



## Effect of dissolved organic matter on adsorption and desorption of mercury by soils

YANG Yongkui<sup>1</sup>, LIANG Li<sup>1</sup>, WANG Dingyong<sup>1,2,\*</sup>

1. College of Resources and Environment, Southwest University, Chongqing 400716, China. E-mail: [petyang000@126.com](mailto:petyang000@126.com)

2. Chongqing Key Lab of Agricultural Resources and Environment, Chongqing 400716, China

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### Abstract

Effects of dissolved organic matter (DOM) on adsorption and desorption of Hg were investigated in two kinds of soils, Xanthi-Udic Ferrallosols (XUF) and Typic Purpli-Udic Cambosols (TPUC). The DOM was obtained from humus soil (DOM<sub>H</sub>), rice straw (DOM<sub>R</sub>), and pig manure (DOM<sub>P</sub>). The presence of DOM obviously reduced Hg maximum adsorption capacity with up to 40% decreases over the control, being an order of DOM<sub>H</sub> (250.00 mg/kg) < DOM<sub>R</sub> (303.03 mg/kg) < DOM<sub>P</sub> (322.58 mg/kg) < CK (control 416.67 mg/kg) for the XUF and DOM<sub>H</sub> (270.27 mg/kg) < DOM<sub>R</sub> (312.50 mg/kg) < DOM<sub>P</sub> (324.23 mg/kg) < CK (476.11 mg/kg) for the TPUC, respectively. The inhibition of DOM on Hg adsorption was slightly weaker in XUF than in TPUC. Meanwhile, the DOM promoted Hg desorption from the soils. The kinetic models, the Two-constant equation and Elovich equation, were applicable to describe the adsorption and desorption processes of Hg in soils. The speed of Hg adsorption was consistently slowed down by the existence of DOM.

**Key words:** mercury (Hg); dissolved organic matter (DOM); adsorption; desorption

### Introduction

Atmospheric cycling of mercury (Hg) plays an important role in global mercury pollution. Atmospheric mercury, arising from both natural sources (surface soils, water bodies, vegetation, wild fires, volcanoes, geothermal sources) and anthropogenic sources (fossil fuel combustion, chlor-alkali plants, incineration of municipal solid wastes, cement manufacturing, metal smelting, refining and manufacturing, gold mining), can reenter the ocean and terrestrial systems via deposition processes, thus resulting in serious harms to the ecosystems. Mercury emission from soils is considered to be a key contributor of global atmospheric Hg inventory (Kim *et al.*, 1995; Ericksen *et al.*, 2006; Wang *et al.*, 2006). Formation and evasion of volatile Hg in soils depend dominantly on soil properties such as dissolved organic matter (DOM), pH, cation exchange capacity (CEC), oxidation-reduction potential (Eh), and so on. Transformation of Hg from adsorbed species to soluble one in the soil system controls over the formation of volatile element Hg, directly impacting Hg emission into the atmosphere (Schroeder *et al.*, 1989; Schroeder and Munthe, 1998). Soil DOM has been demonstrated to be one of the most important soil properties impacting the association of heavy metals with soil particles (Xu and Allard, 1991; Antoniadis and Alloway, 2002). Hence, identifying the effect of DOM on

the adsorption and desorption of Hg is of critical importance for understanding Hg cycling between atmosphere and soils.

Soil DOM can interact with organic contaminants and heavy metals, thus affecting their speciation, transformation, and fate in the environment (Senesi *et al.*, 1989; Benedetti *et al.*, 1996). The Fourier transform infrared spectroscopy (FT-IR) and nuclear magnetic resonance (NMR) studies have shown that DOM derived from different sources contains a myriad of individual compounds with a variety of functional groups from low molecular-weight (MW) compounds such as free amino acids and sugars to chemically heterogeneous macromolecules such as enzymes, amino sugar complexes, polyphenols, and humic substances (Gu *et al.*, 1995; Chefetz *et al.*, 1998; Han and Thompson, 1999; Arias *et al.*, 2005). Dissolved organic matter may enhance Hg sorption by providing additional sorption sites after it is adsorbed on the soil particles, but it may also reduce Hg sorption in soils through formations of stable DOM-Hg in soil solution (Allard and Arsenie, 1991; Skjellberg *et al.*, 2000). Besides, the formation of stable Hg-organic complexes in the solution may cause a reduction to Hg<sup>0</sup> by some processes including an intramolecular electron transfer (Allard and Arsenie, 1991).

