

Recent advances in the study of mercury methylation in aquatic systems

Avnee R. Paranjape^a and Britt D. Hall^{a*}

^aDepartment of Biology, University of Regina, 3737 Wascana Parkway, Regina, SK S4S 0A2, Canada

*britt.hall@uregina.ca

Abstract

With increasing input of neurotoxic mercury to environments as a result of anthropogenic activity, it has become imperative to examine how mercury may enter biotic systems through its methylation to bioavailable forms in aquatic environments. Recent development of stable isotope-based methods in methylation studies has enabled a better understanding of the factors controlling methylation in aquatic systems. In addition, the identification and tracking of the *hgcAB* gene cluster, which is necessary for methylation, has broadened the range of known methylators and methylation-conducive environments. Study of abiotic factors in methylation with new molecular methods (the use of stable isotopes and genomic methods) has helped elucidate the confounding influences of many environmental factors, as these methods enable the examination of their direct effects instead of merely correlative observations. Such developments will be helpful in the finer characterization of mercury biogeochemical cycles, which will enable better predictions of the potential effects of climate change on mercury methylation in aquatic systems and, by extension, the threat this may pose to biota.

Key words: mercury, methylation, methylmercury, biogeochemistry, aquatic systems, *hgcAB* gene cluster, stable isotopes, environmental controls, climate change



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Introduction

Mercury (Hg) is a highly toxic heavy metal present at naturally low concentrations in most environmental systems (Ullrich et al. 2001). Decades of industrial activity such as manufacturing, wastewater treatment plants, and coal burning have resulted in emissions of Hg to concentrations above background levels (Driscoll et al. 2007; Weiss-Penzias et al. 2016; Zhang et al. 2016). Because of its accumulation through aquatic food chains, Hg can be dangerous to organisms even at relatively low concentrations (Boening 2000; Driscoll et al. 2007). Compared with inorganic forms, organic forms of Hg are particularly toxic (Boening 2000; Wiener et al. 2003; Hightower 2009). The methylated form of mercury, monomethylmercury (often simplified to methylmercury; MeHg), is a particularly problematic neurotoxin (Ullrich et al. 2001; Driscoll et al. 2007). This highly bioavailable form of Hg is capable of bioaccumulating to toxic levels at higher trophic levels (Driscoll et al. 2007). In humans, the results of mercury poisoning can be disastrous, including the inhibition of endocrine function (Tan et al. 2009), cardiac effects such as hypertension (Grandjean et al. 2004) and atherosclerosis (Tan et al. 2009), and in severe cases such as in Minamata and Niigata (Japan; Normile 2013), and the Wabigoon-English River system (Ontario, Canada; Takaoka et al. 2014), mercury poisoning can result in fetal death, neurological disorders resembling cerebral palsy, deafness, and visual impairment (Tsubaki and Irukayama 1977).

In the decades since the mercury poisoning of these fishery-reliant communities, research into both understanding the environmental mercury cycle and the development of technologies designed to reduce the release of mercury to ecosystems have culminated in the recent signing of the global treaty, the Minamata Convention on Mercury (Eriksen and Perrez 2014). The genesis of this convention was the 2003 United Nations Environment Programme General Council (UNEP GC), where the Global Mercury Assessment study was presented showing significant global risks to both human health and the environment, prompting calls for action. However, several countries, including the USA, Australia, and Canada, initially opposed a legally binding agreement on reducing anthropogenic mercury sources (Eriksen and Perrez 2014). The agreement gained greater support by 2009 following the election of the Obama administration, which ended the US's opposition to a legally binding instrument, thereby encouraging the UNEP GC to begin negotiations for the convention. The Minamata Convention was formally signed and adopted in October 2013 (Eriksen and Perrez 2014), and contains various provisions targeting key anthropogenic Hg sources such as manufacturing, emissions from power plants, the use of Hg in products, and the management of Hg-contaminated waste (Evers et al. 2016). If properly implemented, the Minamata Convention is expected to continue to reduce anthropogenic emissions of Hg, which have declined by 30% between 1990 and 2010, with significant emission reduction in North America and Europe in particular (Zhang et al. 2016).

However, despite the successful reduction of Hg in emissions since 1990 (Zhang et al. 2016), there are a number of important issues that make Hg a continuing concern. The potential of increased emissions in Asia as well as issues surrounding legacy deposition continue to provide impetus for more fully understanding Hg cycling in environments. In addition, changes in the complex Hg cycling will result from increasing rates of anthropogenic climate change. Thus, the crucial importance of understanding Hg methylation once it enters an aquatic system and the conditions that favour the production of MeHg, allowing its subsequent uptake by biota, continue to be important research subjects.

The objective of this review is to examine key advances in the study of factors that impact and control methylation (particularly bacterial methylation) over the last 10 years. Although there have been several recent studies that focused on contaminated sites (Duran et al. 2008; Gray and Hines 2009; Huguet et al. 2010; Avramescu et al. 2011; Hines et al. 2012; Marvin-DiPasquale et al. 2014; Windham-Myers et al. 2014a, 2014b, 2014c; Eckley et al. 2015; Figueiredo et al. 2016; Liu et al. 2016), these are beyond the scope of this paper; this review focuses exclusively on non-polluted aquatic systems. Recent reviews regarding both Hg methylation and demethylation focused on assessing the bioavailability of particular Hg species and the use of stable isotope techniques in methylation studies (Li and Cai 2013), the molecular mechanisms of mercury uptake and methylation (Hsu-Kim et al. 2013b), and Hg cycling in Arctic (Douglas et al. 2012; Braune et al. 2015) and coastal marine and estuarine (Merritt and Amirbahman 2009) systems. In contrast, our review focuses on advances in the study of biotic methylation, the diverse aquatic environments in which methylation has been identified, and the advancement of understanding abiotic conditions favourable to microbial methylation. In addition, we provide a brief analysis of the impacts of a changing climate on Hg methylation.

Why is net methylmercury production important and how it is determined?

Bioaccumulation of MeHg depends on both the methylation of inorganic mercury and its transfer from sites of methylation to organisms (Boening 2000). Natural demethylation processes occur simultaneously through either the metabolic activity of many microbial species (Oremland et al. 1995; Marvin-DiPasquale et al. 2000) or abiotic photodegradation (Sellers et al. 1996; Hammerschmidt and Fitzgerald 2006a; Lehnher and St. Louis 2009; Bittrich et al. 2011), and it is the net result of these

opposing reactions that regulates concentrations of organic Hg available for uptake (Boening 2000; Ullrich et al. 2001). Abiotic methylation may occur if sufficient methyl donors are present; conditions that may be afforded by an environment rich in organic material (Nagase et al. 1982). However, abiotic methylation is generally thought to be of little importance in most aquatic systems (Ullrich et al. 2001). Another possible avenue of abiotic methylation is photomethylation under UV radiation—a process that has been identified in vitro (Akagi et al. 1974; Hayashi et al. 1977), but the exact mechanism of which remains unclear (Chen et al. 2015). The noted simultaneous production and degradation of MeHg may result in MeHg in sediments being in steady-state (Pak and Bartha 1998); however, levels in biota may be much higher due to bioaccumulation (Ullrich et al. 2001).

