Science of the Total Environment 721 (2020) 137647

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Recent advances in understanding and measurement of mercury in the environment: Terrestrial Hg cycling

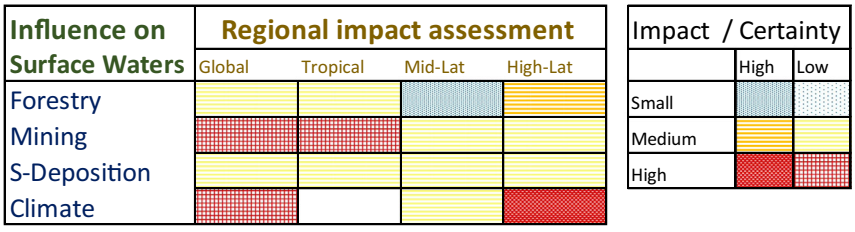
Kevin Bishop [a,*], James B. Shanley [b], Ami Riscassi [c], Heleen A. de Wit [d], Karin Eklöf [a], Bo Meng [e], Carl Mitchell [f], Stefan Osterwalder [g], Paul F. Schuster [h], Jackson Webster [i], Wei Zhu [j]

[a] Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, Box 7050, 75007 Uppsala, Sweden
[b] U.S. Geological Survey, Box 628, Montpelier, VT 05601, USA
[c] Department of Environmental Sciences, University of Virginia, P.O. Box 400123, Charlottesville, VA 22904-4123, USA
[d] Norwegian Institute for Water Research, Gaustadalléen 21, NO-0349, Norway
[e] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
[f] Department of Physical and Environmental Sciences, University of Toronto Scarborough, 1265 Military Trail, Toronto, Ontario M1C 1A4, Canada
[g] Institut des Géosciences de l'Environnement, Université Grenoble Alpes, CNRS, IRD, Grenoble 18 INP, 38000 Grenoble, France
[h] U.S. Geological Survey, 3215 Marine Street, Suite E-127, Boulder, CO 80303-1066, USA
[i] Department of Civil Engineering, California State University, 400 W. 1st Street, 21 95929-0930 Chico, CA, USA
[j] Department of Forest Ecology and Management, Swedish University of Agricultural Sciences, 90183 Umeå, Sweden

HIGHLIGHTS

- Terrestrial Hg cycling influences exposure of humans and biota to this potent neurotoxin.
- Advances in understanding were reviewed with a focus on developments since 2010.
- Decreased Hg emissions may bring recovery sooner than previously expected.
- Arctic warming is likely increasing global Hg fluxes and even direction in some cases.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 7 November 2019
Received in revised form 23 February 2020
Accepted 28 February 2020
Available online 7 March 2020

Editor: Mae Sexauer Gustin

Keywords:
Methylmercury
Climate
Land-use
Land-atmosphere exchange
Streamflow
Food

ABSTRACT

This review documents recent advances in terrestrial mercury cycling. Terrestrial mercury (Hg) research has matured in some areas, and is developing rapidly in others. We summarize the state of the science circa 2010 as a starting point, and then present the advances during the last decade in three areas: land use, sulfate deposition, and climate change. The advances are presented in the framework of three Hg "gateways" to the terrestrial environment: inputs from the atmosphere, uptake in food, and runoff with surface water. Among the most notable advances:

- The Arctic has emerged as a hotbed of Hg cycling, with high stream fluxes and large stores of Hg poised for release from permafrost with rapid high-latitude warming.
- The bi-directional exchange of Hg between the atmosphere and terrestrial surfaces is better understood, thanks largely to interpretation from Hg isotopes; the latest estimates place land surface Hg re-emission lower than previously thought.
- Artisanal gold mining is now thought responsible for over half the global stream flux of Hg.
- There is evidence that decreasing inputs of Hg to ecosystems may bring recovery sooner than expected, despite large ecosystem stores of legacy Hg.

* Corresponding author.
E-mail addresses: kevin.bishop@slu.se (K. Bishop), jshanley@usgs.gov (J.B. Shanley), alr8m@virginia.edu (A. Riscassi), heleen.de.wit@niva.no (H.A. de Wit), karin.eklof@slu.se (K. Eklöf), mengbo@vip.skleg.cn (B. Meng), carl.mitchell@utoronto.ca (C. Mitchell), stefan.osterwalder@univ-grenoble-alpes.fr (S. Osterwalder), pschuste@usgs.gov (P.F. Schuster), jwebster13@csuchico.edu (J. Webster), wei.zhu@slu.se (W. Zhu).

https://doi.org/10.1016/j.scitotenv.2020.137647

- Freshly deposited Hg is more likely than stored Hg to methylate and be incorporated in rice.
- Topography and hydrological connectivity have emerged as master variables for explaining the disparate response of THg and MeHg to forest harvest and other land disturbance.

These and other advances reported here are of value in evaluating the effectiveness of the Minamata Convention on reducing environmental Hg exposure to humans and wildlife.

1. Introduction

The UN Minamata Convention seeks to reduce the threats posed by mercury (Hg) pollution. In the half century since Hg was recognized as the pollutant responsible for the tragic poisoning of Minamata Bay, science has made great strides in better describing the global cycling of Hg (e.g. Obrist et al., 2017). The terrestrial environment is an important component of that global cycle. Land surfaces form extensive, complex interfaces with the atmosphere, and Hg moves in both directions across that interface (Agnan et al., 2016). Within the landscape Hg moves and transforms into different species, including the extremely toxic methylmercury (Kronberg et al., 2016b). Some Hg moves with runoff water from the terrestrial landscape into aquatic ecosystems. Even when the annual runoff flux is combined with the annual atmospheric exchange flux, the total amount is much smaller than the store of Hg in the soils of the landscape (UNEP, 2019). This large soil Hg store is not in itself a particularly large part of the risk posed by Hg to people or biota. Nonetheless, Hg transported from the terrestrial landscape in runoff is a source of the Hg that bioaccumulates in freshwater aquatic ecosystems, (e.g., Wiener et al., 2006; Chen et al., 2016). Methylmercury (MeHg), the methylated fraction of Hg, is of particular interest in this regard. Uptake of MeHg by rice from soils in Hg-contaminated areas also poses a danger to fauna and people since rice bioaccumulates Hg more than many other grains (Qiu et al., 2008). An extra impetus for quantifying Hg exchange between the land surface and atmosphere is provided by emerging questions about the effect of global warming on permafrost (Schuster et al., 2018) and wildfires (Kumar et al., 2018), as well as the atmospheric movements of Hg to and from the Arctic (Soerensen et al., 2016; Sonke et al., 2018). Furthermore, the Minamata Convention on Mercury (UNEP, 2013b) calls for assessing the effects of reductions in anthropogenic emissions as a result of the convention (Article 19), as well as progress towards the goals of "controlling and, where feasible, reducing emissions of mercury and mercury compounds to the atmosphere… and the release to waters." (Articles 8 and 9).

Thus a number of topical issues require an understanding of the terrestrial Hg cycle. Half a century of research has yielded much progress. The ability to measure Hg at environmentally relevant part per trillion levels, and then to subdivide that into different forms, including natural abundance isotopes, has been a key factor in those advances. Recent reviews of the literature of relevance to terrestrial cycling include Shanley and Bishop (2012), Hsu-Kim et al. (2018) and Obrist et al. (2018). To emphasize areas where scientific understanding is developing most rapidly, this paper focuses on advances during the past decade. To provide context for these advances, the paper starts with a concise summary of major features in the understanding of terrestrial Hg cycling as of 2010 (Section 2). This includes how landscapes receive Hg from the atmosphere, Hg cycling within the terrestrial environment, human influences on that cycling, and how Hg ultimately leaves the terrestrial environment. Here, "leave" includes evasion back to the atmosphere, consumption (birds, insects etc.), or delivery with runoff to downstream aquatic ecosystems. Analysis of human influences is focused on spatially extensive factors (forestry, agriculture, and climate) rather than point sources related to mining and industry. This terrestrial cycling is superimposed on the hydrological cycle which moves Hg, and several other terrestrial biogeochemical cycles. One is that of organic matter (OM) which can donate electrons to microbial processes that methylate Hg (Skyllberg et al., 2003). Another key cycle is that of sulfur (S), which influences Hg speciation (e.g., S^{2-}, S_2H^{3-}, SH^-) and can accept electrons (e.g., sulfate) during microbial Hg methylation (Liu et al., 2014b).

Against this background, advances since 2010 in understanding the terrestrial Hg cycle will be presented to focus attention on areas where knowledge has developed most rapidly. This starts by examining three aspects of how human activity alters Hg cycling (Section 3): forest landscape management, atmospheric deposition of sulfur on wetlands, and climate change (including thawing permafrost and fires). Then, impacts of human activity on three "gateways" to the terrestrial cycle will be considered (Section 4): exchange with the atmosphere, utilization of vegetation for food, and exports to surface waters. Key developments are identified at the end of each section. Section 5 then summarizes advances in the conceptualization of terrestrial Hg cycling since 2010, how this new understanding relates to implementing the Minamata Convention to reduce Hg exposure, and areas needing further revision.

2. Terrestrial cycling –baseline understanding circa 2010

A useful starting point for taking up the terrestrial Hg cycle is to consider the Hg mass balance for individual watersheds (Table 1). Alternatively, one can generalize Hg pools and fluxes within specific terrestrial environments (Fig. 1), here including a forested hillslope, a wetland from the nemoboreal zone, and the Arctic tundra. These settings represent only a fraction of the diversity of terrestrial environments, but they are three of the more studied settings (cf. Table 1) owing to the relatively high levels of Hg in the freshwater fish of such landscapes.

2.1. Hg inputs from the atmosphere

There are three potential sources of Hg in the terrestrial environment (Ebinghaus et al., 1999). Some originates in situ, from volcanism, geothermal activity, and near-surface deposits of Hg-bearing rocks. Other Hg can be introduced directly from point sources of human activity such as mining, industrial effluents, or biomedical waste. The rest is deposited from the atmosphere, which is a temporary store of Hg emitted from the earth surface. The net emission of Hg results from the combination of natural sources (10%), ongoing human activities (30%), and re-mobilization of previously deposited Hg from soils or leaves/needles, aquatic ecosystems including oceans, forest fires, and permafrost thawing (60%) (UNEP 2013). The re-mobilized Hg includes "legacy" Hg from earlier human activities.

Atmospheric deposition is the downward component of a bi-directional exchange of Hg between the landscape and the atmosphere. The downward component is composed of both gaseous elemental Hg (GEM, subsequently referred to as Hg^0) and operationally defined gaseous oxidized Hg (Hg(II)) as well as particulate Hg. The upward component is primarily Hg^0, due to the exceptionally high vapor pressure (for a metal) of Hg^0. The presence of gaseous elemental Hg in the atmosphere facilitates long-range transport before deposition to water and land. As of 2010, it was estimated that Hg concentrations in the atmosphere had increased from 2050 Mg to 5600 Mg (Selin et al., 2008).

Table 1
Mercury and methylmercury stream output flux from watersheds, based on whole water (unfiltered), with percent retention based on wet-only and total deposition (when provided). Flux units are μg m^{-2} yr^{-1}. Data summarized from Table S1.

			THg			MeHg			Median % retention[c]			
									THg		MeHg	
Ecosystem type	#studies	#sites[b]	High	Median	Low	High	Median	Low	Wet only	total dep	Wet only	total dep
Arctic[a]	11	18	112.24	1.71	0.13	0.480	0.017	0.010	n/a	n/a	n/a	n/a
Upland Forest	26	53	54.40	1.97	0.16	0.370	0.054	0.010	77.0	92.0	63.4	87.8
Wetland and forest wetland	13	18	5.50	1.45	0.25	0.185	0.055	0.022	75.0	76.3	−5.1	63.3
Agriculture	5	17	4.90	2.69	0.53	0.120	0.063	0.026	n/a	n/a	n/a	n/a
Urban	5	9	22.21	4.77	1.60	0.160	0.080	0.020	n/a	62.6	n/a	52.2

[a] Includes one alpine tundra site.
[b] Some similar sites within a study were lumped as a "single site": Hurley et al. (1995), Domagalski et al. (2016), St Pierre et al. (2018).
[c] Based on wet only deposition and based on total deposition (if available); n/a in % retention implies too few data.

Over the millennia, volcanic eruptions elevated atmospheric Hg concentrations for short periods (years), but human activity is responsible for sustained elevation of background levels, most markedly between circa 1950 and 1985 (Smith-Downey et al., 2010).

Atmospheric Hg deposits to the land surface as both wet and dry deposition. Elemental Hg^0 makes up circa 95% of atmospheric Hg but does not contribute to wet deposition because of its low solubility (i.e. low Henry's law constant) in clouds (Schroeder and Munthe, 1998). Hg^0 can be oxidized to reactive Hg(II), a mix of operationally defined gaseous oxidized Hg and particulate-bound Hg (Landis et al., 2002) which is rapidly scavenged from the air by water droplets and deposited as wet deposition. In contrast, dry deposition consists of Hg(II) and Hg^0 that attaches directly to surfaces, including vegetation and particles in the atmosphere.

The forest canopy takes up Hg^0 from the atmosphere in growing foliage throughout the growing season, such that Hg in litterfall is a measure of dry Hg^0 deposition (Rea et al., 2002; Millhollen et al., 2006). Hg (II) in both gaseous and particle form also deposits to the canopy, but this Hg is readily washed off by throughfall (Graydon et al., 2008). Variation in the different components of Hg in deposition is considerable (Table S1), with dry deposition of Hg^0 particularly uncertain (Fritsche et al., 2008; Zhang et al., 2009). In forested areas, however, litterfall is the single largest component of atmospheric deposition (Lee et al., 2000; Demers et al., 2007). As litterfall decomposes, the remaining OM binds Hg in the soil.

A key factor in terrestrial Hg cycling is that Hg can be re-emitted from the landscape to the atmosphere. However, understanding of re-emission processes has been limited (Lindberg et al., 2007; Hammerschmidt and Fitzgerald, 2008; Brigham et al., 2009). The understanding of Hg re-emission as of 2010 was influenced by awareness of Hg deposition episodes in the Arctic. Photo-oxidation triggered by the Arctic sunrise resulted in rapid dry deposition of Hg (Schroeder et al., 1998). Within days, however, much of this Hg was returned to the atmosphere (Lalonde et al., 2002; Sherman et al., 2010). Studies had also demonstrated re-emission of Hg^0 from other terrestrial surfaces besides snowpack, including forests and wetlands (Lindberg and Zhang, 2000; Gustin and Stamenkovic, 2005; Demers et al., 2007; Fritsche et al., 2008).

The revelation that some of the Hg in atmospheric deposition originates from re-emission from land and ocean surfaces complicates its attribution to natural and anthropogenic sources since re-emission of earlier deposition represents a mixture of these. In 2010, an estimate of global re-emission of Hg^0 from terrestrial ecosystems was 2900 Mg a^{-1} (Smith-Downey et al., 2010), which was similar to re-emission estimates from oceans of about 2800 Mg a^{-1}, and larger than primary anthropogenic emissions of 2200 Mg a^{-1} (Selin et al., 2008).

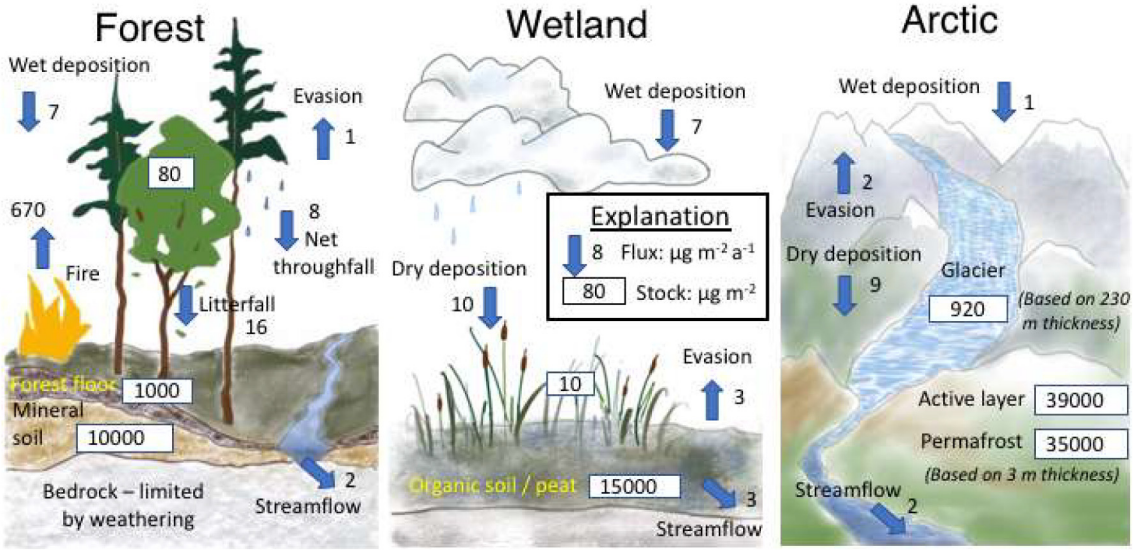


Fig. 1. Typical values for stocks and annual fluxes for THg in the northern temperate/boreal forest and wetland landscapes, and the Arctic landscape. The intent is to show relative magnitudes; actual values and even net flux directions may vary widely among ecosystems, and have high uncertainty. Belowground Hg stocks may consider different depths. Values are specific to that land cover or process. In the case of fire, the value is for a one-time release (not per year); the value is high because it is specific to burned area. Forest and Wetland panels are modified from Shanley and Bishop (2012). Arctic panel values are calculated from data in Schuster et al. (2002), Schuster et al. (2018), Obrist et al. (2017), Sonke et al. (2018), and the GLIMS glacial database (http://glims.colorado.edu/glacierdata/, accessed 2 November 2019). Artwork by Meghan Waskowitz.

2.2. Terrestrial Hg storage

Most Hg in soils is present as oxidized Hg bound to OM. In organic soils, concentrations in the range of 50–250 ng g^{-1} are common (Grigal, 2003). Concentrations are lower in mineral soils due to less OM, but the far greater mass of mineral soil generally makes it the largest pool of Hg in landscapes (Fig. 1, Table 1). Anthropogenic activity has increased Hg pools by a factor of circa three, with much of that increase near the soil surface (e.g. Alriksson, 2001).

Somewhat less than a tenth of the terrestrial pool is held in living vegetation. The concentration range has been reported as 10 to 40 μg kg^{-1} in foliage (Rasmussen et al., 1991; Siwik et al., 2009) and 30 to 90 μg kg^{-1} in roots (Schwesig and Krebs, 2003) at unpolluted sites. In the forested hemiboreal region, this range is somewhat narrower (Fig. 1). As growing vegetation is initially free of Hg, the incorporation of Hg into new vegetation each year is a dynamic part of the terrestrial pool (Smith-Downey et al., 2010).

Environmental archives such as peat, lake sediment and ice hold substantial stores of Hg from thousands of years of atmospheric deposition (Grigal, 2003), and also reveal details about the deposition history. Studies of specific Northern Hemisphere glaciers and ice sheets indicated Hg concentrations rivalling concentrations found in lake sediments and soils, but no global estimates existed prior to 2010 (Schuster et al., 2002; Fitzgerald et al., 2005).

2.3. Hg transformation

Within the terrestrial environment, two transformations are of particular importance, Hg methylation and demethylation. Interest in these counteracting processes relates largely to the extreme toxicity and bioaccumulation of MeHg. Some aquatic ecosystems receive a significant portion of their MeHg load from the terrestrial environment (Wiener et al., 2006; Brigham et al., 2009; Marvin-Dipasquale et al., 2009). High latitude regions with peatlands and fens are an area where Hg outputs from the terrestrial landscape into surface waters commonly contribute to levels of Hg in fish high enough to pose a risk to humans (Lindqvist et al., 1991; Rudd, 1995). MeHg concentrations are also elevated in rice from China (Horvat et al., 2003; Fu et al., 2008; Qiu et al., 2008), the Philippines (Appleton et al., 2006), and Tanzania (Taylor et al., 2005). Wildlife is likewise at risk from terrestrial Hg, from spiders to birds to the Florida panther (Barron et al., 2004; Cristol et al., 2008; Evers et al., 2008). The possibility of direct toxicity from Hg in the soil environment was also recognized (Bringmark and Bringmark, 2001).

By 2010, methylation had been documented for a phylogenetically diverse set of microbial guilds, linked to anoxic conditions where sulfate (SO_4^{2-}) reduction, iron reduction, and fermentation occurs (Fleming et al., 2006). These guilds included both iron- and S-reducing bacteria (Chadwick et al., 2006; Fleming et al., 2006), with S-reducing bacteria most often identified (Gilmour et al., 1992; Branfireun et al., 1999). The importance of atmospheric deposition of anthropogenic SO_4^{2-} in promoting methylation was demonstrated in wetland manipulations (Branfireun et al., 2001; Jeremiason et al., 2006). Sulfur also influences the speciation of inorganic Hg, and thus the availability of Hg for methylation, including the ability to pass through cell membranes (Schaefer and Morel, 2009). When SO_4^{2-} supply and SO_4^{2-} reduction are both elevated, the accumulation of sulfide may inhibit methylation by limiting Hg bioavailability for uptake by microbes (Gilmour et al., 1998; Benoit et al., 1999).

