Mei He, Lei Tian, Hans Fredrik Veiteberg Braaten, Qingru Wu, Jie Luo,
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2019. Mercury–Organic Matter Interactions in Soils and Sediments: Angel or Devil?
Bulletin of Environmental Contamination and Toxicology. 102 (5): 621-627.

Mercury-organic matter interactions in soils and sediments: angel or devil?

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Abstract

Many studies have suggested that organic matter (OM) substantially reduces the bioavailability and risks of mercury (Hg) exposure in soils and sediments; however, recent reports have also found that OM could greatly accelerates Hg methylation and increases the risks of Hg exposure. This study aims to summarize the interactions between Hg and OM in soils and sediments and improve our understanding of the effects of OM on Hg methylation. The results show that component alteration, promotion of the activity of Hg-methylating microbial communities, and the microbial availability of Hg accounted for the acceleration of Hg methylation which increases the risk of Hg exposure. These three key aspects were driven by multiple factors, including the types and content of OM, Hg speciation, desorption and dissolution kinetics and environmental conditions.

Keywords: Organic matter; methyl-mercury; Hg; bioavailability; microbial methylation

1 Introduction

Mercury (Hg) contamination in aquatic and terrestrial environments is a global concern. Soils and sediments may serve as major sinks for Hg in ecosystems present in these environments, and their importance in the biogeochemical cycling of Hg has received recent attention (Eklof et al. 2018; Rajaee et al. 2015; Shu et al. 2016a; Zhang et al. 2018a; Zhu et al. 2018). The constituents and levels of microbial activity in soils and sediments are known as crucial factors in the biogeochemical cycling of Hg (Hang et al. 2018; Ma et al. 2015; Skyllberg 2010). Organic forms of Hg, particularly methyl-mercury (MeHg), are bioaccumulated in food webs (Bloom 1992) and can potentially serve to enhance the risk to ecosystems (Yu et al. 2012; Zhu and Zhong 2015). Among the components of soil, organic matter (OM) is considered to be the most important factor for Hg biogeochemistry, Hg bioavailability, and Hg risks due to its interactions with Hg (Klapstein and O'Driscoll 2018; Liu et al. 2016; Windham-Myers et al. 2014c).

Many studies have suggested that OM substantially decreases the bioavailability and bioaccumulation of Hg and thus significantly reduces the risks associated with Hg in soils or sediments (supplemental file Table S1). The reduced risk of Hg in soils and sediments following interactions with OM stems from three different mechanisms (Ndungu et al 2016). First, OM has a high affinity to Hg and thus strongly affects the partitioning and bioavailability of Hg in soils and sediments. The abundant reduced sulfur sites on OM molecules provide strong binding sites for Hg, resulting in immobilization of Hg and reduced Hg bioavailability in highly Hg-contaminated sediments and soils (Hammerschmidt et al. 2008; Shu et al. 2016b). Second, OM enhances the photodemethylation rates of MeHg and reduces mercury bioavailability (Klapstein and O'Driscoll 2018; Tossell 1998). Even low concentrations of dissolved OM (DOM) are beneficial for the photodemethylation of MeHg (Jeremiason et al. 2015; Qian et al. 2014; Tai et al. 2014; Zhang et al. 2017), which results from the release of radicals from DOM that form an excited triplet state (3DOM*) under UV-radiation and the subsequent breakage of the carbon-Hg bond by the intramolecular charge transfer (Qian et al. 2014). The photodemethylation of Hg has been shown to accelerate in the presence of iron (Hammerschmidt and Fitzgerald 2010; Zhang et al. 2017) and thiolate and aromatic functional groups in DOM (Qian et al. 2014). Hg photodemethylation occurs more easily because the carbon-Hg bond is weakened when Hg binds with reduced thiol functional groups (Zhang et al. 2017), facilitating the absorption of specific radiation wavelengths by aromatic functional groups in DOM (Baker and Spencer 2004). Third, OM can potentially reduce MeHg production in soils and sediments. Enrichment of nitrate, iron, sulfate, cysteine and selenite in OM has been shown to effectively decrease MeHg production and accumulation in rice grown on Hg-contaminated paddy fields, which might be attributed to inhibition of the activity of related bacteria by these additives (Zhang et al. 2018b; Zhong et al. 2018).