The majority of MeHg production occurring in aquatic systems is via biotic mechanisms, namely, bacterial methylation. There are several parameters that may be used as a proxy to measure methylation. The presence of methylation can be inferred from the activity of methylating bacteria, measurements of elevated concentrations of MeHg, changes in Hg mass balance budgets, and the proportion of the total Hg that is in the methylated form (%MeHg). In the past, experimental and descriptive methods were used to assess correlative relationships among MeHg concentrations and environmental controls. However, within the last decade, stable isotopes have been used to measure potential methylation rates (K_{meth}) (Hintelmann et al. 2000) to address questions surrounding rates of, and factors controlling, methylation in aquatic systems (Table 1). Throughout this review, advances in our understanding of MeHg production using this new technique illustrate its value. For example, Jonsson et al. (2012, 2014) used potential methylate measurement in estuarine systems to determine that the input of allochthonous MeHg from terrestrial and atmospheric sources is a substantially greater source than existing sediment Hg pools to aquatic organisms. Another aspect of this newer work is to examine rates of MeHg production in novel environments. Measurements of K_{meth} on the continuum of the saline gradient of Great Salt Lake, Utah, USA has shown that methylation rates are similar to other systems; but more importantly, that seasonal decreases in potential methylation rates in the deep brine layer of the lake corresponds to reduced Hg burdens of aquatic invertebrates, which have been implemented as possible vectors of Hg to avian species using the lake (Johnson et al. 2015). Potential methylation rates in prairie wetlands are similar to other systems, despite elevated concentrations of MeHg in water (Hoggarth et al. 2015), suggesting that the rates of other processes such as demethylation play a proportionally lesser role in these systems compared with others. This stable isotope technique has also been used to test potential mitigation strategies. Bussan et al. (2016) showed that potential methylation rates decreased in sediments amended with activated carbon and biochar, suggesting that adding these materials to contaminated sediments may decrease Hg transfer to aquatic organisms. A similar method of using stable Hg isotopic tracers to determine potential microbial demethylation rates (K_{demeth}) has been used much less frequently due to large errors in differentiating the loss of the tracer isotope from low concentrations of ambient mercury in the sediment. As a result, demethylation rates measured using these techniques are most useful in studies of systems in which the details of mercury cycling are well-characterized.

These short-term measurements of potential rates of methylation and demethylation have been found to be unrelated to gross measures of long-term MeHg accumulation (Drott et al. 2008; Johnson et al. 2016). This suggests that these snapshot measurements may not be valuable in determining long-term process measurements. However, significant positive correlations have been found between K_{meth} and %MeHg in many environments (Drott et al. 2008), and %MeHg values in boreal wetlands tracked the ratio of K_{meth} to K_{demeth} (Tjerngren et al. 2012a), suggesting that if both rates are measured, steady state net MeHg production rates can be estimated. Very recently, measurements of potential rates have been replaced with measurements of the genetic potential of the microbial community (e.g., Gionfriddo et al. 2016) due to the recent discovery of a gene cluster responsible for mercury methylation (see below), and developments in this area will be useful in addressing many of the questions previously addressed using the measurements of potential rates.

Table 1. Methylation ($K_{\text{meth}} \cdot \text{d}^{-1}$) and demethylation ($K_{\text{demeth}} \cdot \text{h}^{-1}$) potentials, methylmercury (MeHg $\text{ng} \cdot \text{g}^{-1}$ dry weight), and percent of total mercury as methylmercury (%MeHg) for various types of freshwater, estuarine, and marine systems.

| Location | Type of system | K_{meth} (d^{-1}) | K_{demeth} (h^{-1}) | Sediment ambient MeHg ($\text{ng} \cdot \text{g}^{-1}$) | Sediment %MeHg | Reference |
|--|---------------------------------------|---------------------------------------|---|---|----------------|--|
| Freshwater | | | | | | |
| Florida Everglades | Wetlands | 0–0.12 | na | ~0.1–4.5 | 0.1–1.7 | Gilmour et al. (1998) |
| Ontario, Canada | Lake sediments | 0.001–0.016 | 0.016–0.022 | 0.43–6.9 | 0.14–4.6 | Hintelmann et al. (2000) |
| Yolo Bypass California | Wetlands | 0.0003–1.3 | na | 0.53–6.17 | 0.31–3.88 | M. Marvin-DiPasquale (personal communication, 2012); Windham-Myers et al. (2009) |
| Boreal Sweden | Wetlands of different nutrient status | 0.0014–0.01 | Negligible–0.007 | 3.5–21 | 2.3–17 | Tjerngren et al. (2012a, 2012b) |
| Ängessjön, Sweden | Lake sediments | 0.002–0.066 | 0.0016–0.028 | na | 0.4–0.9 | Tjerngren et al. (2012a) |
| High Arctic | Wetlands | 0.04–0.16 | 0.021–0.354 | 0.6–18.5 | 1.2–12 | Lehnherr et al. (2012) |
| Northern Great Plains, Saskatchewan | Prairie wetlands | 0.02–0.17 | na | 0.18–2.1 | 0.6–4.8 | Hoggarth et al. (2015) |
| Northeastern Minnesota | Non-sulfate impacted wetlands | <0.01–0.32 | <0.005–0.07 | ~6–15 | ~6–15 | Johnson et al. (2016) |
| Northeastern Minnesota | Sulfate impacted wetlands | <0.01–0.33 | <0.005–0.10 | ~5–13 | ~7–12 | Johnson et al. (2016) |
| Northeastern Minnesota | Non-sulfate impacted lake sediments | 0.01–0.31 | <0.005–0.11 | na | na | N. Johnson (personal communication, 2016); Bailey et al. (in review) |
| Northeastern Minnesota | Sulfate impacted lake sediments | <0.01–0.09 | <0.005–0.15 | na | na | N. Johnson (personal communication, 2016); Bailey et al. (in review) |
| Estuarine and marine | | | | | | |
| Long Island Sound | Marine sediments | 0.014–0.082 | na | 0.2–3.2 | 0.4–1.1 | Hammerschmidt and Fitzgerald (2004) |
| New England coastal shelf | Marine sediments | 0.02–0.21 | na | 0.1–3.3 | 0.4–1 | Hammerschmidt and Fitzgerald (2006b) |
| Kirkpatrick Marsh, Rhode River, Maryland | Estuarine marsh soil | 0.002–0.07 | na | ~110–150 | 0.2–4.6 | Mitchell and Gilmour (2008) |
| San Francisco Bay Delta, California | Estuarine sediments | 0.002–0.122 | na | 0.6–3.7 | 0.1–1.4 | Windham-Myers et al. (2009) |
| Öre River estuary, Sweden | Estuarine sediments | 0.8 | 0.002–0.083 | ~0.1–13 | na | Lambertsson and Nilsson (2006) |
| Mid-Atlantic continental shelf | Marine sediment | 0.002–0.053 | 0.02–0.04 | 0.008–0.096 | 0.7–1.72 | Hollweg et al. (2010) |
| Adour River, France | Estuarine sediment | 0.03 | 0.004 | 2.2 | 0.9 | Martín-Doimeadios et al. (2004) |

(continued)

Table 1. (concluded)

| Location | Type of system | K_{meth} (d^{-1}) | K_{demeth} (h^{-1}) | Sediment ambient MeHg ($\text{ng}\cdot\text{g}^{-1}$) | Sediment %MeHg | Reference |
|--|--|---------------------------------------|---|---|----------------|-------------------------------------|
| San Francisco Bay Delta, California | Estuarine sediment | <0.0003–0.0049 | 0.006–0.009 | na | na | Marvin-DiPasquale et al. (2003) |
| Bay of Fundy, Minnesota, New Brunswick | Estuarine sediment | 0–1.2 | 0.39–0.138 | 1.6 | 0.20 | Heyes et al. (2006) |
| Patuxent River, Maryland | Estuarine sediment | 0–0.276 | na | 0.098 | 0.23 | Heyes et al. (2006) |
| Hudson River, New York | Estuarine sediment | 0.004–0.43 | 0.09–1.59 | 0.22 | 0.70 | Heyes et al. (2006) |
| Long Island Sound, USA | Estuarine sediment | 0.014–0.082 | na | 0.20–3.20 | 0.41–1.05 | Hammerschmidt and Fitzgerald (2004) |
| Gulf of Mexico | Marine sediments | 0.02–0.19 | 1.6–2.6 | 0.016–0.28 | 0.04–1.1 | Liu et al. (2015) |
| Salt lakes | | | | | | |
| Great Salt Lake | Deep brine layer (DBL) water | 0.0000012–0.0011 | Negligible | na | na | Johnson et al. (2015) |
| Great Salt Lake | Sediment slurry below DBL | 0.0000012–0.0031 | Negligible | 0.43–1.60 | 31.6–242.8 | Johnson et al. (2015) |
| Great Salt Lake | Freshwater influenced bays | 0.0000014–0.0011 | na | 0.01–0.41 | 5.9–250.5 | Johnson et al. (2015) |
| Laboratory | | | | | | |
| Intertidal region surrounding University of Connecticut Avery Point campus | Laboratory generated marine snow | <MDL–0.02 | <MDL–0.0125 | na | na | Ortiz et al. (2015) |
| Öre River estuary, Sweden | Estuarine sediment amended with N and P | ~0.03–0.045 | ~0.3–0.43 | 0.28 | 0.72 | Liem-Nguyen et al. (2016) |
| Oxford, Mississippi | Freshwater sediments amended with activated carbon and biochar | 0.00031–0.00366 | 0.002–0.0038 | na | na | Bussan et al. (2016) |

Note: na, not available; MDL, method detection limit.