Demethylation, both biotic and abiotic, proceeds continually as well, but the rate varies less than for Hg methylation (Oremland et al., 1991; Marvin-DiPasquale et al., 2000). The balance of these processes is influenced by the availability of electron acceptors, electron donors (primarily OM), and the amount of Hg within the cell. The latter is a function of the concentration of Hg and its chemical speciation (Benoit et al., 2001). Methylation and demethylation occur simultaneously, and the fraction of MeHg in the pool of Hg on the solid phase has been shown to reflect the balance between these processes in some soils (Skyllberg et al., 2007).

2.4. Co-cycling of Hg with water and organic matter

The physical movement of Hg redistributes its different forms within the terrestrial ecosystem and ultimately removes some of it via gateways out from the terrestrial cycle. The transport and transformation of Hg in the terrestrial ecosystem links to both the hydrological cycle and the biogeochemical cycle of OM (Lindqvist et al., 1991). In the terrestrial environment, Hg moves with water in both particulate (HgP) and dissolved forms (HgD) along hydraulic energy gradients towards the surface water "gateway". Xylem sap also moves Hg(II) from soilwater up to needles (Bishop et al., 1998) and transpiration by vegetation may return Hg^0 from wetland soils to the atmosphere (Lindberg et al., 2002a, 2002b). Water also affects the form of Hg by contributing to suboxic conditions in saturated soils (Mitchell et al., 2008). In suboxic soils anaerobic microbial metabolism can promote Hg methylation and/or reduction, given appropriate electron donors, acceptors, microbes and Hg speciation (Marvin-diPasquale et al., 2000).

Saturated features such as peatlands, riparian fens and other wetlands are hotspots for Hg methylation. Organic surface soils in uplands can also be sources of MeHg, but they often lack "connectivity" to surface waters, making uplands less important for catchment outputs of MeHg. However, increased hydrologic connectivity and MeHg export can be promoted by forest harvest, beaver dams, periods of high flow or other disturbances (Bishop et al., 2009; Roy et al., 2009).

The ability of runoff to move particulate materials is a further hydrological influence on Hg export to surface waters. In some catchments, particulate Hg is the major form of Hg leaving the catchment in runoff (Shanley et al., 2008), whereas in other catchments, particulate Hg is only a minor part of the export (Pettersson et al., 1995). In studies prior to 2010, the proportion of particulate Hg export varied from <1% to 80% (Table S1).

Water moves much more quickly through a catchment to a stream than Hg does. In the small upland catchment in the humid, nemoboreal zone studied by the METAALICUS project, only 1% of the isotopically marked Hg deposited on the uplands reached the lake downslope within three years (Harris et al., 2007). This long lag between Hg inputs from the atmospheric "gateway" to the terrestrial environment and outputs from the surface water "gateway" reinforces concerns from simpler mass balance considerations (low catchment Hg fluxes relative to large Hg storages) that reducing atmospheric emissions will take generations to markedly reduce the loading of Hg from terrestrial catchments to surface waters (Meili et al., 2003). One short-term benefit of Hg deposition reductions is direct lowering of Hg inputs to surface waters (Blanchfield et al., 2004; Munthe et al., 2007), which has an immediate benefit to fish (Harris et al., 2007).

Hg in soils is strongly bound to OM, in particular to the sulfur in thiol groups (Skyllberg et al., 2003; Ravichandran, 2004). Nearly all of the dissolved and particulate Hg moving with water through the terrestrial environment is bound to OM (Bishop et al., 1995; Mitchell et al., 2008; Åkerblom et al., 2008), with the exception of dissolved elemental Hg, which is a minor, unreactive component. The linkage of Hg to OM begins in the atmosphere, where organic matter may promote oxidation of Hg and its subsequent washout in precipitation (Lin et al., 2006). The Hg-OM linkage remains in streamwater leaving the terrestrial environment (e.g. Babiarz et al., 1998; Nelson et al., 2007). Physical coupling of the terrestrial Hg and OM cycles was incorporated in the GEOS global model (Smith-Downey et al., 2010).

The correlation of MeHg to dissolved organic carbon (DOC) is more variable than that of total Hg and DOC from site to site (Brigham et al., 2009). Some areas exhibit a positive MeHg - DOC relationship (e.g. Shanley et al., 2008; Dittman et al., 2009), but other areas, i.e. forested streams of Fennoscandia, show the reverse (Pettersson et al., 1995; Sørensen et al., 2009). The negative correlation may reflect a rate

limitation in the net production of MeHg from soils when flushed rapidly, as during spring snowmelt. This limitation would not limit the co-transport of total Hg (THg) with OM (Bishop and Lee, 1997).

The combustion of forest floor OM by fire reduces the store of Hg (Friedli et al., 2003). At Acadia National Park in Maine, a 1938 forest fire was cited as the reason for a 50% lower THg flux relative to an unburned control catchment (Nelson et al., 2007). Combustion releases Hg primarily as Hg^0, though fuel moisture can increase particulate-bound Hg (Obrist, 2007).

2.5. Human influences on the Hg cycle

Increased levels of Hg in the environment are the most direct human influence, resulting from atmospheric emissions or from contaminated sites. Where rice is grown near areas contaminated by Hg mining, paddy soils are an important site for MeHg production, and consumption of rice from such sites threatens human health (Feng et al., 2008). An extensive human influence on surface waters is forest harvest, which has varying degrees of impact on the export of Hg. Several studies have found that forestry operations resulted in elevated THg and MeHg concentrations in downstream waters (Porvari et al., 2003; Munthe and Hultberg, 2004; Munthe et al., 2007; Skyllberg et al., 2009) and biota (Garcia and Carignan, 1999; Garcia and Carignan, 2000; Desrosiers et al., 2006; Garcia et al., 2007). In contrast, however, several early studies detected little or no forest harvest effects on THg and MeHg in streamwater (Allan et al., 2009; Sørensen et al., 2009) and fish (Rask et al., 1998). Controlled burning in association with harvest was also implicated in Hg methylation hotspots and trophic transfer, but with differing impacts on the biota (Caldwell et al., 2000; Garcia and Carignan, 2000; Kelly et al., 2006).

Forest harvest reduces evapotranspiration and canopy interception (Murray and Buttle, 2003), thereby increasing groundwater levels and soil moisture. The role of increased wetness in mobilizing Hg and promoting methylation was evident from increased Hg concentrations in fish after impoundments created reservoirs (Tremblay et al., 1998). The FLUDEX experiment demonstrated that Hg and MeHg mobilization was not due to wet areas becoming wetter, but rather due to the initial inundation of previously dry areas (Hall et al., 2005, 2009). Similarly, recent beaver ponds show greater methylation than older ponds (Roy et al., 2009).

An estimated 8–23% of all Hg in the fish of Sweden's forest landscape was mobilized into watercourses by forest harvest operations (Bishop et al., 2009). Skyllberg et al. (2009) suggested that elevated MeHg concentrations in streams after forest harvest was sourced mainly from new MeHg formation and less from mobilization of pre-existing MeHg. Best management practices (BMP) for forestry that minimized riparian disturbance were posited as effective countermeasures (Sørensen et al., 2009). The lack of BMPs was identified as a factor increasing MeHg in runoff when forestry machinery crossed a stream (Munthe and Hultberg, 2004).

Sulfur deposition, enhanced by anthropogenic S emissions, was another human influence suspected of increasing methylation in peatlands due to the role of sulfur-reducing bacteria (SRB) in net methylation (Gilmour and Henry, 1991). Sulfate serves as a terminal electron acceptor for SRB (Branfireun et al., 2001; Mitchell et al., 2008). Experimental S addition to wetlands stimulated MeHg production (Gilmour et al., 1998; Branfireun et al., 1999; Jeremiason et al., 2006). Landscape scale effects were harder to discern, although one study linked decreasing Hg in fish to declining S deposition (Drevnick et al., 2007).

Potential implications for future cycling of Hg in a changing climate were initially drawn from findings in studies that captured short-term variability in climate-relevant conditions (flooding/drought cycling, fire, temperature increases/thawing etc.). For example, increased mobilization of Hg from terrestrial storage has been attributed to: (1) thawing of mires in northern Sweden (Klaminder et al., 2008); (2) extreme wet/dry cycling impacts on peat decomposition in a Spanish bog (Cortizas et al., 2007); and (3) forest fires (Nelson et al., 2007; Wiedinmyer and Friedli, 2007; Friedli et al., 2009). Elevated MeHg concentrations in surface waters have also been predicted as a result of future increases in flooding (Balogh et al., 2006). Browning of surface waters, (increased OM, Monteith et al., 2007) has led to speculation that further increases in OM export will increase OM-associated Hg in surface waters (Demers et al., 2010).

Numerous site-specific studies have focused on Hg contamination from point sources, including Hg mining, gold mining, large-scale precious metal production, municipal wastewaters, chlor-alkali production, and other chemical manufacturing processes. These impacts are highly variable (see citations within Table S1, Kocman et al., 2013). Artisanal and small-scale gold mining (ASGM), which use inefficient amalgamation techniques, release Hg. However, as of 2010, ASGM sites had yet to be well-characterized (Veiga et al., 2006; Telmer and Veiga, 2009).

Point-source contaminated environments were the first type of systems in which watershed modeling focused on the transfer of Hg from the terrestrial to the aqueous environment (Carroll et al., 2000; Zagar et al., 2006). Early models of non-point source contaminated systems focused on transport and transformations within aqueous bodies (lakes, ponds and later rivers) (see citations within Knightes and Ambrose, 2007, Knightes et al., 2009). The early model results indicated that an improved understanding of mercury loading from the terrestrial environment, as well as erosion and sediment delivery, were critical to advance process-based model prediction.

3. Recent advances: influences on terrestrial Hg cycling

The preceding overview of terrestrial Hg cycling provides background for the next two sections which explore developments since 2010 on terrestrial Hg cycling. Section 3 addresses the major human influences on that cycling: land-use, SO_4^{2-} pollution and climate.

3.1. Land-use, in particular forest management

Examples of increased Hg export from land to water after forest harvest, the large spatial extent of forestry and expectations that forests will be utilized more intensively in the future have all focused attention on the coupling between Hg and forest management. Since 2010, studies of forestry effects on Hg have more than doubled to over twenty. The new findings vary substantially with respect to forest harvest effects on MeHg concentrations in water (increases of 0–325%) as well as Hg and/or MeHg concentrations in biota (increases of 0–80%) (Fig. 2). Effects on MeHg loads were generally more pronounced due to the increase in discharge after logging. The site-dependent variability highlighted during recent years emphasizes the value of process-based understanding of forestry-induced changes on Hg methylation and mobilization.

Kronberg et al. (2016b) found that the combination of water saturation and availability of electron donors elevated MeHg formation in new discharge areas created when formerly well-drained podzols were inundated by increased groundwater tables. Water-logged soils in driving tracks (Braaten and de Wit, 2016) and water-filled cavities formed after removal of stumps for biofuels (Ukonmaanaho et al., 2016; Eklöf et al., 2018) have been identified as hotspots of Hg methylation, with microbial analyses supporting this interpretation in the latter two studies. Stump harvest may even raise MeHg in groundwater decades later (Magnusson, 2017).

Catchment topography can influence the response to forest harvest. Since the soils outside of the riparian zone may act as a source of MeHg, hilly catchments with narrow riparian zones are at a higher risk of elevated MeHg in runoff than flatter landscapes, where the newly formed discharge area will be located further away from the stream (Kronberg et al., 2016a). While the likelihood of MeHg export after harvest is lowest in the flattest terrain and increases with hilliness, there is a point where increasing catchment steepness precludes the

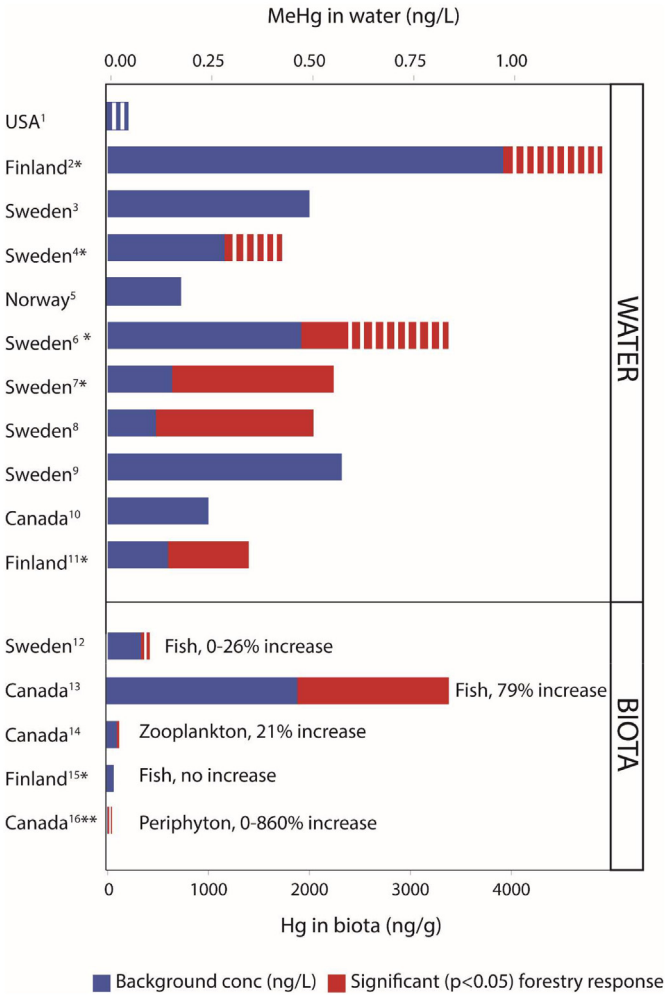


Fig. 2. Background concentrations and significant ($p < 0.05$) forest harvesting responses of MeHg concentrations in water and Hg concentrations in biota from various studies in the Northern Hemisphere. Striped bars represent ranges of background concentrations or forestry responses between different catchments included in the original studies. Only significant ($p < 0.05$) forestry responses on MeHg concentrations, not fluxes, are given in the figure. Background concentrations in biota are highly variable over the range of trophic levels. For the sake of visibility, significant ($p < 0.05$) increases in biota concentrations are stated as a percentage value in the figure. Data originate from the following publications: [1] Eckley et al., 2018, [2] Ukonmaanaho et al., 2016, [3]Kronberg et al., 2016a, [4]Eklöf et al., 2014, [5]de Wit et al., 2014, [6]Eklöf et al., 2012b, [7]Skyllberg et al., 2009, [8]Munthe et al., 2007, [9] Sørensen et al., 2009, [10]Allan et al., 2009, [11]Porvari et al., 2003, [12]Wu et al., 2018, [13]Garcia and Carignan, 2000, [14] Garcia and Carignan, 1999 (numbers in the figure are based on Garcia and Carignan, 1999: Garcia et al., 2007 use the same setup of sample sites showing that the effect remained for several years after logging), [15]Rask et al., 1998, and [16]Desrosiers et al., 2006.

development of saturated, suboxic conditions needed for methylation, such as in the steep terrain of coastal mountain areas in Oregon, USA (stream section gradients ranging from 4 to 20%, Eckley et al., 2018). Streamwater MeHg concentrations in this study were less than detection limit (<0.05 ng/L) both before and after forest harvest. Catchments showing the highest response to forest harvest in terms of topography may thus be intermediate between the steep catchments in Oregon (Eckley et al., 2018) and the flatter, lowland catchments in Scandinavia and Finland (Eklöf et al., 2016; Kronberg et al., 2016a; Ukonmaanaho et al., 2016).

Hydrological connectivity may play an important role in determining the effects of more intensive forest management methods to produce biofuels, such as whole-tree harvest and stump harvest (Eklöf et al., 2018). Removal of logging residues and stumps can create additional soil disturbance, causing erosion and promoting superficial hydrological pathways (Walmsley and Godbold, 2010). However, in some studies where stump harvest promoted MeHg formation in water-filled stump holes (Ukonmaanaho et al., 2016; Eklöf et al., 2018) stream MeHg concentrations did not increase (Eklöf et al., 2012b; Eklöf et al., 2013; Ukonmaanaho et al., 2016). This might be due to limited hydrological connectivity between Hg methylation hotspots and streams (Eklöf et al., 2018).

Several studies with minimal Hg response to forest harvest have attributed this positive outcome to the use of Best Management Practices (BMP) including retention of growing forest in riparian buffers, soil protection for heavy forestry vehicles, and/or harvesting during winter conditions when soil disturbance is less likely (Sørensen et al., 2009; Eklöf et al., 2014; Eckley et al., 2018). However, a Norwegian forest harvest that created extensive wheel ruts and soil disturbance on non-frozen soils yielded no MeHg response in runoff water (de Wit et al., 2014).

Responses of aquatic Hg bioaccumulation to forest harvest needs to consider not only MeHg changes in runoff but also the changes in nutrients that impact aquatic growth, and thereby food-web structure. Wu et al. (2018) found that fish Hg concentrations increased after forest harvest but with a large year-to-year and lake-to-lake variation (−14% to +121%). Decreased MeHg in herbivorous stoneflies after forest harvest in Norway may have resulted from biodilution caused by increased nutrient loadings after harvest (de Wit et al., 2014).

Forest harvesting not only impacts runoff Hg but can also increase Hg evasion to the atmosphere, in particular by changing emission rates and deposition. Canopy removal increases solar radiation reaching the forest floor (Carpi and Lindberg, 1997; Gustin et al., 2002), air and soil temperature (Lin et al., 2010; Kim et al., 2012) and soil moisture (Gustin and Stamenkovic, 2005). More Hg in the soil of old-growth forests compared to second-growth forests in southwest Ohio, USA (Gamby et al., 2015), indicated a net loss of Hg from the forest floor after forest harvest by runoff and/or Hg emission to the atmosphere. Mazur et al. (2014) reported that forest harvest changed net Hg deposition (~150 ng m^{-2} d^{-1} in the unharvested control during the growing season) to net emission. The highest emission rates were from sites with logging residue extraction for biofuels (net emission ~200 ng m^{-2} d^{-1}). Increased photoreduction of Hg in the less shaded forest floor after debris removal may have facilitated Hg emission.

In a changing climate, rising temperatures in high latitude forests shorten snow cover duration, causing forest operations to occur more often on soils unprotected by snow or frost. Where precipitation increases, the extent of water-logged soils after forest harvest will increase, creating more sites with high MeHg. Such climate related challenges facing the forestry sector make understanding and management of water quality issues a priority.

Primary changes in knowledge since 2010:

- The number of published studies has doubled since 2010, including more targeted, process-based studies, but the range of Hg response to forestry remains large.
- Landscapes of intermediate steepness show the greatest response to harvest, through creation of new methylation sites and hydrological connectivity to streams.
- Post-harvest increases in stream MeHg are driven by new MeHg formation in previously well-drained areas.
- Forestry Hg impacts can be mitigated by Best Management Practices, but empirical evidence remains weak.

3.2. Sulfate deposition on wetland systems

Connections between the sulfur cycle and the transformation, transport and bioaccumulation of Hg have been elucidated by a number of recent studies. Archived chironomids linked declining S deposition to decreased Hg in these aquatic insects (Braaten et al., 2020). In peatlands, new lines of evidence support the key role of SRB and other microbial guilds in wetland Hg methylation. Inhibition experiments and targeted gene analyses, showed that SRB are dominant Hg methylators in

Amazonian wetlands during the dry season (Lazaro et al., 2018) and Sphagnum moss mats (Yu et al., 2010). Increased SO_4^{2-} deposition can also alter microbial communities in favor of both SRB abundance and increased MeHg production, such as increases in abundance of *Desulfovibrio*-like bacterial (Yu et al., 2010) and Deltaproteobacterial communities, within which most SRB exist (Strickman et al., 2016).

Given the new awareness that broad microbial guilds beyond SRB have potential Hg methylation capabilities (Gilmour et al., 2013; Podar et al., 2015), it seems plausible that the direct role of SRB in methylating Hg is variable and still relatively poorly characterized. To date, few studies have simultaneously and directly measured MeHg production and methylation specifically by SRB. Rapidly growing access to molecular methods (e.g., Schaefer et al., 2014), promise new discoveries in the microbial ecology of SRB and their role in Hg methylation in wetlands, including microbial syntrophy, which may explain the intricacy of Hg methylation interactions between SRB and other microbial guilds (Yu et al., 2010; Gilmour et al., 2013).

