



## Impacts of water residence time on nitrogen budget of lakes and reservoirs



Yindong Tong<sup>a</sup>, Jiaqi Li<sup>a</sup>, Miao Qi<sup>a</sup>, Xiaoyan Zhang<sup>a</sup>, Mengzhu Wang<sup>a</sup>, Xueyan Liu<sup>b</sup>, Wei Zhang<sup>c</sup>, Xuejun Wang<sup>d</sup>, Yiren Lu<sup>a</sup>, Yan Lin<sup>e,\*</sup>

<sup>a</sup> School of Environmental Science and Engineering, Tianjin University, Tianjin 300072, China

<sup>b</sup> Institute of Surface-Earth System Science, Tianjin University, Tianjin 300072, China

<sup>c</sup> School of Environment and Natural Resources, Renmin University of China, Beijing 100872, China

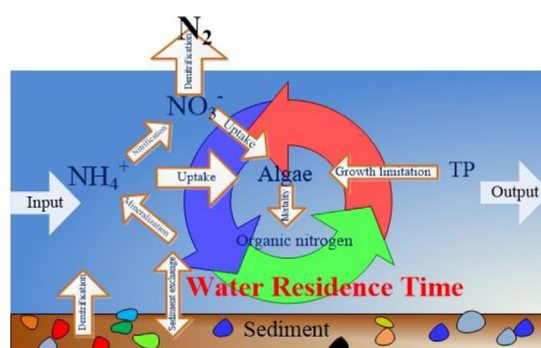
<sup>d</sup> College of Urban and Environmental Sciences, Peking University, Beijing 100871, China

<sup>e</sup> Norwegian Institute for Water Research, Oslo 0349, Norway

### HIGHLIGHTS

- WRT have significant impacts to the TN change rates in waterbodies.
- Longer WRT slows down the TN increasing rates for waterbodies acting as nitrogen sinks.
- Longer WRT accelerates the removal for the waterbodies acting as nitrogen sources.
- Higher water phosphorus concentrations favor faster nitrogen removal from the waterbodies.

### GRAPHICAL ABSTRACT



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### ABSTRACT

As an important factor related to the self-purification capacity (e.g. denitrification, burial rate, and downstream output) in aquatic systems, water residence time (WRT) has great impacts on the nitrogen (N) dynamics and its removal process in lakes and reservoirs. In this study, we have analysed the impacts of WRT on the change rates of total nitrogen (TN) concentrations in 50 waterbodies (including 33 lakes and 17 reservoirs) in China, with different change trends (e.g. increasing trends and decreasing trends) and TN concentrations during 2012–2016. Based on the annual ecosystem-scale N mass balance, TN input and output flux in the waterbodies are estimated. The results showed that the decreases of TN concentrations usually occur in the waterbodies with the relatively high TN concentrations in 2012, and WRT has significant impacts on the TN change rates in the waterbodies. Longer WRT could slow down the TN increasing rates in the waterbodies acting as N sinks, but could accelerate the removal from the waterbodies acting as N sources. Higher water phosphorus (P) concentrations could also be beneficial for the faster N removal from the waterbodies, which is mediated via the coupled processes regulating the N transfer from water column to anoxic sediments. China has recently issued the “lake-chief” systems, addressing the specific and flexible strategies for water pollution control in different lakes. The self-purification capacity through denitrification and burial rate, which are closely related to WRT, should be taken into consideration when making specific water management plans in the future.

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\* Corresponding author at: Norwegian Institute for Water Research, Norway.

E-mail address: [Yan.Lin@niva.no](mailto:Yan.Lin@niva.no) (Y. Lin).

## 1. Introduction

Nitrogen (N) is one of the most important chemical elements for the plants and animals in the world (Vitousek and Howarth, 1991; Galloway et al., 2008; Suddick et al., 2013; Cui et al., 2013). In recent decades, N cycling in the terrestrial systems has been markedly accelerated due to intensive anthropogenic activities and increasing food demands (Galloway et al., 1995; Galloway et al., 2008; Tong et al., 2015). N utilized by human activities, usually in the form of N-containing fertilizers, is mainly fixed from the unreactive N<sub>2</sub> in the atmosphere through the Haber-Bosch reaction (Vojvodic et al., 2014). However, not all the produced N-fertilizers have been utilized efficiently, and large quantities are discharged into aquatic systems (Cui et al., 2013). Human-induced nutrient discharge, including both N and phosphorus (P), has led to unexpected global water problems such as serious eutrophication and the formation of “dead zones” in waterbodies (Ryther and Dunstan, 1971; Smith et al., 1999; Anderson et al., 2002; Diaz and Rosenberg, 2008; Paerl, 2017). Water quality pollution and deterioration are especially an issue in China due to the rapid development in industry and agriculture during the past decades (Smith et al., 1999; Conley et al., 2009). The current N and P concentrations in many of China’s waterbodies still remain at high levels even after years of effort on pollutant control (Zhou et al., 2017; Tong et al., 2017a). In Taihu Lake, the water crisis caused by the extensive outbreak of harmful algal blooms (HABs) in 2007 severely affected the water supplies of millions of people for several weeks (Stone, 2011), and other freshwater lakes in China are also experiencing similar situations (Le et al., 2010).

