



Diurnal characteristics of migration and transformation of mercury and effects of nitrate in Jialing River, Chongqing, China



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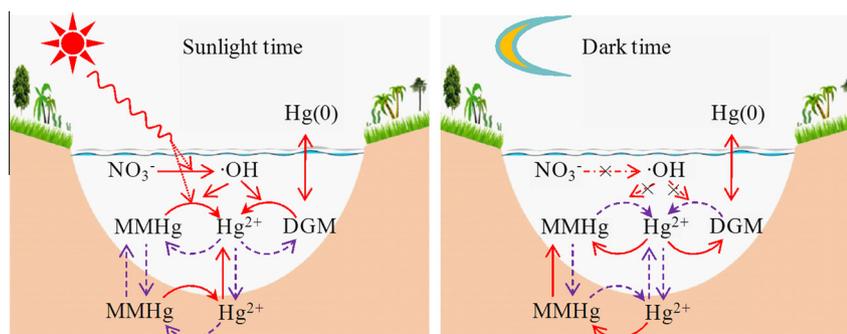
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HIGHLIGHTS

- We identified MMHg, RHg, and THg fate with NO_3^- during daytime and nighttime.
- There were strong diurnal signals of diffusive flux for MMHg, RHg, and THg.
- We estimated the effects of MMHg PD on Hg cycling in aquatic systems.
- We calculated the contribution rate of diffusive flux to overlying water.

GRAPHICAL ABSTRACT



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ABSTRACT

Laboratory incubation experiments were performed to identify diurnal characteristics of migration and transformation of mercury (Hg) and effects of nitrate (NO_3^- , a hydroxyl radical donor by photolysis) in Jialing River, Chongqing, China. It is found that there are strong diurnal signals of [monomethylmercury (MMHg)] and [reactive Hg (RHg)] in sediment, pore water and overlying water, which suggest that solar radiation may be an important variable that involved in aquatic Hg cycling. Photo-degradation (PD) of MMHg plays a key role in Hg cycling in water systems, and the rates are measured to be 38.22% in March, 2012. The presence of NO_3^- has a marked effect on MMHg PD under solar radiation, and affects inorganic species reducing to Hg^0 , resulting in more Hg species available for methylation. So NO_3^- is an important factor for Hg cycling in water systems. Diffusive flux of MMHg, RHg and dissolved Hg (DHg) are 1.92–2.34, 3.43–3.64 and 3.04–5.71 $\text{ng m}^{-2} \text{d}^{-1}$ at daytime, and 6.04–6.92, 3.22–3.25 and 7.79–8.37 $\text{ng m}^{-2} \text{d}^{-1}$ at nighttime, respectively, implying that sediment is a major Hg source for shallow-water area in Jialing River at springtime. These results show great importance for understanding Hg biogeochemical processes in clear, oligotrophic, shallow, sluggish, and agriculturally-impacted waters.

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1. Introduction

Mercury (Hg), a toxic element, is widely distributed in aquatic systems, and has been listed as a priority pollutant by many international agencies (Wang et al., 2004). Inorganic Hg (IHg) can be methylated to monomethylmercury (MMHg), a high bioavailable and toxic Hg form, by biological and/or chemical processes

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(Wang et al., 2009). It can be accumulated through aquatic biota food webs and thus pose a detrimental threat to humans and other piscivorous wildlife (Ullrich et al., 2001; Qureshi et al., 2009). Many studies have identified that sediment is the predominant methylation site, and that the distribution and concentration of Hg in sediment and pore water have significant effects on Hg distribution in water column (Choe et al., 2004; Hines et al., 2004; Rothenberg et al., 2008). Thus, sediment may play an important role in biogeochemical cycling of Hg in water systems.

Methylation, demethylation, reduction, and oxidation processes of Hg induced by photochemical reactions contribute a key portion for Hg cycling in aquatic systems (Seller et al., 1996; Siciliano et al., 2005; Whalin et al., 2007; Lehnher and St. Louis, 2009). The production and destruction of MMHg play a marked role in determining the concentration of bioavailable forms of Hg in ecosystems. Photochemical demethylation (PD) of MMHg has been shown to be the most important geochemical mechanism for MMHg degradation into inorganic forms in freshwater lakes, thus resulting in reducing bio-availability of MMHg for accumulation through aquatic food webs (Black et al., 2012). Meanwhile, the speciation of these inorganic products from MMHg PD is of great importance for estimating the biogeochemical cycling of Hg. Both of reduction and oxidation processes control the production of dissolved gaseous mercury (DGM), which could result in Hg pollution for remote ecosystems via volatilization, transport, and atmospheric deposition (Vost et al., 2012). Although many research works have been done for Hg biogeochemical cycling in water systems, the photochemical characteristics of IHg forms generated from MMHg PD are still unknown. Also, whether the solar radiation has effects on sediment Hg concentration in clear, oligotrophic, shallow, and sluggish waters are unclear.

Hydroxyl radicals ($\cdot\text{OH}$), an extremely reactive oxidant at daytime, which can be mainly produced by nitrate ion (NO_3^-) photolysis (Brezonik and Fulkerson-Brekken, 1998). Previous results confirmed that $\cdot\text{OH}$ is a principal intermediate involved in biogeochemical processes of Hg in natural waters. It hold a capacity for nonselectively attacking C–Hg bond (Chen et al., 2003; Zhang and Hsu-Kim, 2010; Kim and Zoh, 2013), thus improving MMHg PD process, which can generate IHg (Hg(I) and Hg(0)) in aqueous phase (Gårdfeldt et al., 2001; Sun et al., 2013). While $\cdot\text{OH}$ also hold a capacity for oxidizing Hg(I) and Hg(0) to higher valence state (Hg(II)) (Gårdfeldt et al., 2001), which decrease the concentration of DGM, and result in more available Hg for methylation. However, the impact of photochemical reactions of Hg involved by $\cdot\text{OH}$ in overlying water on Hg cycling in water systems is unclear.