The stimulation of Hg methylation by added SO_4^{2-} in field studies (e.g. Bergman et al., 2012; Coleman Wasik et al., 2012) has further supported the well-established role of SRB as Hg methylators (Gilmour et al., 2013). The manipulation of atmospheric SO_4^{2-} inputs in these newer studies demonstrated more clearly that MeHg concentrations follow changes in SO_4^{2-}, with time lags between decreased SO_4^{2-} deposition and significantly decreased MeHg concentrations in peat and biota (e.g., mosquito larvae) on the order of a few years (Coleman Wasik et al., 2012). Stimulation of MeHg production by SO_4^{2-} has also been observed in engineered and wastewater treatment wetlands (Zheng et al., 2013; Oswald and Carey, 2016; McCarter et al., 2017), as well as at the broader landscape scale (Drenner et al., 2011; Gabriel et al., 2014). In the Florida Everglades, the connection between Hg and S has direct policy implications with respect to the use and control of SO_4^{2-} as an agricultural fertilizer (Corrales et al., 2011; Orem et al., 2011).

The connection between changes in SO_4^{2-} deposition and MeHg production can be complicated by impacts on Hg evasion (Fritsche et al., 2014), OM quality, and climatic factors, including drought and temperature (Åkerblom et al., 2013). Water table fluctuations can augment MeHg production in wetlands through the regeneration of SO_4^{2-} via transiently oxidized conditions (Bergman et al., 2012; Feng et al., 2014; Haynes et al., 2017b). Large water table fluctuations often occur during and after drought, which can extend the timeframe of SO_4^{2-}-stimulated MeHg production, though the effect appears to slow over time due to the gradual incorporation of SO_4^{2-} into more recalcitrant organic S pools (Coleman Wasik et al., 2012, 2015). The role of OM quality was identified in rice paddy methylation (Windham-Myers et al., 2014), and the observation that new beaver ponds promote MeHg in water (Levanoni et al., 2015).

Stimulation of MeHg production in wetlands or wetland-dominated landscapes by SO_4^{2-} does not continue beyond an optimum intermediate SO_4^{2-} level, which was first hypothesized by Gilmour and Henry (1991) to be in the 20–50 mg L^{-1} range. High SO_4^{2-} concentrations and/or interactions between S and Fe cycles may influence the duration of elevated MeHg production (Ulrich and Sedlak, 2010; Berndt and Bavin, 2012; Marvin-Dipasquale et al., 2014; Hoggarth et al., 2015; Johnson et al., 2016). In managed wetlands, coagulants such as $Fe_2(SO_4^{2-})_3$ have been used to reduce Hg bioavailability by promoting precipitation and settling of particulate Hg complexes, leading to reductions in fish Hg concentrations (Ackerman et al., 2015). How sulfide accumulation impacts methylation is also complicated by multiple potential inorganic and organic reduced S forms, some promoting bioavailability and some inhibiting Hg uptake (Nagy et al., 2011; Graham et al., 2012; T. Zhang et al., 2012b). Generally, MeHg production enhancement by SO_4^{2-} inputs may be greatest in wetlands with poor to intermediate nutrient levels and biogeochemical SO_4^{2-} limitations (Tjerngren et al., 2012; Johnson et al., 2016), though this may be confounded by plant rhizosphere-microbial interactions and seasonal variations in some wetlands (Windham-Myers et al., 2009; Alpers et al., 2014).

Finally, S cycling can strongly influence Hg solubility and transport within and from wetlands. Low molecular mass thiols, thiols associated with natural OM and nanoparticulate β-HgS affect the solubility of Hg species and possibly the mobility of Hg in hydrological flows or by evasion (Poulin et al., 2016; Liem-Nguyen et al., 2017). Hofacker et al. (2013) suggested that Hg-containing Cu nanoparticles and colloidal transport may be an important mechanism for Hg transport from contaminated riparian wetland soils. Inputs of SO_4^{2-} to wetlands can also result in enhanced soil OM decomposition, which increases soil water concentrations of Hg(II) and MeHg, and possibly impacts downgradient transport (McCarter et al., 2017; Myrbo et al., 2017).

Primary changes in knowledge since 2010:

- The multiple interacting ways through which S influences Hg speciation, bioavailability, and mobility have become clearer.
- Evidence has mounted that SO_4^{2-} additions change bacterial community composition and sustain methylation, though S^{2-} buildup inhibits methylation beyond a certain point.
- An expanded array of microbial guilds is now known to participate in Hg methylation, but SRB remain important, possibly through syntrophy.

3.3. Climate including fires and permafrost thawing

Climate impacts on terrestrial Hg cycling are potentially many-fold. Ecosystem Hg loading, processing rates, lateral transport to aquatic end points, and methylation in particular all are climate-sensitive. IPCC (2018) predicts a highly likely increase in global temperature of 1.5 °C, where warming over land will be higher than over sea and high-latitude ecosystems will likely experience the most warming. Associated changes in precipitation will also be greater in mid- to high-latitude ecosystems, particularly with regard to extremes, both droughts and intense, episodic rainfall. Direct impacts of climate change, related to heat and precipitation, are often distinguished from indirect impacts. Examples of indirect effects, where climate-related ecosystem responses can influence terrestrial Hg cycling in complex ways, include changes in terrestrial productivity, land-atmosphere exchange, and forest fire frequency. A third category of impact is through societal adaption to climate change and mitigation measures, for example intensification of forestry, which could potentially increase Hg release to the environment (cf. Section 3.1).

3.3.1. Atmosphere-soil Hg interactions: circulation patterns, deposition, and re-emission

Large-scale weather patterns impacted wet Hg deposition to forests in both New England, USA, and in the central Himalayas (Mao et al., 2017), resulting in higher Hg deposition during periods of high rainfall, and lower atmospheric Hg concentrations indicative of atmospheric scavenging. Net ecosystem loading, however, also depends on dry deposition and surface emission of Hg.

There is no general consensus about whether terrestrial ecosystems function as a source or sink of atmospheric Hg (Fritsche et al., 2008; Agnan et al., 2016), but re-emission of Hg from soils is usually higher at higher temperatures (Yang et al., 2019; MacSween et al., 2019), possibly because of evaporation and increased microbial activity that reduces Hg^{2+} to Hg^0 (Ma et al., 2018). Yang et al. (2019) observed decreases in soil Hg retention in three different climate manipulation experiments at Hubbard Brook, NH; artificial soil warming, drought (via precipitation exclusion), and simulated ice storms. Vegetation uptake may explain seasonal variation in atmospheric Hg, which suggests that higher terrestrial productivity increases the Hg sink function of vegetation (Jiskra et al., 2018). Greening and browning of the Arctic (Phoenix and Bjerke, 2016) will also alter the areal extent of woody

vegetation interacting with Hg. Section 4.1.3 provides further discussion of climate change influences on land-atmosphere Hg exchange.

3.3.2. Fire

Wildfire contributes to global contamination through atmospheric release and influences Hg cycling in soils, surface waters, sediments, and biota. Current climate predictions indicate global wildfire emissions of Hg will increase 28% by 2050 (Kumar et al., 2018). Recent experiments by Kohlenberg et al. (2018) show that peats release a large percentage of Hg as Hg^0 when heated, suggesting that increased burning at northern latitudes will be a large source of atmospheric Hg. In addition to large Hg^0 emissions, recent global transport models considering particulate-bound Hg emissions from wildfire predict significant increases in deposition in the polar and boreal regions (Simone et al., 2017; Fraser et al., 2018). To assess such model predictions, spatial data on the fate of Hg after a fire are critical. Kolka et al. (2017) demonstrated regional scalability of remotely sensed burn severity to quantify Hg losses from soil during fire.

Post-fire soil cycling of Hg varies with soil characteristics, vegetation, fire severity, and post-fire rainfall. Fire severity dictates release of Hg from soil, and even low severity fires may release significant amounts (Campos et al., 2015; Kolka et al., 2017). Soil Hg concentrations have increased in burned soils following rain (Abraham et al., 2018). It is unclear what causes these increases, but possibilities include exposure of underlying soil, scavenging of atmospheric Hg by soil, or deposition of new litter material (e.g. Engle et al., 2006; Burke et al., 2010).

Following burning, increased surface flow and erosion may transport Hg-containing sediments into surface waters and reservoirs, though only a few studies have quantified aquatic transport of Hg following fire. Jensen et al. (2017) evaluated post-fire Hg transport using paired burned/unburned watersheds and found that burning increased HgP:TSS (Total Suspended Sediment) by an order of magnitude for a range of flow conditions. Thus, in systems where burning was severe enough to cause erosion and increases in TSS, Hg increases can be significant. The fire effects diminished after eight months (Jensen et al., 2017).

Early studies identified burned areas as hotspots of methylation and trophic transfer (cf. Section 2.5). More recent findings in watersheds with low- to moderate-severity burning have not found elevated Hg in the food web (Moreno et al., 2016; Riggs et al., 2017). These contrasting results suggest that post-fire methylation is likely governed by site-specific characteristics.

Patel et al. (2019) studied the long-term effects of fire on Hg cycling by comparing a watershed that burned severely in 1947 to an unburned control in Acadia, Maine. Hg in the burned watershed followed OM recovery in the soil. Mercury export from both watersheds was strongly correlated with DOC. The Hg:DOC was similar in both watersheds, but the burned watershed exported less Hg. Interestingly, MeHg was significantly higher in the O-horizon of the burned watershed, suggesting that fire may provide a lasting boost to MeHg production in soils.

3.3.3. Process rates: methylation, organic matter and erosion

Increasing global temperatures will facilitate the formation of MeHg, which is mediated by temperature-dependent biotic processes (cf. Section 2.3). Additional factors that constrain net MeHg production are availability of Hg and substrates (SO_4, labile OM), soil moisture and redox conditions, and losses from demethylation – all sensitive to water table regime (Haynes et al., 2017a, 2017b). In observations from porewaters (Stern et al., 2012; Creswell et al., 2017) and in a soil warming experiment in Arctic soils (Yang et al., 2016), microbial MeHg production appeared to be more temperature-sensitive than microbial demethylation, suggesting a net increase in MeHg from warming. Multi-factorial manipulations of peatland plots including warming, and S and N addition treatments showed that the S addition effect on methylation disappeared when combined with warming. Warming alone did not affect porewater MeHg (Åkerblom et al., 2013).

Climatic controls on Hg may act through its close link with the C cycle. Experimental soil OM response to climate warming suggest that the pool of labile SOM is rather small and can be rapidly depleted upon warming (Melillo et al., 2002; Knorr et al., 2005; Tang and Riley, 2015), leading to limited Hg mobilization. A notable exception is the case of permafrost thawing, further discussed in Section 3.3.4. Additionally, the browning of surface waters in recent decades, related to reduced acid deposition (Monteith et al., 2007) and climate wetting (de Wit et al., 2016), suggests that soil water DOC concentrations have increased. This has implications for transport of Hg which is bound to DOC, and the influence of DOC on methylation/demethylation (Ravichandran, 2004). Surface water DOC is associated with lower Hg bioaccumulation in fish (Braaten et al., 2018; Wu et al., 2019), possibly reflecting lower bioavailability of Hg through Hg-OM complexation in lakes. The progressive decrease in the Hg:C ratio as water moves from precipitation to soil water to groundwater to runoff suggests a mechanistic link between Hg and C in terrestrial Hg cycling (Demers et al., 2013; Åkerblom et al., 2015).

Climatic shifts in precipitation patterns are likely to promote land-water transport of both MeHg and Hg, leading to higher exposure of the aquatic food web to Hg. Lateral Hg transport is mostly driven by precipitation and hydrologic pathways, but also by disturbance (see Section 3.1). In catchments with bare soils, particulates are an important transport vector of Hg especially during heavy rain events (Baptista-Salazar et al., 2017; Saniewska et al., 2018). Particulates can dominate Hg in runoff from vegetated areas as well (cf. Sections 2.4 and 4.3). Coastal erosion, which is increasing because of more intense rainfall and heavier storms, delivers significant amounts of Hg to marine environments in the Baltic region (Kwasigroch et al., 2018).

3.3.4. Permafrost and glaciers

Permafrost (Schuster et al., 2018) and glacial ice (Schuster et al., 2002) contain significant stores of Hg from thousands of years of natural atmospheric deposition. As the Earth continues to warm, and at a faster rate at higher latitudes, permafrost is thawing (Smith et al., 2010) and glacial ice is melting (Kääb et al., 2007). Warming at northern high latitudes is poised to release new Hg stores from: 1) the large cover of organic-rich (and thus Hg-rich) permafrost soils (Obrist et al., 2017) and glacial ice (Boutron et al., 1998; Schuster et al., 2002; Fain et al., 2008); 2) peaks in atmospheric Hg during summer resulting from atmospheric mixing with ozone that enhance Hg deposition (Sonke and Heimbürger, 2012); 3) processes unique to the Arctic, such as the Polar Sunrise (Lindberg et al., 2002a, 2002b; Fitzgerald et al., 2005; Berg et al., 2008) that trigger Hg deposition; and 4) increases in northern boreal forest fires that release Hg into the atmosphere (Rothenberg et al., 2010; Homann et al., 2015). These processes and conditions unique to the Arctic and Subarctic exert a strong influence on the global Hg cycle.

Model projections estimate a 30–99% reduction in Northern Hemisphere permafrost by 2100, assuming anthropogenic greenhouse gases emissions continue at current rates (Koven et al., 2013). Higher frequencies of freeze-thaw events and thawing permafrost will also deliver more Hg to rivers through increased erosion (Olson et al., 2018). Stores of Hg in Northern Hemisphere permafrost have been estimated to contain 1656 ± 962 Gg Hg, of which 793 ± 461 Gg Hg is frozen in permafrost with the rest residing in the active layer (Schuster et al., 2018). Thus permafrost soils store nearly twice as much Hg as all other soils, the ocean, and the atmosphere combined. Global stores of Hg in glacial ice have yet to be quantified, but site-specific studies document high Hg concentrations and stores there as well (Schuster et al., 2018, Fig. 1).

Most of the world's fisheries are in the Northern Hemisphere, and they feed billions of people. Moreover, indigenous human cultures in the Arctic and Subarctic are dependent on the region's fish for their subsistence lifestyle. Continued warming will likely promote or accelerate the mobilization of Hg from glacial melt and permafrost thaw,

increasing concentrations, exports, yields, and potential Hg methylation at least in the Northern Hemisphere, if not worldwide.

Primary changes in knowledge since 2010:

- Climate-warming induced intensification of water and organic matter cycles has led to a parallel intensification of Hg cycling.
- Global warming is widely anticipated to increase terrestrial Hg net methylation, but experimental results have been mixed.
- Thawing permafrost and increased wildfires are already accelerating the release of Hg to the atmosphere and receiving waters, with the potential to release much more.

4. Recent advances: terrestrial cycle gateways

Having examined advances during the last decade in understanding human influences on the terrestrial Hg cycle, the review moves on to examine three "gateways" to terrestrial cycling of relevance to the impact of Hg pollution on human health and biota: land-atmosphere exchange, food derived from the terrestrial environment, and delivery of Hg in runoff to surface waters.

4.1. Bi-directional land-atmosphere exchange of Hg

Compared to natural levels before 570 BP anthropogenic emissions have increased the global pool of atmospheric Hg by about 450% (Outridge et al., 2018; UNEP, 2019). The current estimate for the total atmospheric Hg pool is between 4400 and 5300 Mg (Amos et al., 2014; Horowitz et al., 2014; Zhang et al., 2014; Streets et al., 2019). Subsequently, the greater atmospheric Hg pool has resulted in an overall increase (~300%) in both wet and dry deposition of Hg to the terrestrial environment. Anthropogenic activities emit between 2000 and 2820 Mg Hg a^{-1} into the atmosphere (Outridge et al., 2018; UNEP, 2019). For Europe, anthropogenic Hg emission estimates have been challenged by a top-down approach based on ambient Hg measurements. The estimated annual European Hg^0 emissions (89 ± 14 Mg a^{-1}) were 17% larger than the inventory estimates, which was within the stated uncertainties (Denzler et al., 2017). Top-down estimates of anthropogenic Hg^0 emissions in Asia, however, were 18–221% larger than inventory assessments (Song et al., 2015). Inventory emission estimates have also been questioned because of a mismatch between the recent rising trends (2010–2015) in anthropogenic emissions (+22%, UNEP, 2019) and decreases in atmospheric Hg^0 concentrations and Hg wet deposition estimates (~30–40%) at Northern Hemisphere background sites since 1990 (Y. Zhang et al., 2016b).

The global atmospheric Hg pool is fed not only by direct anthropogenic emissions (2000–2820 Mg a^{-1}), but also by natural sources, primarily volcanic emissions of circa 500 Mg a^{-1} (0.1–1000 Mg a^{-1}) and terrestrial re-emission of naturally and anthropogenically-derived Hg from vegetation, soil and snow of circa 1000 Mg a^{-1} (Outridge et al., 2018; UNEP, 2019). Most recent estimates of average annual land Hg^0 emissions range between 607 Mg a^{-1} (Agnan et al., 2016) and 1360 Mg a^{-1} (Agnan et al., 2016; Song et al., 2016; Horowitz et al., 2017). Thus, the land-atmosphere exchange of Hg^0 represents a major control on how fast the environment will recover from anthropogenic Hg pollution (Lyman et al., 2020). While wet deposition of Hg(II) is relatively well quantified, Hg^0 dry deposition and land Hg^0 re-emission estimates remain uncertain. This uncertainty is borne out by a recent review of 132 direct Hg^0 flux measurement studies where the central 25% of the distribution ranged between −513 and 1653 Mg a^{-1} (Agnan et al., 2016). Hg^0 dry deposition is not only the dominant pathway of atmospheric Hg deposition to terrestrial surfaces, it is also a global "pump" that drives seasonal patterns in global Hg movement (Jiskra et al., 2018; Sonke et al., 2018).

4.1.1. Deposition of atmospheric Hg to the terrestrial ecosystems

4.1.1.1. Wet deposition. Hg in wet deposition has been monitored by networks in North America, Europe and China as well as the 17 stations of the Global Mercury Observation System (GMOS) project, which ran from 2011 to 2015 (Sprovieri et al., 2017). The Northern Hemisphere results depicted a north to south increase in Hg wet deposition, ranging from <1 μg m^{-2} a^{-1} in Russia to >10 μg m^{-2} a^{-1} in Slovenia (Sprovieri et al., 2017). The European Monitoring and Evaluation Programme (EMEP) network shows a N-S gradient in total Hg deposition from <5 μg m^{-2} a^{-1} in Scandinavia to >40 μg m^{-2} a^{-1} in parts of southern Europe (http://en.msceast.org, accessed 20-01-10). A N-S Hg deposition gradient is also present in Alaska, from 0.2 μg m^{-2} a^{-1} in the northern tundra (68.6° N) (Obrist et al., 2017) to 4.8 μg m^{-2} a^{-1} in the south (57.7° N) (Pearson et al., 2019). Elsewhere in the USA, during 2011–2017, the Mercury Deposition Network (MDN) maps for the USA showed relatively low Hg wet deposition of 2–5 μg m^{-2} a^{-1} in the West (albeit with hotspots in high-precipitation and natural Hg source areas), increasing to 5–10 μg m^{-2} a^{-1} in the Northeast and upper Midwest, 10–15 μg m^{-2} a^{-1} in the remaining Midwest and Mid-Atlantic, and 15–20 μg m^{-2} a^{-1} along the Gulf Coast and Florida (http://nadp.slh.wisc.edu/, accessed 2019-11-03). Shanley et al. (2015) reported wet Hg deposition of 27.9 μg m^{-2} a^{-1} at an unpolluted site at 18° N in Puerto Rico. As in the EMEP and GMOS networks, an increasing north to south west Hg deposition gradient is evident. Other tropical sites, including Mt. Ailao in southwest China (5.4 μg m^{-2} a^{-1}, Zhou et al., 2013) and the Sisal station in Mexico (7.4 μg m^{-2} a^{-1}, Sprovieri et al., 2017), do not fit the trend of higher wet Hg deposition at lower latitudes. Wet Hg deposition measurements in tropical and subtropical latitudes are scarce, and more are needed to clarify this pattern. Also, the Southern Hemisphere has relatively few observational data.

Along with a contemporary global decrease of atmospheric Hg^0 concentrations in North America and Western Europe (1–2% a^{-1}, Streets et al., 2019), Hg wet deposition decreased by about 1.5% and 2.2% a^{-1}, respectively, between 1990 and 2013 (Y. Zhang et al., 2016b). However, strong emission sources in South and East Asia compensated for this decrease in Hg deposition on a global scale (Zhang et al., 2016c). Projected future changes in the Hg cycle with constant Hg emissions between 2015 and 2050 indicate a global increase in Hg deposition of about 30% until 2050 (Amos et al., 2013). The increase occurs because anthropogenic emissions add Hg to the atmospheric pool faster than it can be sequestered into terrestrial and marine reservoirs.