What methylates and how can we use this information going forward?

While methylation may occur abiotically in some environments (Celo et al. 2006; Eckley and Hintelmann 2006; Monperrus et al. 2007a; Perrot et al. 2013), biotic methylation by microbes is the primary source of MeHg in aquatic environments. Sulfate-reducing bacteria (SRB) were the first organisms identified as the primary bacteria responsible for methylation using

SRB inhibitors (Compeau and Bartha 1985; Gilmour and Henry 1991; Kerry et al. 1991; Gilmour et al. 1992). It is important to note that not all SRB can methylate; therefore, the activity of all SRB is not an ideal indicator for methylation (King et al. 1999; Heyes et al. 2006). In addition, some SRB may be more effective methylators than others. For example, SRB that use acetate as a carbon source have been found to methylate Hg at equivalent or higher rates than those that do not use acetate (King et al. 2000; Ekstrom et al. 2003). Particular strains of SRB that are capable of methylation include *Desulfovibrio* (Choi and Bartha 1993; Choi et al. 1994) and identified more recently, 10 of 14 strains in the *Desulfovibrio*, *Desulfotomaculum*, and *Desulfobulbus* genera (Kaschak et al. 2014).

While SRB are important, they are not the sole methylators identified in aquatic environments. In periphyton, sulfate reduction inhibitors were found to have only a partial effect on methylation potential (Ach et al. 2011), indicating the presence of other methylators in the microbial community. In fact, Bravo et al. (2015) found that SRB were not the primary methylators in sewage treatment plants, though specific identification of the methylators was not possible. Correia et al. (2012) asserted that methylation depends on complex interactions among microorganisms, including prokaryotes, algae, fungi, and methanogens, suggesting that syntrophy may have a role in methylation. Iron-reducing bacteria (IRB) have also been implicated in methylation (Yu et al. 2012). In specific tests of the methylating capacity of dissimilatory IRB, it was found that *Geobacter sulfurreducens* produced MeHg through an unknown mechanism, whereas *Shewanella oneidensis* did not (Si et al. 2015). While the reduction of Fe(III) can stimulate MeHg formation, higher concentrations of Fe(III) can suppress Hg methylation by complexing Hg and making it unavailable for methylation (Si et al. 2015). Methanogens (Hamelin et al. 2011), IRB (Yu et al. 2012), and SRB (Compeau and Bartha 1985) are facultative anaerobes (Hintelmann 2010); however, anaerobic bacteria are not significant sources of MeHg to the ocean in oxygen-deficient zones (Malcolm et al. 2010), suggesting that aerobic bacteria may have a greater role than previously thought. The methylating activity of IRB, methanogens, and SRB may function as part of a large community of other closely interacting microorganisms. Several methanogens (e.g., *Methanomethylovorans hollandica* and *Methanobolus tindarius*) have been found to have methylating properties (Gilmour et al. 2013), and contrary to past research that asserted methanogens had only a minor role in methylation (Ullrich et al. 2001), methanogens may in fact be the primary methylators in particular environments such as fluvial lakes (Hamelin et al. 2011).

A developing area of research that is likely to enable significant strides in the future identification of methylating biota is the tracing of the gene pair *hgcAB*, which is necessary for methylation (Gilmour et al. 2013; Parks et al. 2013). Known methylators, including SRB and IRB, possess the genes responsible for methylation (Gilmour et al. 2013; Parks et al. 2013; Sonke et al. 2013; Podar et al. 2015). This gene pair has already enabled researchers to identify the methylating capacities of certain methanogens as well as *Firmicutes* (Parks et al. 2013), particularly syntrophic, acetogenic, and fermentative varieties (Gilmour et al. 2013), several *Proteobacteria*, including five *Deltaproteobacteria* species (Gilmour et al. 2013), and *Euryarchaeota* of the archaea (Parks et al. 2013) (Table 2).

The presence of the *hgc* gene cluster in organisms from environments that are methanogenic (such as rice paddies or animal digestive systems) or with conditions at the extremes of pH and salinity levels (Gilmour et al. 2013) could significantly broaden the range of environments at a high risk for Hg methylation, beyond those that feature sulfate or iron reduction as dominant processes. Potential environments in which methylation may occur, as suggested by the environments of organisms with the *hgcAB* gene cluster, include invertebrate digestive tracts, thawing permafrost soils, coastal dead zones, and extreme environmental conditions (Podar et al. 2015). In fact, studies using molecular

Table 2. Organisms that have been identified as either methylators or having the *hgcAB* homologue.

| Kingdom | Group | Genus and species | Additional info, reference |
|---|---------------------|---|--|
| Presence of <i>hgcAB</i> homologue plus stable isotope incubations | | | |
| Archaea | Methanomicrobia | <i>Methanomethylovorans hollandica</i> <i>Methanolobus tindarius</i> | Gilmour et al. (2013) |
| Bacteria | Firmicutes | <i>Ethanoligenens harbinense</i> <i>Dethiobacter alkaliphilus</i> <i>Desulfosporosinus acidiphilus</i> <i>Desulfotobacterium metallireducens</i> <i>Desulfosporosinus youngiae</i> <i>Desulfotobacterium dehalogenans</i> <i>Acetonema longum</i> | Gilmour et al. (2013) |
| Bacteria | Deltaproteobacteria | <i>Geobacter bemidjensis</i> <i>Desulfonatronospora thiodismutans</i> <i>Desulfomicrobium baculatum</i> <i>Geobacter daltonii</i> <i>Syntrophus acidotrophicus</i> | Lu et al. (2016); Gilmour et al. (2013) |
| Presence of <i>hgcAB</i> homologue plus other tests | | | |
| Bacteria | Deltaproteobacteria | <i>Desulfovibrio aespoeensis</i> <i>Desulfovibrio desulfuricans</i> <i>Desulfovibrio africanus</i> <i>Desulfobulbus propionicus</i> | Sulfate reducing bacteria inhibitors (Compeau and Bartha 1985; Kerry et al. 1991; Gilmour et al. 1992; Choi and Bartha 1993; Choi et al. 1994) Correlations with SO ₄ concentrations (Devereux 1996; King et al. 1999; Eckley and Hintelmann 2006) Presence of <i>hgc</i> gene cluster (Gilmour et al. 2013; Parks et al. 2013; Sonke et al. 2013; Podar et al. 2015) |
| Stable isotope assays only | | | |
| Bacteria | Deltaproteobacteria | <i>Geobacter metallireducens</i> <i>Desulfuromonas palmitatis</i> <i>Geobacter hydrogenophilus</i> <i>Geobacter sulfurreducens</i> | Kerin et al. (2006) |
| Correlations to increased MeHg concentrations | | | |
| Bacteria | Deltaproteobacteria | <i>Geobacter</i> sp. | Fleming et al. (2006) and Yu et al. (2012) |
| Bacteria | Deltaproteobacteria | <i>Desulfotomaculum ruminis</i> <i>Desulfovibrio piger</i> <i>Desulfovibrio giganteus</i> <i>Desulfovibrio termitidis</i> <i>Desulfobulbus propionicus</i> | Kaschak et al. (2014) |
| Bacteria | Deltaproteobacteria | <i>Geobacter sulfurreducens</i> | Si et al. (2015) and Yu et al. (2012) |
| Radiochemical assays | | | |
| Bacteria | Cyanobacteria | <i>Microcystis aeruginosa</i> <i>Synechocystis</i> sp. | Assay using ²⁰³ Hg additions (Coelho-Souza et al. 2006) |
| Presence of <i>hgcAB</i> homologue only | | | |
| Bacteria | Deltaproteobacteria | <i>Nitrospina</i> sp. | Gionfriddo et al. (2016) |

techniques to directly examine potential sites of methylation are currently being published. For example, Gionfriddo et al. (2016) have identified Antarctic sea ice as a possible environment conducive to the growth of a marine microaerophilic bacterium (*Nitrospina*) by isolating the *hgcAB* genes

in environmental samples. Identifying and tracing the *hgcAB* gene cluster will likely be crucial to significant future developments in mercury methylation research in determining both the range of environmental conditions that may support methylation, as well as individual methylating species.