Much work has been focused on the impact of DOM on the Hg reduction and emission in water environment (Xiao *et al.*, 1995; Amyot *et al.*, 1997; Garcia *et al.*, 2005).

\* Corresponding author. E-mail: [dywang@swu.edu.cn](mailto:dywang@swu.edu.cn).

However, little information is available about how DOM influence sorption and desorption behaviors of Hg in soils. The objectives of the present study were just to elucidate the adsorption and desorption characteristics of Hg in soils as a function of different kinds of DOM.

## 1 Materials and methods

### 1.1 Soil samples

Two kinds of soils, the Xanthi-Udic Ferralosols (XUF) and Typic Purpli-Udic Cambosols (TPUC) collected from the farmlands in Chongqing, China, were used for the present study. They were manually crumbled, air dried, and passed through a 2-mm nylon sieve prior to use. The soils were characterized by pH (soil: water ratio, 1:1), CEC (the ammonium acetate method (Sumner and Miller, 1996)), and particle sizes (the pipette method (Day, 1965)). The total organic carbon (TOC) and dissolved organic carbon (DOC) in soils were analyzed by the high temperature platinum-catalyzed combustion method using the Shimadzu® TOC-5000 (Shimadzu Corp., Japan). The contents of Hg were determined by cold vapor atomic absorption spectrometry (CVAAS) (F732-V, Shanghai Huagang, China). These soil characteristics were presented in Table 1.

### 1.2 Preparation of DOM

In the natural ecosystem, the organic matter in soils was the most important source of DOM. As the organic amendments (such as rice straw and pig manure) were used in agriculture to improve the physicochemical characteristics of soils, the DOM from the extra sources cannot be ignored. The humus soil, rice straw, and pig manure were selected to extract DOM for this study. The humus soil was collected in the broad leaved evergreen forest in the Jinyun Mountain, Chongqing, China. The rice straw was obtained from the experimental farm of Southwest University and pig manure from the Beibei Breeding Co., Ltd., Chongqing, China. The humus soil of 40 g was mixed with 200 ml of 10% NaOH solution followed by 24 h shaking at 200 r/min, and then centrifuged at 12,500 r/min for 20 min. The supernatant was immediately passed through a 0.45-μm membrane filter (Stevenson, 1982). DOM<sub>R</sub> from the rice straw was obtained by extracting the straw with deionized water at 1:10 (W/V) for 1 week after the straw was rinsed using deionized water, air-dried, and then cut to approximately 1 cm pieces using a stainless steel knife (Barriuso *et al.*, 1992). For extraction of DOM<sub>P</sub> from pig manure, the manure was air dried, ground to pass a 2-mm sieve, and then rotated continuously with a 1:20

deionized water at 200 r/min for 12 h at room temperature (Baham and Sposito, 1983). The centrifugation and filtering processes for DOM<sub>R</sub> and DOM<sub>P</sub> were the same as those for humus soil (DOM<sub>H</sub>). The deionized water used in the above processes had been displaced with N<sub>2</sub> to minimize the oxidation of these organic compounds. The DOM solutions were immediately put in autoclaved plastic bottles and preserved at 4°C prior to use. Within 48 h of extraction, the concentrations of DOM were represented by DOC. To compare the effect of various types of DOM on Hg adsorption and desorption processes, the DOM solution extracted from different materials was adjusted to about 500 mg DOC/L with deionized water right before the experiments. The basic physicochemical characteristics of adjusted DOM are listed in Table 2.

