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Progress in the study of mercury methylation and demethylation in aquatic environments

LI YanBin & CAI Yong*

Department of Chemistry & Biochemistry, Southeast Environmental Research Center, Florida International University, Miami, Florida 33199, USA

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Mercury (Hg) and its compounds are a class of highly toxic and pervasive pollutants. During the biogeochemical cycling of Hg, methylmercury (MeHg), a potent neurotoxin, can be produced and subsequently bioaccumulated along the food chain in aquatic ecosystems. MeHg is among the most widespread contaminants that pose severe health risks to humans and wildlife. Methylation of inorganic mercury to MeHg and demethylation of MeHg are the two most important processes in the cycling of MeHg, determining the levels of MeHg in aquatic ecosystems. This paper reviews recent progress on the study of Hg methylation and demethylation in aquatic environments, focusing on the following three areas: (1) sites and pathways of Hg methylation and demethylation, (2) bioavailability of Hg species for methylation and demethylation, and (3) application of isotope addition techniques in quantitatively estimating the net production of MeHg.

mercury cycling, methylation, demethylation, bioavailability, methyl mercury

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Mercury (Hg) and its compounds are a class of highly toxic and pervasive pollutants. Over the past several decades, Hg contaminating and poisoning incidents have been reported in many counties (e.g. Japan, America, Canada, Sweden, and Norway) due to the large amount of Hg emitted into the environment, in particular from anthropogenic sources [1]. It was estimated that about 630000 infants born every year in the United States had an unsafe level of mercury in their blood [2]. Hg contamination is a global problem as one of the major Hg species, elemental mercury (Hg^0) has an approximate atmospheric residence time of 6 month to 1 year [3] and therefore it can be transported to ecosystems that are far from the point sources through long range atmospheric transport.

There are five major species of Hg in the environment, including Hg^0, divalent inorganic mercury (Hg^{2+}), monomethylmercury (MeHg), dimethylmercury (DMeHg), and monoethylmercury (EtHg). Inorganic Hg (Hg^0 and Hg^{2+}), in particular Hg^0, are the major chemical forms of Hg input into the environment from anthropogenic or natural sources [4,5]. During the biogeochemical cycling of Hg, organic mercury species (MeHg, DMeHg, and EtHg) can be produced. Although MeHg only accounts for a small fraction of Hg in the environment, it is the species of most concern to humans due to its high toxicity, prevalent existence, and capability of being accumulated and amplified along the food chain. The bioaccumulation of MeHg in the food chain, in particular in aquatic ecosystems, has caused the exposure of both humans and wildlife to MeHg, posing severe health risks [6] and stressing the importance of understanding the key processes controlling the levels of MeHg in aquatic environments.

In most aquatic ecosystems, *in situ* production (methylation of inorganic Hg to MeHg), rather than input from runoff water or atmospheric deposition, is the major source of MeHg. In addition to methylation of inorganic Hg, the reverse process, demethylation of MeHg, simultaneously occurs in the environment. Both processes are important for

*Corresponding author (email: cai@fiu.edu)

MeHg cycling, determining the levels of MeHg in aquatic environments. This paper reviews recent progress on the study of Hg methylation and demethylation, focusing on the following three areas: (1) sites and pathways of Hg methylation and demethylation, (2) bioavailability of Hg species for methylation and demethylation, and (3) application of isotope addition techniques in quantitatively estimating the production and degradation of MeHg in aquatic ecosystems.