4.1.1.2. Dry deposition. In comparison to Hg wet deposition, dry deposition fluxes of Hg^0 and Hg(II) are still poorly constrained (Wright et al., 2016; Cheng and Zhang, 2017). Atmospheric dry deposition of Hg^0 is typically derived from net Hg^0 exchange measurements performed with dynamic flux chambers and micrometeorological techniques (Sommar et al., 2013; Zhu et al., 2015a, 2015b; Zhu et al., 2016; Osterwalder et al., 2018). Based on net Hg^0 deposition events, inferred dry deposition velocities over vegetated surfaces and wetlands ranged from 0.1 to 0.4 cm s^{-1}. Below canopies and over bare soil, deposition velocities were significantly lower (Zhang et al., 2009). However, there are only a few published year-round Hg^0 flux studies that adequately represent the net soil-vegetation-air flux on an ecosystem scale (Fritsche et al., 2008; Castro and Moore, 2016; Obrist et al., 2017; Osterwalder et al., 2017). Thus, in chemical transport models, Hg^0 dry deposition is often parameterized with resistance-based dry deposition schemes (Smith-Downey et al., 2010; Wang et al., 2014b; Song et al., 2016; Wright et al., 2016; Zhang et al., 2016d). The lack of direct evaluations of these models against real-world Hg^0 flux measurements may explain the high uncertainty in the global annual estimates of Hg^0 exchange between the atmosphere and terrestrial surfaces (Song et al., 2015; Horowitz et al., 2017; Khan et al., 2019).

Litterfall Hg is regarded as an approximation for Hg^0 dry deposition in forested areas (Risch et al., 2017). Litterfall accounts for between 65

and 90% of the Hg deposition to terrestrial ecosystems (Demers et al., 2007; Demers et al., 2013; Jiskra et al., 2015; Enrico et al., 2016; Zhang et al., 2016d; Zheng et al., 2016; Yuan et al., 2019a), as confirmed by Hg stable isotope mass balances. Globally, annual Hg^0 deposition through litterfall is estimated between 1020 and 1230 Mg (Risch et al., 2012; Fu et al., 2016a, 2016b; X. Wang et al., 2016; Zhang et al., 2016d). The growing season accumulation of atmospheric Hg^0 in vegetation has been suggested to drive a summertime atmospheric Hg^0 depletion equivalent to 25% of the average annual Hg^0 concentration (1.5 ng m^{-3}) in the Northern Hemisphere (Jiskra et al., 2018). Across the U.S., Hg(II) dry deposition averaged 0.5 (0.1–6.1) μg m^{-2} a^{-1}, which is typically <5% of the total Hg deposition (Lan et al., 2012; L. Zhang et al., 2016a). In these studies, dry deposition was modeled using measurements from active automated Tekran® systems (Gustin et al., 2015). However, on large spatial scales, it has been difficult to model Hg(II) dry deposition since its compounds and physicochemical properties are not well defined (Gustin et al., 2013; Jaffe et al., 2014). When using active and passive samplers including surrogate surfaces, Hg(II) concentrations in the air are 1.3-to-12 times higher than previously thought at locations in the western USA and Florida (Huang et al., 2013; Gustin et al., 2015). These underestimated concentrations suggest that current estimates of Hg(II) deposition are biased low.

Springtime mercury depletion events (AMDEs) in the polar region and subsequent Hg(II) dry deposition to snow and ice are driven by photochemical oxidation of Hg^0 by halogens (Schroeder et al., 1998; Angot et al., 2016). In addition to this process, Hg^0 is deposited to the Arctic ecosystem by (1) reactive oxidative uptake of atmospheric Hg^0 into the snowpack and soils (~9 months), and (2) tundra vegetation uptake (~3 months) (Obrist et al., 2017; Douglas and Blum, 2019). However, a dominant part of the Hg(II) deposited during AMDEs was re-emitted back to the atmosphere through photoreduction. This resulted in minor retention of Hg in the snowpack as a net input to the polar terrestrial ecosystem (Steffen et al., 2008; Sherman et al., 2010; Douglas and Blum, 2019). In the Arctic tundra, only about 2% of the Hg was attributed to wet deposition, while 71% of the soil Hg originated from atmospheric Hg^0 deposition. Dry deposition of Hg(II) occurred mainly during AMDEs and contributed about 27% of the total atmospheric Hg deposition (Obrist et al., 2017).

4.1.2. Re-emission of Hg from the terrestrial environment to the atmosphere

The net Hg^0 flux between land and the atmosphere represents the sum of emissions from land surfaces that comprise primarily emission from soil and vegetation surfaces, geogenic activity and biomass burning as well as Hg^0 dry deposition to surfaces. However, the uncertainty in real-time observations of the net Hg^0 flux (Agnan et al., 2016) is even larger than estimates of total anthropogenic Hg emissions to the atmosphere, which renders model parameterizations difficult (Horowitz et al., 2017).

To date, comprehensive studies of net Hg^0 fluxes on an ecosystem level are scarce, which is exemplified by the major revision of Hg re-emission in the most recent global estimates (UNEP, 2019). A year-round Hg^0 flux measurement on the Alaska tundra revealed re-emission under maximum solar irradiation around noon in the summer growing season besides the AMDEs (Jiskra et al., 2019). Hg^0 re-emission dominates the net Hg^0 flux at a boreal peatland during the growing season (Osterwalder et al., 2017), and that re-emission was likely driven by evasion of dissolved Hg^0 from peat porewater. Jiskra et al. (2015) reported that substantial re-emission resulting from dark abiotic reduction of Hg(II) can occur from peat soils, mobilizing substantial amounts (~ 30%) of historically deposited Hg. However, Hg stable isotope investigations indicated only little photochemically induced re-emission of Hg (Enrico et al., 2016). Hg^0 re-emission from foliage has been observed for more than a decade (Poissant et al., 2008). The re-emission flux has been postulated to originate from instantaneous evasion of recently deposited Hg^0 or Hg(II) on vegetation surfaces (Hintelmann et al., 2002). Recent studies, however, suggested that previously metabolized Hg in the leaf interior was re-emitted after reduction of that sulfur-bound Hg (Manceau et al., 2018; Yuan et al., 2019a). Hg re-emission from previous foliar uptake reached 30% in an evergreen broadleaf subtropical forest (Yuan et al., 2019a).

4.1.3. Accumulation of atmospheric Hg in the terrestrial environment

Although part of deposited Hg is subject to re-emission to the atmosphere, large amounts of atmospheric Hg have been deposited and accumulated in the terrestrial environment (Obrist et al., 2018). Thus polar firn air, peat cores and lake sediments have been extensively used as archives for atmospheric Hg deposition (Biester et al., 2007; Fain et al., 2009; Pérez-Rodríguez et al., 2015; Enrico et al., 2017). Since the onset of industrialization, deposition of atmospheric Hg has increased Hg concentration in the upper organic soil layers by 20% (Smith-Downey et al., 2010). Recent stable Hg isotope studies have pinpointed Hg sources in soil and peat cores and also demonstrated that Hg retained in the organic-rich layers was predominantly derived from atmospheric Hg^0 (Demers et al., 2013; Jiskra et al., 2015; Enrico et al., 2016; X. Wang et al., 2016; Zheng et al., 2016; Obrist et al., 2017). The dominant accumulation of atmospheric Hg^0 over Hg(II) is consistent with the substantially higher litterfall Hg deposition compared to wet deposition of Hg(II) (X. Wang et al., 2016). In mineral soil, source tracing revealed the co-existence of geogenic and atmospheric derived Hg (Jiskra et al., 2015; Zheng et al., 2016; Obrist et al., 2017), suggesting the continuous input of atmospheric Hg to the mineral soil.

Terrestrial storage of atmospheric Hg is a function of (1) atmospheric Hg deposition and (2) Hg re-emission. The major controls on soil Hg accumulation and mobilization are soil OM content, vegetation (e.g., species and leaf area index), precipitation, soil characteristics (e.g., morphology, texture and pH) and climate (Obrist et al., 2018; Wang et al., 2019). With the anticipated decline in anthropogenic Hg emissions under the Minamata Convention, the fate of the terrestrial Hg pool will play a central role in how fast aquatic ecosystems will recover from Hg pollution. A decrease in atmospheric Hg concentrations will reduce uptake of Hg^0 by foliage and associated input of Hg^0 to soil via litterfall.

Climate change has had and will have a broad impact on land-atmosphere exchange of Hg. Plant growth in the Northern Hemisphere, for example, has increased over the past decades (0.13–0.15 Pg C a^{-1}) (Campbell et al., 2017), increasing Hg deposition in vegetation by an estimated 140 Mg a^{-1} since 1990 (Jiskra et al., 2018). On a global scale, however, satellite-based models exhibit a decrease in global vegetation growth due to an increase in the atmospheric vapor pressure deficit (Yuan et al., 2019b). There is strong indication that climate change weakens the vegetation Hg^0 pump by stimulating events such as mid-latitude droughts (Zhao and Running, 2010; Osterwalder et al., 2019), Arctic browning after extreme winter warming and desiccation (Phoenix and Bjerke, 2016; Treharne et al., 2019) or wildfires in the boreal and Arctic region (Mack et al., 2011). Soil warming and permafrost thawing along the northern permafrost gradient are hypothesized to additionally increase future Hg^0 concentrations through accelerated OM decomposition and soil Hg loss to the atmosphere (Obrist, 2007; Smith-Downey et al., 2010; Amos et al., 2013; Jiskra et al., 2015). Furthermore, increased precipitation at northern latitudes driven by climate change could lead to higher groundwater tables and more anoxic conditions favoring Hg reduction and Hg^0 emission from organic soils where these are not counterbalanced by increases in evapotranspiration (cf. Section 3.2, Haynes et al., 2017a, Osterwalder et al., 2017). At the same time soil warming and permafrost thawing support plant growth (Sturm et al., 2001; Jia et al., 2003) and subsequent atmospheric Hg^0 uptake. Deforestation is the main driver of global forest loss (2.3 million km^2 from 2000 to 2012) (Hansen et al., 2013), which is increasing in the tropics (Margono et al., 2014; Mitchard, 2018). Mercury emission from deforested land (Mazur et al., 2014) adds to the atmospheric Hg burden.

Primary changes in knowledge since 2010:

- Elemental Hg has been established as the dominant component of dry deposition in remote areas, with divalent Hg making a minor contribution.
- Hg in foliage and organic soils can be emitted despite strong binding to substrates.
- The estimate of global re-emission was reduced from 2200 Mg a^{-1} (UNEP 2013) to 1000 Mg a^{-1} (UNEP, 2019).
- A latitudinal gradient of increasing wet Hg deposition from Arctic to equator was established.
- Boreal and Arctic systems are large historical sinks of Hg that are becoming strong re-emitters that may counteract efforts to curb anthropogenic Hg emissions.

4.2. Food from terrestrial ecosystems

One gateway through which Hg leaves the terrestrial ecosystem is when plants are used for food by wildlife or people. New reports of Hg contamination in rice, the major food energy source for nearly half of the world's people, have come from Indonesia (Krisnayanti et al., 2012), and different parts of China (e.g. Meng et al., 2011; B. Meng et al., 2014a; M. Meng et al., 2014b; Cheng et al., 2013; Liang et al., 2015; Tang et al., 2015). These confirm that rice bioaccumulates MeHg more than many other plant species, and is an important source of MeHg in terrestrial ecosystems (Zhao et al., 2016a, 2016b). Furthermore, rice consumption was the dominant pathway of MeHg exposure to humans in Hg mining areas and also certain areas of inland southern China (Li et al., 2012; P. Li et al., 2015a; P. Li et al., 2017a; Du et al., 2018). Brombach et al. (2017) reported that 30% of rice products exceeded 10% of the provisional tolerable weekly intake (PTWI) of Hg for toddlers and 13% for adults with a rice-based diet. Abeysinghe et al. (2017) highlighted that MeHg was biomagnified by wildlife in rice-based food webs, with granivorous birds in Hg mining areas accumulating MeHg at levels that are a threat to their health and reproduction.

Paddy soil is the principal source of MeHg in rice plant tissues (Strickman and Mitchell, 2017), which is translocated from the roots to above-ground parts. Most of the MeHg in the plant is transferred to the seed as it ripens (Meng et al., 2011). During grain processing, most of the inorganic Hg (IHg) (~78%) is eliminated, but the majority of the MeHg (~80%) remains in the food product (B. Meng et al., 2014a). Li et al. (2010) indicated that MeHg in rice seeds exists almost exclusively as CH_3Hg-L-cysteinate (CH_3HgCys), a complex that is thought to be responsible for the transfer of MeHg across the blood-brain and placental barriers.

The methylation of IHg in paddy soil primarily occurs under reducing conditions through a process mediated by SRB, iron-reducing bacteria, and methanogens (e.g. Somenahally et al., 2011; Wang et al., 2014a; Liu et al., 2014a, 2014b; X. Liu et al., 2018a; Y.R. Liu et al., 2018b; Vishnivetskaya et al., 2018). The mobility and methylation of Hg in ephemerally flooded soil is determined by a range of factors, such as microbial community structure and diversity, redox potential, pH, DOC, sulfur, iron, and dissolved Hg concentration (Rothenberg and Feng, 2012, Liu et al., 2014a, 2014b, X. Liu et al., 2018a, Wang et al., 2014a, 2014c,). Recently, Zhao et al. (2016b) found that SO_4^{2-} stimulation of SRB was potentially important for Hg methylation in rice paddies. Meng et al. (2010, 2011) suggested that newly deposited Hg is more readily transformed to MeHg and accumulated in rice plants than Hg that has an extended residence time in mining-contaminated soil. Therefore, the concentration of Hg in ambient air was proposed as an indicator for the risk of MeHg accumulation in rice grain (Zhao et al., 2016a, 2016b).

Minimizing the potential for MeHg production in rice paddies is one way to improve the safety of rice in Hg contaminated areas. Intermittent flooding, as opposed to continuous flooding of rice paddies can hinder Hg methylation by creating aerobic conditions in the rhizosphere (Rothenberg et al., 2011; Peng et al., 2012; Wang et al., 2014a, 2014c). Selenium (Se) amendment of soils can also reduce the accumulation of MeHg in rice grain (Wang et al., 2014c, 2016a, 2016b; Zhao et al., 2014; Y.-F. Li et al., 2015b; Tang et al., 2017). Other amendments to paddy soils (elemental S, SO_4^{2-}+Se, SO_4^{2-}+Fe, Fe, Fe+cysteine, and biochar) might also mitigate MeHg accumulation in rice (Shu et al., 2016; Wang et al., 2016a, 2016b; Y. Li et al., 2017b; Zhang et al., 2018, 2019; Zhong et al., 2018). Cultivar selection is another way to reduce MeHg in rice grain grown in Hg-contaminated areas (Peng et al., 2012; Rothenberg and Feng, 2012; Li et al., 2013).

Primary changes in knowledge since 2010:

- The vulnerability of certain wildlife to the Hg in rice has been documented.
- Newly deposited Hg is more likely to be methylated and incorporated in rice than Hg already present in the soil.
- Management strategies to reduce MeHg in rice have been identified.

4.3. Delivery of Hg in runoff to surface waters

In the past decade, research on terrestrial to aquatic transfers of Hg has expanded spatially (e.g. new locations/climates), temporally (e.g. longer-term place-based time series and high frequency in-situ surrogate measurements), and cognitively (e.g. Hg isotopes providing deeper process insight). Furthermore, the proliferation of stream studies has led to synthesis papers that have advanced the science through analysis of differences and commonalities (with respect to Hg fluxes, Hg-carbon relations, Hg:MeHg ratios, etc.) within and among natural and perturbed ecosystems. Here we acknowledge prior synthesis findings and explore the most recent advances in delivery of Hg from the terrestrial landscape to receiving waters.

4.3.1. Arctic studies

One region where riverine Hg work has intensified is the Arctic. Streams in Arctic glaciated and permafrost-dominated systems export higher yields of Hg than other non-contaminated systems (Schuster et al., 2011; Emmerton et al., 2013; Domagalski et al., 2016; Vermilyea et al., 2017; Sondergaard et al., 2015; St Pierre et al., 2018), including large Eurasian rivers (Sonke et al., 2018). High Hg stream yields in the Arctic have been attributed to Hg release from thawing permafrost (Schuster et al., 2018; see Section 3.3.4). Particulate Hg has been shown to dominate Arctic stream Hg flux, (Nagorski et al., 2014, Sondergaard et al., 2015, Vermilyea et al., 2017, St Pierre et al., 2018), while dissolved Hg dominates in Arctic wetland environments (Nagorski et al., 2014; Vermilyea et al., 2017; Poulin et al., 2019). In a small Arctic Alaskan watershed, Douglas et al. (2017) found that more than half of the Hg in the snowpack left the catchment in spring runoff and estimated that 25% of Hg originated from atmospheric mercury depletion events (AMDE) which may become enhanced with future Arctic warming.

4.3.2. Mass balance studies

Hg input-output mass balance at the watershed scale (Table S1) establishes a framework to evaluate processes and assess recovery timescales. Hsu-Kim et al. (2018) analyzed Hg mass balances for different landscapes, and determined that undisturbed forest systems had the lowest stream Hg export as a percentage of total atmospheric deposition (~4–8%). Greater percentages were documented in forested areas with high sediment loads (13%, Shanley et al., 2008; 19%, Riscassi and Scanlon, 2013). Other undisturbed systems that export a relatively high percentage of atmospheric deposition include wetlands, boreal, and alpine tundra (compiled in Shanley and Bishop, 2012), boreal peatlands (33%, Osterwalder et al., 2017), and a subtropical forest in southwestern China (~10%, Ma et al., 2016).

Hg isotope approaches have helped to interpret mass balances. Woerndle et al. (2018) used isotopes to trace sources of Hg deposition,

and its cycling, transport and fate in headwater catchments. Demers et al. (2018) used Hg isotopes in stream suspended sediment to provide new constraints on sources, transport, and transformations of Hg along the flow path. Isotope spikes used in the Mercury Experiment to Assess Atmospheric Loading in Canada and the U.S. (METAALICUS) project revealed that much of the Hg deposited onto vegetation was re-emitted back to the atmosphere (45% in the forest and 71% in the wetland) (Graydon et al., 2012), while Oswald et al. (2014) determined that the morphology and hydrologic behavior of different landscape units in part control the fate and transport of Hg in upland soils.

Net retention of Hg on the terrestrial landscape has generated a large store of legacy Hg in soils, and the conventional wisdom puts stream recovery decades to centuries away (Amos et al., 2014). However, Gerson and Driscoll (2016) reported a decrease in stream Hg (and MeHg) during a 12-year period (2004–2015) in which dry Hg deposition significantly decreased, challenging the notion that stream recovery will take centuries or even decades. Notably, this decrease occurred in spite of expectations that Hg would *increase* as DOC increased in response to the decreased acidity and ionic strength of precipitation. Indeed, experimental liming of an acidic stream by Millard et al. (2018) did result in increased stream Hg concentrations and loads. Measurements from 19 watercourses in Sweden showed that significant OM increases over a decade were also not generally matched by Hg increases (cf. Section 3.3.4) (Eklöf et al., 2012a). Taken together, these studies suggest that decreases in Hg deposition might be "overriding" upward pressure on Hg concentrations expected from acid deposition recovery.

4.3.3. Contaminated and disturbed ecosystems

Recent assessments have drawn attention to the inordinate role of contaminated sites (e.g. industrial and mining operations) to global stream Hg export (Kocman et al., 2013; Kocman et al., 2017). More than half of global stream Hg transport arises from artisanal small-scale gold mining (ASGM) (Obrist et al., 2018), although there is high variability (Gerson et al., 2018). Almost half of the Hg and gold-silver mine-impacted sites ($n = 15$) in western North America showed an order of magnitude greater stream Hg export (16 to 5000 μg m^{-2} a^{-1}) reported in Domagalski et al. (2016) compared to unimpacted forested watersheds (typically <1 to ~10 μg m^{-2} a^{-1}) (Shanley and Bishop, 2012). Disturbed systems also export a relatively high percentage of atmospheric deposition due to mobilization of surficial sediment and a more direct path from deposition to stream runoff due to impervious cover (see Section 3.1 on forestry impacts). Hsu-Kim et al. (2018) estimated ~8–14% Hg export in managed forests and ~50% in urban areas. Elevated Hg export occurred in recently urbanized areas with prior application of Hg-containing pesticides/fungicides (Barringer et al., 2013; Deonarine et al., 2015). As in other streams, high-flow periods generate elevated stream Hg concentrations in systems contaminated from localized sources, including ASGM (Nartey et al., 2011; Diringer et al., 2015), other mining (Holloway et al., 2017), and industrial contamination (Riscassi et al., 2016).

4.3.4. The role of dissolved and particulate organic matter

New empirical studies and compilations (e.g. Lavoie et al., 2019) have reaffirmed the well-established role of OM in the transport of Hg, and this is now supported by Hg isotopic tracing (Jiskra et al., 2017). Strong positive relationships between HgD and DOC were documented for watersheds in North America (Stoken et al., 2016), the Arctic (Nagorski et al., 2014; $r^2 = 0.88$), Sweden (Eklöf et al., 2012a; average $r^2 = 87\%$) and the Czech Republic (Navratil et al., 2015; $r^2 = 0.64–0.93$). Advances in Hg-C interaction research have clarified carbon quality's role with field studies where the optical properties of DOM explained more of the variance in filtered Hg concentration than did bulk DOC (Dittman et al., 2009; Riscassi and Scanlon, 2011; Eklöf et al., 2012a; Burns et al., 2013; Voss et al., 2015; Lescord et al., 2018).

