2</sub>O budgets from global rivers.

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### ABSTRACT

Watershed urbanization, an integrated anthropogenic perturbation, is another considerable global concern in addition to that of global warming and may significantly enrich the N loadings of watersheds, which then greatly influences the nitrous oxide (N<sub>2</sub>O) production and fluxes of these aquatic systems. However, little is known about the N<sub>2</sub>O dynamics in human-dominated metropolitan river networks. In this study, we present the temporal and spatial variations in N<sub>2</sub>O saturation and emission in the Chongqing metropolitan river network, which is undergoing intensified urbanization. The N<sub>2</sub>O saturation and fluxes at 84 sampling sites ranged from 126% to 10536% and from 4.5 to 1566.8 μmol N<sub>2</sub>O m<sup>-2</sup> d<sup>-1</sup>, with means of 1780% and 261 μmol N<sub>2</sub>O m<sup>-2</sup> d<sup>-1</sup>. The riverine N<sub>2</sub>O saturation and fluxes increased along with the urbanization gradient and urbanization rate, with disproportionately higher values in urban rivers due to the N<sub>2</sub>O-rich sewage inputs and enriched *in situ* N substrates. We found a clear seasonal pattern of N<sub>2</sub>O saturation, which was co-regulated by both water temperature and precipitation. Regression analysis indicated that the N substrates and dissolved oxygen (DO) that controlled nitrogen metabolism acted as good predictors of the N<sub>2</sub>O emissions of urban river networks. Particularly, phosphorus (P) and hydromorphological factors (water velocity, river size and bottom substrate) had stronger relationships with the N<sub>2</sub>O saturation and could also be used to predict the N<sub>2</sub>O emission hotspots in regions with rapid urbanization. In addition, the default emission factors (EF<sub>5-r</sub>) used in the Intergovernmental Panel on Climate Change (IPCC) methodology may need revision given the differences among the physical and chemical factors in different rivers, especially urban rivers.

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## 1. Introduction

Nitrous oxide (N<sub>2</sub>O) is an important greenhouse gas with a warming potential approximately 300-fold higher than that of CO<sub>2</sub> and could cataclysmically destroy the ozone layer by reacting with ozone (Ravishankara et al., 2009). The mean atmospheric N<sub>2</sub>O concentration is approximately 328 ppb and continues increasing at a rate of 0.25%–0.31% yr<sup>-1</sup> (IPCC, 2013). The major sources of atmospheric N<sub>2</sub>O are involved with the microbial metabolism of natural systems, including both soil and aquatic ecosystems (oceans, rivers, and lakes), and the anthropogenic emission sources are wastewater treatment, fossil fuel burning and industrial processes. The biogeochemical conversion of anthropogenic N to N<sub>2</sub>O in soils and aquatic ecosystems is considered the most important cause of increasing atmospheric N<sub>2</sub>O concentrations (Beaulieu et al., 2011; IPCC, 2013; Stehfest and Bouwman, 2006). While the N<sub>2</sub>O production in soils with direct local influence has been the focus of intense investigation (Bouwman, 1996; Chapuis et al., 2007; Shepherd et al., 1991), the N<sub>2</sub>O emissions from aquatic ecosystems, especially those from rivers and streams, have received much less attention in the global N<sub>2</sub>O budget models.

Previous studies have demonstrated that rivers and streams not only transport terrestrial N to the ocean but also act as reactors for microbiological nitrification and denitrification (Boyer et al., 2006; Yan et al., 2010), thereby actively degassing a certain mass of N<sub>2</sub>O into the atmosphere (Beaulieu et al., 2011; Rosamond et al., 2012; Wang et al., 2015). Recent estimates indicated that the amount of N<sub>2</sub>O emitted from global river systems was approximately 0.68–0.90 Tg yr<sup>-1</sup>, which is equivalent to 10%–17% of the global agricultural N<sub>2</sub>O emission rate (Beaulieu et al., 2011; Rosamond et al., 2012; Syakila and Kroeze, 2011), making these emissions important to the global N<sub>2</sub>O budget. However, the amounts of N<sub>2</sub>O emitted from rivers and streams remains highly uncertain because of the insufficient amounts of monitoring data and our limited understanding of the mechanisms influencing the spatial-temporal dynamics of riverine N<sub>2</sub>O (Ivens et al., 2011; Rosamond et al., 2012). More regional studies with high spatial and temporal resolution are necessary for a better understanding of the ecological functions of rivers and streams in the global N cycle.