1 Sites of Hg methylation and demethylation in aquatic environments

1.1 Methylation

Adequate attention was not given to the methylation of inorganic Hg in aquatic environments until researchers found that the major species of Hg present in fish was MeHg, rather than inorganic mercury or phenyl mercury, which were the species of Hg in industrial waters [7]. By adding $HgCl_2$ to bottom sediments and incubating for 5–10 d, Jensen and Jernelov firstly found that Hg^{2+} could be methylated to MeHg in sediments of aquatic ecosystems [7]. Since then, a large number of studies have suggested that methylation in sediment was the primary source of MeHg in most aquatic environments [8–11]. A good positive relation was often observed between MeHg concentration and *in situ* methylation potential in sediments of aquatic systems [12–16]. In addition to sediment, Hg methylation has been also observed in water (both fresh water [17–19] and marine water [20,21]) and periphyton (floating mat or macrophyte associated) [22–29] (Figure 1) in recent years. As only a small proportion of MeHg produced in sediment can be transported to water column, methylation of Hg in compartments of water column (water and periphyton) may also contribute significantly to the MeHg pool in pelagic food webs. A recent study showed that methylation in water column can be an important source of MeHg in the Arctic, accounting for around 47% of MeHg present in Arctic waters [21]. Methylation of mercury in periphyton may facilitate the bioaccumulation of MeHg in the food chain since periphyton can be the base of food webs in aquatic ecosystems [22]. However, the relative importance of periphyton or water methylation versus sediment methylation in MeHg levels in water column has yet to be clear. This is mainly due to the lack of quantitative estimation and comparison of the net production of MeHg in water column (periphyton and/or water) and the amount of MeHg diffused from sediment to water. Such estimations are necessary for quantitative evaluation of the importance of methylation in periphyton and water to the cycling of MeHg in aquatic ecosystems.

1.2 Demethylation

MeHg demethylation is a reverse process of Hg methylation. It simultaneously occurs in the sites (sediment, periphyton, and water) that methylation takes place (Figure 1). However, methylation is supposed to overrule the demethylation process and determine the net production of MeHg in sediment

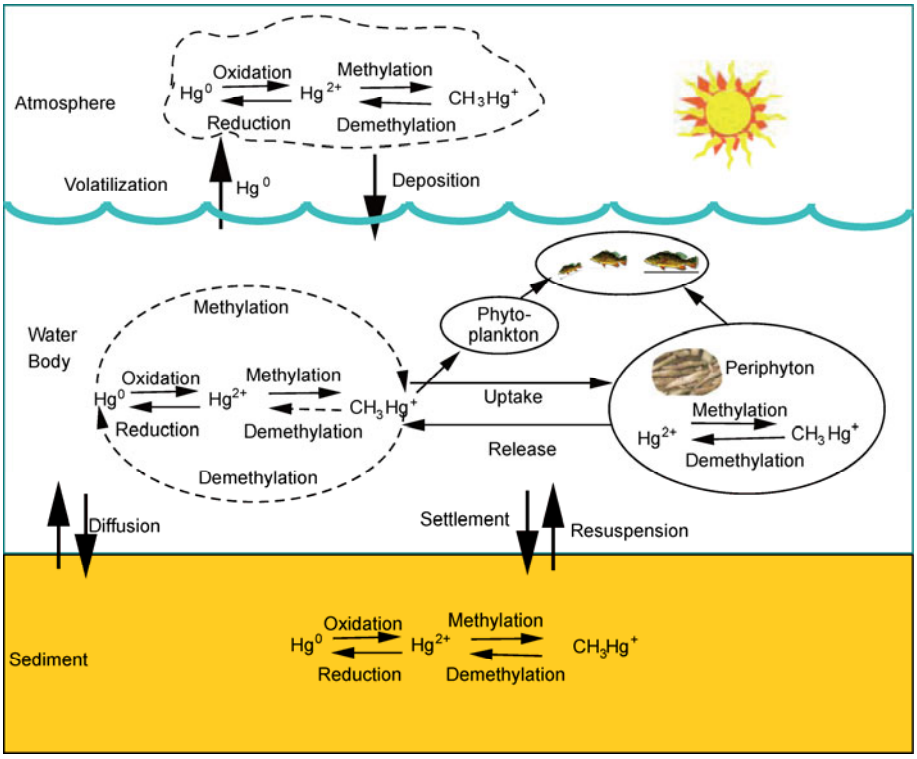

Figure 1 (Color online) Cycling of mercury in aquatic environments.