Where does methylation occur?

Wetlands and sediments

Methylation of Hg occurs in the vast majority of aquatic systems, indicating a wide variety of possible environmental conditions, and by extension, a wide range of geographical locations. Methylation has been extensively studied in boreal lakes and wetlands (Beijer and Jernelov 1979; Regnell 1994; Matilainen and Verta 1995; Pak and Bartha 1998; Benoit et al. 2002; Eckley and Hintelmann 2006), notably at the Experimental Lakes Area in northwestern Ontario, where much of the early work surrounding factors influencing methylation was done (Furutani and Rudd 1980; Ramlal et al. 1986, 1993; Miskimmin et al. 1992; Kelly et al. 1997; St. Louis et al. 2004; Harris et al. 2007). The importance of wetlands as sites of methylation was first determined in boreal wetlands (Zillioux et al. 1993; St. Louis et al. 1994, 1996; Branfireun et al. 1998, 1999; Tjerngren et al. 2012b) and has been recently observed in wetlands in the northern Great Plains (Hoggarth et al. 2015), southern Louisiana (Hall et al. 2008), the high Arctic (Lehnher et al. 2012), in agricultural and non-agricultural wetlands (Marvin-DiPasquale et al. 2014), and temperate forests (Selvendrian et al. 2008). Methylation also occurs in a number of saline, estuarine, and marine environments (e.g., Olson and Cooper 1974; Compeau and Bartha 1985, 1987; Mason et al. 1993), and recently it has been observed in the surface sediments of mudflats (a type of coastal wetland) (Ouddane et al. 2008), mangroves (de Oliveira et al. 2015), and lagoons (Bloom et al. 2004; Coelho-Souza et al. 2006; Monperrus et al. 2007a; Faganeli et al. 2012; Guédron et al. 2012; Hines et al. 2012). The porewaters of peatland have also been found to be a primary source of MeHg to the surrounding environment (Mitchell et al. 2008a), demonstrating the importance of these minuscule interstitial spaces in larger environmental impacts. The sediments of aquatic environments are important sites of methylation, particularly at the interface with water.

Recent studies have confirmed the sediments and porewater of aquatic environments to be key locations of methylation and have elucidated how methylation potential may change in proportion to depth within the sediment. Higher %MeHg in mudflats (Ouddane et al. 2008), higher MeHg concentrations in lagoons (Monperrus et al. 2007b), and higher potential methylation and demethylation rates in Gulf of Mexico sediments (Liu et al. 2015) were found in the surface sediment compared with deep sediment, confirming past research that suggested that methylation occurs primarily in the upper layers of sediment where there is significant microbial activity (Rudd et al. 1983; Korthals and Winfrey 1987; Matilainen 1995). A similar effect has been observed in peatland porewaters, with higher MeHg concentrations found nearer to the surface (Selvendrian et al. 2008). Decreases in methylation potential with increasing distance from the sediment–water interface may be due to bacteria from the sediment moving into the water column once oxygen is depleted (Eckley and Hintelmann 2006), which is further supported by the fact that sulfate reduction rates are positively correlated with methylation rates in terms of sediment depth (King et al. 1999; Merritt and Amirbahman 2009). This may be due to depth-dependent variations in the availability of electron donors (such as organic acids, long-chain fatty acids, hydrogen, aliphatic hydrocarbons, and simple aromatic compounds) that are required for bacterial methylation processes (Devereux 1996; Merritt and Amirbahman 2009). Branfireun et al. (1996) suggested that this effect may be due to greater labile carbon input and exposure, greater nutrient availability, anoxic conditions, abundant methylating microbes, and warmer temperatures nearer to the surface, which all support methylation. High methylation rates at the sediment–water interface with depth-correlated decreases may also be the result of either sulfur(II)-mediated inhibition of methylation or a diminution of the quality of organic substrates for the metabolic activity of SRB (Merritt and Amirbahman 2009).

Benthic surfaces

Methylation has also been found to occur in periphyton (which is primarily composed not only of algae, but also fungi, bacteria, protozoa, and metazoa) (Cleckner et al. 1999; Žižek et al. 2011). Achá et al. (2011) examined periphyton associated with the roots of tropical macrophytes *Eichhornia crassipes* (common water hyacinth) and *Polygonum densiflorum* (knotweed), where anaerobic or microaerophilic bacteria are able to grow within the periphyton biofilm (Enrich-Prast et al. 2004; Achá et al. 2011). Periphyton associated with the floating roots of macrophytes was found to be the primary location of methylation in tropical freshwater environments (Coelho-Souza et al. 2006). In tropical lakes, the phytoplankton responsible for methylation in the periphyton include *Microcystis aeruginosa* and *Synechocystis* species; however, it is unknown whether these phytoplankton are acting in association with other bacteria (Coelho-Souza et al. 2006), indicating a gap in the literature that warrants further exploration. In Canadian fluvial wetlands, Hamelin et al. (2015b) observed levels of net MeHg production in the periphyton that were two orders of magnitude greater than those in the sediment. Furthermore, there was a direct correlation between the methylation rate constant and the MeHg concentration of the periphyton; concentrations that were 10× higher than in the lake's macrophytes (Hamelin et al. 2015a, 2015b). It is evident that significant amounts of methylation occur not only in sediments but also in the periphyton of aquatic environments. Methylation in periphyton could have significant implications for biota, as periphyton can be an important source of food and nursing sites for fish.

Pelagic environments

While sediments are the primary location of methylation, methylation may also occur—albeit to a lesser extent—in the water column of aquatic systems (Olson and Cooper 1974; Korthals and Winfrey 1987; Xun et al. 1987; Ullrich et al. 2001). Recent work has shown that euphotic surface waters of the ocean are a potential location of methylation. Soerensen et al. (2016) found that the majority of net MeHg production occurred at a depth of between 20 and 200 m in the Arctic Ocean. Plankton abundances have been found to fluctuate in correlation with MeHg concentrations in ocean waters (Heimbürger et al. 2010; Kirk et al. 2012), and it has been hypothesized that methylation in oxic ocean waters (Monperrus et al. 2007a), lagoon water columns (Monperrus et al. 2007b), tropical lakes (Coelho-Souza et al. 2006), and hydroelectric reservoirs (Si et al. 2015) is affected by the abundance and activity of pelagic microorganisms like phytoplankton and bacterioplankton. Methylation in Arctic marine ecosystems (Pongratz and Heumann 1998, 1999; Lehnher et al. 2011; Kirk et al. 2012) may be accomplished by macroalgae (Pongratz and Heumann 1998) or marine bacteria (Pongratz and Heumann 1999) in the water column (Lehnher et al. 2011), open polynyas (St. Louis et al. 2007), or shelf sediments and rivers in coastal regions (Fitzgerald et al. 2007; Kirk et al. 2012).

Recent studies have focused on methylation in the water column of still bodies, where higher concentrations of MeHg and spikes in MeHg formation have been strongly associated with anoxic conditions (Eckley and Hintelmann 2006; Johnson et al. 2015). In several Canadian lakes, methylation in the water column was detected only in the lower, cooler, and stagnant anoxic hypolimnia, with the highest methylation potential measured just below the oxycline and decreasing with depth in the water column (Eckley and Hintelmann 2006). However, this contradicts studies of oxygen-deficient zones of marine systems, where anaerobic bacteria were not significant sources of MeHg, which in turn aligns with other studies that demonstrated a positive correlation between MeHg concentration and decomposing organic matter in the oxic water column (Malcolm et al. 2010). Recent research suggests that the water column may prove to be a significant zone of methylation in certain aquatic environments.

