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Influence of silver nanoparticles on benthic oxygen consumption of microbial communities in freshwater sediments determined by microelectrodes☆

Lingzhan Miao [a, b], Chao Wang [a, b], Jun Hou [a, b, *], Peifang Wang [a, b], Yanhui Ao [a, b], Yi Li [a, b], Yu Yao [a, b], Bowen Lv [a, b], Yangyang Yang [a, b], Guoxiang You [a, b], Yi Xu [a, b]

[a] Key Laboratory of Integrated Regulation and Resources Development on Shallow Lakes, Ministry of Education, Hohai University, Nanjing 210098, People's Republic of China
[b] College of Environment, Hohai University, Nanjing 210098, People's Republic of China

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ABSTRACT

Increasing use of silver nanoparticles (NPs) will inevitably result in release of these particles into aquatic environments, with sediments as a substantial sink. Nevertheless it remained largely unknown whether Ag NPs present potential impacts in sediment functioning. In this study, thus, a microcosm approach was constructed and the potential impacts of Ag NPs and PVP-coated Ag NPs on the oxygen consumption in freshwater sediments (collected from Taihu Lake) were determined using oxygen microelectrodes. To our knowledge, this is the first time that microelectrodes have been used to estimate the impacts of Ag NPs in sediments. The steady-state oxygen microprofiles showed that environmental relevant concentration (1 mg/L nano-Ag) did not lead to an apparent change of the oxygen consumption rates of benthic microbial communities in sediment. PVP-coating reduced the nano-toxicity of Ag NPs on the benthic microorganism. While, the addition of 10 mg/L uncoated Ag NPs resulted in remarkable differences in the oxygen concentration profiles within 4–5 h, and significantly inhibited the oxygen consumption of benthic microbial communities in the upper sediment layer after 100 h. Simultaneously, an increase of oxygen consumption in sediment lower zones was observed. These results could be explained by that aerobic microorganisms inhabiting at the upper layer of the sediment reduced metabolic activity to avoid toxic stress of Ag NPs; concomitantly, facultative aerobes harboring below the metabolically active upper layer switched from fermentation or anaerobic respiration to aerobic respiration as an increase of oxygen bioavailability in the lower zones in sediment.

1. Introduction

Silver nanoparticles (Ag NPs) are widely incorporated into many applications including domestic, medical, industrial and environmental products (Nel et al., 2006) for their catalyst or antimicrobial properties. Given their broad use, Ag NPs are expected to be discharged into the environment through waste disposal and other routes (Colman et al., 2014; Zhang et al., 2016). It has already been detected of Ag NPs in wastewater streams (Brar et al., 2010), and with increasing production and use, their concentration in the environment and negative impacts will rise. The toxic effects of Ag NPs have been generally studied on bacteria, yeast, algae, activated sludge and microbial aggregates (Carlson et al., 2008; Fabrega et al., 2009, 2011a), and the potential mechanisms mainly include direct physical membrane disruption (Fabrega et al., 2009), simulation of ROS production (Choi et al., 2010), releasing Ag^{+} and causing cellular enzyme deactivation (Fabrega et al., 2011a), leading to microbial growth inhibition, cell disruption and death.

During their entire life cycle, Ag NPs are inevitably discharged into aquatic environments (Fabrega et al., 2011a; Zhang et al., 2016). It has been documented that the released Ag NPs exhibit high nanotoxicities to microbial communities (Dwivedi et al., 2015), including being inhibitory to phytoplankton (Das et al., 2014), disrupting the natural bacterial communities (Doiron et al., 2012; Gil-Allué et al., 2015) and negatively affecting microbial biofilm development in natural water (Fabrega et al., 2011b). Environmental conditions and nanoparticle properties may alter the physicochemical behavior by

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* Corresponding author. College of Environment, Hohai University, 1 Xikang Road, Nanjing 210098, People's Republic of China.
E-mail addresses: hhuhjyhj@126.com, pfwang2005@hhu.edu.cn (J. Hou).

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promoting surface complexation, and as a result their toxicity (Colman et al., 2012; Gil-Allué et al., 2015). The transport behavior of nanoparticles are mainly dominated by aggregation and sedimentation (Kaegi et al., 2013; Zhang et al., 2016), and nanoparticles were assumed to be mainly accumulated in the sediments after their deposition in the water column (Quik et al., 2014). Indeed, it has been proposed that aquatic sediments are expected to be a substantial sink for most of Ag NPs (Cross et al., 2015; Maurer-Jones et al., 2013). Environmental fate models have predicted low concentrations of ENMs in sediments, i.e., 40–2000 μg/kg (on the basis of dry weight) for worst-case scenarios, which could increase taking into account future emission scenarios (Dale et al., 2015; Gottschalk et al., 2013; Markus et al., 2015). Consequently, the accumulation of Ag NPs in sediments may negatively influence the functioning of sediment routing systems and on its role in fulfilling ecosystem processes (Gao et al., 2011; Jourabchi et al., 2008; Mohanty et al., 2014). Thus, studies relating to the transport behavior of Ag NPs and the potential effects in sediment systems are warranted.