Recently, however, several studies have suggested that additions of OM to soils and sediments significantly accelerate Hg methylation rates through increased microbial activity, subsequently increasing the risk of Hg to the environment. In this paper, a review of the available literature was conducted to summarize the interactions between OM and Hg species in soils and sediments. The objective of the present review is to discuss the effects of these interactions on Hg methylation in soils and sediments, which would improve our understanding of the mechanisms on how OM increases or decreases the environmental risks associated with Hg. This review will also provide guidance on how to manage Hg-contaminated soils and sediments through soil amendments application.

2 Anthropogenic processes where OM can enhance environmental Hg risks

Recently, several studies have demonstrated that OM greatly increases the environmental risks associated with Hg in soils and sediments (a summary of studies is provided in Table S2). The increased risks are mainly from an accelerated methylation processes, which turns available IHg to MeHg in the presence of OM in soils and sediments. An example of an important process that was recently discovered is the production of MeHg in paddy soils during rice cultivation. During this process, MeHg can be translocated to rice grains in the presence of bulk root-derived organic residues during the period from field preparation to post-harvest (Liu et al. 2014; Rothenberg et al. 2014; Windham-Myers et al. 2014a). Seasonal wetting and drying of rice-field sediments leads to a promotion of MeHg production by providing abundant water and nutrients and relatively labile plant-derived carbon (Windham-Myers et al. 2014a; Zhao et al. 2018; Zhu et al. 2015b).

Another process which has recently gained many attentions for its effect on biogeochemical Hg cycling is forestry operations. Such operations accelerate the decomposition of organic residues from logging and other OM in forest soils and creates wetland-like environments with a high abundance of bacterial communities (including sulfur-reducing bacteria (SRB), iron-reducing bacteria (IRB) and Firmicutes families) and increased MeHg production (Eklof et al. 2018). Forestry also increases Hg concentrations in runoff water due to the hydrological connection between methylation hotpots and surface waters (Kronberg et al. 2016). IHg complexed with DOM was usually more available for methylation than dissolved IHg (Mazrui et al. 2016). Relatively high production of MeHg was associated with high OM content in a hydroelectric reservoir sediment (Meng et al. 2016). Anthropogenic processes where OM can enhance environmental Hg risks were also frequently observed in marine sediments (Correia and Guimaraes 2017; Liang et al. 2016), lake and estuarine sediments (Bravo et al. 2017; Liem-Nguyen et al. 2016) and wetlands sediments (Marvin-DiPasquale et al. 2014; Windham-Myers et al. 2014b).

The formation and enhancement of MeHg in soils and sediments following methylation of IHg is a key factor for evaluating Hg risks. The Hg risk is driven by a balance between biotic IHg methylation and biotic and abiotic MeHg demethylation (Zhu et al. 2018). Hg methylation rates in the environment often vary more than demethylation rates (Kronberg et al. 2016). For example, demethylation rates did not differ between an area of clear-cutting and a reference area, although there were quite distinct methylation levels (Kronberg et al. 2016). Therefore, the conversion of IHg to MeHg is usually recognized as the most important factor in this balance, which is predominantly mediated and regulated by microbial methylators under anaerobic conditions (Marvin-DiPasquale et al. 2014; Windham-Myers et al. 2014b; Zhu et al. 2018).