Inundated environments

Increases in MeHg concentrations in flooded environments result from the stimulation of microbial methylators by organic material supplied via decaying vegetation and soils (Bodaly et al. 1984; Kelly et al. 1997; Hall and St. Louis 2004; Hall et al. 2004, 2005), resulting in elevated fish Hg concentrations 10–20 years post impoundment (Bodaly et al. 2007). Revisiting the sites of inundated terrestrial areas nine years after initial flooding has shown that MeHg concentrations in the once flooded soils remain elevated (Rolfhus et al. 2015). Recent studies have focused on the influence of fluctuating inundation on methylation potential, particularly on the enhancement of methylation through the exposure of the sediment to atmospheric oxygen, which may promote sulfate recycling via sulfide oxidation, thus providing additional sulfate resources for SRB (Eckley et al. 2015). In a boreal peatland study, oxidation during droughts encouraged the mobilization of MeHg to peat porewaters when the land was re-flooded, allowing for increased MeHg production, particularly in the areas of peatland that had increased atmospheric sulfate deposition (Coleman Wasik et al. 2015). In addition, inundation and stagnation may reduce oxygen availability in the sediment as pores fill with water, thus encouraging the activity of anaerobic bacteria (Briggs et al. 2015; Singer et al. 2016). In this case, the frequency of inundation should be considered when determining the risks of mercury contamination in particular aquatic environments, as the frequency and duration of inundation have been found to be correlated with estimated MeHg production potential (Singer et al. 2016). Lower frequencies of inundation have been correlated to lower production potential (Singer et al. 2016), which may be linked to the potential of extended inundation to encourage MeHg degradation. Both flow frequency and inundated area have been found to determine production potential, rather than flood magnitude alone (Singer et al. 2016). Temporal flooding also results in an increased volume of sediment in which methylation can take place (Heim et al. 2007; Sizmur et al. 2013). Recent research suggests that prairie wetlands with temporal hydrological patterns of drying and re-flooding can promote methylation over permanently inundated environments, provided that the flooding periods are not sufficiently long to promote MeHg degradation (Sando et al. 2007).

The influence of flooding is also demonstrated through the disproportionately higher levels of MeHg found in rice compared with other crops, which is a result of its cultivation in flooded conditions that increases the anoxic environment in which methylating organisms thrive (Qiu et al. 2008). The majority of recent studies on methylation in rice paddies have examined contaminated systems (Rothenberg et al. 2014), which are beyond the scope of this paper. However, methylation has been found to occur in non-contaminated rice cultivation sites around the world including Brazil (da Silva et al. 2010; Batista et al. 2012; Silva et al. 2012), China (Rothenberg et al. 2011), and Spain (da Silva et al. 2013). The use of stable isotope tracers recently confirmed that MeHg produced in saturated soils is subsequently observed in the plant's rice grains (Strickman and Mitchell 2017), suggesting that the prevalence of methylation in rice paddies is a significant concern due to its potential to cause mercury poisoning directly through a common staple food source. However, Strickman and Mitchell (2017) also found significant MeHg losses between flowering and maturity; evidence of in planta demethylation that has also been observed in the roots rice exposed to more elevated Hg concentrations typical of contaminated rice paddies (Xu et al. 2016). Understanding the relationships of the plant life cycle and changes in mercury speciation could have implications for the management of rice production, for example, timing the flooding of fields to reduce the uptake of MeHg from soil before flowering to reduce the translocation of MeHg into grains (Peng et al. 2012). Exposing rice paddies to aerobic conditions reduced the uptake of MeHg by plants, perhaps due to a reduction in the amount of SRB in aerobic conditions and water management strategies that include exposing soil to the air during particular phases of growth may lessen the risks of methylation (Wang et al. 2014). However, it is unclear whether the effects of re-flooding, which seem to promote methylation in other environments, would negate the benefits of aerobic cultivation. While a complete picture of the effects

of temporal flooding on methylation in rice paddies is unclear, its effects in other aquatic environments have been more intensively studied.

What abiotic factors control mercury methylation?

The factors controlling biotic methylation are those that influence the activity of methylating bacteria, affect Hg bioavailability, or both (Benoit et al. 1999, 2002; Heyes et al. 2006), and include the concentration of bioavailable Hg, temperature, pH, redox potential, and the presence of complexing agents (presented in more detail below) (Celo et al. 2006). It is in the study of these factors that the newer techniques of stable isotope and genomic methods have begun to clarify some of the confounding issues of multiple controls on MeHg production.

Oxygen availability

Anoxic environments have been established over decades of research as the primary location of methylation (Compeau and Bartha 1985; Watras et al. 1995b; Warner et al. 2003; Sonke et al. 2013) and contemporary research has continued to observe the importance of anoxic conditions to methylation potential due to the fact that most sediments are anoxic. However, there is growing evidence that methylation can occur in both reducing and oxic environments such as the oxic surface waters of the ocean (Monperrus et al. 2007a; Heimbürger et al. 2010; Sonke et al. 2013) and polar marine waters (Lehnher et al. 2011; Sonke et al. 2013). Liu et al. (2015) found no relationship for MeHg concentrations and trends in water column hypoxia in the Gulf of Mexico, although potential rates of both methylation and demethylation were higher than in other coastal systems (Table 1). Methylation in macrophyte roots of tropical lakes was observed only in lakes with oxic conditions (Correia et al. 2012). Another study detected methylation in the proximity of marine snow, which are aggregates of exopolymers, planktonic and bacterial cells, and organic matter that form differently sized particles 300 µm or greater (Ortiz et al. 2015). While the authors suggest that anoxic areas around these aggregations provide ideal conditions for methylation (Ortiz et al. 2015), others consider the marine snow itself an oxic environment in which methylation may be occurring (Sonke et al. 2013). Other research has demonstrated positive correlations with oxygenic conditions such as those that exist in photosynthetic blooms in marine waters and MeHg concentrations (Cossa et al. 2009; Sunderland et al. 2009; Heimbürger et al. 2010). Oxic conditions may even influence methylation in the sediment, as high methylation has been observed in the upper layers of sediment below oxygenated water (Korthals and Winfrey 1987; Eckley and Hintelmann 2006), in contrast to lower methylation in sediment below anoxic water (Watras et al. 1995b; Eckley and Hintelmann 2006). One possible explanation of the association of oxic conditions with methylation is that some methylating bacteria are not anaerobic, but in fact require oxygen (Eckley and Hintelmann 2006). Recent research suggests that methylation occurs not only in anoxic environments, as has been well established, but may occur in oxic conditions as well. The role of microbial organisms able to methylate in oxic environments will become clearer as the use of the gene pair to determine methylators and the metabolic requirements of these organisms expands.

Organic matter

It has been well-established that dissolved organic matter (DOM) stimulates microbial activity, and by extension, methylation (Furutani and Rudd 1980; Hall et al. 2004, 2005; Ravichandran 2004). Organic matter produced in wetlands complexes MeHg (Ravichandran 2004), thereby enabling its transport to surface waters, but it can also complex inorganic Hg, thereby decreasing Hg bioavailability (Miskimmin et al. 1992; Driscoll et al. 1995; Watras et al. 1995a; Barkay et al. 1997; Ullrich et al. 2001; Eckley and Hintelmann 2006). A number of recent studies have confirmed significant correlations with organic matter and MeHg concentrations. In freshwater wetlands in the Adirondack Mountain region of New York, the highest concentrations of MeHg were observed during growing periods, which were also periods of high decomposition (Selvendrian et al. 2008). In subarctic and boreal lakes, DOM

was the variable most strongly associated with MeHg concentration regardless of climate, deposition patterns, or system size (Braaten et al. 2014). Faganeli et al. (2012) found that organic matter in lagoons was important in stimulating microbial methylating activity and that rapid degradation of organic matter by microbial activity is a seasonally fluctuating environmental condition that affects Hg bioavailability. In Louisiana wetlands within a salinity gradient, Hall et al. (2008) observed that concentrations of MeHg were correlated with the fraction of highly reactive dissolved organic carbon (DOC). While concentrations of DOM in marine systems are lower than in freshwater systems, the availability of organic carbon has been correlated to MeHg distribution in marine surface and intermediate waters in the Arctic (Cossa et al. 2009; Sunderland et al. 2009; Kirk et al. 2012), north Pacific (Sunderland et al. 2009), and Mediterranean (Cossa et al. 2009), as well as in estuarine (Lambertsson and Nilsson 2006) and marine (Mazrui et al. 2016) sediment. In rice paddies, the organic matter from rice waste both acts a nutrient source for methylating microbes and promotes the partitioning of MeHg to the dissolved phase (Rothenberg et al. 2011), thereby potentially increasing its bioavailability (Fleck et al. 2014). In tundra lakes with varying levels of DOC due to slumping permafrost, MeHg uptake to aquatic invertebrates was stimulated as DOC concentrations increased, until $\sim 8.5 \text{ mg C}\cdot\text{L}^{-1}$, after which there was an inhibitory effect (French et al. 2014).

