JOURNAL OF ENVIRONMENTAL SCIENCES 26 (2014) 1445–1452

Available online at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/jes

A simulation study of mercury release fluxes from soils in wet–dry rotation environment

Peng Liang[1,3], *Cheng Zhang*[1], *Yongkui Yang*[1], *Dingyong Wang*[1,2,*]

1. *College of Resources and Environment, Southwest University, Chongqing 400715, China. E-mail: pengliang2727@gmail.com*
2. *Chongqing Key Laboratory of Soil Multi-scale Interfacial Process, Chongqing 400715, China*
3. *School of Environmental and Resource Sciences, Zhejiang A&F University, Lin'an, Zhejiang 311300, China*

ARTICLE INFO

Article history:
Received 10 August 2013
Revised 14 November 2013
Accepted 31 December 2013
Available online 13 June 2014

Keywords:
Mercury
Flux
Dynamic flux chamber
Wet–dry rotation

ABSTRACT

A simulative mesocosm study was conducted to evaluate the influence of wet–dry rotation on mercury (Hg) flux from soil/water to air and the distribution of Hg species in water as well as Hg chemical fractions in soil. Three types of soil were employed including two kinds of paddy soil, Typic Purpli-Udic Cambosols (TPUC) and Xanthi-Udic Ferralosols (XUF), as well as the Alluvial Soil (AS) from Three Gorge reservoir area in Chongqing, China. The results showed that Hg fluxes in wetting periods were significantly higher than that in drying periods. It might be due to the formation of a layer of stable air over the water surface in which some redox reactions promote evasion processes over the water surface. This result indicated that more Hg would be evaporated from the Three Gorge reservoir and paddy soil field during the flooding season. Hg fluxes were positively correlated with air temperature and solar irradiation, while negatively correlated with air humidity and the electronic conductivity of water. Hg fluxes from AS and TPUC were significantly higher than that from XUF, which might be due to the higher organic matter (OM) contents in XUF than TPUC and AS. The reduction processes of oxidized Hg were restrained due to the strong binding of Hg to OM, resulting in the decrease in Hg flux from the soil.

Introduction

Mercury (Hg) is one of the most toxic pollutants in the world. Its geochemical cycling and health perspectives have been paid great attention to by scientists, governments and the general public since last century. Hg is a global contaminant since it can stay in the atmosphere for about 0.5–2 years (Fu et al., 2010), and transport to remote areas without Hg emission sources (Fitzgerald et al., 1998). Both natural and anthropogenic sources have contributed to the global Hg pool in the environment. Natural resources generally include the emissions from natural reservoirs (e.g., volcanic activity and weathering of rocks) and the re-emission of Hg deposited previously from anthropogenic and natural sources. The emission of Hg from soil/water surface accounted 85% of the total natural release of Hg (Pirrone et al., 2010).

Hg flux at the interface between atmosphere and surface water is a function of the equilibrium between the atmospheric and surface water concentrations of dissolved Hg^0 and a turbulent enhanced transfer rate (Rolfhus and Fitzgerald, 2001). The concentration gradient of Hg between the water and air, the solar irradiation and the temperature of water and air are three main factors influencing the release of Hg^0 (Pirrone et al., 2010). The Hg^0 primarily comes from the photoreduction of aqueous Hg^{2+} and particulate-bound Hg (Feng et al., 2002). The release of Hg from soil to air was found to be influenced by many factors, including Hg concentrations in soil, soil organic matter content, soil water

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

http://dx.doi.org/10.1016/j.jes.2014.05.010
1001-0742/

content and soil temperature (Gustin and Stamenkovic, 2005; Rinklebe et al., 2010; Yang et al., 2007). Rising soil water content can promote the reduction of Hg^{2+} to Hg^0 and subsequent Hg volatilization exponentially (Song and Van Heyst, 2005). Increasing soil temperatures due to solar irradiation are also promoted Hg^0 emissions (Wang et al., 2006). The vapor pressure of volatile Hg species was increased with rising soil temperatures and decreasing Hg sorption to the soil.