Tight linkages between DOC and Hg in runoff suggest that large-scale browning may have increased Hg loading to surface waters, but changes in DOM quality also mediate how changes in DOM quantity influence Hg export. In a study of 19 Swedish watercourses by Eklöf et al. (2012a) filtered Hg concentration remained unchanged despite a decade of increase in DOC. However, as DOC increased, absorbance at 420 nm remained unchanged, suggesting that Hg responded to the amount of humic matter rather than bulk DOM. In a cross-European study, Bravo et al. (2018) showed that stream THg was terrestrially-derived based on the high fluorescence index of stream DOM. However, Bravo et al. (2017) found that terrestrial DOM was less likely than other DOM fractions to promote MeHg formation in lakes.

As dissolved Hg tracks DOC, particulate Hg (PHg) tracks particulate organic carbon (POC). TSS can serve as a proxy for POC when POC is a consistent fraction of TSS in streams. Riscassi and Scanlon (2011) compiled studies that documented strong relations of PHg (and/or THg) to POC (and/or TSS) for 15 mostly forested watersheds. Similarly strong relations were reported in the Yukon River (Schuster et al., 2011), downstream of ASGM activity (Diringer et al., 2015), during freshet in a large Canadian river (Voss et al., 2015), in a recently de-glaciated stream (Nagorski et al., 2014), and at a mine site in Idaho (Holloway et al., 2017). Departures from a consistent Hg:TSS ratio were used to infer a new source of Hg (Diringer et al., 2015) and an increase in the THg:TOC ratio was related to forestry site preparation (Eklöf et al., 2014).

Increasingly, researchers are linking strong Hg-OM relations to high frequency in-stream optical measurements (Vermilyea et al., 2017). A strong THg-turbidity relation has been found in Alaskan glacial streams ($r^2 = 0.82$, Vermilyea et al., 2017) and mid-latitude forested streams ($r^2 = 0.79–0.98$, Riscassi and Scanlon, 2013). Both of these studies were thus able to use high-frequency turbidity to calculate an annual PHg flux.

4.3.5. Methylation of Hg

The past decade has brought increasing awareness that the Hg methylation process is not restricted to wetlands and impoundments. Methylation occurs in anoxic soils, even in microzones of anoxia within otherwise oxygenated soil (Shanley et al., 2019). Thus a significant fraction of the MeHg carried by streams was formed in the terrestrial landscape (Burns et al., 2014; Bravo et al., 2017; Rodenhouse et al., 2019). The dynamics of MeHg transfer from the terrestrial to aquatic environment share some commonalities and contrasts to THg. High flows and corresponding DOM increases that drive THg export can also mobilize MeHg, but the link is often weaker (Eklöf et al., 2014; Lescord et al., 2018). High-flow conditions that connect the stream to the broader watershed can mobilize MeHg from methylation hotspots such as wetlands, but may dilute MeHg if such hotspots are absent or less active during spring when soils are cold and snowmelt flows are high (Schelker et al., 2011; Eklöf et al., 2013; Riscassi et al., 2016).

4.3.6. Watershed Hg modeling

Development of watershed models that capture the complexity of Hg biogeochemistry and accurately predict stream Hg and MeHg concentrations and fluxes remains a major unmet research need (Hsu-Kim et al., 2018). Used within a single watershed, multiple mechanistic and empirical models that each capture the dynamics of a particular component of Hg have proved useful (Golden et al., 2012). However, this ensemble approach can be time consuming, and no one model exists that captures both the DOC-driven watershed THg fluxes and sediment-bound THg fluxes. Simulating MeHg in those same watersheds has proved even more difficult (Knightes et al., 2014). Carroll and Warwick (2016) successfully modeled both Hg and MeHg in a contaminated system in which soil bank erosion was the dominant process contributing to in-stream fluxes. In this case, in-depth a priori knowledge of the system was pivotal, as four different flow regimes were needed to capture differences in Hg loading mechanisms. Evaluation of empirical data and hydrology within a watershed will be necessary to guide the appropriate application of process-based watershed

specific models in the future (Oswald and Branfireun, 2014; Zhu et al., 2018; Eklöf et al., 2015; Berndt et al., 2016). While Hg and MeHg modeling is still developmental, modeling that tests the effects of climate change is critical to guide future watershed management (Golden et al., 2013).

Primary changes in knowledge since 2010:

- Increased focus on the Arctic has revealed high Hg concentrations and fluxes.
- "Recovery" of streams to lower Hg concentration may be more rapid than expected.
- Artisanal small-scale gold mining is responsible for more than half of global stream Hg export, but such sites have not been studied well.
- DOM character is a critical factor in Hg mobility and in MeHg formation.
- Much of the MeHg carried by streams originated from the terrestrial landscape.

5. Summary and implications for the Minamata Convention

Terrestrial cycling is an area of Hg science that is developing rapidly. Part of the development relates to new measurements. Natural abundance isotopes that trace sources and processes have been particularly useful, for instance in quantifying re-emission from strongly binding substrates in soils and foliage at some sites (Jiskra et al., 2017; Yuan et al., 2019a), accumulation at others (Obrist et al., 2017), or defining the importance of newly deposited Hg for bioaccumulation in rice (Meng et al., 2011). Microbial techniques are also opening up new avenues in understanding the biological foundations for Hg transformations and interactions with other environmental variables (Eklöf et al., 2018; Jones et al., 2019).

Another factor contributing to progress is the increased efficiency of routine analyses that facilitates study of more sites (cf. Tables 1, S1). Given the complexity of Hg, the addition of new sites is essential for conceptual generalizations and process understanding that can be tested experimentally or in model predictions. The suggestion of a topographical optimum between steep and gentle terrain in the vulnerability of forests to releasing Hg after harvesting would not have been possible without an increasing number of catchment studies (Kronberg et al., 2016a). Even more long-term studies, however, are needed to resolve the causes of the variability in response to forest harvest (cf. Section 3.1 and Fig. 2). Advances in micrometeorological techniques and their deployment around the globe from the Arctic (Kamp et al., 2018) to the tropics (Zhang et al., 2016c) are also helping to address the complex interface between land and atmosphere where the strength and even the overall direction of Hg exchange are critical issues.

With so many developments on many fronts, this review has focused on recent (post-2010) developments in understanding influences on the terrestrial cycle, in particular the three "gateways" through which impacts arise for the health of people and wildlife, as well as the global cycling of Hg. A comparison of the impact severity and level of certainty about these impacts reveals clear patterns, despite much uncertainty and/or paucity of data for many regions and influences (Fig. 3). The most severe and certain impacts follow from the rapidly warming climate in the high latitudes. Substantial amounts of Hg that have accumulated in boreal and Arctic soils are being mobilized by permafrost thawing and wildfire. Research documenting the return of these stores to the atmosphere is reshaping our understanding of the global Hg cycle by calling into question previously accepted ideas that Hg would tend to move from the tropics and temperate zone towards higher latitudes (Jiskra et al., 2018).

The impacts on the global Hg cycle depend to a large extent on how changing land cover, both intentional as in afforestation and deforestation, as well as unintentional related to climate change (e.g. greening and browning) will influence net ecosystem exchange of Hg with the atmosphere in the mid-latitudes and the tropics. The effectiveness of measures to reduce anthropogenic Hg emissions and subsequently atmospheric Hg concentrations may be offset by a combination of climate warming induced environmental changes such as more frequently occurring wildfires (Abatzoglou and Williams, 2016), droughts (Allen et al., 2010), decreasing global vegetation growth (Yuan et al., 2019b) and release of Hg from thawing permafrost soils (Schuster et al., 2018). Wet Hg deposition and vegetation cover increase towards the tropics, but this is where measurements are fewest and the potential for changes in forest cover is particularly high. This lack of data highlights the need to improve our measurements of Hg mobility across the land-atmosphere gateway in representative environments. Advances with in-situ Hg flux measurements, such as using eddy covariance (Osterwalder et al., 2019) and integration of several techniques (e.g., stable Hg isotopes and micrometeorology) to constrain fluxes will be central to better quantification of land-atmosphere Hg exchange. This will help integrate bi-directional surface-atmosphere exchange in global Hg chemistry and transport models.

Another critical Hg impact on people and biota is exposure to Hg in artisanal gold mining (ASGM). While focused on a relatively small land area, ASGM has an inordinate global influence on the export of Hg to surface waters and the atmosphere as well as a regional influence on the food supply where rice is grown near areas contaminated by mining (Ha et al., 2017). More measurements are needed from ASGM sites, and the tropics in general, to better define the cycling of Hg in these areas.

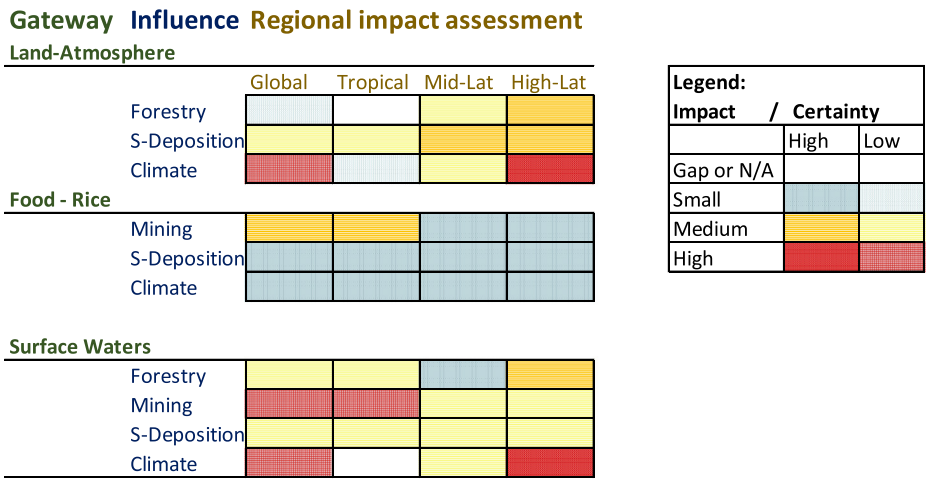


Fig. 3. Severity and certainty of impacts on the "gateways" of the terrestrial Hg cycle that influence people, the biota and global cycling.

Documentation of Hg in wildlife that eat rice has added to the importance of studies on people and biota in areas new to Hg research (Abeysinghe et al., 2017).

Recognition that new sources of Hg are being mobilized in high latitudes, together with increased awareness of the impacts from ASGM will help keep Hg on the research agenda as a pervasive issue for human and ecosystem health. A key issue for implementing the Minamata Convention will be further improvements in understanding how the anthropogenic spread of Hg, and our efforts to control this spread, affects the exposure of humans and the rest of the world's biota to Hg. Decadal declines of atmospheric Hg bring into focus the time scale of recovery, especially as the compounding factor of anthropogenic S deposition has also abated.

If and when these factors will translate into reduced Hg loadings to surface waters via catchment outputs remains a major question mark. This recovery was generally believed to be very slow (centennial scale) prior to 2010, but there are now tantalizing, albeit inconclusive indications that recovery may be faster. Regional recovery observed in the fish of some regions has been complemented by a handful of decadal stream time series where Hg has decreased, or has not increased in step with increases in OM (Eklöf et al., 2012a; Gerson and Driscoll, 2016). Whether this recovery can be attributed solely to decreased Hg and S deposition, or whether other factors such as climate or land use change contribute, remains to be answered. The possibility that some wetlands might evade legacy Hg back to the atmosphere if and when atmospheric concentrations decline might also contribute to more rapid aquatic recovery than currently expected (Osterwalder et al., 2017). Advances in understanding the microbial basis for Hg transformation, with consequences for both bioavailability and mobility within catchments will be a key part of resolving recovery time scales for both downstream aquatic ecosystems and land-based food webs. Part of the challenge for the future will be revealing how changes in nutrient availability and climate interact to influence microbial methylation both directly and indirectly through effects such as enhanced syntrophy, Hg(II) bioavailability impacts from S^{2-} molecules, and Hg(II) solubility (e.g. Hu et al., 2020). Methodological advances in molecular biology combined with microbial ecology, as well as better analytical sensitivity in probing atomic and molecular interactions among Hg, S and DOM, will be critical tools for meeting this challenge.

The cycling of Hg in terrestrial ecosystems has an important bearing on how actions in accordance with the Minamata Convention will affect the global cycling of Hg, and ultimately the risks of Hg exposure to people and other biota. This importance is due partly to the sink strength of terrestrial ecosystems for atmospheric Hg, but also to how quickly terrestrial ecosystems transfer anthropogenic Hg into food webs or back to the atmosphere. In the years ahead further advances in isotopic, microbial and micrometeorological techniques will play key roles in distinguishing the effect of international efforts to reduce Hg emissions on the terrestrial Hg cycle from the effect of climate, land-use and other anthropogenic influences.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.137647.

CRediT authorship contribution statement

Kevin Bishop: Conceptualization, Formal analysis, Methodology, Supervision, Validation, Writing - original draft, Writing - review & editing. **James B. Shanley:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Supervision, Validation, Writing - original draft, Writing - review & editing. **Ami Riscassi:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Writing - original draft, Writing - review & editing. **Heleen A. de Wit:** Formal analysis, Investigation, Methodology, Validation, Writing - original draft, Writing - review & editing. **Karin Eklöf:** Formal analysis, Investigation, Validation, Writing - original draft, Writing - review & editing. **Bo Meng:** Formal analysis, Investigation, Validation, Writing - original draft, Writing - review & editing. **Carl Mitchell:** Formal analysis, Investigation, Validation, Writing - original draft, Writing - review & editing. **Stefan Osterwalder:** Formal analysis, Investigation, Validation, Writing - original draft, Writing - review & editing. **Paul F. Schuster:** Formal analysis, Investigation, Validation, Writing - original draft, Writing - review & editing. **Jackson Webster:** Formal analysis, Investigation, Validation, Writing - original draft, Writing - review & editing. **Wei Zhu:** Formal analysis, Investigation, Validation, Writing - original draft, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors. We thank Heather Golden for help with the text on Hg modeling, and Doug Burns and three anonymous reviewers for useful comments on the paper. The use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

References

Abatzoglou, J.T., Williams, A.P., 2016. Impact of anthropogenic climate change on wildfire across western us forests. Proc. Natl. Acad. Sci. U. S. A. 113 (42), 11770–11775. https://doi.org/10.1073/pnas.1607171113.

Abeysinghe, K.S., Qiu, G., Goodale, E., Anderson, C.W.N., Bishop, K., Evers, D.C., et al., 2017. Mercury flow through an Asian rice-based food web. Environ. Pollut. 229, 219–228. https://doi.org/10.1016/j.envpol.2017.05.067.

Abraham, J., Dowling, K., Florentine, S., 2018. Effects of prescribed fire and post-fire rainfall on mercury mobilization and subsequent contamination assessment in a legacy mine site in Victoria, Australia. Chemosphere 190, 144–153. https://doi.org/10.1016/j.chemosphere.2017.09.117.

Ackerman, J.T., Kraus, T.E., Fleck, J.A., Krabbenhoft, D.P., Horwath, W.R., Bachand, S.M., et al., 2015. Experimental dosing of wetlands with coagulants removes mercury from surface water and decreases mercury bioaccumulation in fish. Environ. Sci. Technol. 49 (10), 6304–6311. https://doi.org/10.1021/acs.est.5b00655.

Agnan, Y., Le Dantec, T., Moore, C.W., Edwards, G.C., Obrist, D., 2016. New constraints on terrestrial surface-atmosphere fluxes of gaseous elemental mercury using a global database. Environ. Sci. Technol. 50 (2), 507–524. https://doi.org/10.1021/acs.est.5b04013.

Åkerblom, S., Meili, M., Bringmark, L., Johansson, K., Kleja, D.B., Bergkvist, B., 2008. Partitioning of hg between solid and dissolved organic matter in the humus layer of boreal forests. Water Air Soil Pollut. 189 (1–4), 239–252. https://doi.org/10.1007/s11270-007-9571-1.

Åkerblom, S., Bishop, K., Björn, E., Lambertsson, L., Eriksson, T., Nilsson, M.B., 2013. Significant interaction effects from sulfate deposition and climate on sulfur concentrations constitute major controls on methylmercury production in peatlands. Geochim. Cosmochim. Acta 102, 1–11. https://doi.org/10.1016/j.gca.2012.10.025.

Åkerblom, S., Meili, M., Bishop, K., 2015. Organic matter in rain: an overlooked influence on mercury deposition. Environ. Sci. Technol. Letters. 2 (4), 128–132. https://doi.org/10.1021/acs.estlett.5b00009.

Allan, C.J., Heyes, A., Mackereth, R.J., 2009. Changes to groundwater and surface water Hg transport following clearcut logging: a Canadian case study. In: *Does Forestry Contribute to Mercury in Swedish Fish?* Royal Swedish Academy of Agriculture and Forestry (KSLA) Report 148, 50–54.

Allen, C.D., Macalady, A.K., Chenchouni, H., Bachelet, D., McDowell, N., Vennetier, M., et al., 2010. A global overview of drought and heat-induced tree mortality reveals emerging climate change risks for forests. For. Ecol. Manag. 259 (4), 660–684. https://doi.org/10.1016/j.foreco.2009.09.001.

Alpers, C.N., Fleck, J.A., Marvin-DiPasquale, M., Stricker, C.A., Stephenson, M., Taylor, H.E., 2014. Mercury cycling in agricultural and managed wetlands, Yolo Bypass, California: spatial and seasonal variations in water quality. Sci. Total Environ. 484, 276–287. https://doi.org/10.1016/j.scitotenv.2013.10.096.

Alriksson, A., 2001. Regional variability of Cd, Hg, Pb and C concentrations in different horizons of Swedish forest soils. Water Air Soil Pollut. Focus. 1, 325–341.

Amos, H.M., Jacob, D.J., Streets, D.G., Sunderland, E.M., 2013. Legacy impacts of all-time anthropogenic emissions on the global mercury cycle. Glob. Biogeochem. Cy. 27 (2), 410–421. https://doi.org/10.1002/gbc.20040.

Amos, H.M., Jacob, D.J., Kocman, D., Horowitz, H.M., Zhang, Y., Dutkiewicz, S., et al., 2014. Global biogeochemical implications of mercury discharges from rivers and sediment burial. Environ. Sci. Technol. 48 (16), 9514–9522. https://doi.org/10.1021/es502134t.

