



# Response of dissolved organic matter optical properties to net inflow runoff in a large fluvial plain lake and the connecting channels

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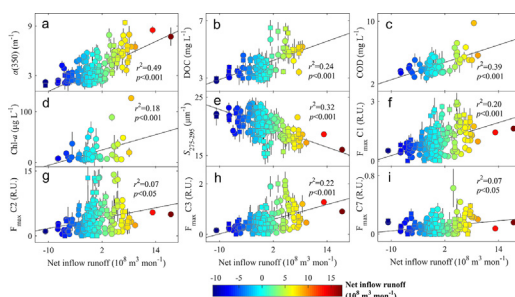
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## HIGHLIGHTS

- Net water inflow varied greatly in different sub-watersheds of the Lake Taihu Basin.
- We found significant relationships between  $Q_{net}$  and CDOM optical properties.
- We found strong catchment and anthropogenic effects in various regions of Lake Taihu.
- Net water inflow controls the optical properties of CDOM molecules in Lake Taihu.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Fluvial plain lake watersheds are usually highly urbanized and have high concentrations of chromophoric dissolved organic matter (CDOM). CDOM derived from the connecting urban channels usually share strong terrestrial and anthropogenic signals and net inflow runoff ( $Q_{net}$ ) to the lake serves as a proxy of residential household sewage input. We investigate how  $Q_{net}$  controls the optical characteristics of CDOM in fluvial plain Lake Taihu and the connecting channels. CDOM absorption coefficient  $a(350)$ , dissolved organic carbon (DOC), the fluorescence intensity ( $F_{max}$ ) of seven PARAFAC components C1–C7, and  $\delta^{15}N$ -TDN were higher in the northwestern relative to the other lake regions, and  $a(250)/a(365)$ , spectral slope  $S_{275-295}$ , and  $\delta^{13}C$ -DOM relative low in the northwestern lake, all indicating strong terrestrial and anthropogenic effects. Conversely, the urban land cover (%Cities), gross domestic product (GDP), and population density were relatively low in the western sub-watersheds and high in the eastern sub-watersheds. This apparent paradox reflects variations in  $Q_{net}$  from different sub-watersheds. Thus, significant positive relationships were found between  $Q_{net}$  and  $a(350)$ , DOC, chemical oxygen demand (COD), chlorophyll-*a* (Chl-*a*),  $F_{max}$  of C1–C3 and C6–C7, and %C2–%C3 in the five hydraulic sub-watersheds. We revealed significant positive relationships between mean  $a(350)$ , DOC, COD, Chl-*a*, C1–C3 and C6, %C2–%C3, and the products of  $Q_{net} \times \%Cities$ ,  $Q_{net} \times GDP$ , and  $Q_{net} \times$  population density. We further found dominant contribution of lignin to the total number of assigned formulas for the samples

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collected from the channels in the Huxi watershed and the central lake using high resolution mass spectroscopy. We conclude that  $Q_{\text{net}}$  is of key importance for the optical properties of CDOM molecules in the various regions of Lake Taihu and the connecting channels.

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## 1. Introduction

Urbanization and industrialization usually take place in large fluvial plains and coastal lake watersheds, which support the livelihood of >75% of the human population worldwide (Paerl et al., 2013). An increase in the urban and agricultural land use results in increasing amounts of household sewage and agricultural humic-like substances that are discharged downstream (Williams et al., 2016; Williams et al., 2010). Fluvial plains are usually located at low altitude with elevations < 10 m, and lake watersheds drainage within these areas can be complex as the connecting river systems include natural rivers, semi-artificial-reformed rivers, man-made channels, and hundreds of floodgates. The resulting effect on the carbon loading of lakes can be complex and represents a challenge to unravel the sources of organic matter. Sewage derived from intensive residential areas in these ecosystems often contains high concentrations of dissolved organic matter (DOM).

Chromophoric DOM (CDOM) is the colored fraction of DOM that can strongly absorb light in the ultraviolet (UV) and blue regions. CDOM originates from terrestrial and anthropogenic organic matter, microbial degradation of aquatic organisms, and extracellular secretion released by algae, and consists of a complex mixture of organic molecules. Its translocation and transformation strongly impact the inland water carbon cycling and the outgassing of greenhouse gases (Davidson et al., 2018; Le et al., 2015; Stedmon and Markager, 2005a; Zhao et al., 2017). Numerous studies have elucidated how discharge impacts the dynamics of CDOM in streams, rivers, mountainous lakes, and estuaries (Guo et al., 2014; Huang et al., 2017; Li et al., 2016; Stedmon and Markager, 2005a; Yang et al., 2013; Zhao et al., 2016). In fluvial plain lake watersheds, however, the direction of flow in the lake and the connecting river networks are broadly similar to that of lagoons and can be reversed (backflow) under certain anthropogenic, climatic, and tidal conditions, and especially during cyclone events (Qin et al., 2007; Zhu et al., 2014). The hydraulic linkages between lakes and the connecting channels in fluvial plain lake watersheds are therefore much more complicated than mountainous lake ecosystems with fixed flow direction. However, there have been few attempts to characterize how net inflow runoff ( $Q_{\text{net}}$ , calculated as backflow runoff subtracting from inflow runoff) from connecting channels affects the variation in the optical properties of CDOM in fluvial plain lake ecosystems.