and periphyton. This opinion is supported by the phenomenon that MeHg concentrations were often positively correlated to the methylation potentials [12–16], other than the demethylation potentials in these compartments. Unlike in sediment and periphyton, demethylation in water, in particular photodemethylation was suggested to be more important than the methylation process [30–32]. The importance of demethylation in water was firstly reported in the mid-1990s by Sellers et al. [30] They found that MeHg in water could be rapidly demethylated and about 83% of MeHg flowing into the lake would be removed by photodemethylation. A number of recent studies using various means have confirmed the importance of photodemethylation in a wide range of aquatic ecosystems. Based on measuring the rates of photodemethylation in water, this process was estimated to account for about 80% and 31.4% of the MeHg mobilized annually from sediments in a lake [31] and a wetland [32], respectively. In addition, a good inverse relationship was observed between MeHg concentrations and photodemethylation potentials in water column [32]. Mass independent fractionation (MIF) of Hg isotopes in fish could also be used to quantify the loss of MeHg by photodemethylation as this process leads to significant MIF of odd-mass isotopes [33]. About 25%–68% of MeHg in lakes were estimated to be lost via photodemethylation by measuring MIF in fish [33]. In addition, a study [34] on the diurnal cycling of methylmercury in a wetland found that dissolved MeHg concentrations in water consistently decreased during daylight periods and increased during nondaylight periods. These studies on MeHg demethylation have suggested that MeHg photodemethylation in water can be a major sink of MeHg in various aquatic ecosystems.

2 Pathways of Hg methylation and demethylation in aquatic environments

2.1 Methylation

Inorganic Hg can be methylated to MeHg in water phase through biotic pathways [35–50] or abiotic pathways (photo-mediated or non photo-mediated chemical methylation) [51–66]. In natural aquatic environments, biotic process was generally suggested to be the dominant pathway of Hg methylation due to the fact that sterilization procedure could prohibit the methylation of Hg [7,67,68]. Sulfate reducing bacteria (SRB) were deemed to be responsible for the methylation of Hg^{2+} in most aquatic systems [69–73]. Methylation of Hg^{2+} by SRB in aquatic sediments was firstly confirmed by Compeau and Bartha [74]. In that study, they found that Hg^{2+} methylation in anoxic salt marsh sediment decreased more than 95% in presence of sodium molybdate, a specific inhibitor of sulfate reducers. They isolated a strain of SRB, *Desulfovibrio desulfuricans*, from the sediment and found that it could vigorously methylate Hg^{2+}. Since then, SRB has been proposed to dominate the methylation of Hg in a variety of aquatic ecosystems based on the sodium molybdate inhibition [22,26,40,69] or good relations observed between sulfate reducing rates and Hg methylation rates [70]. Other microorganisms may also dominate the methylation of Hg, although this may only occur in limited aquatic ecosystems. For instance, iron-reducing bacteria were found to have the capability of methylating Hg^{2+} in both natural sediment [75] and pure culture [76]. A recent study [27] found that methylation rates in periphyton were totally inhibited by a methanogenesis inhibitor and highly stimulated by molybdate, indicating the involvement of methanogens in Hg methylation.

Many chemicals, e.g., amino acids [77], humic substances [53,55,58], silicones [78], and low molecular weight organic acids [66], were found to be able to methylate inorganic mercury in lab settings. Irradiance is necessary for some of these reactions [66,77]. However, importance of these reactions has yet to be confirmed in natural environments. In a recent study, methylation of Hg^{2+} by dissolved organic matter (DOM) under sunlight was proposed to occur in lake waters and cause the increase in MeHg concentration during sunlight hours [19]. However, photomethylation or other chemical pathways of Hg methylation was rarely found to contribute significantly to MeHg production in other ecosystems. A recent study [79] indicated that methylation of Hg under sunlight played a minor role in a wetland as its rate was much slower than that of MeHg photodemethylation. Most previous studies only focused on the methylation of Hg^{2+} in the environment, while there is a lack of knowledge on the methylation of Hg^0. Our recent study implied that photomethylation of Hg^0 by CH_3I can occur in Florida Everglades water and a pond water (Yin et al., in submission). Contribution of Hg^0 methylation to MeHg pool also needs to be considered as Hg^{2+} can provides sufficient and continuous source of Hg^0 through photo-mediated reduction in natural water.

2.2 Demethylation

Similar to methylation of Hg, demethylation of MeHg can also proceed through biotic [8,80–82] or abiotic pathways (photodemethylation [30–32,34,83–91] or non photo-mediated demethylation [92,93]). Biotic process was suggested to be the dominant pathway of Hg demethylation in sediment and periphyton. Both SRB and methanogens could be the primary microorganisms for this process [27,80,81,94]. Although microbial demethylation of MeHg was also observed to occur in water column [18,20,95], photodemethylation was commonly deemed to be the most important in water column [30–32,89]. Non photo-mediated demethylation of MeHg can also occur in lab settings (e.g., by selenoamino acids [92,93]), however this process has not been confirmed in natural environments.