Angot, H., Dastoor, A., De Simone, F., Gardfeldt, K., Gencarelli, C.N., Hedgecock, I.M., et al., 2016. Chemical cycling and deposition of atmospheric mercury in polar regions:

review of recent measurements and comparison with models. Atmos. Chem. Phys. 16 (16), 10735–10763. https://doi.org/10.5194/acp-16-10735-2016.
Appleton, J.D., Weeks, J.M., Calvez, J.P., Beinhoff, C., 2006. Impacts of mercury contaminated mining waste on soil quality, crops, bivalves, and fish in the Naboc River area, Mindanao, Philippines. Sci. Total Environ. 354 (2–3), 198–211. https://doi.org/10.1016/j.scitotenv.2005.01.042.
Babiarz, C.L., Hurley, J.P., Benoit, J.M., Shafer, M.M., Andren, A.W., Webb, D.A., 1998. Seasonal influences on partitioning and transport of total and methylmercury in rivers from contrasting watersheds. Biogeochemistry 41 (3), 237–257. https://doi.org/10.1023/A:1005940630948.
Balogh, S.J., Swain, E.B., Nollet, Y.H., 2006. Elevated methylmercury concentrations and loadings during flooding in Minnesota rivers. Sci. Total Environ. 368 (1), 138–148. https://doi.org/10.1016/j.scitotenv.2005.09.045.
Baptista-Salazar, C., Richard, J.H., Horf, M., Rejc, M., Gosar, M., Biester, H., 2017. Grain-size dependence of mercury speciation in river suspended matter, sediments and soils in a mercury mining area at varying hydrological conditions. Appl. Geochem. 81, 132–142. https://doi.org/10.1016/j.apgeochem.2017.04.006.
Barringer, J.L., Szabo, Z., Reilly, P.A., Riskin, M.L., 2013. Variable contributions of mercury from groundwater to a first-order urban coastal plain stream in New Jersey, USA. Water Air Soil Pollut. 224 (4). https://doi.org/10.1007/s11270-013-1475-7.
Barron, M.G., Duvall, S.E., Barron, K.J., 2004. Retrospective and current risks of mercury to panthers in the Florida Everglades. Ecotoxicology 13 (3), 223–229. https://doi.org/10.1023/b:ectx.0000023567.42698.38.
Benoit, J.M., Gilmour, C.C., Mason, R.P., Heyes, A., 1999. Sulfide controls on mercury speciation and bioavailability to methylating bacteria in sediment pore waters. Environ. Sci. Technol. 33 (6), 951–957. https://doi.org/10.1021/es9808200.
Benoit, J.M., Gilmour, C.C., Mason, R.P., 2001. Aspects of bioavailability of mercury for methylation in pure cultures of *Desulfobulbus propionicus* (1pr3). Appl. Environ. Microbiol. 67 (1), 51–58. https://doi.org/10.1128/AEM.67.1.51-58.2001.
Berg, T., Aspmo, K., Steinnes, E., 2008. Transport of Hg from atmospheric mercury depletion events to the mainland of Norway and its possible influence on Hg deposition. Geophys. Res. Lett. 35 (9). https://doi.org/10.1029/2008gl033586.
Bergman, I., Bishop, K., Tu, Q., Frech, W., Akerblom, S., Nilsson, M., 2012. The influence of sulphate deposition on the seasonal variation of peat pore water methyl Hg in a boreal mire. PLoS One 7 (9), e45547. https://doi.org/10.1371/journal.pone.0045547.
Berndt, M.E., Bavin, T.K., 2012. Methylmercury and dissolved organic carbon relationships in a wetland-rich watershed impacted by elevated sulfate from mining. Environ. Pollut. 161, 321–327. https://doi.org/10.1016/j.envpol.2011.06.006.
Berndt, M.E., Rutelonis, W., Regan, C.P., 2016. A comparison of results from a hydrologic transport model (HSPF) with distributions of sulfate and mercury in a mine-impacted watershed in northeastern Minnesota. J. Environ. Manag. 181, 74–79. https://doi.org/10.1016/j.jenvman.2016.05.067.
Biester, H., Bindler, R., Martinez-Cortizas, A., Engstrom, D.R., 2007. Modeling the past atmospheric deposition of mercury using natural archives. Environ. Sci. Technol. 41 (14), 4851–4860. https://doi.org/10.1021/es0704232.
Bishop, K.H., Lee, Y.H., 1997. Catchments as a source of mercury/methylmercury in boreal surface waters. In: Sigel, A., Sigel, H. (Eds.), Metal Ions in Biological Systems, Vol 34: Mercury and its Effects on Environment and Biology. 34, pp. 113–130.
Bishop, K., Lee, Y.H., Pettersson, C., Allard, B., 1995. Terrestrial sources of methylmercury in surface waters - the importance of the riparian zone on the Svartberget catchment. Water Air Soil Pollut. 80 (1–4), 435–444. https://doi.org/10.1007/Bf01189693.
Bishop, K.H., Lee, Y.H., Munthe, J., Dambrine, E., 1998. Xylem sap as a pathway for total mercury and methylmercury transport from soils to tree canopy in the boreal forest. Biogeochemistry 40 (2–3), 101–113. https://doi.org/10.1023/A:1005983932240.
Bishop, K., Allan, C., Bringmark, L., Garcia, E., Hellsten, S., Hogbom, L., et al., 2009. The effects of forestry on Hg bioaccumulation in nemoral/boreal waters and recommendations for good silvicultural practice. Ambio 38 (7), 373–380. https://doi.org/10.1579/0044-7447-38.7.373.
Blanchfield, P.J., Paterson, M., Podemski, C.L., Hintelmann, H., 2004. The movement of newly deposited mercury through an aquatic food web. In: Horvat, M., Ogrinc, N., Faganeli, J., Kotnik, J. (Eds.), RMZ Materials and Geoenvironment: Mercury as a Global Pollutant. 51, pp. 838–840.
Boutron, C.F., Vandal, G.M., Fitzgerald, W.F., Ferrari, C.P., 1998. A forty year record of mercury in central Greenland snow. Geophys. Res. Lett. 25, 3315–3318. https://doi.org/10.1029/98GL02422.
Braaten, H.F.V., de Wit, H.A., 2016. Effects of disturbance and vegetation type on total and methylmercury in boreal peatland and forest soils. Environ. Pollut. 218, 140–149. https://doi.org/10.1016/j.envpol.2016.08.029.
Braaten, H.F.V., de Wit, H.A., Larssen, T., Poste, A.E., 2018. Mercury in fish from Norwegian lakes: the complex influence of aqueous organic carbon. Sci. Total Environ. 627, 341–348. https://doi.org/10.1016/j.scitotenv.2018.01.252.
Braaten, H.F.V., Lindholm, M., De Wit, H.A., 2020. Five decades of declining methylmercury concentrations in boreal foodwebs suggest pivotal role for sulphate deposition. Sci. Total Environ. https://doi.org/10.1016/j.scitotenv.2020.136774.
Branfireun, B.A., Roulet, N.T., Kelly, C.A., Rudd, J.W.M., 1999. In situ sulphate stimulation of mercury methylation in a boreal peatland: toward a link between acid rain and methylmercury contamination in remote environments. Glob. Biogeochem. Cy. 13 (3), 743–750 (doi:Doi 10.1029/1999gb900033).
Branfireun, B.A., Bishop, K., Roulet, N.T., Granberg, G., Nilsson, M., 2001. Mercury cycling in boreal ecosystems: the long-term effect of acid rain constituents on peatland pore water methylmercury concentrations. Geophys. Res. Lett. 28 (7), 1227–1230. https://doi.org/10.1029/2000gl011867.
Bravo, A.G., Bouchet, S., Tolu, J., Bjorn, E., Mateos-Rivera, A., Bertilsson, S., 2017. Molecular composition of organic matter controls methylmercury formation in boreal lakes. Nat. Commun. 8, 14255. https://doi.org/10.1038/ncomms14255.
Bravo, A.G., Kothawala, D.N., Attermeyer, K., Tessier, E., Bodmer, P., Ledesma, J.L.J., et al., 2018. The interplay between total mercury, methylmercury and dissolved organic matter in fluvial systems: a latitudinal study across europe. Water Res. 144, 172–182. https://doi.org/10.1016/j.watres.2018.06.064.
Brigham, M.E., Wentz, D.A., Aiken, G.R., Krabbenhoft, D.P., 2009. Mercury cycling in stream ecosystems. 1. Water column chemistry and transport. Environ. Sci. Technol. 43 (8), 2720–2725. https://doi.org/10.1021/es802694n.
Bringmark, L., Bringmark, E., 2001. Lowest effect levels of lead and Hg on decomposition of mor layer samples in a long-term experiment. Water Air Soil Pollut. Focus. 1 (3–4), 425–437.
Brombach, C.C., Manorut, P., Kolambage-Dona, P.P.P., Ezzeldin, M.F., Chen, B., Corns, W.T., et al., 2017. Methylmercury varies more than one order of magnitude in commercial European rice. Food Chem. 214, 360–365. https://doi.org/10.1016/j.foodchem.2016.07.064.
Burke, M.P., Hogue, T.S., Ferreira, M., Mendez, C.B., Navarro, B., Lopez, S., et al., 2010. The effect of wildfire on soil mercury concentrations in southern California watersheds. Water Air Soil Pollut. 212 (1–4), 369–385. https://doi.org/10.1007/s11270-010-0351-y.
Burns, D.A., Aiken, G.R., Bradley, P.M., Journey, C.A., Schelker, J., 2013. Specific ultra-violet absorbance as an indicator of mercury sources in an Adirondack river basin. Biogeochemistry 113 (1–3), 451–466. https://doi.org/10.1007/s10533-012-9773-5.
Burns, D.A., Nystrom, E.A., Wolock, D.M., Bradley, P.M., Riva-Murray, K., 2014. An empirical approach to modeling methylmercury concentrations in an Adirondack stream watershed. J. Geophys. Res.-Biogeosci. 119 (10), 1970–1984. https://doi.org/10.1002/2013jg002481.
Caldwell, C.A., Canavan, C.M., Bloom, N.S., 2000. Potential effects of forest fire and storm flow on total mercury and methylmercury in sediments of an arid-lands reservoir. Sci. Total Environ. 260 (1–3), 125–133. https://doi.org/10.1016/s0048-9697(00)00554-4.
Campbell, J.E., Berry, J.A., Seibt, U., Smith, S.J., Montzka, S.A., Launois, T., Belviso, S., Bopp, L., Laine, M., 2017. Large historical growth in global terrestrial gross primary produciton. Nature 544, 84–87. https://doi.org/10.1038/nature22030.
Campos, I., Vale, C., Abrantes, N., Keizer, J.J., Pereira, P., 2015. Effects of wildfire on mercury mobilisation in eucalypt and pine forests. Catena 131, 149–159. https://doi.org/10.1016/j.catena.2015.02.024.
Carpi, A., Lindberg, S.E., 1997. Sunlight-mediated emission of elemental mercury from soil amended with municipal sewage sludge. Environ. Sci. Technol. 31 (7), 2085–2091. https://doi.org/10.1021/es960910+.
Carroll, R.W.H., Warwick, J.J., 2016. Modeling the highly dynamic loading of mercury species in the Carson River and Lahontan Reservoir system, Nevada. J. Am. Water Res. Assoc. 52 (5), 1207–1222. https://doi.org/10.1111/1752-1688.12448.
Carroll, R.W.H., Warwick, J.J., Heim, K.J., Bonzongo, J.C., Miller, J.R., Lyons, W.B., 2000. Simulation of mercury transport and fate in the Carson River. Nevada. Ecol. Model. 125 (2–3), 255–278. https://doi.org/10.1016/S0304-3800(99)00186-6.
Castro, M., Moore, C., 2016. Importance of gaseous elemental mercury fluxes in western Maryland. Atmosphere 7 (9), 110. https://doi.org/10.3390/atmos7090110.
Chadwick, S.P., Babiarz, C.L., Hurley, J.P., Armstrong, D.E., 2006. Influences of iron, manganese, and dissolved organic carbon on the hypolimnetic cycling of amended mercury. Sci. Total Environ. 368 (1), 177–188. https://doi.org/10.1016/j.scitotenv.2005.09.039.
Chen, J.B., Hintelmann, H., Zheng, W., Feng, X.B., Cai, H.M., Wang, Z.H., et al., 2016. Isotopic evidence for distinct sources of mercury in lake waters and sediments. Chem. Geol. 426, 33–44. https://doi.org/10.1016/j.chemgeo.2016.01.030.
Cheng, I., Zhang, L., 2017. Uncertainty assessment of gaseous oxidized mercury measurements collected by atmospheric mercury network. Environ. Sci. Technol. 51 (2), 855–862. https://doi.org/10.1021/acs.est.6b04926.
Cheng, J., Zhao, W., Wang, Q., Liu, X., Wang, W., 2013. Accumulation of mercury, selenium and PCBs in domestic duck brain, liver and egg from a contaminated area with an investigation of their redox responses. Environ. Toxicol. Pharmacol. 35 (3), 388–394. https://doi.org/10.1016/j.etap.2013.01.011.
Coleman Wasik, J.K., Mitchell, C.P.J., Engstrom, D.R., Swain, E.B., Monson, B.A., Balogh, S.J., et al., 2012. Methylmercury declines in a boreal peatland when experimental sulfate deposition decreases. Environ. Sci. Technol. 46 (12), 6663–6671. https://doi.org/10.1021/es300865f.
Coleman Wasik, J.K., Engstrom, D.R., Mitchell, C.P.J., Swain, E.B., Monson, B.A., Balogh, S.J., et al., 2015. The effects of hydrologic fluctuation and sulfate regeneration on mercury cycling in an experimental peatland. J. Geophys. Res. Biogeosci. 120 (9), 1697–1715. https://doi.org/10.1002/2015jg002993.
Corrales, J., Naja, G.M., Dziuba, C., Rivero, R.G., Orem, W., 2011. Sulfate threshold target to control methylmercury levels in wetland ecosystems. Sci. Total Environ. 409 (11), 2156–2162. https://doi.org/10.1016/j.scitotenv.2011.02.030.
Cortizas, A.M., Biester, H., Mighall, T., Bindler, R., 2007. Climate-driven enrichment of pollutants in peatlands. Biogeosciences 4 (5), 905–911.
Creswell, J.E., Shafer, M.M., Babiarz, C.L., Tan, S.Z., Musinsky, A.L., Schott, T.H., et al., 2017. Biogeochemical controls on mercury methylation in the Allequash Creek wetland. Environ. Sci. Pollut. Res. Int. 24 (18), 15325–15339. https://doi.org/10.1007/s11356-017-9094-2.
Cristol, D.A., Brasso, R.L., Condon, A.M., Fovargue, R.E., Friedman, S.L., Hallinger, K.K., et al., 2008. The movement of aquatic mercury through terrestrial food webs. Science 320 (5874), 335. https://doi.org/10.1126/science.1154082.
de Wit, H.A., Granhus, A., Lindholm, M., Kainz, M.J., Lin, Y., Braaten, H.F.V., et al., 2014. Forest harvest effects on mercury in streams and biota in Norwegian boreal catchments. For. Ecol. Manag. 324, 52–63. https://doi.org/10.1016/j.foreco.2014.03.044.
de Wit, H.A., Valinia, S., Weyhenmeyer, G.A., Futter, M.N., Kortelainen, P., Austnes, K., et al., 2016. Current browning of surface waters will be further promoted by wetter climate. Environ. Sci. Technol. Letters. 3 (12), 430–435. https://doi.org/10.1021/acs.estlett.6b00396.

Demers, J.D., Driscoll, C.T., Fahey, T.J., Yavitti, J.B., 2007. Mercury cycling in litter and soil in different forest types in the Adirondack region, New York, USA. Ecol. Appl. 17 (5), 1341–1351. https://doi.org/10.1890/06-1697.1.

Demers, J.D., Driscoll, C.T., Shanley, J.B., 2010. Mercury mobilization and episodic stream acidification during snowmelt: role of hydrologic flow paths, source areas, and supply of dissolved organic carbon. Water Resour. Res. 46. https://doi.org/10.1029/2008wr007021.

Demers, J.D., Blum, J.D., Zak, D.R., 2013. Mercury isotopes in a forested ecosystem: implications for air-surface exchange dynamics and the global mercury cycle. Glob. Biogeochem. Cy. 27 (1), 222–238. https://doi.org/10.1002/gbc.20021.

Demers, J.D., Blum, J.D., Brooks, S.C., Donovan, P.M., Riscassi, A.L., Miller, C.L., et al., 2018. Hg isotopes reveal in-stream processing and legacy inputs in East Fork Poplar Creek, Oak Ridge, Tennessee, USA. Environ. Sci. Process Impacts. 20 (4), 686–707. https://doi.org/10.1039/c7em00538e.

Denzler, B., Bogdal, C., Henne, S., Obrist, D., Steinbacher, M., Hungerbuhler, K., 2017. Inversion approach to validate mercury emissions based on background air monitoring at the high altitude research station Jungfraujoch (3580 m). Environ. Sci. Technol. 51 (5), 2846–2853. https://doi.org/10.1021/acs.est.6b05630.

Deonarine, A., Hsu-Kim, H., Zhang, T., Cai, Y., Richardson, C.J., 2015. Legacy source of mercury in an urban stream-wetland ecosystem in central North Carolina, USA. Chemosphere 138, 960–965. https://doi.org/10.1016/j.chemosphere.2014.12.038.

Desrosiers, M., Planas, D., Mucci, A., 2006. Short-term responses to watershed logging on biomass mercury and methylmercury accumulation by periphyton in boreal lakes. Can. J. Fish. Aquat. Sci. 63 (8), 1734–1745. https://doi.org/10.1139/F06-077.

Diringer, S.E., Feingold, B.J., Ortiz, E.J., Gallis, J.A., Araujo-Flores, J.M., Berky, A., et al., 2015. River transport of mercury from artisanal and small-scale gold mining and risks for dietary mercury exposure in Madre de Dios, Peru. Environ. Sci. Process. Impacts. 17 (2), 478–487. https://doi.org/10.1039/c4em00567h.

Dittman, J.A., Shanley, J.B., Driscoll, C.T., Aiken, G.R., Chalmers, A.T., Towse, J.E., 2009. Ultraviolet absorbance as a proxy for total dissolved mercury in streams. Environ. Pollut. 157 (6), 1953–1956. https://doi.org/10.1016/j.envpol.2009.01.031.

Domagalski, J., Majewski, M.S., Alpers, C.N., Eckley, C.S., Eagles-Smith, C.A., Schenk, L., et al., 2016. Comparison of mercury mass loading in streams to atmospheric deposition in watersheds of western North America: evidence for non-atmospheric mercury sources. Sci. Total Environ. 568, 638–650. https://doi.org/10.1016/j.scitotenv.2016.02.112.

Douglas, T.A., Blum, J.D., 2019. Mercury isotopes reveal atmospheric gaseous mercury deposition directly to the arctic coastal snowpack. Environ. Sci. Technol. Letters. 6 (4), 235–242. https://doi.org/10.1021/acs.estlett.9b00131.

Douglas, T.A., Sturm, M., Blum, J.D., Polashenski, C., Stuefer, S., Hiemstra, C., et al., 2017. A pulse of mercury and major ions in snowmelt runoff from a small arctic Alaska watershed. Environ. Sci. Technol. 51 (19), 11145–11155. https://doi.org/10.1021/acs.est.7b03683.

Drenner, R.W., Chumchal, M.M., Wente, S.P., McGuire, M., Drenner, S.M., 2011. Landscape-level patterns of mercury contamination of fish in north Texas, USA. Environ. Toxicol. Chem. 30 (9), 2041–2045. https://doi.org/10.1002/etc.589.

Drevnick, P.E., Canfield, D.E., Gorski, P.R., Shinneman, A.L., Engstrom, D.R., Muir, D.C., et al., 2007. Deposition and cycling of sulfur controls mercury accumulation in Isle Royale fish. Environ. Sci. Technol. 41 (21), 7266–7272. https://doi.org/10.1021/es0712322.

Du, B., Feng, X., Li, P., Yin, R., Yu, B., Sonke, J.E., et al., 2018. Use of mercury isotopes to quantify mercury exposure sources in inland populations, China. Environ. Sci. Technol. 52 (9), 5407–5416. https://doi.org/10.1021/acs.est.7b05638.

Ebinghaus, R., Tripathi, R.M., Wallschläger, D., Lindberg, S.E., 1999. Natural and anthropogenic mercury sources and their impact on the air-surface exchange of mercury on regional and global scales. editors. In: Ebinghaus, R., Turner, R.R., de Lacerda, L.D., Vasiliev, O., Salomons, W. (Eds.), Mercury Contaminated Sites: Characterization, Risk Assessment and Remediation. Springer, Berlin, Heidelberg, pp. 3–50.

Eckley, C.S., Eagles-Smith, C., Tate, M.T., Kowalski, B., Danehy, R., Johnson, S.L., et al., 2018. Stream mercury export in response to contemporary timber harvesting methods (Pacific coastal mountains, Oregon, USA). Environ. Sci. Technol. 52 (4), 1971–1980. https://doi.org/10.1021/acs.est.7b05197.

Eklöf, K., Folster, J., Sonesten, L., Bishop, K., 2012a. Spatial and temporal variation of THg concentrations in run-off water from 19 boreal catchments, 2000-2010. Environ. Pollut. 164, 102–109. https://doi.org/10.1016/j.envpol.2012.01.024.

Eklöf, K., Kraus, A., Weyhenmeyer, G.A., Meili, M., Bishop, K., 2012b. Forestry influence by stump harvest and site preparation on methylmercury, total mercury and other stream water chemistry parameters across a boreal landscape. Ecosystems 15 (8), 1308–1320. https://doi.org/10.1007/s10021-012-9586-3.

Eklöf, K., Meili, M., Akerblom, S., von Bromssen, C., Bishop, K., 2013. Impact of stump harvest on run-off concentrations of total mercury and methylmercury. For. Ecol. Manag. 290, 83–94. https://doi.org/10.1016/j.foreco.2012.05.039.

Eklöf, K., Schelker, J., Sorensen, R., Meili, M., Laudon, H., von Bromssen, C., et al., 2014. Impact of forestry on total and methyl-mercury in surface waters: distinguishing effects of logging and site preparation. Environ. Sci. Technol. 48 (9), 4690–4698. https://doi.org/10.1021/es404879p.

Eklöf, K., Kraus, A., Futter, M., Schelker, J., Meili, M., Boyer, E.W., et al., 2015. Parsimonious model for simulating total mercury and methylmercury in boreal streams based on riparian flow paths and seasonality. Environ. Sci. Technol. 49 (13), 7851–7859. https://doi.org/10.1021/acs.est.5b00852.

Eklöf, K., Lidskog, R., Bishop, K., 2016. Managing Swedish forestry's impact on mercury in fish: defining the impact and mitigation measures. Ambio 45 (Suppl. 2), 163–174. https://doi.org/10.1007/s13280-015-0752-7.

Eklöf, K., Bishop, K., Bertilsson, S., Bjorn, E., Buck, M., Skyllberg, U., et al., 2018. Formation of mercury methylation hotspots as a consequence of forestry operations. Sci. Total Environ. 613-614, 1069–1078. https://doi.org/10.1016/j.scitotenv.2017.09.151.

