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Identifying the sources and fate of anthropogenically impacted dissolved organic matter (DOM) in urbanized rivers

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ABSTRACT

Anthropogenic activities have dramatically changed the loads and compositions of dissolved organic matter (DOM) in urbanized streams. In this study, the spatial and temporal variations of DOM in the anthropogenically impacted Zhujiang River were investigated by analyzing the water samples in an upstream, urbanized area and downstream of the rivers on different days of one year. The results indicated that the levels of dissolved organic carbon (DOC) and total phosphorus (TP) were unaffected by seasonal changes, but the specific UV_{254} absorbance (SUVA) values and the total nitrogen (TN) content were greater in the winter than those in the summer. Parallel factor (PARAFAC) analysis of the excitation emission matrices (EEM) revealed the presence of three anthropogenically derived components [tryptophan-like (C1) and tyrosine-like proteins (C3) and anthropogenic humic substances (C5)] in the urbanized rivers, and they had greater seasonal and spatial variability than the terrestrial and microbial humic substances (C2 and C4). Cluster analysis revealed that treated wastewater was an important source of DOM in the urbanized streams. Photodegradation experiments indicated that the DOM in the populous area of the rivers had greater photodegradation potentials than that in the downstream region or in the natural waters. Interestingly, that the anthropogenic humic substances (C5) were considerably more photoreactive than the other four PARAFAC components, which exhibited a decrease of 80% after exposure to sunlight for 0.5 d. This study suggests that the treated wastewater could be an important input to the DOM in the urbanized rivers and the naturally occurring photodegradation could help in eliminating the anthropogenic DOM during their transport.

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1. Introduction

Dissolved organic matter (DOM) plays important roles in both natural waters and engineered water systems (Borisover et al., 2009; Ishii and Boyer, 2012). DOM is a pool of organic molecules that have various origins, e.g., autochthonous and allochthonous (Coble, 2007; Murphy et al., 2008; Stedmon et al., 2007). As a result of their heterogeneity, the DOM can be transported and transformed in natural waters (Ogawa et al., 2001). DOM also causes problems for water treatment facilities, such as the occurrence of membrane fouling (Huang et al., 2007; Jermann et al., 2007; Neubrand et al., 2010), the increase of coagulant usage (Baghoth et al., 2011) and the generation of disinfection byproducts (Pifer and Fairey, 2012; Yang et al., 2007). Thus, further understanding of the characteristics, sources and fate of DOM in natural waters would be helpful for water management.

However, one challenge to the understanding of DOM is its various and heterogenous inputs. The DOM in remote rivers or lakes is normally less affected by anthropogenic activities, which can be defined to be natural organic matter (NOM). In contrast, the DOM in urbanized rivers is strongly affected by the inputs of treated and/or untreated wastewater (Greenwood et al., 2012; Kalscheur et al., 2012). In addition, agricultural and industrial land use has substantially contributed to the increase of DOM levels in waters (Kaushal et al., 2011; Petrone et al., 2011). In particular, wastewater-derived DOM was shown to exhibit a greater organic nitrogen content and was more aliphatic than the DOM in natural waters (Chen et al., 2011; Czerwionka et al., 2012; Mesfioui et al., 2012; Sirivedhin and Gray, 2005). In general, the previous attempts to understand the characteristics of DOM have revealed that urbanized rivers have been greatly changed as a result of the anthropogenic input (Baker, 2001; Felipe-Sotelo et al., 2007; Kaushal et al., 2011; Mostofa et al., 2010; Osburn et al., 2012; Petrone et al., 2011). However, little is known about the sources and fate of anthropogenically impacted DOM in urbanized rivers. More detailed research is required to answer the following questions. (1) What are the main origins of the anthropogenic DOM? (2) How does the anthropogenic DOM contribute to the total DOM in the urbanized rivers? (3) What will happen to the anthropogenic DOM during its transport in the rivers? The goal of this work is to answer these questions.

In recent years, the combined use of three-dimensional excitation-emission matrix (EEM) and parallel factor analysis (PARAFAC) have greatly advanced the characterization of DOM (Stedmon and Bro, 2008; Zhang et al., 2011). Utilizing PARAFAC enables the deconvolution of the overlapping EEM spectral peaks into their independent components, each of which represents a class of DOM that has similar fluorophores. However, the PARAFAC method is limited to the classification of a pool of FDOM, and the assignment of the categories of FDOM is based on the previously identified PARAFAC components (Ishii and Boyer, 2012). In fact, the FDOM is comprised of thousands of molecules, which could yield a wealth of EEM vectors in the datasets. Utilizing all of the EEM vectors enables us to capture more of the signatures of the FDOM. To utilize the EEM features and improve the reliability of the datasets, both PARAFAC and cluster analysis of the EEM datasets were performed in this study. The PARAFAC method herein was used to decompose the FDOM into several classes of components; the cluster analysis was used to assess the similarities or differences of the DOM samples collected from the urbanized streams. The multivariate analysis can assist in the identification of the origins, transport and transformation of the DOM in the investigated area.

Because DOM molecules are very photoreactive (Benner and Kaiser, 2011; Dittmar et al., 2007), photodegradation is an important mechanism for the transformation DOM in rivers (Benner and Kaiser, 2011), lakes (Borisover et al., 2009; Cory et al., 2007; Grzybowski, 2003; Zhang et al., 2009b) and oceans (Cory et al., 2007; Dittmar et al., 2007; Hedges et al., 1997; Hernes and Benner, 2003; Mopper et al., 1991). The photochemical transformation of DOM in estuaries has been demonstrated to cause decreases in its molecular size, lead to significant carbon mineralization, and cause changes its optical properties (Miller and Zepp, 1995; Moran et al., 2000). DOM is highly complex, with source-specific photodegradation characteristics; hence, photodegradation potentials could aid in DOM source determination.

The primary objective of this study was to understand the origins and fate of DOM in the heavily anthropogenically impacted Zhujiang River, in order to reveal how and to what extent urbanization has an impact on DOM in rivers and to resolve the different roles of the anthropogenic impacts and the seasonal patterns on DOM. The bulk quality of the DOM samples, such as the amount of dissolved organic carbon (DOC) and the UV absorbance at 254 nm, was monitored. EEM characterization, in combination with PARAFAC and cluster analysis, was conducted to examine the DOM fingerprint in the urbanized rivers. Photodegradation incubations were conducted to understand how exposure to sunlight contributes to the transformation of DOM and to help track the photodegradation potentials of the DOM originating from different sources.

2. Experimental section

2.1. Sampling

The studied Zhujiang River runs through the city of Guangzhou, with an urban population of more than 12 million, before it flows into the South China Sea. Fourteen sampling sites were set along the rivers at upstream (before urbanization area) to downstream (after urbanization area) locations during the summer and winter (see Fig. 1). The water in sites 1 and 2 (S1 and S2) were less impacted by human activities, so they were considered to be upstream sites in this study. Site S13 and particularly site S14 were located in the estuary to the South China Sea, both of which were considered to be downstream sites. In fact, the sample at S14 exhibited a conductivity value that was as high as that of seawater (see Figure S1 of Supporting Material (SM) file), indicating the influence of salt water in the estuarine mixing zone. The other 10 sites (S3-S12) were located in the populous area of Guangzhou city. The Shijing River is a stream that has been heavily polluted by the discharge of untreated domestic

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