Since the initial demonstration of the importance of MeHg photodemethylation in aquatic ecosystems [30], a

number of studies have attempted to investigate the mechanisms underlying this process [90,91]. UV radiations (UV-A and UV-B) have been confirmed to be the primary driver of MeHg photodegradation in both a northern temperate lake and a subtropical wetland [32,89]. However, the chemical processes governing MeHg photodemethylation remain unclear. By reviewing the pathways of Hg photoreactions in the literature, five potential pathways could be responsible for MeHg photodemethylation (Figure 2). Although direct photodemethylation of MeHg species such as MeHgOH and MeHgCl (Pathway 1) is theoretically possible, recent studies showed that MeHg present in DI water cannot be demethylated under sunlight [90,91]. Free oxygen radical-induced degradation of MeHg (Pathway 2 or 3) was speculated to be the primary pathway of MeHg photodegradation in previous studies [87,88,90,91]. Both hydroxyl radical (·OH) and singlet oxygen ($^1O_2$) have been proposed to dominate the photodemethylation of MeHg in aquatic ecosystems. By adding scavengers of singlet oxygen, superoxide anion, hydrogen peroxide and hydroxyl radical, Suda et al. [85] found that singlet oxygen could be responsible for the degradation of MeHg and EtHg in sea water. Another study suggested that $^1O_2$ induced degradation of methylmercury-DOM complexes (Pathway 2 in Figure 2) was the major pathway of MeHg photodemethylation [91]. A recent work in Arctic lakes found that MeHg photodemethylation was driven by hydroxyl radical produced from photo-Fenton reactions [90]. Some researchers proposed that Hg^0 is the product of MeHg photodemethylation [33,88], while the others reported Hg^{2+} as the main product of this reaction [87,91]. If Hg^0 is the main product in the aquatic environment, photodemethylation of MeHg could also be treated as a reduction process of Hg. Direct transfer of electrons from photosensitized DOM to Hg within Hg-DOM complexes have been proposed to be one of the possible pathways for Hg^{2+} photoreduction [96]. Thus, direct degradation of DOM-MeHg complexes (Pathway 4) may also be a possible pathway of MeHg photodemethylation in some aquatic ecosystems. A recent study using DOM isolated from natural waters [97] found that MeHg photodemethylation rate was not significantly decreased after adding scavengers of ·OH and $^1O_2$, implying that Pathway 4 may be the dominant pathway in this ecosystem. These results indicate that the pathways of MeHg photodemethylation may vary in different aquatic ecosystems, as evidenced by inconsistent results reported in previous studies [97]. The variation of MeHg photodemethylation pathway in different aquatic systems may be caused by their differences in chemical characteristics, e.g., DOM. A recent study showed that MIF of MeHg by photodemethylation was significantly affected by the amount of reduced organic sulfur [98], implying that concentrations and characterization of DOM may play an important role in determining the dominant pathway of MeHg photodemethylation in water.

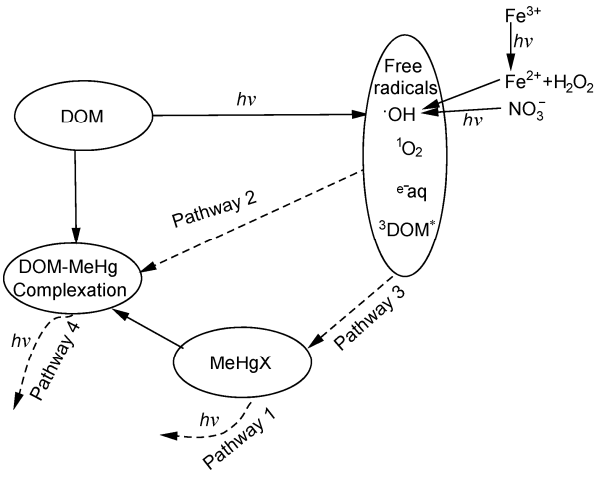

Figure 2 Possible pathways of MeHg photodemethylation in aquatic environments. By reviewing the pathways of Hg photoreactions in the literature, we proposed that five potential pathways could be responsible for MeHg photodemethylation: (1) direct photodemethylation of MeHg, (2) photodemethylation of MeHg-DOM complexes by free radicals, (3) photodemethylation of MeHg by free radicals, (4) direct photodemethylation of MeHg-DOM complexes.