From the knowledge gaps outlined above, our objectives were to (1) investigate Hg cycling/transformation characteristics during daytime and nighttime in the presence of NO_3^- , (2) identify the impact of MMHg PD induced by $\cdot\text{OH}$ on Hg cycling, and (3) calculate Hg diffusive flux across sediment–water interface and estimate their contribution-rate to overlying water for sunlight time and nighttime with NO_3^- .

2. Methods

2.1. Chemicals and materials

Methylmercury chloride (MMHg) standard solution was purchased from Brooks Rand LLC (Seattle, WA, USA). Potassium nitrate (KNO_3), benzoic acid (BA), nitric acid, and hydrochloric acid were reagent grade and purchased from Guangfu Chemical Company (Tianjin, China). The details of experimental set-up and materials are provided in [Supplementary Information \(SI\)](#).

2.2. Experimental design and procedures

Diurnal characteristics of migration and transformation of Hg and effect of NO_3^- in Jialing River, Chongqing, China were investigated March 18–27, 2012. Experiments were performed on a building roof in Southwest University (29°48′51.49″ N, 106°24′42.60″ E), which is about 3 km distant from sampling site (29°50′16.89″ N, 106°25′58.47″ E). [Table 1](#) shows the amendment, experimental set-up, objective, and explanation for each treatment.

For experiment Nos. 1 and 2, three bottles of each treatment were taken into laboratory every incubation time. Surficial sediment samples were collected by a mini-sampler to an ice-cooled container, and were stored in a refrigerator at -20°C . Pore water was extracted from wet sediment within 30 min by centrifugation at 3000 r min^{-1} for 30 min in lab, and then filtered through a $0.45\ \mu\text{m}$ polyvinylidene fluoride (PVDF) membrane (Millipore). The resulting pore waters and overlying waters were collected to borosilicate glass bottles and acidified using a 0.5% HCl solution (He et al., 2007), and were stored in a refrigerator at 4°C . The operation procedures of experiment No. 3 are provided in SI.

2.3. Mercury analysis

Prior to the analyses, sediment samples were lyophilization, ground, and sieved to minus 100 meshes particle sizes. Total Hg (THg) in sediment was determined by thermal decomposition

Table 1
Details of experimental design.

Experiment No.	Amendment and treatment	Experimental set-up	Objective and explanation
1	(a) 2 mg BA L^{-1} , (b) 2 mg BA L^{-1} + 15 mg $\text{NO}_3^- \text{L}^{-1}$, (c) nothing (CK), (d) 2.5 mg $\text{NO}_3^- \text{L}^{-1}$, (e) 10 mg $\text{NO}_3^- \text{L}^{-1}$, and (f) 15 mg $\text{NO}_3^- \text{L}^{-1}$	Fig. S1	To identify Hg cycling characteristics and effects of $\cdot\text{OH}$ in water system. The natural water and undisturbed sediment were collected from Jialing River by gloved hands to fill Teflon reactors
2	(a) 2 mg BA L^{-1} , (b) 2 mg BA L^{-1} + 15 mg $\text{NO}_3^- \text{L}^{-1}$, (c) nothing (CK), (d) 2.5 mg $\text{NO}_3^- \text{L}^{-1}$, (e) 10 mg $\text{NO}_3^- \text{L}^{-1}$, and (f) 15 mg $\text{NO}_3^- \text{L}^{-1}$	Fig. S2	To clarify MMHg PD characteristics and roles of $\cdot\text{OH}$, also, to elucidate the role of MMHg PD in Hg cycling in water system. Both of unfiltered and filtered ($0.22\ \mu\text{m}$) water were incubated to characterize the effects of particulate and microorganism on MMHg PD
3	(a) Nothing, (b) MMHg, (c) MMHg + 2 mg BA L^{-1} , (d) MMHg + 2 mg BA L^{-1} + 15 mg $\text{NO}_3^- \text{L}^{-1}$, (e) MMHg + 2.5 mg $\text{NO}_3^- \text{L}^{-1}$, (f) MMHg + 10 mg $\text{NO}_3^- \text{L}^{-1}$, and (j) MMHg + 15 mg $\text{NO}_3^- \text{L}^{-1}$	Fig. S3	To investigate mechanism of MMHg PD in the presence of $\cdot\text{OH}$. These experiments were conducted at midday to get similar light intensities ($\sim 5.8\ \text{E m}^{-2}\ \text{h}^{-1}$). MMHg was spiked to a final concentration of $20\ \text{ng L}^{-1}$

Note: the light conditions of experiment Nos. 1, 2 and 3 were natural sunlight and dark. KNO_3 was amended as NO_3^- donor, which can generate $\cdot\text{OH}$ by photolysis. BA was used as $\cdot\text{OH}$ scavenger. For experiment Nos. 1 and 2, all treatments were allowed to equilibrate for >12 h under dark at 18°C prior to incubation. Each treatment was distributed into eight incubation times (01:00, 04:00, 07:00, 10:00, 13:00, 16:00, 19:00, and 22:00). Every time-point was performed in triplicate. For experiment No. 3, each treatment was performed in triplicate.

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