

Effect of dissolved organic matter on mercury release from water body

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Abstract

Dissolved organic matter (DOM) plays an important role in the process of mercury release from water body. In this study, the influence of DOM from different sources (DOM_R, DOM_S and DOM_H, extracted from rice straw, compost and humic soil respectively) on mercury reduction was investigated. The molecular weight distribution and chemical composition of DOM from each source were determined using ultrafiltration membrane technique and elemental analysis respectively. The result showed that mercury release from DOM-added samples was much lower than the control; the lowest mercury release flux was observed in the treatment of DOM_H, 25.02% of the control, followed by DOM_S and DOM_R, 62.46% and 64.95% of the control, respectively. The higher saturation degree and lower molecular weight of DOM_H was responsible for the highest inhibition degree on the mercury release. The link between DOM_H, concentration and mercury flux was also estimated and the result showed that mercury flux was increased with DOM_H at lower concentration, while decreased with DOM_H at higher concentration. Different mechanism dominated the influence of DOM on mercury release with variation of DOM concentration.

Key words: dissolved organic matter; mercury release; inhibition; molecular weight

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Introduction

The release of mercury from water in the form of dissolved gaseous mercury (DGM) plays an important role in the global Hg cycling and this process has important implications not only because it may decrease mercury pool in water but also due to its wider geographic dispersion to other environments. It was reported that 93.7% of gaseous mercury can be deposited to the earth by wet and dry deposition (Fitzgerald, 1995) and does great harm to the ecosystem.

It is well acknowledged that dissolved organic matter (DOM) is of great importance in the biogeochemical cycling of mercury in the environment (Gill and Bruland, 1990; Mason et al., 1993; Driscoll et al., 1994; Benes and Havlik, 1979; Lindberg and Harriss, 1974). Many studies indicated that there was a positive correlation between the concentration of mercury and DOM in an aquatic environment (Mierle and Ingram, 1991; Watras et al., 1995; Babiarz et al., 1998). However, this is only a net result of many complicated interactions between mercury

and DOM. The detailed mechanisms remain unclear.

Studies in recent years have cast some light on this topic; however, the results of these studies still disagree with each other to some extent. Many studies suggested a higher rate of mercury reduction in the presence of DOM especially when photochemistry was involved (Allard and Arsenie, 1991; Xiao et al., 1995; Costa and Liss, 1999). Ravichandran et al. (2000) reported that when experiments were conducted under biotic and dark environment no mercury reduction was observed even in the presence of DOM. These studies suggested that the importance of direct reduction by DOM was probably overestimated; there may be some intermediates produced from photolysis of DOM mediated mercury reduction. It had been hypothesized quinone or semiquinone in humic acid initially or intermediates were involved in the electrons transfer in mercury reduction (Alberts et al., 1974). In field experiments, O'Driscoll et al. (2003) determined DGM concentration in lake surface water in southern Nova Scotia and found it was about 2–4 times higher in a high dissolved organic carbon (DOC) lake than that in a low DOC lake. In addition, DOM can supply microorganism

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with more bioavailable nutrients and increase the transport of mercury from watershed or sediment to water body (Loux, 1998). Note that the result of the field experiment was just a net result that could be influenced by many combined factors such as the microbe and the initial mercury concentration in aqueous environment.

In contrast to these results, Amyot et al. (1997) observed that DGM concentration was 1.7 and 8.8 times higher in the lower DOC lake than in two higher DOC lakes respectively. It was hypothesized that complex between mercury and DOM decreased available Hg^{2+} for reduction; in addition, the high DOC reduced the light penetration resulting in limited photons for mercury reduction. These results indicated that the mechanism of DOM influence on mercury release is very complicated and combined factors are involved in this process. The reason why these results appear so conflicting is that the DOM used in these experiments may differ from each other not only in concentration but also in structure and composition.

