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Effect of Inorganic and Organic Suspended Particles on Nitrogen Transformation in Freshwater Column

BACKGROUND AND OBJECTIVES OF THE STUDY

Nitrogen (N) is the most abundant chemical element of the Earth's atmosphere (almost 80%), and also one of the essential components of many key biomolecules (e.g., amino acids, nucleotides). It ranks fourth behind carbon, oxygen and hydrogen as the commonest chemical element in living tissues (Campbell 1990). An increase in the environmental availability of bioavailable N usually boosts life production, firstly increasing the abundance of primary producers. In freshwater system, N is a major nutrient that affects the productivity of freshwater and it is the limiting factor for the growth of many microorganisms.

During the past two centuries, and especially over the last five decades, humans have substantially altered the global nitrogen cycle (as well as the global cycles of other chemical elements), increasing both the availability and the mobility of nitrogen over large regions of Earth (Galloway, Cowling 2002). Higher rates of urbanization have represented huge amount of N to streams, rivers, and estuaries (Howarth et al., 1996; Grimm et al., 2008; Kaushal et al., 2008). Serious N pollution (nitrate concentration: 10–50 mg-N/L) has occurred in areas with significant agricultural activities and small precipitation surpluses (He et al., 2011). Moreover, it is difficult to estimate the lever of N pollution because its sources are widely spread. Increased N loading has been associated with eutrophication and declines in water quality (Vitousek et al., 1997), symptoms of which include harmful algal blooms, hypoxia (low O_2) and anoxia (no O_2) events, fish kills and trophic disruption (Rosenberg, 1985; Kirshtein et al., 1991; Paerl and Carolina, 1997; Paerl et al., 1998; Boesch, 2001).

Although there are many researches investigating N transformation, existing knowledge of some pathways is still fragmentary, which are microbial-mediated N transformation under high suspended sediment (SS) and instream release of dissolved nitrogen (DN) from particulate organic nitrogen (PON). Consequently, the purpose of this study is to elucidate the interaction between organic and inorganic suspended particles and N dynamics in freshwater column. The necessary and details of each pathway will be discussed in the following parts.

Pathway 1: Effect of inorganic suspended solid on nitrogen transformation in freshwater columns

Many rivers in the world (e.g., Asian) contain inorganic suspended solid (ISS) at relatively high concentrations and ISS is often noticed as a source of surface areas for the attachment of microorganisms and substrates (Wang et al. 2005, Xia et al. 2009). Although dam construction has recently reduced the amount of SS transport, about 12.6 Gt/year of SS has transported to the ocean by global river system (Syvitski et al. 2005) and its concentration varies from 5 to 29000 mg/L (Meybeck et al. 2003). The major part of the SS load is silt and clay with minor contributions of sands. In freshwater, such suspended clay has been noticed to affect microorganism growth and N transformation although the results are still complex and detailed mechanisms remain unclear. In N cycles, nitrification is a key biochemical transformation in freshwater and as the initial step of nitrification-denitrification process, it plays a critical role in nitrogen (N) biological removal from both freshwater and wastewater (Kim et al. 2013) to prevent eutrophication, oxygen depletion, and toxic effects to aquatic life in water column (Piehler et al. 2004).

The present study, therefore, investigated the effect of suspended clay on nitrification as a typical transformation in N dynamics in freshwater columns with two specific objectives:

1) Elucidation of detailed nitrification processes under the presence of ISS; and
2) Confirmation of mechanisms under the observed effects of ISS on nitrification.

Pathway 2: Instream release of dissolved nitrogen from particulate organic matter

Microbial decomposition of particulate organic matter (POM) is a key transformation process in N dynamics. Some fragments of that complex process have been pointed out, such as POM provides large surface areas for the attachment of bacteria and stimulates the growth of bacteria and bacterial hydrolytic enzyme activity and is the source of dissolved organic matter (DOM). Fine POM (FPOM) is believed to be more refractory than coarse POM (CPOM) (Yoshimura et al. 2008); hence, its role in N dynamics is often considered negligible. Furthermore, a few researches have been conducted to investigate the transformation of POM to dissolved nitrogen (DN), although it can be an important pathway of the N dynamics in streams which produce DN to water column. Consequently, we investigated the transformation to DN of 16 types of POM, each of which was produced from a single defined source. The materials were classified by size into three groups: CPOM (>1 mm, 6 types), L-FPOM (100–250 µm, 5 types), and M-FPOM (250–500 µm, 5 types). Using these POMs, we conducted DN release experiments. There are two purposes in this study:

1) FPOM, having a relatively large surface area, plays a significant role as DN source in streams, despite being relatively refractory; and

2) DN release rates from CPOM and FPOM is significantly affected by their sources and generation processes.

