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Dissolved organic nitrogen transformation in river water: Effects of suspended sediment and organic nitrogen concentration

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SUMMARY

High suspended sediment (SPS) concentration exists in many Asian rivers. In addition, human activities and climate change can change river runoff, leading to the variation of SPS and pollutant concentrations. In this research, the effects of SPS and dissolved organic nitrogen (DON) concentration on DON transformation in river systems were studied through simulation experiments with samples collected from the Yellow River which is famous for its high SPS concentration. The results indicated that high DON concentration resulted in a longer retention time of NH_4^+—N and NO_2^-—N in the system due to the inhibition effect of ammonia on nitrification. The re-suspension of sediment accelerated DON transformation, and both the ammonification and nitrification rates increased with SPS concentration. The ammonification rate constants obtained from the first-order kinetics were 0.286, 0.332, 0.538 day^{-1}; the nitrification rate constants obtained from the Logistic model were 0.0018, 0.0038, 0.005 day^{-1} μmol^{-1} L^{-1} for the systems with SPS concentration of 0, 5, 10 g L^{-1}, respectively. Bacteria tended to attach onto SPS, and the specific growth rate in the systems with SPS was approximately two orders of magnitude higher than that without SPS in the first 3 days of cultivation, which resulted in an increase of DON transformation rate with SPS concentration. This study implied that DON transformation rate may be lower in the dry season than that in the wet season, and nitrogen transformation will be affected by the variation of river runoff and SPS concentration.

1. Introduction

Dissolved organic nitrogen (DON) comprises the largest part (60–69%) of the total dissolved nitrogen in aquatic systems such as lakes, rivers, estuarine and surface ocean waters (Hedin et al., 1995; Berman and Bronk, 2003; Worsfold et al., 2008). DON, mainly originating from animal feedlots, industrial wastewater, municipal wastewater and other waste disposal sites, serves as both an important mineralization substrate (Jones, 1999; Jones and Hodge, 1999; Bronk et al., 2007), and an important sink and source of nitrogen cycle in aquatic systems (Berman and Bronk, 2003). However, excessive discharge of DON might cause the diminution of dissolved oxygen, overgrowth of phytoplankton and eutrophication of water bodies (Seitzinger and Sanders, 1997; Rabalais, 2002). Consequently, many studies of DON in marine and freshwater systems have been conducted in recent decades (Zehr et al., 1988; Knicker et al., 1996; Cornell et al., 2003; Worsfold et al., 2008).

DON transformation is an important process of nitrogen cycle that controls the levels of nitrogenous compounds in water bodies. DON transformation includes two steps: firstly, DON is converted to ammonium through ammonification; secondly, ammonium nitrogen is transformed into nitrite, then into nitrate by nitrification (Anthonisen et al., 1976; Revsbech et al., 2006). Berman and Bronk (2003) pointed out that DON is a dynamic participant in the nitrogen cycle in aquatic systems. According to Takahashi's study (Takahashi and Saijo, 1981), the daily decomposition rate of DON is 8.6% in Lake Kizaki, implying a DON pool turnover time of about 12 day. Hasegawa et al. (2000) and Berg et al. (2002) indicated that microzooplankton and phytoplankton are important sources of DON release in coastal waters. In addition, Zehr et al. (1988) showed that the sediment release is also an important source of DON.

As one process of DON transformation, nitrification has received the most attention of researchers (Rysgaard et al., 1996; Williams and Tonnessen, 2000; Xia et al., 2008). The effects of organic carbon, pH, nitrogen available and suspended sediment (SPS) on nitrification in river systems have been studied intensively (Strauss and Lamberti, 2002; Xia et al., 2009). However, the knowledge about the DON transformation in river systems remains inadequate. Most of the previous studies on DON transformation in fresh water systems considered only transformation occurred in water and/or the bed-sediment phases (Richey et al., 1985; Zehr et al.,

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1988; Revsbech et al., 2006). The effect of SPS, which is affected by the river flows and exists at high levels in many Asian rivers (Shi et al., 2002), on DON transformation process remains unknown. For example, the Yellow River is known for its high SPS concentration, which is reported in g L^{-1} rather than the usual mg L^{-1}, with an average SPS concentration of 20 g L^{-1} (WCC, 2006). Our previous studies indicated that the nitrification rate increases with the concentration of SPS, which is due mainly to the fact that SPS can provide a large surface area for nitrification process (Xia et al., 2009). Similar to nitrification, we assumed that SPS can also influence DON transformation process. Furthermore, little has been reported about how the transformation of DON is influenced by its initial concentration which shows significant variation with season (Banoub and Williams, 1973; Hasegawa et al., 2000).

In addition, anthropogenic changes, particularly the increases of greenhouse gas in the last century, have induced global climate change (Crowley, 2000). The change of river runoff, floods, and droughts caused by climate change (Jha et al., 2004; Bond et al., 2008; Chang and Jung, 2010), will lead to the variations of SPS and DON concentration in river systems (Walling and Fang, 2003; Wang et al., 2007). Therefore, it is necessary to study the effects of SPS and initial DON concentration on DON transformation; especially under the condition that climate change will continue and may accelerate (Cox et al., 2000).