In modern agriculture, more and more straw returning and compost were used to minimize the harmful effect of chemical fertilizer to soil. The use of straw returning and compost increased the content of organic matter in farmland soil. Subsequently, the organic matter entered water body with flood and constituted the main component of DOM in aqueous system (DeGrandpre et al., 1996; Del Castillo et al., 1999; Rochelle and Fisher, 2002), which affected the transport and transformation of mercury speciation significantly. Hence, the study of effect of DOM from exotic source on mercury redox was of great meaning to estimate the transport and fate of mercury in the environment. The chemical properties of DOM also play an important role in the transport and transformation of mercury in the environment. The molecular weight distribution and chemical composition of DOM were dominating factors to the interaction between DOM and metal ions possibly (Peter, 2003).

The objective of this study was to estimate the influence of DOM from exotic source on mercury release from water body by laboratory experiments. DOM extracted from rice straw, compost and humic soil was chosen as the main exotic sources of DOM in water body. The function of their chemical properties in the mercury release process was estimated. The molecular weight distribution and chemical composition of DOM from each source was detected and the relationship between mercury reduction and these chemical characteristics was discussed. In addition, the relationship between DOM_H , concentration and mercury release was investigated.

1 Materials and method

1.1 Preparation and chemical properties of DOM

Strict clean-laboratory techniques were employed through the whole study. The glassware was soaked in 25% (V/V) nitric acid for 24 hr and was rinsed sufficiently with ultra pure water in steps. All the reagents used in this study were analytical grade.

Rice straw was collected from the Southwest University farm in the autumn of 2008. Compost was sampled after 20 days incubation of sludge. The humic soil was sampled below evergreen broad-leaved trees in Jinyun Mountain. All the samples were delivered to the lab immediately and kept at 4°C.

Rice straw was cut into pieces about 1 cm in length and washed using ultra pure water (18.2 MΩ·cm). Then the rice straw, together with compost and humic soil samples was dried at 250°C for 24 hr to remove most of mercury and autoclaved to avoid bio-reduction. After that, ultra pure water was added to the rice straw by a ratio of 1:10 (W/W) and incubated for a week at 25°C. Finally, the mixture went through 0.45 μm microporous membrane (Sartorius Stedim Biotech, Germany) and the filtrate was kept at 4°C until analysis.

To extract DOM from compost and humic soil, ultra pure water was added by the ratio 1:10 (W/W) to compost and 1:2 (W/W) to humic soil. The mixture of compost was centrifuged at 12,000 r/min for 20 min, and the mixture of humic soil was centrifuged at 4000 r/min for 10 min. Then the supernatant went through a 0.45 μm microporous membrane and the filtrate was placed in a refrigerator at 4°C until analysis.

The basic chemical properties of the filtrates are summarized in Table 1. Microprocessor pH meter (pH 211, Hanna, Italy) was employed to measure pH, total organic carbon (TOC) was detected using a multi N/C analyzer (multi N/C 2100, Analytic Jena, German), total mercury (THg) was measured using cold atomic absorption analyzer (F732-V, Shanghai Huaguang, China), and an electric conductivity meter (DDS-11A, Sichuan, China) was employed to detect electric conductivity (EC).

Elemental analysis was performed on freeze-dried samples of each kind of DOM using elemental analyzer (Elementar Company, Hanau, Germany). The elemental composition and atomic ratio are listed in Table 2.

The distribution of molecular weight was determined with ultrafiltration membrane technique. The membranes of polyacrylonitrile and polyether sulphone (Shanghai, China) were used for the molecular weight above 30 kDa, from 30 to 10 kDa and below 10 kDa. The DOC

Table 1 Chemical properties of the DOM solutions

	pH	TOC (g/L)	THg (μg/L)	EC (mS/cm)
DOM_H	5.63	0.944	0.001	3.31
DOM_R	5.82	3.243	0.005	2.80
DOM_S	6.62	0.802	0.003	3.26

DOM_H , DOM_R , and DOM_S : dissolved organic matter extracted from humic soil, rice straw and compost, respectively; TOC: total organic carbon; THg: total mercury; EC: electric conductivity.