**Table 2** Basic physicochemical characteristics of adjusted DOM

Category	pH	DOC (mg/L)	EC (mS/cm)	Hg (μg/ml)
DOM <sub>H</sub>	5.52	500	3.14	0.005
DOM <sub>R</sub>	5.72	500	2.90	0.003
DOM <sub>P</sub>	6.45	500	3.06	0.006

DOM<sub>H</sub>: DOM extracted from humus soil; DOM<sub>R</sub>: DOM extracted from rice straw; DOM<sub>P</sub>: DOM extracted from pig manure. EC: soil electric conductivity.

### 1.3 Adsorption and desorption of Hg

Soil samples of 1.000 g were put into 50 ml of polypropylene centrifuge tubes. Then, 10 ml of 0.01 mol/L NaNO<sub>3</sub> solution containing Hg<sup>2+</sup> 1, 5, 10, 14, 18, 22, 26, 30, 35, and 40 μg/ml (prepared using Hg(NO<sub>3</sub>)<sub>2</sub>) was added to the tubes followed by the addition of 10 ml of DOM from different sources. The pH was adjusted to 7 using 0.1 mol/L HCl and NaOH. The tubes were shaken (250 r/min) at 25°C for 4 h in a thermostatic water bath, and then centrifuged at 4,000 r/min for 5 min. The supernatant liquid was decanted into a volumetric flask and diluted to 50 ml with deionized water for Hg content analysis. The amount of adsorption was calculated based on the difference of Hg concentrations in the solution before and after adsorption according to the mass balance equation (Eq.(1)):

$$Q = \frac{(C_0 - C_t)V}{m} \quad (1)$$

where,  $Q$  (mg/kg) is the amount of Hg adsorbed,  $C_0$  (mg/L) and  $C_t$  (mg/L) are the initial and equilibrium Hg concentration at time  $t$  (min),  $V$  (L) is the volume of solution, and  $m$  (kg) was the mass of the adsorbent.

Following adsorption, the tubes with soils were immediately washed using deionized water to remove any

**Table 1** Basic physicochemical properties of soils used in the experiment

Soil sample	pH	Hg (mg/kg)	TOC (g/kg)	DOC (mg/kg)	CEC (cmol/kg)	Particle size (%)		
						> 0.02 mm	0.02–0.002 mm	< 0.002 mm
XUF	5.21	0.096	15.73	18.48	12.02	35	41	24
TPUC	6.20	0.042	19.53	23.60	13.72	57	30	13

XUF: Xanthi-Udic Ferralosols; TPUC: Typic Purpli-Udic Cambosols; TOC: total organic carbon; DOC: dissolved organic carbon; CEC: cation exchange capacity.

unadsorbed Hg. And then 20 ml of NaNO<sub>3</sub> solution was added into the tubes. The tubes were reshaken at 250 r/min (25°C) for 4 h. The extractants were then isolated by centrifugation at 4,000 r/min for 5 min. The contents of Hg in the solution were measured and desorbed Hg was calculated accordingly and expressed as a percentage of previously adsorbed Hg.

#### 1.4 Kinetics of adsorption and desorption of Hg

For adsorption test, exact 1.000 g of soil sample and 10 ml of Hg solution (26 µg/ml) were transferred into 50 ml of polypropylene centrifuge tubes followed by addition of DOM. The tubes were agitated at 250 r/min, 25°C. The supernatant liquid was collected at 10, 30, 60, 120, 240, 360, 480, 600, and 720 min for the analysis of Hg contents, respectively. The desorption process was in a manner identical to the adsorption procedure.

All the analyses were conducted in duplicate and mean values were presented. Double deionized water was used for preparing the solutions and dilutions. All chemicals were at least of analytical grade. The glassware and plastic material used were soaked in 10% (V/V) HNO<sub>3</sub> (overnight) and rinsed using double deionized water.