Paddy field for rice growth plays an important role in agriculture production. It has been demonstrated that methylmercury can be accumulated in the grain of rice, which poses big threat to the human health through rice consumption (Kosmulski, 2002). Whereas soil is the potential source of methylmercury in the tissues of rice plants, methylmercury in soil is easy to be absorbed by roots and then translocated to the above-ground parts (Meng et al., 2011). The water fluctuating area is an important area since it is the last riparian zone of the reservoir, which works on blocking the pollutants and elements enter into the reservoir. The 632 km^2 of water fluctuating system area was formed following the completion of the Three Gorges Dam, which promoted the formation of Alluvial Soil (AS) in this area.

Both paddy fields and riparian zone of the reservoir are alternatively controlled by aquatic and terrestrial system, which is so called as the wet–dry rotation environment (WDRE). The WDRE would change the chemical–physical properties of soil, e.g., pH, electrical conductivity (EC), microbe and the concentration of dissolved organic carbon (Lundquist et al., 1999). All these factors can influence the Hg species and transportation from soil to air (Ullrich et al., 2001; Yang et al., 2007). Therefore, it is hypothesized that the cycling of wetting and drying conditions in paddy filed and riparian zone of the reservoir would change the Hg species and Hg flux at these areas. A number of studies have been conducted on Hg species and the air/surface exchange of Hg and in different environment (Gabriel et al., 2011; Marsik et al., 2005; Zhu et al., 2011), but significantly less at WDRE, e.g., paddy filed and riparian zone of the reservoir. Thus, the purposes of this study were to investigate: (1) the variation trend of Hg flux and Hg species in soil and water at different periods of WDRE; (2) the correlation between Hg flux and Hg species in matrix; and (3) the influence of meteorological factors on Hg flux and Hg species in soil and water during WDRE.

1. Materials and methods

1.1. Soil collection

Three types of soil, i.e., Typic Purpli-Udic (TPUC), Xanthi-Udic Ferralosols (XUF), and AS, were collected from surface part (0–10 cm) of paddy soil and Three Gorge reservoir area in Chongqing. Among them, TPUC and XUF were collected from Beibei and Caoshang, respectively. AS was collected from Kaixian, located in the Three Gorge reservoir area (Fig. 1). All the soil samples were air-dried and grind to pass through the 2 mm sieve.

1.2. Experiment setup

The overlying water was prepared according to the composition of the Yangtze River since soil samples were collected from the Three Gorge reservoir areas, flooded by the Yangtze River perennially. A proportion of $NaHCO_3$, KCl, $CaCO_3$, $MgCO_3$, $MgSO_4$, and $MgCl_2$ were added to the 1 L of deionizer water orderly, and puffed with air for 48 hr in order to make the system balance (Table 1).

Fig. 1 – Sampling sites in Chongqing City.

The $Hg(NO_3)_2$ solution was prepared by dissolving $Hg(NO_3)_2$ powder (230,421, Sigma-Aldrich, Germany) into deionizer water. Ten kilogram soils were enclosed to a plastic basin (60 cm × 20 cm × 20 cm) that was placed on the platform of one building. One liter of Hg^{2+} solution (10 μg/L) and deionizer water were added to each basin. All of soils were sealed for 30 days, and was followed by the twice wet–dry rotation period. The first wet–dry rotation period lasted from Jul 24 to Sep 2. Twenty liters simulating the Yangtze River water were added to each basin, which represent the first flooding period (from Jul 24 to Aug 2). With the evaporation of overlying water, the simulation experiment was in the first air-dried period (from Aug 2 to Sep 4), followed by flooding period (from Sep 4 to Sep 19), when, 20 L simulating the Yangtze River water was added to each basin as before. After the overlying water evaporated again, the experiment was in the second air-dried period (from Sep 19 to Oct 20).

1.3. Sample measurement

1.3.1. Hg species in water

Three Hg species, i.e., dissolved gaseous mercury (DGM), reactive mercury (RM) and THg in overlying water, were determined. For the DGM, water samples were purged with

Table 1 – Prepared method of simulated river.

Main ion in the Yangtze River	Concentration (mg/L)	Reagent	Concentration (mg/L)
Ca^{2+}	28.9	$CaCO_3$	66.4
Mg^{2+}	9.6	$MgCO_3$	21.84
Na^+	5.3	$NaHCO_3$	16.8
K^+	3.3	$KHCO_3$	10.3
SO_4^-	13.4	$MgSO_4$	16.80
Cl^-	4.2	$CaCl_2$	6.55
HCO_3^-	128.9		