Table 2 Elemental composition and atomic ratio of the DOM samples from different sources

	Elemental composition (%)				Atomic ratio		
	C	H	O	N	H/C	N/C	O/C
DOM_H	42.3	8.32	44.3	3.98	2.36	0.08	0.79
DOM_R	50.6	5.32	39.6	3.18	1.26	0.07	0.59
DOM_S	52.3	6.21	35.7	4.26	1.42	0.07	0.51

concentration of each filtrate in the process was determined with a multi N/C analyzer (multi N/C 2100, Analytic Jena, German). For the filtration of each step, the DOC content was calculated and their percentage in the TOC content in the unfiltered solution was used to demonstrate the molecular weight distribution of each kind of DOM (Table 3).

Table 3 Molecular weight distribution of each kind of DOM

	Percentage of each molecular weight range		
	< 10 kDa (%)	10–30 kDa (%)	> 30 kDa (%)
DOM _H	48.8	31.1	20.1
DOM _R	29.1	32.6	38.3
DOM _S	28.9	37.5	33.6

1.2 Description of the reactor device

The experiments were conducted in a quartz cylinder reactor (Fig. 1). The reactor has a diameter of 15 cm a height of 25 cm, and a chamber volume of 4.4 L. Argon was used as carrier gas and purged into the reactor through the downside Teflon tube at a constant flow rate of 1.2 L/min. A gold trap amalgam was employed to remove trace level mercury in the argon. Gaseous elemental mercury (GEM) was puffed out of the water by argon and passed through soda lime, which was used to eliminate moisture in the purge gas. Real-time GEM concentration in the purge gas was detected by an R-915⁺ multifunctional mercury analyzer (Lumex Ltd., Russia) which was connected to a computer, and the data was analyzed using SPSS software.

1.3 Influence of DOM from different sources on mercury release from water body

A standard solution of 0.2 µg Hg²⁺/mL was prepared from mercury nitrate (Shanghai Products, China) prior to each experiment. Two liter ultra pure water and 5 mL standard mercury solutions were added into the reactor vessel, yielding a concentration of 0.5 µg Hg²⁺/L. To assure the influence of DOM from different sources on mercury release was estimated at the same TOC concentration, 34 mL DOM_H, filtrate, 40mL DOM_S filtrate and 10 mL DOM_R filtrate were added into the quartz cylinder reactor containing 2 L ultra pure water, therefore, the TOC concentration of all the treatments was at 16 mg/L. No DOM was added to the control. Then argon was purged

into solution and data was collected for 20 min with 10 sec interval.

1.4 Effect of DOM concentration on mercury release from water body

To simulate the natural environment, the mercury concentration of the reaction solution was reduced to 0.1 µg Hg²⁺/L; and reaction time was prolonged to 90 min. To estimate the relationship between concentration and mercury release flux from water body, the mercury solution was treated with a concentration gradient of DOM_H, and the TOC concentrations were 0.5, 1.0, 1.5, 2.0 and 2.5 mg/L. The control experiment was conducted without DOM added.

Duplicated experiments runs for each treatment were conducted. All the experiments were carried out in an air conditioned room at a temperature of (20 ± 3)°C. To obtain similar light intensity for each treatment, experiments were conducted under natural light at noon.

1.5 Mercury release flux calculations

Since the flow rate of argon was constant at 1.2 L/min, the release flux can be calculated using the following equation:

$$F = \bar{C} \times V \times T$$

where, F (ng) is mercury release flux, \bar{C} (ng/m³) is mean concentration in argon within detecting time, V (m³/min) is flow rate of purge gas and T (min) is detecting time.

