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Photodegradation of methylmercury in Jialing River of Chongqing, China

Rongguo Sun[1,2], *Dingyong Wang*[1,3,*], *Wen Mao*[1], *Shibo Zhao*[1], *Cheng Zhang*[1,3]

1. *Chongqing Key Laboratory of Soil Multiscale Interfacial Process, College of Resource and Environment, Southwest University, Chongqing 400715, China. E-mail:* sunrongguo88@163.com
2. *School of Chemistry and Material, Guizhou Normal University, Guiyang 550001, China*
3. *Chongqing Key Laboratory of Agricultural Resources and Environment, Chongqing 400716, China*

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ABSTRACT

Photodegradation (PD) of methylmercury (MMHg) is a key process of mercury (Hg) cycling in water systems, maintaining MMHg at a low level in water systems. However, we possess little knowledge of this important process in the Jialing River of Chongqing, China. *In situ* incubation experiments were thus performed to measure temporal patterns and influencing factors of MMHg PD in this river. The results showed that MMHg underwent a net demethylation process under solar radiation in the water column, which predominantly occurred in surface waters. For surface water, the highest PD rate constants were observed in spring ($12 \times 10^{-3} \pm 1.5 \times 10^{-3}$ m^2/E), followed by summer ($9.0 \times 10^{-3} \pm 1.2 \times 10^{-3}$ m^2/E), autumn ($1.4 \times 10^{-3} \pm 0.12 \times 10^{-3}$ m^2/E), and winter ($0.78 \times 10^{-3} \pm 0.11 \times 10^{-3}$ m^2/E). UV-A radiation (320–400 nm), UV-B radiation (280–320 nm), and photosynthetically active radiation (PAR, 400–700 nm) accounted for 43%–64%, 14%–31%, and 16%–45% of MMHg PD, respectively. PD rate constants varied substantially with the treatments that filtered the river water and amended it with chemicals (*i.e.*, Cl^-, NO_3^-, dissolved organic matter (DOM), Fe(III)), which reveals that suspended particulate matter and water components are important factors in affecting the PD process. For the entire water column, the PD rate constant determined for each wavelength range decreased rapidly with water depth. UV-A, UV-B, and PAR contributed 27%–46%, 6.2%–12%, and 42%–65% to the PD process, respectively. PD flux was estimated to be 4.7 μg/(m^2·year) in the study site. Our results are very important to understand the cycling characteristics of MMHg in the Jialing River of Chongqing, China.
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Introduction

Mercury (Hg), a potent neurotoxin, is widely distributed in the environment, and has been listed as a priority pollutant by many international agencies (Wang et al., 2004; Wu et al., 2008). Inorganic forms of Hg (IHg), once released into the environment (especially aquatic systems), can be methylated to methylmercury (MMHg), a highly bioavailable and toxic Hg form, by biological and/or chemical processes (Wang et al., 2009). It can be accumulated through aquatic biota food webs and pose a serious threat to humans and piscivorous wildlife (Ullrich et al., 2001; Yang et al., 2007; Qureshi et al., 2009;

* *Corresponding author*. E-mail: dywang@swu.edu.cn (Dingyong Wang).

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Zheng et al., 2010; Zhang et al., 2011; Wang and Zhang, 2013). Thus, MMHg cycling characteristics in water systems should be given careful consideration.

Photodegradation (PD) is the dominant sink of MMHg in surface waters, resulting in a low level of MMHg and reducing the availability of Hg for bioaccumulation in aquatic food webs (Seller et al., 1996; Li et al., 2010). This process plays an important role in Hg cycling in ecosystems, and nearly 59%–80% of MMHg in lake waters can be removed by the PD mechanism (Seller et al., 1996; Lehnherr and St. Louis, 2009). The inconsistent results for MMHg PD obtained from different water systems show that the pathway of MMHg PD in natural waters may vary in different ecosystems, and that many environmental factors are involved in the PD process (Hammerschmidt and Fitzgerald, 2006; Lehnherr and St. Louis, 2009; Li et al., 2010; Black et al., 2012). The MMHg PD process can be achieved *via* a direct pathway by ultraviolet (UV) radiation (185–254 nm) and/or an indirect pathway involving .OH, $^1O_2$ in surface waters (Inoko, 1981; Zepp et al., 1987; Suda et al., 1993; Gårdfeldt et al., 2001; Chen et al., 2003; Hammerschmidt and Fitzgerald, 2010; Zhang and Hsu-Kim, 2010). In addition, the PD process can be inhibited by dissolved organic matter (DOM), salinity, and suspended particulate matter (SPM) through complexation of MMHg with DOM or Cl^-, or through the influence of DOM on photo-penetration (Ravichandran, 2004; Siciliano et al., 2005; Sun et al., 2013). Although previous studies have illustrated some mechanisms of MMHg PD in surface waters, the entire suite of environmental variables affecting MMHg PD has yet to be fully elucidated.