Emmerton, C.A., Graydon, J.A., Gareis, J.A., St Louis, V.L., Lesack, L.F., Banack, J.K., et al., 2013. Mercury export to the arctic ocean from the Mackenzie River, Canada. Environ. Sci. Technol. 47 (14), 7644–7654. https://doi.org/10.1021/es400715r.

Engle, M.A., Sexauer Gustin, M., Johnson, D.W., Murphy, J.F., Miller, W.W., Walker, R.F., et al., 2006. Mercury distribution in two sierran forest and one desert sagebrush steppe ecosystems and the effects of fire. Sci. Total Environ. 367 (1), 222–233. https://doi.org/10.1016/j.scitotenv.2005.11.025.

Enrico, M., Roux, G.L., Marusczak, N., Heimburger, L.E., Claustres, A., Fu, X., et al., 2016. Atmospheric mercury transfer to peat bogs dominated by gaseous elemental mercury dry deposition. Environ. Sci. Technol. 50 (5), 2405–2412. https://doi.org/10.1021/acs.est.5b06058.

Enrico, M., Le Roux, G., Heimburger, L.E., Van Beek, P., Souhaut, M., Chmeleff, J., et al., 2017. Holocene atmospheric mercury levels reconstructed from peat bog mercury stable isotopes. Environ. Sci. Technol. 51 (11), 5899–5906. https://doi.org/10.1021/acs.est.6b05804.

Evers, D.C., Mason, R.P., Kamman, N.C., Chen, C.Y., Bogomolni, A.L., Taylor, D.L., et al., 2008. Integrated mercury monitoring program for temperate estuarine and marine ecosystems on the North American Atlantic coast. Ecohealth 5 (4), 426–441. https://doi.org/10.1007/s10393-008-0205-x.

Fain, X., Ferrari, C.P., Dommergue, A., Albert, M., Battle, M., Arnaud, L., Barnola, J.M., et al., 2008. Mercury in the snow and firn at Summit Station, central Greenland, and implications for the study of past atmospheric mercury levels. Atmos. Chem. Phys. 8, 3441–3457. https://doi.org/10.5194/acp-8-3441-2008.

Fain, X., Ferrari, C.P., Dommergue, A., Albert, M.R., Battle, M., Severinghaus, J., et al., 2009. Polar firn air reveals large-scale impact of anthropogenic mercury emissions during the 1970s. Proc. Natl. Acad. Sci. U. S. A. 106 (38), 16114–16119. https://doi.org/10.1073/pnas.0905117106.

Feng, X., Li, P., Qiu, G., Wang, S., Li, G., Shang, L., et al., 2008. Human exposure to methylmercury through rice intake in mercury mining areas, Guizhou province, China. Environ. Sci. Technol. 42 (1), 326–332. https://doi.org/10.1021/es071948x.

Feng, S.L., Ai, Z.J., Zheng, S.M., Gu, B.H., Li, Y.C., 2014. Effects of dryout and inflow water quality on mercury methylation in a constructed wetland. Water Air Soil Pollut. 225 (4). https://doi.org/10.1007/s11270-014-1929-6.

Fitzgerald, W.F., Engstrom, D.R., Lamborg, C.H., Tseng, C.M., Balcom, P.H., Hammerschmidt, C.R., 2005. Modern and historic atmospheric mercury fluxes in northern Alaska: global sources and arctic depletion. Environ. Sci. Technol. 39 (2), 557–568. https://doi.org/10.1021/es049128x.

Fleming, E.J., Mack, E.E., Green, P.G., Nelson, D.C., 2006. Mercury methylation from unexpected sources: molybdate-inhibited freshwater sediments and an iron-reducing bacterium. Appl. Environ. Microbiol. 72 (1), 457–464. https://doi.org/10.1128/AEM.72.1.457-464.2006.

Fraser, A., Dastoor, A., Ryjkov, A., 2018. How important is biomass burning in Canada to mercury contamination? Atmos. Chem. Phys. 18 (10), 7263–7286. https://doi.org/10.5194/acp-18-7263-2018.

Friedli, H.R., Radke, L.F., Prescott, R., Hobbs, P.V., Sinha, P., 2003. Mercury emissions from the August 2001 wildfires in Washington State and an agricultural waste fire in Oregon and atmospheric mercury budget estimates. Glob. Biogeochem. Cy. 17 (2). https://doi.org/10.1029/2002gb001972.

Friedli, H.R., Arellano, A.F., Cinnirella, S., Pirrone, N., 2009. Initial estimates of mercury emissions to the atmosphere from global biomass burning. Environ. Sci. Technol. 43 (10), 3507–3513. https://doi.org/10.1021/es802703g.

Fritsche, J., Obrist, D., Zeeman, M.J., Conen, F., Eugster, W., Alewell, C., 2008. Elemental mercury fluxes over a sub-alpine grassland determined with two micrometeorological methods. Atmos. Environ. 42 (13), 2922–2933. https://doi.org/10.1016/j.atmosenv.2007.12.055.

Fritsche, J., Osterwalder, S., Nilsson, M.B., Sagerfors, J., Åkerblom, S., Bishop, K., et al., 2014. Evasion of elemental mercury from a boreal peatland suppressed by long-term sulfate addition. Environ. Sci. Technol. Letters. 1 (10), 421–425. https://doi.org/10.1021/ez500223a.

Fu, J., Zhou, Q., Liu, J., Liu, W., Wang, T., Zhang, Q., et al., 2008. High levels of heavy metals in rice (*Oryza sativa L.*) from a typical e-waste recycling area in southeast China and its potential risk to human health. Chemosphere 71 (7), 1269–1275. https://doi.org/10.1016/j.chemosphere.2007.11.065.

Fu, X.W., Yang, X., Lang, X.F., Zhou, J., Zhang, H., Yu, B., et al., 2016a. Atmospheric wet and litterfall mercury deposition at urban and rural sites in China. Atmos. Chem. Phys. 16 (18), 11547–11562. https://doi.org/10.5194/acp-16-11547-2016.

Fu, X.W., Zhu, W., Zhang, H., Sommar, J., Yu, B., Yang, X., et al., 2016b. Depletion of atmospheric gaseous elemental mercury by plant uptake at Mt. Changbai, northeast China. Atmos. Chem. Phys. 16 (20), 12861–12873. https://doi.org/10.5194/acp-16-12861-2016.

Gabriel, M.C., Howard, N., Osborne, T.Z., 2014. Fish mercury and surface water sulfate relationships in the Everglades protection area. Environ. Manag. 53 (3), 583–593. https://doi.org/10.1007/s00267-013-0224-4.

Gamby, R.L., Hammerschmidt, C.R., Costello, D.M., Lamborg, C.H., Runkle, J.R., 2015. Deforestation and cultivation mobilize mercury from topsoil. Sci. Total Environ. 532, 467–473. https://doi.org/10.1016/j.scitotenv.2015.06.025.

Garcia, E., Carignan, R., 1999. Impact of wildfire and clear-cutting in the boreal forest on methyl mercury in zooplankton. Can. J. Fish. Aquat. Sci. 56 (2), 339–345. https://doi.org/10.1139/f98-164.

Garcia, E., Carignan, R., 2000. Mercury concentrations in northern pike (*Esox lucius*) from boreal lakes with logged, burned, or undisturbed catchments. Can. J. Fish. Aquat. Sci. 57 (S2), 129–135. https://doi.org/10.1139/f00-126.

Garcia, E., Carignan, R., Lean, D.R., 2007. Seasonal and inter-annual variations in methyl mercury concentrations in zooplankton from boreal lakes impacted by deforestation or natural forest fires. Environ. Monit. Assess. 131 (1–3), 1–11. https://doi.org/10.1007/s10661-006-9442-z.

Gerson, J.R., Driscoll, C.T., 2016. Is mercury in a remote forested watershed of the Adirondack mountains responding to recent decreases in emissions? Environ. Sci. Technol. 50 (20), 10943–10950. https://doi.org/10.1021/acs.est.6b02127.

Gerson, J.R., Driscoll, C.T., Hsu-Kim, H., Bernhardt, E.S., 2018. Senegalese artisanal gold mining leads to elevated total mercury and methylmercury concentrations in soils, sediments, and rivers. Elem. Sci. Anth. 6 (1). https://doi.org/10.1525/elementa.274.

Gilmour, C.C., Henry, E.A., 1991. Mercury methylation in aquatic systems affected by acid deposition. Environ. Pollut. 71 (2–4), 131–169. https://doi.org/10.1016/0269-7491(91)90031-q.

Gilmour, C.C., Henry, E.A., Mitchell, R., 1992. Sulfate stimulation of mercury methylation in freshwater sediments. Environ. Sci. Technol. 26 (11), 2281–2287. https://doi.org/10.1021/es00035a029.

Gilmour, C.C., Riedel, G.S., Ederington, M.C., Bell, J.T., Gill, G.A., Stordal, M.C., 1998. Methylmercury concentrations and production rates across a trophic gradient in the northern Everglades. Biogeochemistry 40 (2/3), 327–345. https://doi.org/10.1023/a:1005972708616.

Gilmour, C.C., Podar, M., Bullock, A.L., Graham, A.M., Brown, S.D., Somenahally, A.C., et al., 2013. Mercury methylation by novel microorganisms from new environments. Environ. Sci. Technol. 47 (20), 11810–11820. https://doi.org/10.1021/es403075t.

Golden, H.E., Knightes, C.D., Conrads, P.A., Davis, G.M., Feaster, T.D., Journey, C.A., et al., 2012. Characterizing mercury concentrations and fluxes in a coastal plain watershed: insights from dynamic modeling and data. J. Geophys. Res.-Biogeosci. 117 (G1). https://doi.org/10.1029/2011jg001806.

Golden, H.E., Knightes, C.D., Conrads, P.A., Feaster, T.D., Davis, G.M., Benedict, S.T., et al., 2013. Climate change and watershed mercury export: a multiple projection and model analysis. Environ. Toxicol. Chem. 32 (9), 2165–2174. https://doi.org/10.1002/etc.2284.

Graham, A.M., Aiken, G.R., Gilmour, C.C., 2012. Dissolved organic matter enhances microbial mercury methylation under sulfidic conditions. Environ. Sci. Technol. 46 (5), 2715–2723. https://doi.org/10.1021/es203658f.

Graydon, J.A., St Louis, V.L., Hintelmann, H., Lindberg, S.E., Sandilands, K.A., Rudd, J.W., et al., 2008. Long-term wet and dry deposition of total and methyl mercury in the remote boreal ecoregion of Canada. Environ. Sci. Technol. 42 (22), 8345–8351. https://doi.org/10.1021/es801056j.

Graydon, J.A., St. Louis, V.L., Lindberg, S.E., Sandilands, K.A., Rudd, J.W.M., Kelly, C.A., et al., 2012. The role of terrestrial vegetation in atmospheric Hg deposition: pools and fluxes of spike and ambient Hg from the METAALICUS experiment. Glob. Biogeochem. Cy 26 (1). https://doi.org/10.1029/2011gb004031.

Grigal, D.F., 2003. Mercury sequestration in forests and peatlands: a review. J. Environ. Qual. 32 (2), 393–405. https://doi.org/10.2134/jeq2003.3930.

Gustin, M.S., Stamenkovic, J., 2005. Effect of watering and soil moisture on mercury emissions from soils. Biogeochemistry 76 (2), 215–232. https://doi.org/10.1007/s10533-005-4566-8.

Gustin, M.S., Biester, H., Kim, C.S., 2002. Investigation of the light-enhanced emission of mercury from naturally enriched substrates. Atmos. Environ. 36 (20), 3241–3254. https://doi.org/10.1016/s1352-2310(02)00329-1.

Gustin, M.S., Huang, J., Miller, M.B., Peterson, C., Jaffe, D.A., Ambrose, J., et al., 2013. Do we understand what the mercury speciation instruments are actually measuring? Results of RAMIX. Environ. Sci. Technol. 47 (13), 7295–7306. https://doi.org/10.1021/es3039104.

Gustin, M.S., Amos, H.M., Huang, J., Miller, M.B., Heidecorn, K., 2015. Measuring and modeling mercury in the atmosphere: a critical review. Atmos. Chem. Phys. 15 (10), 5697–5713. https://doi.org/10.5194/acp-15-5697-2015.

Ha, E., Basu, N., Bose-O'Reilly, S., Dorea, J.G., McSorley, E., Sakamoto, M., et al., 2017. Current progress on understanding the impact of mercury on human health. Environ. Res. 152, 419–433. https://doi.org/10.1016/j.envres.2016.06.042.

Hall, B.D., Louis, V.L.S., Rolfhus, K.R., Bodaly, R.A., Beaty, K.G., Paterson, M.J., et al., 2005. Impacts of reservoir creation on the biogeochemical cycling of methyl mercury and total mercury in boreal upland forests. Ecosystems 8 (3), 248–266. https://doi.org/10.1007/s10021-003-0094-3.

Hall, B.D., Cherewyk, K.A., Paterson, M.J., Bodaly, R.A., 2009. Changes in methyl mercury concentrations in zooplankton from four experimental reservoirs with differing amounts of carbon in the flooded catchments. Can. J. Fish. Aquat. Sci. 66 (11), 1910–1919. https://doi.org/10.1139/f09-123.

Hammerschmidt, C.R., Fitzgerald, W.F., 2008. Methylmercury in arctic Alaskan mosquitoes: implications for impact of atmospheric mercury depletion events. Environ. Chem. 5 (2), 127–130. https://doi.org/10.1071/en08003.

Hansen, M.C., Potapov, P.V., Moore, R., Hancher, M., Turubanova, S.A., Tyukavina, A., et al., 2013. High-resolution global maps of 21st-century forest cover change. Science 342 (6160), 850–853. https://doi.org/10.1126/science.1244693.

Harris, R.C., Rudd, J.W., Amyot, M., Babiarz, C.L., Beaty, K.G., Blanchfield, P.J., et al., 2007. Whole-ecosystem study shows rapid fish-mercury response to changes in mercury deposition. Proc. Natl. Acad. Sci. U. S. A. 104 (42), 16586–16591. https://doi.org/10.1073/pnas.0704186104.

Haynes, K.M., Kane, E.S., Potvin, L., Lilleskov, E.A., Kolka, R.K., Mitchell, C.P.J., 2017a. Gaseous mercury fluxes in peatlands and the potential influence of climate change. Atmos. Environ. 154, 247–259. https://doi.org/10.1016/j.atmosenv.2017.01.049.

Haynes, K.M., Kane, E.S., Potvin, L., Lilleskov, E.A., Kolka, R.K., Mitchell, C.P.J., 2017b. Mobility and transport of mercury and methylmercury in peat as a function of changes in water table regime and plant functional groups. Glob. Biogeochem. Cy. 31 (2), 233–244. https://doi.org/10.1002/2016gb005471.

Hintelmann, H., Harris, R., Heyes, A., Hurley, J.P., Kelly, C.A., Krabbenhoft, D.P., et al., 2002. Reactivity and mobility of new and old mercury deposition in a boreal forest ecosystem during the first year of the METAALICUS study. Environ. Sci. Technol. 36 (23), 5034–5040. https://doi.org/10.1021/es025572t.

Hofacker, A.F., Voegelin, A., Kaegi, R., Kretzschmar, R., 2013. Mercury mobilization in a flooded soil by incorporation into metallic copper and metal sulfide nanoparticles. Environ. Sci. Technol. 47 (14), 7739–7746. https://doi.org/10.1021/es4010976.

Hoggarth, C.G., Hall, B.D., Mitchell, C.P., 2015. Mercury methylation in high and low-sulphate impacted wetland ponds within the prairie pothole region of North America. Environ. Pollut. 205, 269–277. https://doi.org/10.1016/j.envpol.2015.05.046.

Holloway, J.M., Pribil, M.J., McCleskey, R.B., Etheridge, A.B., Krabbenhoft, D.P., Aiken, G.R., 2017. Mobilization of mercury and arsenic from a carbonate-hosted ore deposit, central Idaho, USA. In: Marques, J.M., Chambel, A. (Eds.), 15th Water-Rock Interaction International Symposium, WRI-15. 17, pp. 610–613.

Homann, P.S., Darbyshire, R.L., Bormann, B.T., Morrissette, B.A., 2015. Forest structure affects soil mercury losses in the presence and absence of wildfire. Environ. Sci. Technol. 49 (21), 12714–12722. https://doi.org/10.1021/acs.est.5b03355.

Horowitz, H.M., Jacob, D.J., Amos, H.M., Streets, D.G., Sunderland, E.M., 2014. Historical mercury releases from commercial products: global environmental implications. Environ. Sci. Technol. 48 (17), 10242–10250. https://doi.org/10.1021/es501337j.

Horowitz, H.M., Jacob, D.J., Zhang, Y., Dibble, T.S., Slemr, F., Amos, H.M., et al., 2017. A new mechanism for atmospheric mercury redox chemistry: implications for the global mercury budget. Atmos. Chem. Phys. 17 (10), 6353–6371. https://doi.org/10.5194/acp-17-6353-2017.

Horvat, M., Nolde, N., Fajon, V., Jereb, V., Logar, M., Lojen, S., et al., 2003. Total mercury, methylmercury and selenium in mercury polluted areas in the province Guizhou, China. Sci. Total Environ. 304 (1–3), 231–256. https://doi.org/10.1016/S0048-9697(02)00572-7.

Hsu-Kim, H., Eckley, C.S., Acha, D., Feng, X., Gilmour, C.C., Jonsson, S., Mitchell, C.P.J., 2018. Challenges and opportunities for managing aquatic mercury pollution in altered landscapes. Ambio 47 (2), 141–169. https://doi.org/10.1007/s13280-017-1006-7.

Hu, H., Wang, B., Bravo, A.G., Bjorn, E., Skyllberg, U., Amouroux, D., et al., 2020. Shifts in mercury methylation across a peatland chronosequence: from sulfate reduction to methanogenesis and syntrophy. J. Hazard. Mat. 387, 121967. https://doi.org/10.1016/j.jhazmat.2019.121967.

Huang, J., Miller, M.B., Weiss-Penzias, P., Gustin, M.S., 2013. Comparison of gaseous oxidized Hg measured by KCl-coated denuders, and nylon and cation exchange membranes. Environ. Sci. Technol. 47 (13), 7307–7316. https://doi.org/10.1021/es4012349.

Hurley, J.P., Benoit, J.M., Babiarz, C.L., Shafer, M.M., Andren, A.W., Sullivan, J.R., Hammond, R., Webb, D.A., 1995. Influences of watershed characteristics on mercury levels in Wisconsin rivers. Environ. Sci. Technol. 29 (7), 1867–1875.

IPCC. 2018. Global Warming of 1.5°C. an IPCC Special Report on the Impacts of Global Warming of 1.5°C above Pre-Industrial Levels and Related Global Greenhouse Gas Emission Pathways, in the Context of Strengthening the Global Response to the Threat of Climate Change, Sustainable Development, and Efforts to Eradicate Poverty. Masson-Delmotte V., Zhai P., Pörtner H.-O., Roberts D., Skea J., Shukla P.R., Pirani A., Moufouma-Okia W., Péan C., Pidcock R., Connors S., Matthews J.B.R., Chen Y., Zhou X., Gomis M.I., Lonnoy E., Maycock T., Tignor M., Waterfield T., editors. (In Press).

Jaffe, D.A., Lyman, S., Amos, H.M., Gustin, M.S., Huang, J., Selin, N.E., et al., 2014. Progress on understanding atmospheric mercury hampered by uncertain measurements. Environ. Sci Technol. 48 (13), 7204–7206. https://doi.org/10.1021/es5026432.

Jensen, A.M., Scanlon, T.M., Riscassi, A.L., 2017. Emerging investigator series: the effect of wildfire on streamwater mercury and organic carbon in a forested watershed in the southeastern United States. Environ. Sci. Process. Impacts. 19 (12), 1505–1517. https://doi.org/10.1039/c7em00419b.

Jeremiason, J.D., Engstrom, D.R., Swain, E.B., Nater, E.A., Johnson, B.M., Almendinger, J.E., et al., 2006. Sulfate addition increases methylmercury production in an experimental wetland. Environ. Sci. Technol. 40 (12), 3800–3806. https://doi.org/10.1021/es0524144.

Jia, G.J., Epstein, H.E., Walker, D.A., 2003. Greening of arctic Alaska, 1981-2001. Geophys. Res. Lett. 30 (20). https://doi.org/10.1029/2003gl018268.

Jiskra, M., Wiederhold, J.G., Skyllberg, U., Kronberg, R.M., Hajdas, I., Kretzschmar, R., 2015. Mercury deposition and re-emission pathways in boreal forest soils investigated with Hg isotope signatures. Environ. Sci. Technol. 49 (12), 7188–7196. https://doi.org/10.1021/acs.est.5b00742.