Sediments microorganism play a crucial role in many biogeochemical cycling, including carbon, nitrogen, Sulphur and phosphorous cycles (Battin et al., 2008). Due to the close contact with sediment-particles and pore water, benthic communities are very vulnerable to sediment contamination, and considered as a highly relevant and sensitive agent for monitoring artificial influence (Antizar-Ladislao et al., 2015). Several studies have been performed to evaluate the effects of Ag NPs on microbial communities in aquatic sediment, however, little consensus was achieved regarding the degree or even the presence of impacts. For example, exposure of Ag NPs was found to have minor or no negative impact on the metabolic function (Echavarri-Bravo et al., 2015), enzymatic activity (Colman et al., 2012) nor on the microbial community structure in the sediment systems (Bradford et al., 2009; Moore et al., 2016). As the biogeochemical conditions in sediment are controlled by the benthic microbial communities and exhibit high heterogeneity (predominant in vertical direction) with the metabolically most active zone close to the sediment-water interface (Jourabchi et al., 2008). Therefore, methods with high-resolution were necessary to better characterize the potential impacts of Ag nanoparticles on microbial communities in surface sediment in situ.

In this study, the potential impacts of Ag NPs on benthic microbial communities in term of oxygen concentration distribution and oxygen consumption were investigated using oxygen microelectrodes. Benthic O_2 availability plays a vital role in many important biogeochemical processes and has essential influence on the biology and ecology of benthic communities (Hondzo and Steinberger, 2008). Microelectrodes are effective and promising research tools allowing the high-resolution determination of the oxygen diffusion in sediments under almost in situ conditions. Similar approaches have been performed to study the inhibitory effects of heavy metal in sediment (Viret et al., 2006) and in microbial mats (Nguyen et al., 2012). In order to achieve this goal, freshwater sediments were collected from Taihu Lake and a microcosm approach was established, and then the inhibitory effects of Ag NPs on the oxygen consumption in freshwater sediments were determined using oxygen microelectrodes. This study will give new insights into the potential impacts of Ag NPs on the benthic microbial communities in freshwater sediment.

2. Materials and methods

2.1. Sample collection and microcosm set up

The sediment and overlying water were collected from Lake Taihu (31°45′97″ N, 120°04′22″ E), the third largest freshwater lake in China. With the development of economy in Taihu Lake catchment in the last two decades, a mass of industrial waste water and domestic sewage were discharged into the Lake (Tao et al., 2012). Sediment samples were collected from between 0 and 8 cm using a gravity corer (Rigo Co., Japan), and brought back to the lab on ice in a polyethylene bag. According to the procedures provided in our previous study (Wang et al., 2016), the sediment core was sectioned at 2 cm intervals and then sediment samples from homologous depths were mixed to avert heterogeneity. The natural sediment was wet-sieved through <0.6 mm sieve to remove gravel and large particles. Finally, they were transferred into cylindrical 6 L polypropylene (PP) containers (18 cm inner diameter) up to their original depths (yielding a ~6 cm layer of sediment), and carefully overlaid with 10 cm of filtered overlying water from the sampling site.

All the containers used for sampling were thoroughly acid washed (10% v/v HNO_3, overnight) and rinsed once with lake water before use. Containers were always covered with parafilms and controlled at a 12 h of dark cycle. O_2 concentration in the overlying water was managed using a bubbling air diffusor, which maintained a well-mixed overlying water phase (oxygen-saturated environment). The sediments were cultured in a constant-temperature room at 20 °C, as samples were collected in the summer (August, 2015). The physicochemical parameters in the water, including pH, conductivity, DO, and ORP were measured once every two days and three weeks of equilibration were needed for the stabilization of these parameters.

2.2. Characterization of silver nanoparticles

Two types of commercial Ag NPs (uncoated Ag NPs and PVP coated Ag NPs) were purchased in the form of dry powder from Sigma-Aldrich (St. Louis, MO, USA). The average size of these two NPs, as determined by transmission electron microscopy (TEM), was 80 ± 16 nm and 105 ± 24 nm in diameter, respectively, which is consistent with the manufacturer's description. Stock solutions of 100 mg/L uncoated Ag NPs and PVP-Ag NPs were prepared by adding 50 mg Ag NPs to 500 mL filtered and autoclaved Milli-Q water (18.2 MΩ cm^{-1}, Milipore). The nano-Ag suspension was placed in an ultrasonic ice water bath and sonicated for 30 min at 400 W (Miao et al., 2016). Test nanoparticle solutions were prepared immediately prior to NPs exposure. The primary morphology and particle size of the nanoparticle suspension were analyzed using scanning electron microscopy (Hitachi S-4800). The hydrodynamic diameter (HDD) and zeta potential were measured using a Malvern Zetasizer Nano ZSP (Malvern Instruments, U.K.).

Prior to the exposure experiment, the colloidal behavior of the two NPs in the filtered lake-water (through 0.22 μm) were studied in order to better understand the transport behaviors of NPs in the water column. In this study, the sediment samples were incubated in a static system, and the suspended solids in the water column were determined at relatively low level (0.52 ± 0.2 mg/L) during the pseudo steady-state conditions in the containers. Thus, the hetero-aggregation processes (aggregation between nanoparticles and suspended particles in the aqueous solution) in the microcosms were not taken consideration in this study. Homo-aggregation of the nano-Ag suspensions was studied at 10 mg/L (which can give a good detected signal according to our preliminary test) in the filtered overlying water (through 0.22 μm) using Malvern Zetasizer Nano ZSP. Experiments were carried out in the container (described in Section 2.1) without sediment, under daylight conditions at 20 °C temperature. Two nano-Ag stock suspensions were added into the container containing 2.6 L filtered overlying water (yielding a ~10 cm layer of water), and then, the particle size distributions and zeta potentials of two NPs in the water column (at ~5 cm from the air/water interface) were

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