Many previous studies have been carried out on the recognition and control of human-induced N discharges into aquatic systems (Xu et al., 2010; Cui et al., 2013; Liu et al., 2016). Excess N could come from agricultural fertilizer applications, residential sewages, industrial wastewaters, atmospheric depositions, etc. (Cui et al., 2013; Liu et al., 2016). Compared with P, N sources are more diffusive in the watersheds, increasing the difficulty in source quantification and control (Finlay et al., 2013). Strategies for the control of point and non-point N sources have also been widely developed to reduce the human-induced N discharges into waterbodies (Ministry of Environmental Protection, China, 2015). For instance, Waste Water Treatment Plants (WWTPs) have been widely built in China, and they have been proven to be quite effective in the control of nutrient discharges into waterbodies (Tong et al., 2017a, 2017b). So far, most of these efforts are devoted to the control of N transports from lands to water, but after the N is discharged into the water, its removal process from the waterbodies could be only achieved by the function of the ecosystem itself, acting in a “self-purification capacity” (Finlay et al., 2013). Downstream outflows, denitrification, sedimentation and plant uptake in the aquatic ecosystems are believed to be the most important pathways to remove N from the waterbodies (Schlesinger, 2009; Finlay et al., 2013). Among all the internal removal processes, N removal from the water through denitrification is much higher than the loss through the sedimentation and plant uptake (Saunders and Kalff, 2001). For instance, in experimental wetlands in New Zealand, N removal by denitrification (3.0–3.3 g N/(m<sup>2</sup>·d)) is far greater than either sedimentation (0.16–0.27 g N/(m<sup>2</sup>·d)) or plant uptake (0.19–0.33 g N/(m<sup>2</sup>·d)) (Oostrom, 1995).

With regard to the internal N cycling in the waterbodies, water residence time (WRT) plays a crucial role since it is related to almost all the natural processes (e.g. denitrification, sedimentation and downstream output) that could affect the nutrient concentrations in the waterbodies (Brett and Benjamin, 2008; Kõiv et al., 2011; Bruesewitz et al., 2012). Based on the nutrient input and output budget in the waterbodies, Finlay et al. (2013) reported that the N removal efficiency through denitrification or the permanent burial in the waterbodies could increase significantly with the increase of WRT. A similar mechanism has also been observed for the P concentrations in the waterbodies, and the

longer WRT could accelerate the settling down of particulate P from the water phase (Brett and Benjamin, 2008). For the organic carbons, Catalán et al. (2016) confirmed the negative relationship between the decay rates of dissolved organic carbon (DOC) and WRT across the aquatic systems. Evans et al. (2017) further confirmed that WRT could impose different impacts on organic carbon decays in the waterbodies acting as sources or sinks. For the waterbodies acting as carbon sinks, the longer WRT might be beneficial for the improvement of internal carbon removal efficiency, while for the waterbodies acting as the carbon sources, the longer WRT could accelerate the carbon loss (Evans et al., 2017). These studies confirmed the significant impacts of WRT on the internal nutrient (e.g. P and C) cycling in the waterbodies, but these impacts could also vary considerably among different nutrient types and waterbody types (i.e. nutrient sources or sinks).

As the human activities continue to alter the global nitrogen cycling, the ability to predict the WRT’s impacts on the internal N cycling in the waterbodies is becoming more and more important. Internal N removal from the waterbodies is of particular interest because it is through the combined processes (e.g. denitrification, sedimentation and plant uptake) that the local and downstream N concentrations are related (Finlay et al., 2013). In this study, we have analysed the annual ecosystem-scale mass balance to explore the TN input and output budget in 50 waterbodies with different hydrological characteristics in China. Impacts of WRT on TN change rates in the waterbodies acting as N sinks or sources have been addressed. To identify the dominant processes in affecting the TN concentrations, TN input flux and output flux are estimated based on the annual ecosystem-scale N mass balance, respectively. The better understanding of the N removal pathway from the waterbodies is important to make effective management decisions to maintain and enhance the N removal processes in the face of agricultural intensification, urbanization and overall population growth that could increase the human-induced nutrient discharges into the water.

## 2. Materials and methods

### 2.1. Selected waterbodies

In this study, all of the water quality data are part of the randomized, unequally weighted probability surveys overseen by the Ministry of Environmental Protection, China, with the goal of creating unbiased assessments of water quality in the freshwater lakes across different provinces in China (Tong et al., 2017a, 2017b). In detail, we have applied the following three guidelines during the selections of waterbodies. 1) With the continuous monitoring data, the lakes or reservoirs with over 40 continuous monthly TN monitoring points starting from 2012 have been selected. 2) The inclusion of various hydrological characteristics. To make the result representative, we have selected the lakes or reservoirs with different hydrological characteristics (considering lake depth, surface area, and water volume). 3) Large geographic coverage. We have selected the lakes or reservoirs in different regions to cover different climate conditions and economic development stages. In summary, a total of 50 freshwater waterbodies (including 33 lakes and 17 reservoirs, respectively), which are distributed in the 19 provinces in China, have been selected, and the detailed locations of the selected waterbodies are provided in Fig. S1. Considering that the nutrient concentrations in the waterbodies could be easily affected by the human activities, the land use types (e.g. farmland, grassland, waterbody, built areas and desert), population density and annual precipitation in the belonging catchments have been summarized in Table S1. The division of catchments is provided by the Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences, with data available at <http://www.resdc.cn/> (shown in Fig. S1). The hydrological conditions of the selected lakes and reservoirs are obtained from the Hydro Lakes database developed by the Global Hydro Laboratory (Messenger et al., 2016) and reported values from Wang and Dou (1998). The detailed information for each waterbody, including WRT, surface area, water

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