Structure of the dissertation

Chapter 1 introduces the overall background, the importance of this study, and summary of the main contents and purposes of this study.

Chapter 2 provides a comprehensive literature review about N dynamics and impact factors of N transformations as well as characteristics of SS in global rivers including SS sources, concentration, size, and chemical quality. Furthermore, the interaction between N dynamics and SS has also reviewed.

Chapter 3 and 4 reports on the effect of ISS mineral characteristics and concentration on nitrite oxidation and ammonia oxidation process, respectively. Furthermore, the possible mechanisms under the observed effects were also discussed and compared with results reported previously in other studies.

Chapter 5 provides a comparison about DN release rate from POM varied in size, source, and chemical quality. Focus of this study was placed on the role of FPOM as a DN source and key POM chemical qualities influencing DN release process in freshwater.

Chapter 6 outlines the main findings and provides recommendations for further research and applications.

MATERIAL AND METHODS

Effect of ISS mineral characteristic and concentration on nitrification in freshwater columns

In the present study, the effect of ISS on nitrification was investigated by using dominant mineral types as representatives. Batch experiments were set up in the laboratory to investigate the effect of ISS characteristics and their concentrations on nitrification. There dominant ISS types was montmorillonite, sericite, and kaolinite and concentration range is 0–1000 mg/L. Nitrogen species concentration (i.e., NO_2^-) was measured at several time steps to estimate nitrification rate.

Instream release of dissolved nitrogen from particulate organic matter

We investigated total DN (TDN) and nitrate and nitrite (NO_3^-+ NO_2^-) releases from POM by incubating POM of various sizes and chemical characteristics in stream water over a two-week period. Large- and medium-sized fine POM (L-FPOM, 250–500 μm, and M-FPOM, 100–250 μm) were prepared by feeding five types of coarse POM (CPOM) to shredding amphipods in laboratory aquaria. The DN release experiments were initiated within 3 h of FPOM collection. Each of the 16 POM types with different sizes and sources (6 types of CPOM, 5 types of L-FPOM, and 5 types of M-FPOM) was added separately to 100 mL of filtered stream water (GF/F filter, pore size 0.7 μm) in Erlenmeyer flasks and was incubated in 12°C water for 14 days. Each sample solution was stirred gently at 100 rpm to minimize physical fragmentation due to the shearing of the POM sample. To quantify the DN release rate, water samples were taken from flasks at specified intervals (i.e., 1, 3, 7, and 14d), filtered, and then the DN concentrations were measured.

RESULT AND CONCLUSIONS

Effect of ISS mineral characteristic and concentration on nitrification in freshwater columns

ISS and ammonium oxidation

In the concentration range from 200–1000 mg/L, the introduction of sericite significantly improve ammonium oxidation compared to control experiment (i.e., clear medium). There was no effect of sericite concentration on transformation rates (0.066–0.072 L mg^{-1} d^{-1}). Stimulation effect was also observed with the introduction of montmorillonite although the transformation rate (0.042–0.046 L mg^{-1} d^{-1}) was much lower than those in sericite system. The transformation in the system treated with kaolinite was negligible with kaolinite higher than 200 mg/L.

The mechanism of the result observed was the enhancing effect of sericite on the growth of *Nitrosomonas europaea*, the specific bacterial species for NH_4^+ oxidation. This bacterial cell density was found to be higher in the system treated with sericite and low in other systems. The possibility of sericite as iron source (because of high iron composition) for *Nitrosomonas europaea* growth and enzymatic activity was the proposed to be the main mechanism of this stimulation effect.

ISS and nitrite oxidation

Both of stimulation and inhibition effect of three clay minerals on nitrite oxidation was observed. Under pH= 8.2 and the introduction of sodium ion (i.e., clay particles was dispersed), the higher concentration of montmorillonite and sericite resulted in higher nitrite oxidation rate while kaolinite concentration did not significantly affect the oxidation rate. The role of clay particles as physical supporter (sercite) and source of minerals was expected as the mechanism of the result observed. Under the condition of pH= 7.2 and without the addition of sodium (clay particles was in natural flocculation state), in the system treated with montmorillonite and sericite, the highest nitrite oxidation rate was found when these clays concentration were around 200–500 mg/L and strongly reduced when clay concentration increased to 800–1000 mg/L. Probably the presence of high clay aggregation concentration in the system changed the viscosity of water environment which negatively affected bacterial growth and activities. Similar to previous condition, kaolinite did not significantly affect nitrite oxidation rate.