The Jialing River is the largest tributary of the Yangtze River, where the biogeochemical cycling, output, and input of Hg have been of great concern (Wang and Zhang, 2013). However, we possess little knowledge of these possesses. Thus the investigation of MMHg PD is very important for understanding the cycling characteristics of Hg in the Jialing River, and knowing the input and output of Hg from the Jialing River to the Yangtze River. Meanwhile, some lakes have been extensively studied regarding MMHg PD, but much less is known about rivers (Tsui et al., 2013). The environmental conditions of the Jialing River are significantly different from the ecosystems whose MMHg PD has been extensively studied (Seller et al., 1996; Hammerschmidt and Fitzgerald, 2006; Lehnherr and St. Louis, 2009). Therefore, it is necessary to research the PD characteristics of MMHg in the Jialing River.

The objectives of this study were to (1) measure rate constants, flux, and temporal patterns of MMHg PD in the Jialing River of Chongqing, China; and (2) identify the effects of environmental factors, such as light intensity and wavelength range, anions and cations, DOM, and SPM, on MMHg PD in this area.

1. Materials and methods

1.1. Site description

The monitoring site is located in Beibei district, Chongqing, China, with a north latitude of 29°50′16.89″ and an east longitude of 106°25′58.47″. The location of the incubation site is illustrated in Fig. 1. The physicochemical characteristics of the study site are provided in Table 1.

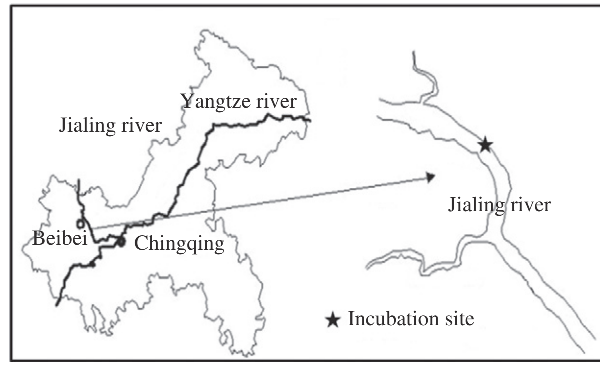

Fig. 1 – Schematic diagram of incubation site, Jialing River of Chongqing, China.

1.2. Materials and methods

The characteristics of the MMHg PD rate were investigated by incubating river water in borosilicate glass bottles (diameter 45 cm, height 8 cm) in October 2012 (autumn) and in January (winter), April (spring), and July (summer) 2013. Borosilicate glass bottles wrapped by various films were incubated at different depths to investigate vertical patterns of PD rate constants in various wavelength ranges (280–700 nm, UV-A, UV-B, photosynthetically active radiation (PAR), and darkness). Detailed descriptions of the films have been presented by Lehnherr and St. Louis (2009). The average transmittances of UV-B, UV-A, and PAR through the borosilicate bottles were 71%, 77%, and 81%, respectively.

The effects of DOM and other chemicals on MMHg PD in surface water were studied in July 2013. DOM was amended to be 6.0, 10, and 20 mg/L to investigate the role of DOM in MMHg PD, and a DOM solution was isolated from vegetation on

Table 1 – Physicochemical characteristics of study site.

Environmental parameters	Date			
	2012–10	2013–01	2013–04	2013–07
Light intensity (E/(m^2·day))				
PAR	31	19	76	100
UV-A (320–400 nm)	2.4	1.3	5.8	7.6
UV-B (280–320 nm)	0.13	0.067	0.31	0.40
Water temperature (°C)	21	12	17	25
DO (mg/L)	7.1	9.4	8.6	7.8
pH	7.6	7.3	7.5	8.0
Cl^- (mg/L)	1.2	1.1	1.5	5.7
NO_3^- (mg/L)	0.32	0.47	0.46	1.5
SPM (g/L)	0.24	0.19	0.011	17
Fe(III) (μg/L)	31	24	29	36
Unfiltered: THg (ng/L)	9.5	11	8.3	17
Unfiltered: MMHg (ng/L)	0.14	0.10	0.16	0.37
Filtered: THg (ng/L)	8.3	9.7	7.4	8.2
Filtered: MMHg (ng/L)	0.11	0.080	0.15	0.12
DOM (mg/L)	2.5	2.0	2.2	3.3
Transparency (m)	1.6	1.8	1.2	0.22

PAR: photosynthetically active radiation.

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