2 Results and discussion

2.1 Mercury release flux varied with DOM from different sources

In the reaction, free Hg²⁺ was reduced to Hg⁰, and Hg⁰ released from water body with carrier gases subsequently. GEM concentration in deferent gases is shown with 5 sec intervals for 20 min in Fig. 2. Mercury release rate increased gradually with time. The samples treated by all the three kinds of DOM are much lower than the control. Among them, DOM_H treated samples had the lowest emission rate while samples treated by DOM_R and DOM_S had similar emission rates. Because of the positive correlation between Hg⁰ release rate and DGM concentration in water, the increase of release rate suggested an accumulation of DGM in the reaction solution. The result indicates that when TOC concentration was 16 mg/L, all the three kinds of DOM can inhibit mercury release from water, but inhibit efficiencies were different with DOM sources.

Emission flux of the treatments and control was calculated (Fig. 3). In 20 min, the mercury emission flux of control was 80.34 ng. The emission flux is 52.18, 50.80, and 20.10 ng for samples treated by DOM_R, DOM_S and DOM_H, and they are 64.95%, 62.46% and 25.02% of control, respectively.

The order of mercury emission of samples treated with DOM from different source as: control > DOM_R > DOM_S > DOM_H. Amyot et al. (1997) observed lower Hg⁰ produc-

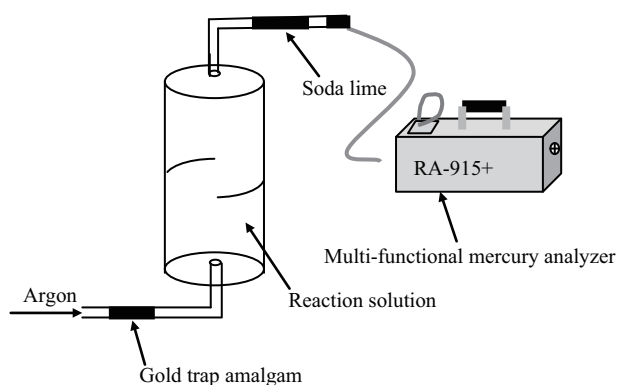


Fig. 1 Schematic of simulation reactor.

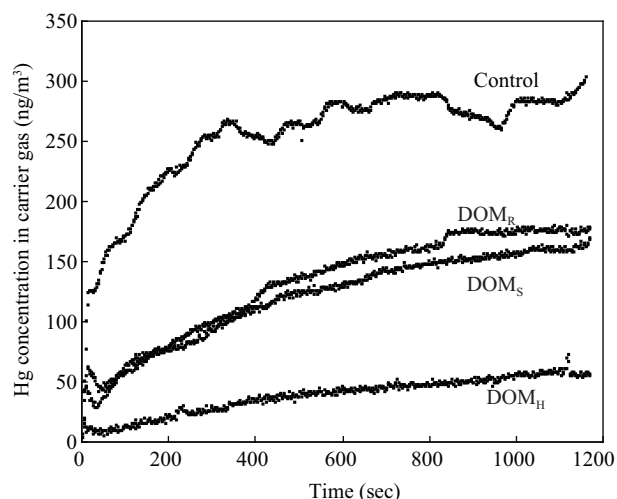


Fig. 2 Variation of mercury concentration in control and samples treated with DOM_R, DOM_S and DOM_H in 20 min.