#### 1.5 Models of adsorption and desorption isotherms

The equilibrium of adsorbate between liquid and solid phases was commonly described by Langmuir and Freundlich isotherms. The Langmuir model supposed a monolayer sorption with a homogenous distribution of sorption sites and sorption energies without interactions between the adsorbed molecules (Langmuir, 1918) (Eq.(2)). The Freundlich model assumed heterogeneous energetic distribution of sites due to the diversity of sorption sites or the diverse nature of the metal ions adsorbed (Freundlich, 1906) (Eq.(3)).

$$S = \frac{k_1 S_m C}{1 + k_1 C} \quad (2)$$

$$S = k_2 C^{1/n} \quad (3)$$

where,  $S$  (mg/kg) and  $C$  (mg/L) are the amount of metal adsorbed and concentration in the solution at equilibrium,  $S_m$  (mg/kg) and  $k_1$  are Langmuir constants denoting the maximum adsorption capacity and free energy of adsorption,  $k_2$  and  $n$  are Freundlich constants indicating the adsorption capacity and the intensity of adsorption, respectively.

Several kinetics models have been widely used to examine the controlling mechanisms of adsorption pro-

cesses such as chemical reaction, diffusion control, and mass transfer (Senthilkumaar *et al.*, 2005; Gupta and Bhattacharyya, 2006; Namasivayam and Prathap, 2007). In the current study, the Two-constant equation (Eq.(4)), Elovich equation (Eq.(5)), First-order equation (Eq.(6)), and Parabolic diffusion equation (Eq.(7)) were used (Crank, 1975; Chien and Clayton, 1980; Sparks, 1986).

$$\ln S = a \ln t + b \quad (4)$$

$$S = a \ln t + b \quad (5)$$

$$\ln (S/S_m) = at + b \quad (6)$$

$$S/S_m = at^{1/2} + b \quad (7)$$

where,  $t$  (min) is the time,  $a$ , and  $b$  are the constants with different meanings for each model.

## 2 Results and discussion

### 2.1 Effect of DOM on Hg adsorption

According to the high values of correlation coefficient ( $r$ ) (Table 3), both Langmuir and Freundlich equations can fit well with the experimental data. Moreover, the Langmuir equation fitted slightly better than the Freundlich one as shown by the higher  $r$  values. According to the Freundlich constant ( $1/n$ ), sorption isotherm might be L-type ( $1/n < 1$ ), indicating minimum competition between Hg and water molecules for sorption sites on soils (Giles *et al.*, 1960). The  $S_m$  of Hg, increased with the increasing Freundlich constant ( $k_2$ ), especially for the TPUC. It was well in agreement with the accepted theory that  $k_2$  can indicate the adsorption capacity (Freundlich, 1906). Compared with CK (control), additions of DOM<sub>H</sub>, DOM<sub>R</sub>, and DOM<sub>P</sub> decreased Hg adsorption by 40.0%, 27.3%, and 22.6% for the XUF, and 43.2%, 34.4%, and 31.9% for TPUC, respectively. The association of DOM with Hg in the solution has been proposed as a mechanism by which DOM can reduce the sorption of Hg by soils (Allard and Arsenie, 1991). The DOM sorption by soil may also contribute to the observed decrease in Hg sorption via competition for sites (Lee *et al.*, 1990; Celis *et al.*, 1998). The  $S_m$  of Hg was in sequence of DOM<sub>H</sub> < DOM<sub>R</sub> < DOM<sub>P</sub> < CK, for both soils with the smallest  $S_m$  occurred in the DOM<sub>H</sub> treatments. Since  $S_m$  values for all treatment of XUF were smaller than of TPUC, DOM had greater impact on the Hg sorption in TPUC than in XUF. Accordingly, interaction of DOM with heavy metals depended on DOM and soil types due to different binding

**Table 3** Fitted isotherm models for adsorption of Hg in soils

Soil	Treatment	Langmuir			Freundlich		
		$S_m$ (mg/kg)	$k_1$	$r$	$k_2$	$1/n$	$r$
XUF	CK	416.67	12.00	0.999	353.11	0.46	0.977
	DOM <sub>H</sub>	250.00	1.60	0.975	269.41	0.72	0.950
	DOM <sub>R</sub>	303.03	3.30	0.983	269.30	0.71	0.985
	DOM <sub>P</sub>	322.58	1.48	0.991	168.38	0.74	0.973
TPUC	CK	476.11	3.50	0.999	376.55	0.63	0.992
	DOM <sub>H</sub>	270.27	3.50	0.976	140.92	0.82	0.945
	DOM <sub>R</sub>	312.50	8.00	0.993	296.85	0.65	0.994
	DOM <sub>P</sub>	324.23	4.83	0.996	232.72	0.52	0.959