3 Mechanisms whereby OM affects Hg methylation

OM has a direct or indirect link with the conversion of Hg in soil and sediment ecosystems

(Liem-Nguyen et al. 2016). Factors and conditions affecting soil and sediment OM and the subsequent effects on Hg methylation processes and Hg risks must be clarified. In this paper, three aspects that relates to the effects of OM on IHg methylation in soils and sediments are summarized and discussed, including the following: (1) The activity of microbial Hg methylators; and (2) the microbial availability of IHg.

3.1 Effects from OM on the microbial activity of Hg methylators

The Hg-methylating microbial community are restricted to specific anaerobic methylators carrying the gene hgcA and hgcB, which encode a corrinoid protein and a ferredoxin required in the corrinoid cofactor reduction (Parks et al. 2013). SRB, IRB, syntrophic and acetogenic bacteria, and methanogens are important Hg methylators in soils and sediments (Eklof et al. 2018; Kronberg et al. 2016; Marvin-DiPasquale et al. 2014; Mazrui et al. 2016; Zhu et al. 2018).

Stimulated microbial activity of Hg methylators appears to be the main control of OM on accelerating Hg methylation in soils and sediments. Substantial and variable types of OM were widely distributed in natural soils and sediments. The activity of Hg methylating microbes was significantly higher as a result of the existence of OM in soils and sediments. The OM usually acted as a source of energy, available nutrition and an electron donor, which furnished plentiful substrate, and mediated microbial activity, for Hg methylators in the biotic Hg methylation process (Eklof et al. 2018; Frohne et al. 2012; Kronberg et al. 2016; Marvin-DiPasquale et al. 2014; Windham-Myers et al. 2014a). However, the effects of OM on the microbial activity of Hg methylators depend on the characteristic and availability of OM and the environmental conditions (temperature, redox conditions, water saturation, nutrients, light, etc.). These issues are discussed in detail below.

3.1.1 The characteristic and availability of OM

Microbial Hg transformation and microbial activity of Hg methylators are controlled by the sources and characteristics of OM (Kronberg et al. 2016; Zhu et al. 2018). Labile OM including organic carbon, rice straw decay products, logging residue, natural OM (NOM), DOC, which are easily decomposed and utilized by microorganisms, play an important role in limiting and mediating the activity of Hg-methylating microbes (e.g., IRB, SRB, Firmicutes and methanogens) in different types of soils and sediments (Table S3) (Meng et al. 2016; Zhu et al. 2015a; Zhu et al. 2016). Autochthonous NOM was more prone to be utilized as an electron donor by methylating bacteria than allochthonous NOM, which might be an important factor affecting the microbial activity of Hg methylators (Liem-Nguyen et al. 2016). Organic compounds originating from fresh chlorophyll, proteins and phyto-derived cell wall lipids were important labile NOM for enhancing microbial activity in lake sediments and rice paddy soils for methylation of inorganic mercury (Bravo et al. 2017; Zhao et al. 2018). Root exudation of labile organic carbon appeared to be the primary factor that enhanced microbial activity and methylation in the presence of vegetated soils and sediments (Windham-Myers et al. 2014b; Windham-Myers et al. 2014c; Zhao et al. 2018). For example, pore-water acetate supplied abundant labile carbon as a key electron donor for improving the activity of microbial methylators in soils and sediments (Windham-Myers et al. 2014b; Zhao et al. 2018).