The influence of DOM on methylation is often confounded by sulfur geochemistry. In environments where sulfate is not limited, methylation is primarily determined by the availability of organic matter; however, in environments with a sulfide concentration of over $0.01 \text{ }\mu\text{M}$, mercury sulfide complexes form preferentially, making binding with DOC less significant in determining methylation (Benoit et al. 2001). An additional effect of DOM, particularly aromatic carbon, is its potential to enhance the release of Hg from insoluble cinnabar (Waples et al. 2005), which can serve as a source of mercury to the food web through methylation. Given that cinnabar is the most commonly occurring form of mercury (Boening 2000), this can pose a significant risk for elevated MeHg in aquatic organisms. Organic matter functions in multiple ways to promote and enhance Hg methylation through its capacity to stimulate microbial activity, provide methyl groups for methylation, and release Hg from cinnabar, potentially enabling its methylation.

Sulfur

The presence of sulfur has repeatedly been found to have strong correlations to MeHg levels in several types of aquatic environments. As the activity of SRB is controlled in part by the presence of sulfate, increased sulfate can stimulate methylation (Gilmour et al. 1992; King et al. 1999). Low sulfide concentrations allow for the formation of neutral mercury sulfide complexes capable of diffusing through cell membranes, thereby promoting methylation, whereas higher concentrations can result in charged mercury sulfide complexes that decrease Hg bioavailability (Benoit et al. 1999). The influence of sulfate was recently demonstrated in an experiment that manipulated atmospheric sulfate loading to a small boreal peatland, demonstrating that both MeHg concentrations and %MeHg in porewaters increased within a week of sulfate addition and declined as sulfate disappeared (Jeremiason et al. 2006; Coleman Wasik et al. 2012). The stimulation of MeHg production decreased once the addition of sulfate was discontinued; however, even four years later, MeHg concentrations and %MeHg were still higher than in control systems (Coleman Wasik et al. 2012). In a related mesocosm study, Mitchell et al. (2008b) found that while additions of labile organic carbon to peat did not stimulate methylation, sulfate, both alone and in combination with some forms of organic C, increased methylation production significantly. They also found that MeHg concentrations in treatments receiving both sulfate and carbon additions were similar to those previously measured in MeHg “hot spots” found near the interface of upland and peatland systems.

It is worth noting that not all environments with high sulfate concentrations are MeHg production “hotspots”. Prairie wetland sites with naturally high sulfate values (up to thousands of $\text{mg}\cdot\text{L}^{-1}$) well

within the range of concentrations that have been found to be inhibitory to methylation (Gilmour et al. 1998; Benoit et al. 2001) had higher MeHg proportions in the sediment and higher MeHg concentrations in surface water, but not higher potential methylation rates (Hoggarth et al. 2015). High electrical conductivities (driven by high sulfate concentrations) may have resulted in low partitioning coefficients in the high sulfate sites suggesting that porewater sulfide accumulation may have been preferentially pulling Hg into the porewater phase. While there continue to be clear correlations between the presence of sulfate and methylation potential, and developing research suggests methylation is positively correlated with the presence of sulfides, Hg cycling and methylation in naturally high sulfate sites have not been adequately investigated and may be an area of future research.

Temperature

Previous research has suggested that warmer water temperatures may promote bacterial methylation (Bacci 1989; Bodaly et al. 1993; Ramlal et al. 1993), which is supported by recent studies that identified a positive correlation for MeHg production rates and temperature (Monperrus et al. 2007a; Johnson et al. 2016). In some cases, methylation could only be detected in lake hypolimnia (which are perpetually cold due to a lack of thermal circulation) (Eckley and Hintelmann 2006), but this also is likely due to the anoxic nature of hypolimnia; the link between methylation and cool temperatures is ultimately unclear. Lower MeHg concentrations and %MeHg in subarctic lakes compared with boreal lakes were believed to be due to lower temperatures not suitable for methylation, but differing Hg deposition patterns between sites was also presented as an explanation (Braaten et al. 2014). The influence of temperature on methylation may, therefore, be dependent on the climatic conditions of the specific aquatic environment under study. In addition, confounding factors such as increased anoxia in hypolimnetic areas make it difficult to fully assess the impact of temperature on methylation in situ.

Salinity, pH, and trophic status

Previous research has found low-salinity environments to be linked to higher levels of net methylation (Blum and Bartha 1980; Compeau and Bartha 1987; Barkay et al. 1997); however, recent studies have shown salinity to both stimulate, and to have no correlation with, methylation potential (e.g., Braaten et al. 2014). In laboratory conditions at a constant pH of 5.3, MeHg concentrations were found to be highest at a salinity of $0.5 \text{ mol}\cdot\text{L}^{-1}$, and concentrations declined as salinity rose (Chen et al. 2015). No correlations were observed for salinity and MeHg concentrations in natural mangrove ecosystems (de Oliveira et al. 2015) or hypersaline lakes (Johnson et al. 2015). The effect of pH conditions on methylation is equally unclear. While prior research demonstrated that the highest levels of methylation were typically associated with acidic environments (Ramlal et al. 1986; Bloom et al. 1991; Gilmour and Henry 1991; Spry and Wiener 1991; Ullrich et al. 2001), recent studies have observed methylation to occur only in tropical lakes with a neutral pH (Correia et al. 2012) and in prairie wetlands with pH above 8 (Hoggarth et al. 2015). In sub-Arctic and boreal lakes the opposite was observed, in that both MeHg concentration and %MeHg were positively correlated to pH and alkalinity (Pennanen et al. 1998; Braaten et al. 2014).

The trophic status of a lake may also be an important consideration. In oligotrophic systems, nitrogen availability was positively correlated with MeHg concentration and %MeHg, suggesting that in some systems methylation may be stimulated by N availability (Braaten et al. 2014). Eutrophic conditions are also believed to influence methylation (Todorova et al. 2009), though this may be due to the enhancement of methylation in the presence of organic matter. In an eutrophic and hypolimnetic reservoir in Idaho sediments that were rich in organic matter, low in dissolved oxygen, under redox conditions, and with increased nutrient content were suggested to lead to elevated methylation rates compared with other systems (Gray and Hines 2009). This is in contrast to other recent studies that have observed negative correlations of nitrate and MeHg concentrations in the water column of

eutrophic lakes (Todorova et al. 2009). A mesocosm study of estuarine sediments that were amended with various levels of N and P loadings to surface water showed that increased nutrients led to increased phytoplankton biomass productivity with stimulated microbial activity in sediments with Hg tracers that were considered highly bioavailable. However, low bioavailability Hg tracers showed no response, suggesting both increased sediment productivity and bioavailability-controlled MeHg production (Liem-Nguyen et al. 2016). The influence of salinity, pH, and eutrophic conditions on methylation remains unclear and may only form part of a set of interconnected environmental variables that determine methylation.

Iron and manganese oxides

Iron and manganese oxides have been found to significantly influence Hg cycling and methylation. In sewage treatment plants that use hydrated ferric oxide for water purification, the highest levels of methylation were found in areas rich in dissolved Fe^{2+} and organic matter (Bravo et al. 2015). The presence of compounds containing hydroxyl radicals has been found to enhance methylation at low concentrations due to their ability to function as methyl donors during photomethylation, but these compounds degrade MeHg when present in excess (Chen et al. 2015). A study of a variety of lagoon systems found that regardless of Hg source, Hg was influenced by the transformations of other elements, particularly redox-sensitive and microbially important elements like sulfur, iron, and manganese, and by the interactions of those elements with organic material (Faganeli et al. 2012). The correlation of methylation and the presence of particular elements and compounds in a number of aquatic environments may be linked to the importance of those elements in the mediation of microbial activity, which in turn determines methylation potential.