**Table 4** Fitted isotherm model for desorption of Hg in soils

Soil	Treatment	Langmuir			Freundlich		
		$S_m$ (mg/kg)	$k_1$	$r$	$k_2$	$1/n$	$r$
XUF	CK	10.97	5.60	0.982	1,590.00	0.88	0.962
	DOM <sub>H</sub>	8.25	2.50	0.979	421.95	0.73	0.938
	DOM <sub>R</sub>	9.25	2.90	0.916	786.52	0.36	0.974
	DOM <sub>P</sub>	9.10	2.70	0.998	388.41	0.68	0.988
TPUC	CK	12.65	1.40	0.997	1,527.90	0.91	0.987
	DOM <sub>H</sub>	8.17	11.00	0.996	1,187.80	0.78	0.985
	DOM <sub>R</sub>	9.04	5.00	0.998	1,232.20	0.69	0.970
	DOM <sub>P</sub>	9.18	5.00	0.994	756.86	0.58	0.974

capacity (McBride *et al.*, 1997; Han and Thompson, 1999; Bäckström *et al.*, 2003).

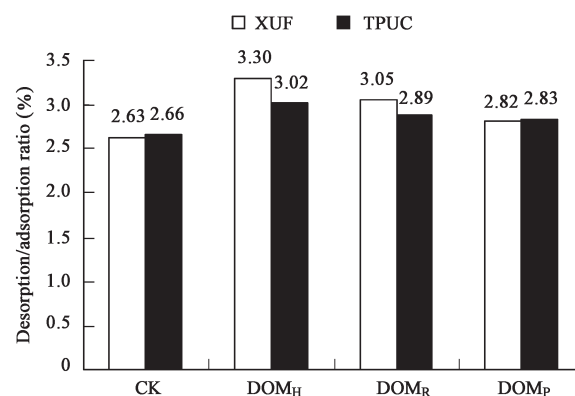
## 2.2 Effect of DOM on Hg desorption

In comparison with Hg adsorption by the soils, desorption of Hg from the soils was invariably quite small (Table 4). Low desorption has been attributed to the formation of Hg complexes with high binding energies (Arias *et al.*, 2002). The desorption processes were also fitted by Langmuir and Freundlich equations and Langmuir equation was little better than Freundlich equation, with greater  $r$  values.  $S_m$  obtained from the Langmuir equation was less in the presence of DOM<sub>H</sub>, DOM<sub>R</sub>, and DOM<sub>P</sub> than in CK treatment. However, the desorption/adsorption ratio was promoted obviously (Fig.1), which might be due to Hg retention by DOM.

## 2.3 Kinetics of adsorption and desorption of Hg

The adsorption kinetics normally includes two stages: a rapid stage followed by a much slower stage before the equilibrium is established (Chen and Wang, 2004). The kinetics of adsorption and desorption in soils could be modeled (Table 5). The Two-constant equation and Elovich equation provided much better fit for adsorption and desorption processes as shown by higher correlation coefficients ( $r$ ). Low  $r$  values for the first-order kinetic

model obtained at all concentrations suggested that Hg adsorption was not a simple first-order reaction (Bäckström *et al.*, 2003; Inbaraj and Sulochana, 2006). The high  $r$  values from the Elovich equation can suggest that the sorption system studied belonged to the second-order kinetic model. It assumed that the Hg<sup>2+</sup> were strongly held to the soil surfaces by chemisorptive bonds, involving valence forces through sharing or exchange of electrons between adsorbent and adsorbate (Cheung *et al.*, 2000; Rudzinski and Panczyk, 2002). The values of  $a$  in all the equations could represent the speed of adsorption reactions. The

**Fig. 1** Desorption/adsorption ratio of Hg in soil.**Table 5** Characteristics of kinetics equations stimulated for Hg adsorption and desorption