3 Bioavailability of Hg species for Hg methylation and demethylation

Bioavailability of Hg species is among the most important factors that determine the production of MeHg in aquatic ecosystems. Previous studies showed that measured Hg methylation rates were usually positively correlated not to the total Hg^{2+} concentrations, but to the calculated concentrations of Hg^{2+} available for methylation [99–101]. It is also known that the newly deposited Hg^{2+} are more bioavailable for methylation compared to the legacy Hg^{2+} in sediment [102]. The bioavailability of Hg species in aquatic ecosystems is mainly determined by two processes: (1) distribution of Hg between solid and aqueous phase, and (2) speciation of Hg species in water phase (Figure 3).

Distribution of Hg species between solid and aqueous phase is expected to significantly affect the bioavailability of Hg species as only dissolved Hg species can transport through cell membranes and subsequently be methylated or demethylated. The importance of adsorption/desorption on Hg bioavailability has been confirmed by both laboratory and field studies. When *Pseudomonas fluorescens* was tested for its ability to methylate dissolved Hg^{2+} and Hg^{2+} absorbed on mineral colloids, the results showed that methylation rate of dissolve Hg^{2+} was much larger than that of Hg^{2+} absorbed on solids [103]. In addition, Hg methylation rates in surface sediments were observed to be inversely related to the distribution coefficients (K(D)) of Hg^{2+} and positively correlated to concentrations of Hg^{2+} in pore waters in some aquatic ecosystems [99,100]. The adsorption/desorption of Hg species is known to be affected by a variety of factors in aquatic environments, e.g., pH, redox potential, salinity,

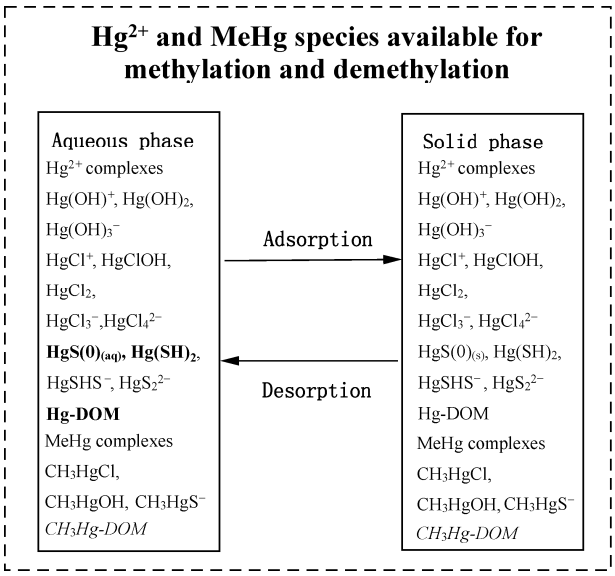

Figure 3 Hg^{2+} and MeHg complexes present in aquatic environments. Bold: species that have been reported to be the major species available for biotic methylation and demethylation. Italic: species that have been reported to be the major Hg species available for photodemethylation.

organic and inorganic complexing reagents, composition of solid. These factors are also expected to play an important role in controlling the bioavailability of Hg^{2+} in aquatic environments. Unlike the biotic methylation and demethylation, photodemethylation rates of MeHg were observed to be similar in filtrated and un-filtrated waters [89], implying that adsorption/desorption of MeHg in water column plays a minor role in the photodemethylation of MeHg.