The availability of OM is believed to be an important driver regulating microbial MeHg formation in soils and sediments. A significant positive correlation between OM content and the microbial abundance of Hg methylators was observed in rice paddy soils due to the contribution of OM to microbial growth and reproduction (Liu et al. 2014; Zhao et al. 2018). Microbial activity and Hg methylation rates were the highest in locations with more organic content in estuarine sediment (Schartup et al. 2013). Organic matter favoured microbial methylators and subsequently enhanced their activity for Hg methylation (Zhao et al. 2018). An abundant amount of labile organic carbon was a significant variable in directly stimulating the activity of Hg methylators, which contributed to methylation differences in marine sediments and rice paddies, especially during the post-harvest period (Mazrui et al. 2016; Windham-Myers et al. 2014b; Zhao et al. 2018). The stimulating effect of high labile organic carbon concentrations on the microbial activity of Hg methylators is exemplified by rice paddy soils. In the practice of rice cultivation, a large amount of structural and exuded labile organic carbon from root tissue enters into the rice paddy soils after decay and would be readily consumed by secondary microorganisms (Rothenberg et al. 2014; Zhao et al. 2018). This root-derived organic carbon, including acetate, aliphatic hydrocarbons and simple aromatic compounds, provides plentiful energy and carbon as well as electron donors for Hg methylators that facilitate microbial activity (Rothenberg et al. 2014; Windham-Myers et al. 2014b; Windham-Myers et al. 2014c; Zhao et al. 2018). If the content of OM was lower than the threshold for utilization, the microbial activity of Hg methylators would be limited. Primary methylators would compete for electron donors (e.g., acetate and hydrogen) with each other and with other microbes in soils and sediments (Rothenberg and Feng 2012). The lack of electron donors led to a decrease in the activity of microbial methylators. These results suggested that differences in the availability of OM might hold the key to explaining the large variability in the activity of microbial methylators.

3.1.2 Effects of environmental factors on OM related methylation

The effects of OM on microbial activity pertaining to Hg methylation are dependent on selected environmental variables (water saturation, redox condition, temperature, nutrients, light, et al.) resulting from anthropogenic activities or natural biogeochemical environmental changes (Eklof et al. 2018; Kronberg et al. 2016; Zhu et al. 2016; Windham-Myers et al. 2014a). The effects of water saturation, redox state, temperature, and nutrient and light availability could be illustrated as examples of forest and rice cultivation practices.

Forest practices can enhance microbial Hg-methylation activity in the presence of logging residue by influencing water saturation, redox conditions, temperature, nutrition and light conditions in soils (Eklof et al. 2018; Eklöf et al. 2016; Kronberg et al. 2016). The increased microbial activity from logging activity is reflected by the overall bacterial diversity and relative abundance of microbial methylator families (e.g., SRB *Desulfovibrio, Desulfobacteraceae*; IRB *Geobacteraceae*; Firmicutes *Peptococcaceae, Ruminococcaceae, Veillonellaceae*) in soils (Eklof et al. 2018). The decomposition of organic residue and microbial methylating activity is closely associated with the amount of water, the saturation time, temperature and light conditions in soils and sediments (Eklof et al. 2018; Kronberg et al. 2016). Wet, low-oxygen, and high-temperature conditions are more likely to result from stump and stem logging practices, which create favourable environments for anaerobic Hg methylators (Eklof et al. 2018; Rothenberg et al. 2014). Solar radiation exposure to OM in soils quickly increased after plant-cutting (Kronberg et al. 2016). Organic residue from logging and soil OM were prone to decompose and degrade under these environments, resulting in bulk production of fresh organic carbon sources, which provides abundant and excellent substrate (as electron donors) for enhancing the microbial activity of Hg methylators (Eklöf et al. 2016; Kronberg et al. 2016).

Rice cultivation practices can affect the activity of Hg methylators in the presence of rice straw decay by altering water saturation, nutrients, and redox conditions (Zhu et al. 2016; Windham-Myers et al. 2014a). Aerenchyma tissue for enhancing gas transport between soil and plants was more likely to alter the surrounding environment in the plant rhizosphere under anaerobic conditions, which might be a good habitat for anaerobic microbial communities capable of methylating Hg (Rothenberg et al. 2014). Periodic flooding and drying produced high water saturation resulting from more frequent water movement and relatively oxic to suboxic conditions due to long-time cultivation in standing water (Windham-Myers et al. 2014a). The activity of native Hg-methylating microbes was triggered by enhancing the proliferation of microorganisms living at the oxic-anoxic boundaries (Eklof et al. 2018; Windham-Myers et al. 2014a). The decomposition of post-harvest rice straw residue was accelerated under these environments, resulting in a large pool of labile OM that provided microbial electron acceptors (sulfate and ferric iron) and electron donors (e.g., acetate) for the Hg-methylating microbial community (Liem-Nguyen et al. 2016; Marvin-DiPasquale et al. 2014; Windham-Myers et al. 2014a; Zhao et al. 2018). Moreover, the release of a large amount of labile OM also led to the alteration of the ambient redox potential in rice fields (Zhu et al. 2018).