Light

Light plays a role in regulating the opposing processes of methylation and demethylation (Sellers et al. 1996; Monperrus et al. 2007a). In conditions such as those in the euphotic zones of coastal and marine waters, light can stimulate phototrophic organisms to reduce HgII to Hg0 , thus reducing the pool of inorganic Hg available to methylation organisms and exerting a crucial role in regulating levels of MeHg in surface waters (Grégoire and Poulain 2014). However, light also stimulates photosynthetic activity, which in turn influences the release of labile DOM that encourages methylating microbial activity. Monperrus et al. (2007a) found that peak methylation corresponded to seasonal conditions later in the summer, which were conducive to higher planktonic content suggesting that methylation was primarily mediated by heterotrophic activity. Recently, both MeHg concentration and %MeHg were found to be negatively correlated with both lake size and catchment area (Braaten et al. 2014). As photodemethylation is considered the most important sink of MeHg in freshwater lakes (Lehnher and St. Louis 2009), this negative correlation is likely due to the larger area of surface water in the catchment exposed to UV light degradation compared with smaller lakes, thereby decreasing the net amount of MeHg produced in situ or that has leached from catchment soils and wetlands (Lehnher and St. Louis 2009; Braaten et al. 2014). As a result, organisms in larger lakes with greater catchment areas may be less subject to the deleterious effects of methylated Hg, as photodemethylation processes may actively be breaking down this highly bioavailable Hg species. However, catchment wetlands are believed to be the primary location of methylation (St. Louis et al. 1994; Tjerngren et al. 2012a; Braaten et al. 2014), suggesting that such effects may be mitigated if the catchment wetland area is sufficiently large.

Bioavailability of Hg

Production of MeHg requires inorganic Hg to be available to methylating organisms. Deposition of inorganic Hg, although decreasing globally, is not expected to decline to zero. In addition, legacy Hg deposits currently sequestered in sediments, wetland soils, permafrost, and forests may become

mobile under a number of scenarios such as forest fires, and the disturbance of soils, sediment, and permafrost, thus increasing the inorganic Hg available for MeHg production. However, the uptake of this critical substrate has been found to be limiting in the methylation process, especially in the absences of certain complexing thiols (Schaefer et al. 2011). Hsu-Kim et al. (2013b) provide an excellent review of the factors controlling bioavailable Hg and summarize four main hypotheses for the uptake through both outer and inner membranes. Inorganic Hg is generally complexed with chloride, sulfide, or DOM and can be taken up either by diffusion or active transport by transmembrane proteins (Hsu-Kim et al. 2013a). As well as dissolved forms, nanoparticles, colloids, and crystalline particles may also be bioavailable (Aiken et al. 2011). Because of the complexity of the interactions among the various possible sources of bioavailable Hg, chemical equilibrium modelling contains high degrees of uncertainty and therefore does not confidently predict bioavailability and perhaps a kinetics-based approach would be more illuminating (Hsu-Kim et al. 2013b). Regardless, experiments examining the impact of different environmental parameters on MeHg production in complex microbial communities simultaneously will hopefully allow for the development of models more descriptive of methylation.

The use of a genetically engineered bacterium that exhibits bioluminescence in response to the uptake of Hg into the cell (Selifonova et al. 1993) has provided some information on the influence of Hg speciation on bioavailability. This “bioreporter” has been successful in showing enhanced uptake of inorganic mercury when newly deposited (Chiasson-Gould et al. 2014), under low base cation concentrations (Daguené et al. 2012), and at lower pH (Kelly et al. 2003). The same bioreporter has been used to assess the bioavailability of MeHg, showing that increasing chloride concentration resulted in increased MeHg uptake; the presence of both humic acids and cysteine reduced MeHg bioavailability (Ndu et al. 2012). This approach has also been used to show that dissolved organic carbon both mobilizes inorganic Hg and alters cell walls to facilitate uptake (Chiasson-Gould et al. 2014). The use of this technique, however, has challenges. For one, the bioreporter must be constructed, which can be time consuming. For example, it took Ndu et al. (2012) 20 cycles before the *lux* reporter proteins were synthesized by the bacterium of interest. In addition, significant care must be taken to ensure that the growth medium is not contaminated with low levels of Hg that will promote light production. Finally, the bacterium is generally not representative of methylating organisms, with experiments generally performed using *Escherichia coli* or *Vibrio anguillarum*, both Gram-negative bacteria capable of aerobic metabolism.

How do we deal with so many confounding factors?

It is clear that the vast majority of studies of the environmental factors exerted by DOM on methylation have been correlative rather than direct observations. However, molecular techniques have allowed research to explore the direct impact of environmental factors on the activity of methylating organisms. For example, using the model Hg-methylating bacterium *Desulfovibrio desulfuricans* ND132, Graham et al. (2013) examined Hg bioavailability in cell assays at gradients of DOM (~ 1 – 8 ng-Hg \cdot mg $^{-1}$ C) and sulfide concentrations (1–1000 μ M). Methylation was significantly greater in DOM-amended assays (especially those with highly aromatic DOM isolates) as compared with control absent of DOM. In addition, they found that assays amended with high sulfur content were particularly effective at enhancing methylation. In another example of the use of molecular techniques it was observed that the greater relative abundance of organisms with known methylator gene sequences in the sediment microbiome of high carbon DOM, as opposed to oligotrophic, mesocosms (Graham et al. in review). Goñi-Urriza et al. (2015) showed that *hgcAB* gene expression varied in *Desulfovibrio dechloroacetivorans* BerOc1 when grown under sulidogenic conditions and different carbon sources. However, they were unable to relate gene expression to potential methylation rates. Although in its infancy, with the advent of new molecular techniques, including the recent development of clade-specific quantitative molecular probes (Christensen et al. 2016), future studies should

be able to test the impact of differences in amounts and types of DOM directly on methylating organisms.

Until recently, research has focused on one or two environmental factors as a means of understanding the controls of MeHg production. Recent developments of the use of stable isotope tracers and geonomic work have begun to examine the synergies of multiple factors. For example, [Keucharzyk et al. \(2015\)](#) found that methylation rates in marine sediments were highest when mixed cell cultures were amended with carbon, regardless of whether inorganic Hg was added as dissolved nitrate salt or as nanoparticles of HgS (as also observed by [Graham et al. \(2013\)](#); see above). Similarly, nutrient loading that stimulated microbial activity indirectly by increasing phytoplankton biomass was as important as bioavailability of Hg available to methylators ([Liem-Nguyen et al. 2016](#)). The conclusion was that MeHg production is limited by microbial productivity, regardless of Hg bioavailability, and that there is a threshold over which Hg speciation becomes a contributing control on methylation.

How will Hg methylation be impacted by a changing climate?

As the chemistry of the earth's atmosphere continues to change with anthropogenic emissions of greenhouse gases, global circulation models predict substantial atmospheric warming of 2–4.5 °C (Intergovernmental Panel on Climate Change (IPCC) 2014). The most significant impact predicted to accompany increased radiative forcing is the increased warming of the earth's surface, which is predicted to be greatest at the poles. As a result of this increased warming, a number of indirect impacts are predicted such as increased sea level rises and intensified hydrological cycles, which are anticipated to increase the frequency of both drought and deluge, both of which have ecological impacts of their own. Many of these impacts have the potential to change rates of net Hg production (Fig. 1), and our current and future understanding of the factors that control the Hg biogeochemical cycle will allow us to make better informed predictions on what these changes may be.