The role of organic suspended particles as nitrogen source in freshwater columns

FPOM was produced by feeding conditioned CPOM (i.e., conditioned leaf litters, twig) to amphipod in laboratory aquaria. Two FPOM size fractions were produced (i.e., L-FPOM 250–500 µm, and M-FPOM 100–250 µm). Compared to CPOM chemical quality, FPOM fractions were more recalcitrant (i.e., higher linocellulose index) but they have larger surface area than CPOM (4–40 fold for L-FPOM and CPOM and 6–75 fold for F-POM and CPOM). The release of DN from L-FPOM was low during the first day but comparable or even higher than that of CPOM during the second week of the experiment. The contribution of L-FPOM on DN release on the first day and the second week were 17 and 52%, respectively. Although FPOM is relatively recalcitrant, it has high surface area and accounts for a dominant budget of stream POM. This study confirmed that DN release from FPOM especially L-POM was comparable with that from CPOM. Thus, FPOM should be considered as an important N source in stream.

In summary, this study provided deep understanding about the interaction between suspended particle and nitrogen transformation, which aimed to support water quality management in freshwater column. Nitrification is one of the key transformation and the first step in N removal process in river and one of component of water quality model. The estimation of nitrification rate is one of the crucial step in water quality calibration and validation steps. The overestimate or underestimate of transformation rate can result in poor performance of the model and affect decision making in water quality planning. The result from this study about the effect of clay, which is very common with high concentration in many turbid rivers in Asia countries, on nitrification showed that transformation rate of both sub processes of nitrification was strongly affected by the appearance of clay characteristics and concentration. So the investigation of suspended sediment quality and concentration is necessary for the estimation of nitrification rate. Information about N source is also a critical part of N model. Hence the underestimation of FPOM as a nitrogen source can be a potential cause of eutrophication which resulted in water quality degradation and fish killed.

In the future study, the effect of clay on other transformation in N dynamics is needed to investigate to have a comprehensive understanding about inorganic suspended particle and N cycle relationship. Furthermore, the investigation of DN release from FPOM formed from different processed (i.e., not amphipod-produced FPOM) is needed to have a more general conclusion about the role of FPOM as DN source in stream.

Moreover, in this study, sericite was observed to stimulate both ammonium and nitrite oxidation the most among three clay minerals. The application of montmorillonite and kaolinite in waste water treatment plan to improve bacterial growth, activities and nitrification rate has been reported in some researches before. Thus, this study results suggest the application of sericite in waste water treatment plant to increase the efficiency of N treatment and removal.

List of Publications

List of Journal Papers

1. Quynh Nga Le, Manabu Fujii, Chihiro Yoshimura and Klement Tockner. Dissolved nitrogen release from coarse and amphipod-produced fine particulate organic matter in freshwater column. **Limnology** (in press).

2. Quynh Nga Le, Manabu Fujii and Chihiro Yoshimur. Effect of concentration and size of inorganic suspended solids on microbial-mediated nitrogen transformation in freshwater column. **Sustainable Environment Research** (accepted).

3. Quynh Nga Le, Manabu Fujii and Chihiro Yoshimura 2013. Effect of concentration and size of suspended sediment on nitrogen dynamics in freshwater ecosystem. **Journal of Science & Technology, Vietnam** 51(2B):108-115 (Printed).

Conferences papers

1. Quynh Nga Le, Manabu Fujii and Chihiro Yoshimura. Effect of clay particles on nitrification process in freshwater. 7th ASEAN Environmental Engineering Conference, Palawan, Philippine, Nov. 2014

2. Quynh Nga Le, Manabu Fujii and Chihiro Yoshimura. Effect of concentration and size of suspended sediment on nitrogen dynamics in freshwater ecosystem. Water and Environment Technology Conference, Jun. 2013

3. Quynh Nga Le, Manabu Fujii and Chihiro Yoshimura. Effect of concentration and size of suspended sediment on nitrogen dynamics in freshwater ecosystem. The 5th ASEAN Civil Engineering Conference (ACEC), the 5th ASEAN Environmental Engineering Conference (AEEC) and the 3rd Seminar on Asian Water Environment (Asian Core Program of JSPS, NRCT and ERDT)-Ho Chi Minh City, Vietnam Oct. 2012.

List of Journal Papers in Preparation

1. Quynh Nga Le, Chihiro Yoshimura and Manabu Fujii. Effect of suspended clay minerals on nitrification in freshwater column (Target journal: Water Science and Technology)

2. Quynh Nga Le, Chihiro Yoshimura and Manabu Fujii. Effect of inorganic suspended solids concentration and mineral characteristic on nitrification process in freshwater column (Target journal: Biogeochemistry)