3.2 Effects of OM on the microbial availability of IHg

The microbial availability of IHg was demonstrated to be responsible for the effects of OM on enhancing the risk of Hg from soils and sediments. However, compared with the spatial-temporal variation of the activity of Hg methylators, Hg methylation was less affected by variations in the microbial availability of IHg in some cases (Marvin-DiPasquale et al. 2014). For example, the effects of the former on Hg methylation were 100 times larger than the latter in wetland sediments, whereas the latter appeared to be the main limiting factor in permanently flooded wetlands (Marvin-DiPasquale et al. 2014). The pool of IHg available to methylating microbes was controlled by the speciation of IHg and the desorption and dissolution kinetics of IHg from much more abundant sediment and soil pools (Jonsson et al. 2012; Liem-Nguyen et al. 2016). The corresponding speciation or the desorption and dissolution of IHg were summarized and discussed to understand the effects of OM on the microbial availability of IHg.

3.2.1 The speciation of IHg

The chemical speciation of Hg in solid/absorbed phases potentially limited Hg availability for microbial uptake because of the control on aqueous concentrations of IHg (Liem-Nguyen et al. 2016). Some aqueous Hg forms, such as Hg-sulfide complexes and low-molecular-mass Hg-thiol complexes, were more bioavailable to microbial methylators (Liem-Nguyen et al. 2016).

The effects of OM on the microbial availability of IHg varied according to Hg species. Hg-S complexes affected the interactions between OM and Hg due to the high aqueous solubility and the presence of sulfur (Gerbig et al. 2011; Graham et al. 2013; Liem-Nguyen et al. 2016). Recent studies suggested that bioavailable neutral Hg-S species may be nanoparticulate β-HgS(s) or polynuclear Hg-S clusters, rather than aqueous HgS^0 monomers. DOM can strongly react with β-HgS(s) (Miller Carrie et al. 2009) and inhibit the aggregation of β-HgS(s) particles (Gerbig et al. 2011; Graham et al. 2013). HgS-DOM polynuclear clusters and Hg nanoparticles were more bioavailable for Hg methylators, and transformation was enhanced (Graham et al. 2012). Low-molecular-weight Hg-thiol complexes (LMMC) were another aqueous Hg species that could readily become bioavailable to Hg methylators in soils and sediments. It was demonstrated that NOM was important for the complexation of LMMC due to the formation of NOM and thiol ligand complexes and thus subsequently influenced the microbial availability of IHg (Liem-Nguyen et al. 2017).

3.2.2. The desorption and dissolution kinetics of IHg

The desorption and dissolution kinetics of IHg affects the role of OM on the microbial availability of IHg in soils and sediments. In this paper, the effects of the desorption and dissolution kinetics of IHg are illustrated as the interactions between IHg and DOM. Two theories were suggested and supported to explain enhanced Hg availability by DOM. One view held that IHg complexed with DOM was part of the dissolved Hg pool, which could directly facilitate the bacterial uptake of Hg and act as a shuttle molecule to transport Hg from the environment to metal transporters (Jonsson et al. 2012; Mazrui et al. 2016). Another view suggested that Hg was first bound with DOM and subsequently transported into bacterial cells with DOM as a nutrition source (Mazrui et al. 2016). To the contrary, dissolved IHg complexes were readily absorbed by the sediment matrix and unavailable (Mazrui et al. 2016). Recent studies showed that IHg complexed with DOM was more readily dissolved and more available for microbial methylation in sediments (Frohne et al. 2012; Mazrui et al. 2016; Zhao et al. 2018). Hg complexes with DOM facilitated rapid Hg bio-uptake and methylation by Hg methylators, which might be attributed to the presence of the thiol ligand in DOM (Graham et al. 2017; Kronberg et al. 2016).