The N<sub>2</sub>O production in streams and rivers is known to mainly derive from the *in situ* nitrification and denitrification of terrestrial N, added to aquatic systems via leaching, runoff, sewage, and N<sub>2</sub>O-supersaturated groundwater (Beaulieu et al., 2011). The Intergovernmental Panel on Climate Change (IPCC) put forward a methodology for estimating the N<sub>2</sub>O emitted by rivers and streams, where the product of the annual N lost to leaching and runoff and the emission factor (*EF5-r*) derived from N<sub>2</sub>O yields the amount of nitrification or denitrification (IPCC, 2006). However, the value of *EF5-r* (0.25%) is highly uncertain due to limited data availability (Beaulieu et al., 2011; Rosamond et al., 2012; Wang et al., 2015), as it depends on the exogenous N<sub>2</sub>O input, the magnitude of the N<sub>2</sub>O yields and human activity densities in watersheds. Therefore, more direct measurements in aquatic river environments should be conducted for the further revision of the *EF5-r* value.

Human activities, having multiple effects on streams and rivers, have been considered the main driving force of large increases in future N<sub>2</sub>O emissions from streams and rivers (Burgos et al., 2015; Kroeze and Seitzinger, 1998; Seitzinger et al., 2000; Wang et al., 2015; Yu et al., 2013). However, understanding the mechanisms of human influences other than that of N loading and estimating the contributions of these influences are particularly challenged by the limited number of available studies. Over the past few decades, countries and regions all over the world have faced the challenges of urbanization, which are a composite of the impacts of human activities and bring a series of environment problems, including

large-scale land use changing from natural to agricultural or urban, hydrologic process changes caused by an increasing amount of impermeable surfaces, water quality deterioration and the input of labile carbon (Astarai-Imani et al., 2012; Sickman et al., 2007; Williams et al., 2016), all of which might affect fluvial N<sub>2</sub>O dynamics. Prior studies have shown that urban rivers present disproportionately high N<sub>2</sub>O emissions (Beaulieu et al., 2011; Rosamond et al., 2012). Fast urbanization provides us with an opportunity to explore the ecology of riverine N<sub>2</sub>O and to formulate N<sub>2</sub>O mitigation strategies for sustainable urban development. However, the responses of riverine N<sub>2</sub>O dynamics to urbanization are not as fully researched as those of agricultural systems; therefore, the N<sub>2</sub>O emissions from expanding urban-impacted rivers can cause large uncertainties in the current N<sub>2</sub>O budget (Beaulieu et al., 2010; Ivens et al., 2011; Yu et al., 2013). In addition, phosphorus (P) additions, which have been strongly linked to N<sub>2</sub>O production in soil via their stimulation on nitrifying or denitrifying bacterial activities (Mori et al., 2010; Wang et al., 2014), have never been confirmed in aquatic ecosystems. Given the expansion of urban areas and the domestic pollution and sewage loads that follow (Seto et al., 2012), more studies quantifying and improving the database of N<sub>2</sub>O emissions of urban river systems are needed to better understand the potential impact of urbanization on nitrogen cycling in rivers.

With its economic development, China is facing unprecedented and substantial urbanization, accompanied by a series of environmental problems and research hotspots. Rivers in China, such as the Changjiang River (Wang et al., 2015), Jiulong River (Chen et al., 2015) and Pearl River (Wang et al., 2012), have been found to have high N<sub>2</sub>O saturation and fluxes, but few studies have mentioned the effect of urbanization on the riverine N<sub>2</sub>O (Chen et al., 2015; Yu et al., 2013). In this study, we chose to investigate the N<sub>2</sub>O saturation and fluxes from the river network of the Chongqing metropolitan area, where the urbanization rate has been especially fast due to the Western Development Strategy in China. The objectives of this study were to (1) determine the spatial and temporal variability of N<sub>2</sub>O saturation and emissions in basin-wide river networks across an urbanization gradient and (2) examine the potential predictors and controls of N<sub>2</sub>O production in urban rivers.

## 2. Materials and methods

### 2.1. Study area

The study was conducted in the Chongqing metropolitan area, located in the western Chongqing Municipality (28°10′–32°13′ N, 105°11′–110°11′ E) (Fig. 1). The Chongqing metropolitan area features a subtropical monsoon climate and four distinct seasons, with winter temperatures of approximately 9 °C and summer temperatures of approximately 35 °C. The mean rainfall is 1089 mm, most of which (approximately 85%) occurs in the wet season, from May to October. With its mountainous and hilly geomorphology, there are over 700 rivers and streams and a water area totalling >160 km<sup>2</sup> in the study area. The Changjiang River and Jialing River, the two largest rivers in our study area, carve through the whole metropolitan area and act as the backbone of the river network (Fig. 1).

### 2.2. Land use and sampling site design

In this study, the Landsat TM/ETM + images from August 2014 (30-m pixels) were downloaded, and Landsat7 software was used as the data source for generating the land-use estimates and basic information about the river network through employing ESRI ARCGIS 9.2 software. The optimization and verification of the

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