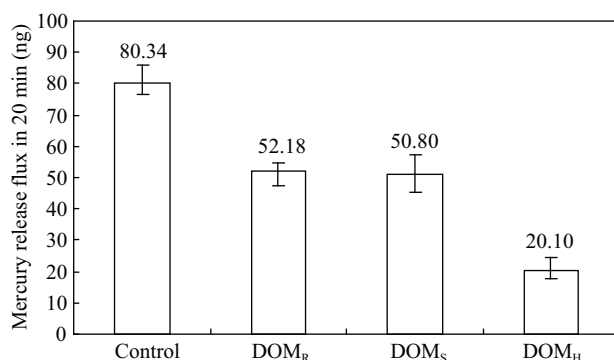


Fig. 3 Mercury release flux of control and samples treated with DOM_R, DOM_S and DOM_H in 90 min.

tions in high DOC lakes and hypothesized that high DOC reduced light penetration and increased Hg complex with DOM. Their result agreed with our observation. Note that their result was based on a field experiment and it was a net result of combined factors. Complex between mercury ion and active functional group such as hydroxyl, carboxyl and amino group in DOM is of great importance for DOM to inhibit mercury release from water body probably. As shown in Table 2, the elemental compositions of H and N are higher in DOM_H than that in DOM_S and DOM_R, indicating that DOM_H is rich in oxygen and hydrogen compared with the other two kinds of DOM. The atomic ratio of H/C, N/C and O/C are also higher in DOM_H than that in DOM_S and DOM_R, indicating that DOM_H has a higher saturation level and containing more functional groups. Although TOC in the reaction solution was at the same concentration level, 16 mg/L, type and quantity of active functional groups in different DOM molecule differed with each DOM sources, resulting in different complex efficiencies consequently.

Note that average molecular weight of DOM and stability of complex reaction product may also be responsible for the inhibition to mercury reduction. As shown in Table 3, the percentage of molecular weight below 10 kDa in DOM_H is 48.8%, which is significantly higher than that in DOM_S and DOM_R, 29.1% and 28.9% respectively. These results indicate that the mean molecular weight of

DOM_H is lower than the other two kinds of DOM. As TOC concentration of all the DOM solution was at the same level, 16 mg/L DOM_H solution would have a higher molecule concentration for its lower average molecular weight. Compared with control, DOM_R and DOM_S, the higher molecule concentration in DOM_H solution not only increased the rate of bonded Hg²⁺ but also decreased the light penetration. Considering the stability constant of Hg-DOM complex, Ravichandran (2004) represented a summary to the conditional stability constant of Hg-DOM complex. In his report, the stability constant of complex varied over large range according to different DOM sources. For example, the stability constant for complex of Hg and DOM from peat was 10^{32.2}, which were about 28 magnitudes orders higher than the DOM from humic soil. His result indicated that the complex efficient of DOM from different sources varied over a large rang.

Probably, all the above factors are of some importance in the process of mercury release from water body in the presence of DOM when TOC was at a comparatively high concentration level 16 mg/L in this study.

2.2 Influence of DOM_H concentration gradient on mercury release from water

Many publications have proved that the concentration of DOM is of great importance in the mercury release from water (Allard and Arsenie, 1991; Xiao et al., 1995; Watras et al., 1995; Costa and Liss, 1999). In this study, DOM_H, for its more significant inhibition to mercury release from water than DOM_R and DOM_S, was chosen to estimate the relationship between DOM concentration and mercury release. The total mercury release flux in 90 min for each treatment was calculated and shown in Fig. 4.

After 90 min, the mercury total flux was 9.18, 16.06, 16.26, 4.80, 1.70 and 1.19 ng for control, 0.5, 1.0, 1.5, 2.0 and 2.5 mg/L TOC treatments respectively. It showed that, compared with control, DOM_H increased mercury release when TOC concentration was lower than 1 mg/L and decreased mercury release when TOC concentration was higher than 1.5 mg/L.