Hg^{2+} and MeHg in aquatic environments are generally not free ions, but complexed to various inorganic or organic ligands, including hydroxide, chloride, sulfides, and DOM [104]. The dominant species of Hg^{2+} and MeHg in water or porewater depends upon various physical and chemical parameters, e.g., pH, Eh, sulfide, Cl^-, and DOM [104–108]. As not all of these complexes are available for methylation and demethylation, speciation of Hg species in water and porewater is another important factor determining bioavailability of Hg species. Neutral Hg-sulfide complexes (e.g. $HgS(0)_{(aq)}$ and $Hg(SH)_2$) have been suggested to be the major species of Hg that are available for methylation since the late 1990s [101,105,109–112]. It was proposed that these neutral Hg species could be transported to microorganism cells by passive diffusion and then methylated [109–110]. This opinion was supported by the positive relationship between Hg methylation rates and calculated concentrations of neutral Hg-sulfide complex in solution, which was observed both in laboratory pure culture of SRB [101,111] and field investigations [112,113]. Hg^{2+}-thiol complexes are another Hg^{2+} species that have been proposed to be available for methylation [114,115]. In a recent study, methylation of Hg^{2+} by *Geobacter sulfurreducens* was found to be greatly enhanced in the presence of low concentrations of cysteine. The authors proposed that the formation of Hg^{2+}-cysteine complexes, which could be transported to cells by active transport [115], facilitated both the uptake and methylation of Hg^{2+} [114]. However, the importance of Hg-thiol complexes in the methylation of Hg has yet to be confirmed in natural environments. As mercury in aquatic ecosystems is expected to preferentially bind with thiol and other sulfur-containing groups in organic matter [116], it is necessary to investigate whether this process plays an important role in Hg methylation in natural environments.

Similar to the biotic methylation and demethylation of Hg, photodemethylation of MeHg can also be affected by the speciation of MeHg in water column. A previous study reported that sunlight-induced MeHg photodegradation could not occur when MeHgCl, MeHgOH or MeHg ion was the dominant species of MeHg in water, while phenyl and sulfur bonded MeHg species could be decomposed [117]. A recent study suggested that methylmercury species bound to sulphur-containing ligands such as glutathione and mercaptoacetate had a much higher demethylation rates than methylmercury-chloride complexes [91]. Despite these findings in laboratory studies, it is still unclear which species of MeHg dominate the photodemethylation process in natural waters and more attention should be paid to this area.

4 Application of isotope addition technique in the study of Hg methylation and demethylation and estimating the net production of MeHg in aquatic environments

Since the finding of the importance of methylation and demethylation in aquatic ecosystems, efforts have been made to precisely measure the Hg methylation and demethylation rates, which are important for quantitatively estimating the production of MeHg. Isotope addition technique has been applied in this field since the late 1970s [17,118] due to its high accuracy and precision, short incubation time, and ability of simultaneously determining the methylation and demethylation rates. Both radio [17,118–120] and stable isotope addition methods [15,20,32,89,102,121–128] have been applied in this field. When radio isotope addition method was adopted, a radio isotope of Hg^{2+} (e.g., $^{203}Hg^{2+}$) is spiked into samples to monitor the methylation process. C-14 labeled MeHg is usually used to examine the demethylation process. Stable isotope addition methods have been widely using in studying Hg methylation and demethylation since the mid-1990s [122]. By using this technique, stable isotope labeled Hg^{2+} (e.g., $^{199}Hg^{2+}$) and/or MeHg (e.g., $Me^{201}Hg$) were added into samples to monitor methylation and/or demethylation process, respectively. Although both techniques promise a precise measurement of Hg methylation and demethylation rates, application of radio isotope addition techniques are limited due to its disadvantage of utilizing highly radioactive material and usually requiring

specific safety measures [129].

Net MeHg production rate is a crucial parameter for estimating the net production of MeHg in various compartments of aquatic ecosystems, which is necessary for identifying the major source and sink of MeHg in aquatic ecosystems. This parameter can be calculated by incubating the samples without the addition of any Hg species and measuring the changes in ambient MeHg. However, changes in ambient MeHg are usually difficult to be accurately measured as variation in ambient MeHg is often too small to be detected [119]. By utilizing isotope addition techniques, a method based on measuring the specific methylation and demethylation rates has been developed and used to estimate the net MeHg production rate [14,27,130]. In these studies, methylation (k_m) and demethylation (k_d) rate constants were measured and calculated (eqs. (1) and (2)) by double isotope addition methods. Then, the net production rates of MeHg were calculated by the difference of potential methylation rate ($k_m \times C_{Hg^{2+}}$(ambient) and potential demethylation rate ($k_d \times C_{MeHg}$(ambient)) (eq. (3)). However, such calculation does not take consideration of the differences in bioavailability of ambient and newly spiked Hg species [102]. A significant error could occur with this omission. For example, a recent study showed that the net production rate of MeHg in Everglades sediment would be overestimated by a factor of 20 without the consideration of the difference of newly spiked and ambient Hg species in methylation and demethylation efficiency [79]. Thus, it is necessary to take consideration of this difference (α and β in eq. (4)) when calculating the net production of MeHg using isotope addition techniques. In previous studies [102], α and β were obtained by measuring the methylation and demethylation rates of ambient and newly spiked Hg species, which is also based on the accurate measurement of changes in ambient MeHg concentrations. Due to the difficulty of accurate measuring the variation in ambient MeHg, it is necessary to develop a method that can accurately measure the differences of ambient and newly spiked mercury species in methylation and demethylation efficiency.