Soil	Treatment	Two-constant equation $\ln S = a \ln t + b$			Elovich equation $S = a \ln t + b$			First-order equation $\ln(S/S_m) = at + b$			Parabolic diffusion equation $S/S_m = at^{1/2} + b$		
		$a$	$b$	$r$	$a$	$b$	$r$	$a$	$b$	$r$	$a$	$b$	$r$
XUF	Adsorption												
	CK	0.0149	6.12	0.959	0.0717	451.81	0.960	0.0007	0.14	0.748	0.0027	1.31	0.863
	DOM <sub>H</sub>	0.0084	6.03	0.975	0.0363	416.43	0.983	0.0005	6.09	0.924	0.0025	1.70	0.971
	DOM <sub>R</sub>	0.0075	6.09	0.939	0.0339	439.39	0.929	0.0003	0.40	0.715	0.0017	1.48	0.826
TPUC	DOM <sub>P</sub>	0.0085	6.09	0.898	0.0391	441.23	0.886	0.0003	0.72	0.625	0.0026	2.04	0.758
	CK	0.0196	6.12	0.957	0.0963	451.50	0.959	0.0009	0.023	0.738	0.0031	1.00	0.854
	DOM <sub>H</sub>	0.0045	6.05	0.915	0.0193	422.55	0.916	0.0002	0.47	0.654	0.0010	1.59	0.782
	DOM <sub>R</sub>	0.0050	6.08	0.930	0.0222	438.46	0.941	0.0002	0.36	0.688	0.0011	1.42	0.819
Mean	DOM <sub>P</sub>	0.0059	6.08	0.970	0.0036	155.70	0.971	0.0003	0.26	0.787	0.0012	1.29	0.893
				0.942			0.943			0.735			0.846
XUF	Desorption												
	CK	0.0005	6.25	0.955	0.0025	519.90	0.949	0.0002	0.22	0.761	0.0001	1.25	0.880
	DOM <sub>H</sub>	0.0012	6.24	0.935	0.0061	514.66	0.945	0.0005	0.86	0.705	0.0004	2.36	0.826
	DOM <sub>R</sub>	0.0008	6.25	0.947	0.0042	520.54	0.957	0.0004	0.54	0.761	0.0002	1.71	0.873
TPUC	DOM <sub>P</sub>	0.0061	5.19	0.966	0.0112	179.37	0.959	0.0003	5.21	0.801	0.1559	1.82	0.959
	CK	0.0022	6.26	0.962	0.0012	521.73	0.929	0.0001	0.084	0.811	0.0004	1.09	0.896
	DOM <sub>H</sub>	0.0013	6.25	0.940	0.0066	517.63	0.939	0.0006	0.65	0.730	0.0004	1.92	0.845
	DOM <sub>R</sub>	0.0006	6.25	0.960	0.0032	520.12	0.959	0.0003	0.51	0.797	0.0006	1.66	0.897
Mean	DOM <sub>P</sub>	0.0018	6.25	0.942	0.0092	519.58	0.953	0.0008	0.40	0.754	0.0004	1.50	0.863
				0.951			0.949			0.765			0.880

results also showed declines in the adsorption speed by the DOM<sub>H</sub>, DOM<sub>R</sub>, and DOM<sub>P</sub>. In contrast, the desorption speeds were obviously increased by DOM<sub>H</sub>, DOM<sub>R</sub>, and DOM<sub>P</sub> for both the XUF and TPUC.

### 3 Conclusions

The maximum adsorption capacity of Hg declined in both XUF and TPUC at the presence of the DOM from different sources, in an order of DOM<sub>H</sub> < DOM<sub>R</sub> < DOM<sub>P</sub> < CK. Dissolved organic matter had greater impact on Hg adsorption in TPUC than XUF. In contrast, the existence of DOM increased Hg desorption from the soils. The desorption/adsorption ratio was enhanced by DOM as compared with CK. Langmuir and Freundlich equations could be used to describe the isotherms of Hg adsorption/desorption in soils. Also, DOM slowed down the adsorption speed of Hg by soils. Two-constant equation and Elovich equation satisfactorily fit the process of adsorption and desorption of Hg in soils.

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