When the samples were treated with DOM_H, at a concentration lower than 1 mg/L TOC, compared with control, the mercury emission flux was increased almost two times. It has been concluded that photolytic reduction of Hg was substantially enhanced in the presence of DOM (Allard

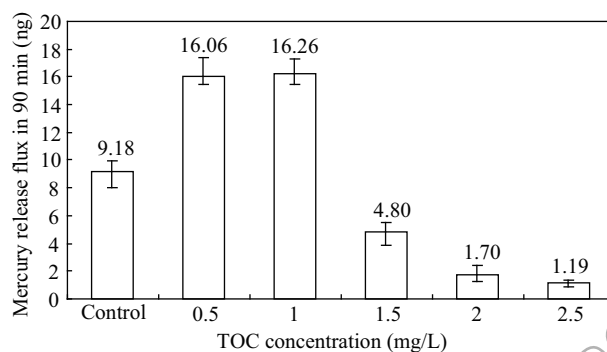


Fig. 4 Mercury release flux of control and samples treated with DOM_H at different TOC concentrations.

and Arsenie, 1991; Costa and Liss, 1999). Our results were in agreement with that to some extent. Many mechanisms were proposed to explain the enhanced mercury reduction in the presence of DOM. Published articles suggested that mercury reduction was likely coupled to the oxidation of DOM especially when photochemistry was involved (Amyot et al., 1997; Costa and Liss, 1999). Maybe some function groups in DOM molecule, such as carbonyl and aldehyde, could be an active reductant of Hg^{2+} . In addition, Alberts et al. (1974) proposed another hypothesis to the enhanced mercury reduction in the presence of DOM and they suggested that maybe quinone or semiquinone function groups in DOM molecule mediated electron transport and thus reduction of Hg^{2+} was accelerated. The result of this study tends to support the mechanism proposed by Alberts et al. (1974), since increasing TOC concentration from 0.5 to 1 mg/L did not bring significant influence to mercury emission flux.

However, when samples were treated with DOM_H , at higher TOC concentration (> 1 mg/L) an unexpected result was observed that mercury release flux decreased rapidly with increasing TOC concentration. Most probably, the complex between DOM and Hg^{2+} should be responsible for the decreased mercury flux. As it is well known that not all the mercury species can be reduced and the mercury reduction was governed by the concentration of reducible mercury species. The complexes formed by DOM and Hg^{2+} are difficult to be reduced for their remarkable stability. Hence, when the concentration of DOM was higher than a certain level, mercury reduction was dominated by complex reaction. Previously publications suggested that more than 90% of Hg^{2+} was complexed by DOM in freshwater (Hudson et al., 1994; Meili, 1997). However, the extent of complex reaction was influenced significantly by the ligands concentration. When the ligand concentration was very low, complex reaction was not remarkable and DOM functioned as an accelerator to mediate electron transfer. On the contrary, when the concentration of ligand was high, most of the Hg^{2+} was complexed and mercury reduction was decreased for the decreasing reducible Hg^{2+} concentration.

The result of this study seemed to conflict with some field experiments in which higher mercury releasing was observed in the presence of high DOM concentration (Allard and Arsenie, 1991; Costa and Liss, 1999). However, in most of the aquatic environment containing high DOM concentration, concentration of the total mercury was also high. Maybe floods containing higher DOM can deliver more mercury from land to aquatic environment. Although DOM concentration in natural water varies in a large range, it was higher than 5 mg C/L in most water systems (Gigliotti et al., 2002; Garcia et al., 2004). Hence, most probably the inhibition of DOM on mercury reduction in the environment was underestimated.

The results of these experiments indicated that complicated mechanism was involved in the influence of DOM on mercury release from water. Most probably, when DOM concentration varied, different reactions between DOM and Hg^{2+} dominated the mercury reduction.

3 Conclusions

DOM plays an important role in mercury release from water body. In this study, when TOC concentration was 16 mg/L, the order of mercury release flux was control $> \text{DOM}_\text{R} > \text{DOM}_\text{S} > \text{DOM}_\text{H}$. It seemed that the strongest inhibition of DOM_H to mercury reduction is due to its lower molecular weight and higher saturation degree. Concentration of DOM is another important influencing factor to mercury release from water. Mercury release was increased with DOM_H when TOC concentration was lower than 1 mg/L, and decreased when TOC concentration was higher than 1.5 mg/L.

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