It is difficult to trace the concentration and compositional dynamics of CDOM due to the complex mix of CDOM molecules. UV-Vis spectral absorption and fluorescence measurements are important tools to evaluate the optical concentration and composition of CDOM molecules. CDOM fluorescent (FDOM) measurements of excitation (Ex) and emission (Em) matrices (EEMs) are easy to obtain and yield high optical resolution by enabling distinction of materials that absorb at the same wavelength but emit in different spectral regions (Murphy et al., 2008). Parallel factor analysis (PARAFAC) decomposes EEMs of complex mixture into distinct trilinear components and is useful in interpreting EEMs data arrays (Murphy et al., 2013; Stedmon and Bro, 2008). The stable isotopes  $\delta^{13}\text{C}$ -DOM and  $\delta^{15}\text{N}$ -TDN can be used to trace the sources of bulk DOM as, typically,  $\delta^{13}\text{C}$ -DOM of terrestrial humic-rich DOM ranges between  $-29\%$  and  $-26\%$ , and is similar to that of terrestrial C3 plants with  $\delta^{13}\text{C}$ -DOM ranges from  $-34\%$  to  $-23\%$  (Hood et al., 2009; Vizzini et al., 2005). In comparison,  $\delta^{13}\text{C}$ -DOM of biological autochthonous DOM (microbial degradation of phytoplankton) typically ranges between  $-25\%$  and  $-20\%$  and is roughly similar to values for C4 plants ( $-23\%$  to  $-7\%$ ) (Hood et al., 2009; Vizzini et al., 2005).  $\delta^{15}\text{N}$ -TDN values were highest for riverine and anthropogenic input (Vizzini et al., 2005; Zhou et al., 2015b). Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS) can be used to trace the

composition of DOM from the molecular level. FT-ICR MS enables the assignment of thousands of molecular formulas of DOM in various aquatic ecosystems (Kellerman et al., 2015; Spencer et al., 2014; Stubbins et al., 2010).

The Lake Taihu watershed is the most developed area in China with high population density, urbanization, and economic development (Qin et al., 2007). It covers 36,500 km<sup>2</sup>, and the lake has an area of 2338.1 km<sup>2</sup> with a mean depth of 1.89 m. The lake bottom has a mean elevation of  $\sim 1.1$  m above sea level and a total of 172 rivers or man-made channels are connected to the lake (Qin et al., 2007). The summed length of channels in the Lake Taihu watershed is 120,000 km, i.e.  $\sim 3.2$  km km<sup>-2</sup> (Qin et al., 2007), and the water retention time of the lake is about 300 days (Tang et al., 2010). Water runoff usually drains from the west, passes through the lake and to the east, and finally empties into the East China Sea (Fig. 1). However, the water flow in the whole lake watershed can be reversed, i.e. backflow runoff; this is especially pronounced during the dry season (from late autumn to late spring) and occurs particularly in the eastern part of the lake watershed (Qin et al., 2007). Water flow directions of the channels connecting to Lake Taihu are largely controlled by tidal exchange, extreme events (typhoon passages), and anthropogenic disturbance (e.g. hundreds of floodgates), and are broadly similar to that of lagoon systems.  $Q_{\text{net}}$  draining to the lake from different hydraulic sub-watersheds can therefore be used to trace anthropogenic effluents from the corresponding sub-watersheds. High urban land cover (%Cities) in the Lake Taihu watershed result in a high nutrient and CDOM input, which boosts the outbreak of algal blooms, as well as the accumulation of high concentrations of autochthonous CDOM (Duan et al., 2014; Zhou et al., 2015a). High concentrations of CDOM can cause an unpleasant odor and taste of the water and produce undesirable disinfection byproducts during water treatment processes (Tomlinson et al., 2016). The lability of CDOM to degradation and mineralization is largely determined by its source and composition (Stedmon et al., 2007). Although there have been studies unraveling the spatial and temporal variations of CDOM optical properties in Lake Taihu (Yao et al., 2011; Zhang et al., 2011; Zhou et al., 2015b), how the  $Q_{\text{net}}$  from individual hydraulic sub-watersheds may influence the sources and optical properties of CDOM in the lake watersheds remains, however, largely unknown. It was found that the  $Q_{\text{net}}$  varied greatly between the individual hydraulic sub-watersheds of the Lake Taihu watershed (Qin et al., 2007).

In this study, we attempt to unravel how  $Q_{\text{net}}$  draining to the lake from individual hydraulic sub-watersheds may quantitatively control the optical properties of CDOM in the fluvial plain Lake Taihu watershed. A total of 2277 field samples (128 samples collected in 2011 were previously reported in reference Zhou et al. (2015b)) collected from the lake watershed from February 2008 to November 2016 and long-term data on the monthly (August 2009–December 2016) inflow and backflow runoff (net inflow,  $Q_{\text{net}}$ ) in individual hydraulic sub-watersheds of the lake were used to elucidate how  $Q_{\text{net}}$  may influence the sources and optical properties of CDOM in the lake watersheds. We hypothesized that the  $Q_{\text{net}}$  from individual sub-watersheds controls the optical properties of CDOM and its variation in the lake and the connecting channels.

## 2. Materials and methods

### 2.1. Hydraulic sub-watersheds of Lake Taihu and hydrological and socioeconomic data collection

The Lake Taihu watershed can be divided into seven individual hydraulic sub-watersheds according to the Taihu Basin hydrological

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