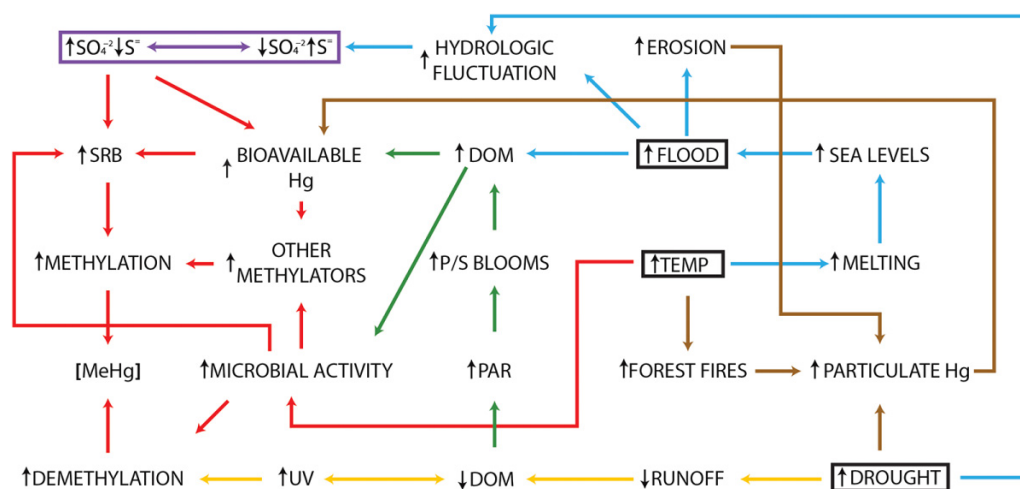


Fig. 1. A summary of the probable impacts of climate change on the changes in rates of methylation and demethylation resulting in changes in methylmercury concentrations ([MeHg]). Arrows up and down represent, respectively, positive and negative responses for variables including (but not limited to): temperature (TEMP), dissolved organic matter (DOM), sulfate and sulfide concentrations (SO_4^{2-} $\text{S}^{=}$), photosynthetic blooms (P/S blooms), and the activity of sulfate-reducing bacteria (SRB).

Increased temperature

Reviews of the impact of climate change on the Arctic have been published (Douglas et al. 2012; Stern et al. 2012); however, we touch briefly on a few here. Because the impacts of climate change are already being experienced in the Arctic, there has been recent work examining changes in Hg biogeochemistry with increased temperature. There have been a number of studies examining the impact of warming-induced ecosystem change from forested peat permafrost to treeless and permafrost-free thermokarst wetlands that is accompanied not only by increases of labile organic carbon (Jansson and Tas 2014; Yang et al. 2016b) but also by the release of inorganic Hg sequestered over decades of Hg deposition (Gordon et al. 2016). These changes, together with increased microbial activity as a result of warmer temperatures, have been hypothesized to result in greater rates of methylation and MeHg bioaccumulation in the Arctic (Podar et al. 2015; Yang et al. 2016a). Melting ice and permafrost may also increase river discharge to estuarine systems, with accompanying greater loads of bioavailable Hg and DOM, as well as increases in riparian area flooded, both factors in stimulating Hg methylation. However, increased temperature may also result in greater rates of microbial demethylation. Increased temperature will also result in the loss of sea ice and larger areas of open ocean susceptible to increased rates of photochemically mediated reactions in the Hg biogeochemical cycles (Stern et al. 2012). Interestingly, Girard et al. (2016) found that the rates of photodemethylation were higher in high DOC thaw ponds than in oligotrophic lakes, suggesting that some level of DOC is necessary for photodegradation to occur. Finally, increased temperatures will likely result in increased forest fires, which are a source of atmospheric deposited Hg sequestered in plants and soils (Kelly et al. 2006).

Increased periods of deluge and drought

Fluctuations in the hydrologic cycles will result in periods of deluge and drought. In the former, flooding of terrestrial matter will mimic reservoir creation and result in higher rates of methylation as well as an increase in the release of previously sequestered Hg in eroded soils. While increased drought may have a substantial impact on the release of sequestered Hg in dried soils that may be transferred from sources to lakes and wetlands, the most significant impact of drought on Hg cycling will be in those methylating systems (for a comprehensive examination of the impact of climate change on lakes and wetlands, see Schindler 1997). For example, decreases in hydrologic inputs to lakes and wetlands result in decreases in DOM and Hg inputs. Dissolved organic carbon is natural “sunscreen” for lakes, and declines would result in higher rates of photodemethylation in these systems. Higher rates of photodemethylation could counter higher rates of methylation that may result from increased temperatures or concentrations of microbial substrates such as DOM. Increases in lake residence time with decreased water inputs may result in increased productivity, also stimulating methylation; however, this may be countered by decreased inputs of DOM and Hg. Finally, the interplay between periods of drought and deluge will result in hydrological fluctuation, influencing the oxidation of sulfur species and thus impacting the activity of SRB. While the inherent complexity of ecological systems and the interacting factors within them necessarily complicate exact prediction of the effects of climate change on Hg methylation, increased temperature and subsequently enhanced patterns of deluge and drought are likely to promote Hg methylation beyond levels of any potential mitigating effects.

Conclusion

While research in mercury methylation over the past 10 years has largely confirmed the past understanding of Hg cycling, there have been a number of advances, perhaps most significantly the identification of the *hgcAB* gene cluster, which is necessary for methylation, in many diverse organisms. This vastly broadens the scope of knowledge about methylating organisms and the environments in

which they may reside. It also contradicts prior research asserting that SRB are the primary biotic methylators, because many microorganisms, including IRB, methanogens, and archaea, have been found to contain this gene pair. Recent studies have found that organisms containing this cluster may live in extreme environmental conditions, which illuminates the potential for a much broader environmental scope of methylating organisms. This innovation has the potential to develop not only our understanding of which organisms are capable of methylation, but also of the diversity of abiotic environmental conditions that may support methylation. It is, however, important to note that the mere presence of methylating capacities in organisms does not indicate that methylation will necessarily occur. Therefore, further study that not only identifies organisms with the capacity to methylate, but also examines the role of these organisms in situ, is required.

Recent research has identified the presence of methylation in numerous environments, including freshwater and brackish wetlands, which are thought to be more prominent in terms of methylation than marine wetlands and marine lakes, and estuarine systems and open marine waters, which have been identified as potential areas of methylation. The presence of MeHg in the open ocean may be due to methylation in the water column or may originate from the sediments of coastal regions. Research into the methylation potential of the open ocean requires further development, particularly in the polar regions, which is an ongoing concern due to increased atmospheric Hg deposition in the region. Inundated aquatic systems may also act as sources of MeHg to the surrounding environment, and include hydroelectric reservoirs, rice paddies, and more recently identified sewage treatment plants due to the stimulation of methylation by the use of ferric oxide. The examination of inundated anthropogenic systems adds another dimension to the conversation about methylation in aquatic systems, considering not only how anthropogenic Hg pollution can be biologically mobilized, but also how man-made environments may contribute to MeHg contamination via changes in net methylation.

Recent research has developed the notion that methylation may occur in oxic environments, both in pelagic environments and in the sediment exposed to oxygenated water or temporary droughts that expose sediments to air, both of which have been found to enhance methylation. There is mounting evidence that systems that experience temporal hydrological patterns may be loci of greater MeHg production. With the use of stable isotope tracers and the ability to measure potential methylation and demethylation rates, the understanding of environmental factors as controls on MeHg production is mainly in the form of correlations among rates and physiochemical factors. Synergies, both positive and negative, among environmental controls are just recently being examined, in part due to the use of molecular techniques made possible by the *hgcAB* discovery. In the future, a better understanding is required of the rates of demethylation, which, in some environs, may be very important in determining net MeHg.

Finally, the incredibly complex responses of the earth's ecosystems to a changing climate will result in a myriad of possible changes to factors that control methylation and demethylation. Increased understanding of the mercury biogeochemical cycle will allow us better predictive abilities to assess responses in Hg cycling due to a changing climate. Recent molecular technology will likely guide the most significant strides in future research and enable a broadened understanding of the organisms that possess methylating capacities, the environments that these organisms can inhabit, and by extension, which environments may be at risk of MeHg contamination.

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Author contributions

Conceived and designed the study: ARP, BDH. Performed the experiments/collected the data: ARP. Analyzed and interpreted the data: ARP, BDH. Contributed resources: BDH. Drafted or revised the manuscript: ARP, BDH.

Competing interests

The authors have declared that no competing interests exist.

Data accessibility statement

All relevant data are within the paper.

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