Therefore, the goal of this study was to explore the effects of SPS and initial DON concentration on DON transformation. Experimental studies were conducted under laboratory conditions with samples collected from the Yellow River. The processes of DON transformation were analyzed through the features of ammonification and nitrification kinetics. The effects of DON concentration and SPS on DON transformation rate were investigated, and the influence mechanism was examined. In addition, the possible variations of nitrogen transformation in the Yellow River were analyzed under the context of climate change.

2. Materials and methods

2.1. Sampling site and sampling regime

As the second longest river in China, the Yellow River provides critical water resources to northern and northwestern China (Chen et al., 2004). The Yellow River watershed contains 15% of plow land and 12% of population of the country. However, the river is suffering from severe nitrogen pollution due mainly to high concentration of ammonium nitrogen (NH_4^+—N), which is higher in the dry season than that in the wet season (Xia et al., 2002). Some reaches of the river cannot be used as drinking water sources because their NH_4^+—N concentrations are higher than 1.0 mg L^{-1} with the water quality worse than Grade III of the Chinese water quality grade scale in which Grade 1 is best and Grade V is worst (Xia et al., 2004a). The nitrogen pollutant in the Yellow River comes from point and nonpoint sources (Xia et al., 2002), and the latter are mainly composed of inorganic nitrogen (Zhang et al., 2010). That is to say, organic nitrogen mainly comes from point sources; approximately 4.0 billion tons of industrial and municipal wastewater has been discharged into the Yellow River in 2003, and the concentration of organic nitrogen ranged from 7 to 24 mg L^{-1} in the wastewater (WCC, 2003).

The sampling site was located at the Huayuankou Hydrological Station near Zhengzhou which is in the middle reach of the Yellow River. The monthly average SPS concentration varied between 0.4 and 15 g L^{-1} in the Huayuankou Station in 1999 (WCC, 1999). Water and SPS samples were collected with a TC-Y water sampler (produced by TECH Instrument of Shenyang, China) at a depth of 0.2 m in the central stream point in 2002, 2003 and 2004. All samples were kept under 4 °C in a cooler, and then transported to the laboratory for experimental analysis. The cultivation experiments were conducted within 72 h after the sample collection. All the chemical reagents used in the experiments were of analytical grade.

2.2. Laboratory simulation experiment

A simulation experiment system was designed to study the effects of DON concentration and SPS on DON transformation. The collected water and SPS were used as media for DON transformation experiments; indigenous bacteria in the samples remained active, which was testified by cultivation of bacteria from samples. Peptone had been commonly used as organic nitrogen for environmental simulation experiments (Jansson et al., 1955; Bauhus et al., 1993; Ishiguro et al., 1994). Therefore, peptone (Beijing Chemical Works, 13.5% N) was used as organic nitrogen in this study. The simulation experiment systems were composed of a series of flasks which contained 400 ml water samples, and a known amount of SPS and DON. Then the flasks were covered by eight layers of gauze to exclude external bacteria. Then they were cultivated in the dark at 20 °C with magnetic stirrer to make the suspension of sediment, and the stirring rate was adjusted to ensure all sediment suspended. Aliquots were withdrawn and examined for the levels of nitrogen species and bacteria in water and sediment phases at pre-determined intervals. Ammonification rate was calculated based on the variation of DON concentration in the reaction systems including both water and sediment phases, while nitrification rate was calculated based on the variation of nitrate concentration in the water phase. Each experimental set was cultivated in triplicate with a set of controls. The control experiments were carried out with the sterilized water and SPS samples, and microbial activities were inhibited by adding 0.5% mercuric chloride.

The effect of initial DON concentration on DON transformation was examined with samples collected in 2002. The SPS concentration in flasks were all set at 11 g L^{-1}, and two known amounts of organic nitrogen were added to the flasks to obtain two initial DON concentrations of 5.5 and 8.6 mg L^{-1}, respectively. These concentrations are in the range of dissolved organic nitrogen concentration near the wastewater discharge outlet in the Yellow River.

The impact of state of sediment (suspension, deposition and resuspension) on DON transformation in natural water systems was investigated with samples collected in 2004. Three different stirring regimes, i.e., no stirring, intermittent stirring (stirring for 12 h and without stirring for 12 h), and continuous stirring, were studied. The initial DON concentration was 6.0 mg L^{-1} and the SPS concentration was 5.0 g L^{-1} in each system.

To study the effect of SPS concentration on DON transformation, for the samples collected in 2003, a known amount of sediment was added to a series of flasks, obtaining SPS concentrations of 0, 3.62, 6.32 g L^{-1}, respectively. Then a known amount of organic nitrogen was added to obtain an initial DON concentration of 3 mg L^{-1}. For the samples collected in 2004, the initial DON concentration was 4 mg L^{-1}, and SPS concentrations were 0, 5, 10 g L^{-1}, respectively. The variables of these above experiment conditions were summarized and shown in Table 1.

2.3. Chemical and biological analysis

For the determination of DON, NH_4^+—N, NO_2^-—N, NO_3^-—N in water phase of cultivated systems, aliquots were filtered through 0.45 μm filters before the analysis was conducted. The Nesslerization colorimetric method was applied to the ammonium analyses in water phase; NO_2^-—N was determined with colorimetric method through the formation of a reddish-purple azo dye produced at pH 2.0–2.5 by coupling diazotized sulfanilamide with