DOM with low-molecular-weight organic acids (LMWOAs) and low-molecular-weight thiols (LMWTs) enhance microbial Hg methylation in soils and sediments. LMWOAs led to a lower pH and thus facilitated desorption of Hg from soil solid phases and increased Hg availability to microbial methylators (Zhao et al. 2018). At the same time, LMWOAs provided a carbon source for utilization by Hg methylators (You et al. 2016), which promoted microbial methylation. Increased numbers of carboxylic groups in LMWOAs led to increased Hg desorption from soils and sediments, which can be beneficial to the net production of MeHg (Yin et al. 2018). Two types of LMWOAs, Suwannee River humic acid and Williams Lake hydrophobic acid, increased the bioavailability of Hg

(2 to 38-fold) to sulfate-reducing bacteria under sulfidic conditions and subsequently enhanced the methylation of Hg. MeHg production by sulfate-reducing bacteria showed a linear relationship with DOM concentration (Zhao et al. 2017). LMWTs can enhance Hg bioavailability via Hg-S-DOM complexation and provide a source of energy for Hg methylators, contributing to an indirect uptake of Hg (Chiasson-Gould et al. 2014; French et al. 2014; Graham et al. 2012; Moreau et al. 2015). However, complexation of IHg with NOM provided less available Hg for methylation in an organic forest soil due to the effect of the thiol groups in NOM on IHg speciation (Kronberg et al. 2016).

4 Conclusions and implications

The interactions between Hg and OM are illustrated on Figure S1. Component alteration and the stimulated activity of the Hg-methylating microbial community, as well as the microbial availability of IHg, account for the impact of OM on Hg risks associated with soils and sediments. The characteristic and availability of OM, the speciation, desorption and dissolution kinetics of Hg, as well as environmental conditions, are important factors controlling the three key processes. Firstly, OM with Fe, thiolate, S, cysteine, selenite and aromatic functional groups and some strong Hg binding sites greatly reduced the risks associated with Hg in soils and sediments, which resulted from the decrease in the bioavailability of IHg and MeHg. However, labile OM (acetate, hydrogen, etc.) promotes the activity of microbial Hg methylators and accelerate Hg methylation, which contributed to the enhanced risks of Hg from soils and sediments. Secondly, a large amount of labile OM led to the bulk production of energy, nutrition and electron donors, which regulated the microbial activity of Hg methylators. If the content of OM was lower than a threshold for utilization, the microbial activity of Hg methylators would be limited. Third, mercury methylation was prone to be triggered in environments with a low oxygen supply, sufficient water saturation, and high temperatures and solar radiation. Lastly, OM, which is beneficial for the desorption of Hg from soils and sediments and the formation of aqueous Hg complexes, will increase the risk of Hg from soils and sediments.

This study showed that enhanced Hg related risks from soils and sediments are controlled by multiple factors, which should be fully considered in applying organic amendments to Hg-contaminated soils or sediments. Background mercury concentrations cannot be ignored in the amendments.

Acknowledgments

This study was supported by the National Natural Science Foundation of China (Grant No. 41472124), PetroChina Innovation Foundation (Grant No. 2015D-5006-0210, No. 2016D-5007-0702), Natural Science Foundation of Hubei Province (Grant No. 2016CFB178, 2016CFB601), the Yangtze Youth Fund (Grant No. 2016cqr14) and China Scholarship Council (Grant No. 201708420108, No. 201708420260).

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