$$k_m = \frac{\Delta C_{Me^mHg}}{C_{{}^mHg^{2+}} \times t}, \quad (1)$$

$$k_d = \frac{\ln \dfrac{C_{Me^nHg}}{C_{Me^nHg} - \Delta C_{Me^nHg}}}{t}, \quad (2)$$

$$\text{Net MeHg production rate (ng g}^{-1}\text{ d}^{-1}) = k_m \times C_{Hg^{2+}}(\text{ambient}) - k_d \times C_{MeHg}(\text{ambient}), \quad (3)$$

$$\text{Net MeHg production rate (ng g}^{-1}\text{ d}^{-1}) = k_m \times \alpha \times C_{Hg^{2+}}(\text{ambient}) - k_d \times \beta \times C_{MeHg}(\text{ambient}), \quad (4)$$

where k_m is the specific methylation rate constant of spiked $^mHg^{2+}$ (d^{-1}); k_d is the specific demethylation rate constant of spiked Me^nHg (d^{-1}); t is the incubation time (d); $C_{{}^mHg^{2+}}$ and C_{Me^nHg} are the spiked concentrations of $^mHg^{2+}$ and Me^nHg (ng g^{-1}), respectively; α is the ratio of methylation rate constant of ambient to newly spiked Hg^{2+}; β is the ratio of demethylation rate constant of ambient to newly spiked MeHg.

5 Concluding remarks

Methylation and demethylation of mercury are a significant part of MeHg cycling, determining the levels of MeHg in aquatic ecosystems. They can occur in various compartments of aquatic ecosystems, including sediment, periphyton and water. Biotic methylation in sediment and photodemethylation in water are suggested to be the major source and sink of MeHg in aquatic environments. Recent studies found that biotic methylation in periphyton and water could also contribute significantly to the MeHg pool, in particular for the pelagic food webs. The relative importance of methylation in periphyton and/or water versus sediment in MeHg levels in pelagic food webs has yet to be clear. This is mainly due to the lack of quantitative estimation and comparison of the net production of MeHg in water column (periphyton and/or water) and the amount of MeHg diffused from sediment to water. Such estimations are necessary for evaluating the importance of methylation in periphyton and water to the cycling of MeHg in aquatic ecosystems.

Much work is also required in the study of bioavailability of Hg species for methylation and demethylation. Distribution of Hg between solid and water and speciation of Hg species in aqueous phase are the two factors that determine the bioavailability of Hg species. Both neutral Hg^{2+}-sulfide complexes and Hg^{2+}-thiol complexes have been proposed as the species of Hg^{2+} that can be transported to bacteria cells, and subsequently being methylated. More work is required to identify which is the dominant species of Hg^{2+} that associate with the biotic methylation of Hg in natural environments. In comparison to Hg^{2+}, bioavailability of MeHg for both biotic demethylation and photodemethylation has been poorly studied and requires more attentions.

More efforts should also be made on estimating the net MeHg production rate, which is important for identifying the major source of MeHg in aquatic ecosystems. Isotope addition technique is a useful technique in this area due to its high accuracy and precision, short incubation time, and ability of simultaneously determining the methylation and demethylation rates. The difference of newly spiked and ambient mercury species in methylation and demethylation efficiency should be taken into account when estimating this rate using isotope addition techniques. There is an urgent need to develop a method that can accurately measure the differences between ambient and newly spiked mercury

species in methylation and demethylation efficiency.

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