Jiskra, M., Wiederhold, J.G., Skyllberg, U., Kronberg, R.M., Kretzschmar, R., 2017. Source tracing of natural organic matter bound mercury in boreal forest runoff with mercury stable isotopes. Environ. Sci. Process. Impacts. 19 (10), 1235–1248. https://doi.org/10.1039/c7em00245a.

Jiskra, M., Sonke, J.E., Obrist, D., Bieser, J., Ebinghaus, R., Myhre, C.L., et al., 2018. A vegetation control on seasonal variations in global atmospheric mercury concentrations. Nat. Geosci. 11 (4), 244–250. https://doi.org/10.1038/s41561-018-0078-8.

Jiskra, M., Sonke, J.E., Agnan, Y., Helmig, D., Obrist, D., 2019. Insights from mercury stable isotopes on terrestrial–atmosphere exchange of Hg(0) in the arctic tundra. Biogeosciences 16 (20), 4051–4064. https://doi.org/10.5194/bg-16-4051-2019.

Johnson, N.W., Mitchell, C.P., Engstrom, D.R., Bailey, L.T., Coleman Wasik, J.K., Berndt, M.E., 2016. Methylmercury production in a chronically sulfate-impacted sub-boreal wetland. Environ. Sci. Process. Impacts. 18 (6), 725–734. https://doi.org/10.1039/c6em00138f.

Jones, D.S., Walker, G.M., Johnson, N.W., Mitchell, C.P.J., Coleman Wasik, J.K., Bailey, J.V., 2019. Molecular evidence for novel mercury methylating microorganisms in sulfate-impacted lakes. ISME J 13, 1659–1675. https://doi.org/10.1038/s41396-019-0376-1.

Kääb, A., Chiarle, M., Raup, B., Schneider, C., 2007. Climate change impacts on mountain glaciers and permafrost. Glob. Planet. Change. 56 (1–2), vii–ix. https://doi.org/10.1016/j.gloplacha.2006.07.008.

Kamp, J., Skov, H., Jensen, B., Sørensen, L.L., 2018. Fluxes of gaseous elemental mercury (GEM) in the high arctic during atmospheric mercury depletion events (AMDEs). Atmos. Chem. Phys. 18 (9), 6923–6938. https://doi.org/10.5194/acp-18-6923-2018.

Kelly, E.N., Schindler, D.W., St Louis, V.L., Donald, D.B., Vladicka, K.E., 2006. Forest fire increases mercury accumulation by fishes via food web restructuring and increased mercury inputs. Proc. Natl. Acad. Sci. U. S. A. 103 (51), 19380–19385. https://doi.org/10.1073/pnas.0609798104.

Khan, T.R., Obrist, D., Agnan, Y., Selin, N.E., Perlinger, J.A., 2019. Atmosphere-terrestrial exchange of gaseous elemental mercury: parameterization improvement through direct comparison with measured ecosystem fluxes. Environ. Sci.: Processes Impacts 21, 1699–1712. https://doi.org/10.1039/C9EM00341J.

Kim, K.H., Yoon, H.O., Jung, M.C., Oh, J.M., Brown, R.J., 2012. A simple approach for measuring emission patterns of vapor phase mercury under temperature-controlled conditions from soil. Sci. World J. 940413. https://doi.org/10.1100/2012/940413.

Klaminder, J., Yoo, K., Rydberg, J., Giesler, R., 2008. An explorative study of mercury export from a thawing palsa mire. J. Geophys. Res.-Biogeosci. 113 (G4). https://doi.org/10.1029/2008jg000776.

Knightes, C.D., Ambrose Jr., R.B., 2007. Evaluating regional predictive capacity of a process-based mercury exposure model, regional-mercury cycling model, applied to 91 Vermont and New Hampshire lakes and ponds, USA. Environ. Toxicol. Chem. 26 (4), 807–815. https://doi.org/10.1897/06-317r.1.

Knightes, C.D., Sunderland, E.M., Craig Barber, M., Johnston, J.M., Ambrose, R.B., 2009. Application of ecosystem-scale fate and bioaccumulation models to predict fish mercury response times to changes in atmospheric deposition. Environ. Toxicol. Chem. 28 (4), 881–893. https://doi.org/10.1897/08-242R.1.

Knightes, C.D., Golden, H.E., Journey, C.A., Davis, G.M., Conrads, P.A., Marvin-DiPasquale, M., et al., 2014. Mercury and methylmercury stream concentrations in a coastal plain watershed: a multi-scale simulation analysis. Environ. Pollut. 187, 182–192. https://doi.org/10.1016/j.envpol.2013.12.026.

Knorr, W., Prentice, I.C., House, J.I., Holland, E.A., 2005. Long-term sensitivity of soil carbon turnover to warming. Nature 433 (7023), 298–301. https://doi.org/10.1038/nature03226.

Kocman, D., Horvat, M., Pirrone, N., Cinnirella, S., 2013. Contribution of contaminated sites to the global mercury budget. Environ. Res. 125, 160–170. https://doi.org/10.1016/j.envres.2012.12.011.

Kocman, D., Wilson, S.J., Amos, H.M., Telmer, K.H., Steenhuisen, F., Sunderland, E.M., et al., 2017. Toward an assessment of the global inventory of present-day mercury releases to freshwater environments. Int. J. Environ. Res. Public Health 14 (2). https://doi.org/10.3390/ijerph14020138.

Kohlenberg, A.J., Turetsky, M.R., Thompson, D.K., Branfireun, B.A., Mitchell, C.P.J., 2018. Controls on boreal peat combustion and resulting emissions of carbon and mercury. Environ. Res. Lett. 13 (3). https://doi.org/10.1088/1748-9326/aa9ea8.

Kolka, R.K., Sturtevant, B.R., Miesel, J.R., Singh, A., Wolter, P.T., Fraver, S., et al., 2017. Emissions of forest floor and mineral soil carbon, nitrogen and mercury pools and relationships with fire severity for the Pagami Creek fire in the boreal forest of northern Minnesota. Int. J. Wild. Fire. 26 (4), 296–305. https://doi.org/10.1071/wf16128.

Koven, C.D., Riley, W.J., Stern, A., 2013. Analysis of permafrost thermal dynamics and response to climate change in the CMIP5 earth system models. J. Clim. 26 (6), 1877–1900. https://doi.org/10.1175/jcli-d-12-00228.1.

Krisnayanti, B.D., Anderson, C.W., Utomo, W.H., Feng, X., Handayanto, E., Mudarisna, N., et al., 2012. Assessment of environmental mercury discharge at a four-year-old artisanal gold mining area on Lombok Island, Indonesia. J. Environ. Monit. 14 (10), 2598–2607. https://doi.org/10.1039/c2em30515a.

Kronberg, R.-M., Drott, A., Jiskra, M., Wiederhold, J.G., Björn, E., Skyllberg, U., 2016a. Forest harvest contribution to boreal freshwater methyl mercury load. Glob. Biogeochem. Cy. 30 (6), 825–843. https://doi.org/10.1002/2015gb005316.

Kronberg, R.M., Jiskra, M., Wiederhold, J.G., Bjorn, E., Skyllberg, U., 2016b. Methyl mercury formation in hillslope soils of boreal forests: the role of forest harvest and anaerobic microbes. Environ. Sci. Technol. 50 (17), 9177–9186. https://doi.org/10.1021/acs.est.6b00762.

Kumar, A., Wu, S., Huang, Y., Liao, H., Kaplan, J.O., 2018. Mercury from wildfires: global emission inventories and sensitivity to 2000–2050 global change. Atmos. Environ. 173, 6–15. https://doi.org/10.1016/j.atmosenv.2017.10.061.

Kwasigroch, U., Beldowska, M., Jedruch, A., Saniewska, D., 2018. Coastal erosion-a "new" land-based source of labile mercury to the marine environment. Environ. Sci. Pollut. Res. Int. 25 (28), 28682–28694. https://doi.org/10.1007/s11356-018-2856-7.

Lalonde, J.D., Poulain, A.J., Amyot, M., 2002. The role of mercury redox reactions in snow on snow-to-air mercury transfer. Environ. Sci. Technol. 36 (2), 174–178. https://doi.org/10.1021/es010786g.

Lan, X., Talbot, R., Castro, M., Perry, K., Luke, W., 2012. Seasonal and diurnal variations of atmospheric mercury across the US determined from AMNet monitoring data. Atmos. Chem. Phys. 12, 10569–10582. https://doi.org/10.5194/acp-12-10569-2012.

Landis, M.S., Stevens, R.K., Schaedlich, F., Prestbo, E.M., 2002. Development and characterization of an annular denuder methodology for the measurement of divalent inorganic reactive gaseous mercury in ambient air. Environ. Sci. Technol. 36 (13), 3000–3009. https://doi.org/10.1021/es015887t.

Lavoie, R.A., Amyot, M., Lapierre, J.F., 2019. Global meta-analysis on the relationship between mercury and dissolved organic carbon in freshwater environments. J. Geophys. Res.-Biogeosci. 124 (6), 1508–1523. https://doi.org/10.1029/2018jg004896.

Lazaro, W.L., Diez, S., da Silva, C.J., Ignacio, A.R.A., Guimaraes, J.R.D., 2018. Seasonal changes in peryphytic microbial metabolism determining mercury methylation in a tropical wetland. Sci. Total Environ. 627, 1345–1352. https://doi.org/10.1016/j.scitotenv.2018.01.186.

Lee, Y., Bishop, K.H., Munthe, J., 2000. Do concepts about catchment cycling of methylmercury and mercury in boreal catchments stand the test of time? Six years of atmospheric inputs and runoff export at Svartberget. northern Sweden. Sci. Total Environ 260 (1–3), 11–20. https://doi.org/10.1016/s0048-9697(00)00538-6.

Lescord, G.L., Emilson, E.J.S., Johnston, T.A., Branfireun, B.A., Gunn, J.M., 2018. Optical properties of dissolved organic matter and their relation to mercury concentrations in water and biota across a remote freshwater drainage basin. Environ. Sci. Technol. 52 (6), 3344–3353. https://doi.org/10.1021/acs.est.7b05348.

Levanoni, O., Bishop, K., McKie, B.G., Hartman, G., Eklof, K., Ecke, F., 2015. Impact of beaver pond colonization history on methylmercury concentrations in surface water. Environ. Sci. Technol. 49 (21), 12679–12687. https://doi.org/10.1021/acs.est.5b03146.

Li, P., Feng, X., Qiu, G., 2010. Methylmercury exposure and health effects from rice and fish consumption: a review. Int. J. Res. Public Health. 7 (6), 2666–2691. https://doi.org/10.3390/ijerph7062666.

Li, P., Feng, X., Yuan, X., Chan, H.M., Qiu, G., Sun, G.X., et al., 2012. Rice consumption contributes to low level methylmercury exposure in southern China. Environ. Int. 49, 18–23. https://doi.org/10.1016/j.envint.2012.08.006.

Li, B., Shi, J.B., Wang, X., Meng, M., Huang, L., Qi, X.L., et al., 2013. Variations and constancy of mercury and methylmercury accumulation in rice grown at contaminated paddy field sites in three provinces of China. Environ. Pollut. 181, 91–97. https://doi.org/10.1016/j.envpol.2013.06.021.

Li, P., Feng, X., Chan, H.M., Zhang, X., Du, B., 2015a. Human body burden and dietary methylmercury intake: the relationship in a rice-consuming population. Environ. Sci. Technol. 49 (16), 9682–9689. https://doi.org/10.1021/acs.est.5b00195.

Li, Y.-F., Zhao, J., Li, Y., Li, H., Zhang, J., Li, B., et al., 2015b. The concentration of selenium matters: a field study on mercury accumulation in rice by selenite treatment in Qingzhen, Guizhou, China. Plant Soil 391 (1–2), 195–205. https://doi.org/10.1007/s11104-015-2418-4.

Li, P., Du, B., Maurice, L., Laffont, L., Lagane, C., Point, D., et al., 2017a. Mercury isotope signatures of methylmercury in rice samples from the Wanshan mercury mining area, China: environmental implications. Environ. Sci. Technol. 51 (21), 12321–12328. https://doi.org/10.1021/acs.est.7b03510.

Li, Y., Zhao, J., Guo, J., Liu, M., Xu, Q., Li, H., et al., 2017b. Influence of sulfur on the accumulation of mercury in rice plant (*Oryza sativa L.*) growing in mercury contaminated soils. Chemosphere 182, 293–300. https://doi.org/10.1016/j.chemosphere.2017.04.129.

Liang, P., Feng, X., Zhang, C., Zhang, J., Cao, Y., You, Q., et al., 2015. Human exposure to mercury in a compact fluorescent lamp manufacturing area: by food (rice and fish) consumption and occupational exposure. Environ. Pollut. 198, 126–132. https://doi.org/10.1016/j.envpol.2014.12.036.

Liem-Nguyen, V., Skyllberg, U., Bjorn, E., 2017. Thermodynamic modeling of the solubility and chemical speciation of mercury and methylmercury driven by organic thiols and micromolar sulfide concentrations in boreal wetland soils. Environ. Sci. Technol. 51 (7), 3678–3686. https://doi.org/10.1021/acs.est.6b04622.

Lin, C.-J., Pongprueksa, P., Lindberg, S.E., Pehkonen, S.O., Byun, D., Jang, C., 2006. Scientific uncertainties in atmospheric mercury models I: model science evaluation. Atmos. Environ. 40 (16), 2911–2928. https://doi.org/10.1016/j.atmosenv.2006.01.009.

Lin, C.J., Gustin, M.S., Singhasuk, P., Eckley, C., Miller, M., 2010. Empirical models for estimating mercury flux from soils. Environ. Sci. Technol. 44 (22), 8522–8528. https://doi.org/10.1021/es1021735.

Lindberg, S.E., Zhang, H., 2000. Air/water exchange of mercury in the Everglades II: measuring and modeling evasion of mercury from surface waters in the Everglades nutrient removal project. Sci. Total Environ. 259 (1–3), 135–143. https://doi.org/10.1016/s0048-9697(00)00586-6.

Lindberg, S.E., Brooks, S., Lin, C.J., Scott, K.J., Landis, M.S., Stevens, R.K., et al., 2002a. Dynamic oxidation of gaseous mercury in the arctic troposphere at polar sunrise. Environ. Sci. Technol. 36 (6), 1245–1256. https://doi.org/10.1021/es0111941.

Lindberg, S.E., Dong, W., Meyers, T., 2002b. Transpiration of gaseous elemental mercury through vegetation in a subtropical wetland in Florida. Atmos. Environ. 36 (33), 5207–5219.

Lindberg, S., Bullock, R., Ebinghaus, R., Engstrom, D., Feng, X., Fitzgerald, W., et al., 2007. A synthesis of progress and uncertainties in attributing the sources of mercury in deposition. Ambio 36 (1), 19–32. https://doi.org/10.1579/0044-7447(2007)36[19:asopau]2.0.co;2.

Lindqvist, O., Johansson, K., Bringmark, L., Timm, B., Aastrup, M., Andersson, A., et al., 1991. Mercury in the Swedish environment? Recent research on causes, consequences and corrective methods. Water Air Soil Pollut. 55 (1–2), xi-261. https://doi.org/10.1007/bf00542429.

Liu, Y.R., Yu, R.Q., Zheng, Y.M., He, J.Z., 2014a. Analysis of the microbial community structure by monitoring an hg methylation gene (*hgcA*) in paddy soils along an Hg gradient. Appl. Environ. Microbiol. 80 (9), 2874–2879. https://doi.org/10.1128/AEM.04225-13.

Liu, Y.R., Zheng, Y.M., Zhang, L.M., He, J.Z., 2014b. Linkage between community diversity of sulfate-reducing microorganisms and methylmercury concentration in paddy soil. Environ. Sci. Pollut. Res. Int. 21 (2), 1339–1348. https://doi.org/10.1007/s11356-013-1973-6.

Liu, X., Ma, A., Zhuang, G., Zhuang, X., 2018a. Diversity of microbial communities potentially involved in mercury methylation in rice paddies surrounding typical mercury mining areas in China. Microbiol. Open. 7 (4), e00577. https://doi.org/10.1002/mbo3.577.

Liu, Y.R., Johs, A., Bi, L., Lu, X., Hu, H.W., Sun, D., et al., 2018b. Unraveling microbial communities associated with methylmercury production in paddy soils. Environ. Sci. Technol. 52 (22), 13110–13118. https://doi.org/10.1021/acs.est.8b03052.

Lyman, S.N., Cheng, I., Gratz, L.E., Weiss-Penzias, P., Zhang, L., 2020. An updated review of atmospheric mercury. Sci. Total Environ. 707, 135575. https://doi.org/10.1016/j.scitotenv.2019.135575.

Ma, M., Wang, D., Du, H., Sun, T., Zhao, Z., Wang, Y., et al., 2016. Mercury dynamics and mass balance in a subtropical forest, southwestern China. Atmos. Chem. Phys. 16 (7), 4529–4537. https://doi.org/10.5194/acp-16-4529-2016.

Ma, M., Sun, T., Du, H., Wang, D., 2018. A two-year study on mercury fluxes from the soil under different vegetation cover in a subtropical region, south China. Atmosphere 9 (1). https://doi.org/10.3390/atmos9010030.

Mack, M.C., Bret-Harte, M.S., Hollingsworth, T.N., Jandt, R.R., Schuur, E.A., Shaver, G.R., et al., 2011. Carbon loss from an unprecedented arctic tundra wildfire. Nature 475 (7357), 489–492. https://doi.org/10.1038/nature10283.

MacSween, K., Edwards, G.C., Howard, D.A., 2020. Up-scaling mercury emissions from terrestrial surfaces as a response to sustained temperature increase. Atmos. Environ. 223, 117190.

Magnusson, T.P.T., 2017. Long-term effects of stump harvest on mercury and general chemistry in discharge water. Scandinavian J. For. Res. 32 (3), 230–238. https://doi.org/10.1080/02827581.2016.1244287.

Manceau, A., Merkulova, M., Murdzek, M., Batanova, V., Baran, R., Glatzel, P., et al., 2018. Chemical forms of mercury in pyrite: implications for predicting mercury releases in acid mine drainage settings. Environ. Sci. Technol. 52 (18), 10286–10296. https://doi.org/10.1021/acs.est.8b02027.

Mao, H., Ye, Z., Driscoll, C., 2017. Meteorological effects on Hg wet deposition in a forested site in the Adirondack region of New York during 2000–2015. Atmos. Environ. 168, 90–100. https://doi.org/10.1016/j.atmosenv.2017.08.058.

Margono, B.A., Potapov, P.V., Turubanova, S., Stolle, F., Hansen, M.C., 2014. Primary forest cover loss in Indonesia over 2000–2012. Nat. Clim. Chang. 4 (8), 730–735. https://doi.org/10.1038/nclimate2277.

Marvin-DiPasquale, M., Agee, J., McGowan, C., Oremland, R.S., Thomas, M., Krabbenhoft, D., et al., 2000. Methyl-mercury degradation pathways: a comparison among three mercury-impacted ecosystems. Environ. Sci. Technol. 34 (23), 4908–4916. https://doi.org/10.1021/es0013125.

Marvin-Dipasquale, M., Lutz, M.A., Brigham, M.E., Krabbenhoft, D.P., Aiken, G.R., Orem, W.H., et al., 2009. Mercury cycling in stream ecosystems. 2. Benthic methylmercury production and bed sediment-pore water partitioning. Environ. Sci. Technol. 43 (8), 2726–2732. https://doi.org/10.1021/es802698v.

Marvin-DiPasquale, M., Windham-Myers, L., Agee, J.L., Kakouros, E., Kieu le, H., Fleck, J.A., et al., 2014. Methylmercury production in sediment from agricultural and non-agricultural wetlands in the Yolo Bypass, California, USA. Sci. Total Environ. 484, 288–299. https://doi.org/10.1016/j.scitotenv.2013.09.098.

Mazur, M., Mitchell, C.P.J., Eckley, C.S., Eggert, S.L., Kolka, R.K., Sebestyen, S.D., et al., 2014. Gaseous mercury fluxes from forest soils in response to forest harvesting intensity: a field manipulation experiment. Sci. Total Environ. 496, 678–687. https://doi.org/10.1016/j.scitotenv.2014.06.058.

McCarter, C.P.R., Branfireun, B.A., Price, J.S., 2017. Nutrient and mercury transport in a sub-arctic ladder fen peatland subjected to simulated wastewater discharges. Sci. Total Environ. 609, 1349–1360. https://doi.org/10.1016/j.scitotenv.2017.07.225.

Meili, M., Bishop, K., Bringmark, L., Johansson, K., Munthe, J., Sverdrup, H., et al., 2003. Critical levels of atmospheric pollution: criteria and concepts for operational modelling of mercury in forest and lake ecosystems. Sci. Total Environ. 304 (1–3), 83–106. https://doi.org/10.1016/S0048-9697(02)00559-4.

Melillo, J.M., Steudler, P.A., Aber, J.D., Newkirk, K., Lux, H., Bowles, F.P., et al., 2002. Soil warming and carbon-cycle feedbacks to the climate system. Science 298 (5601), 2173–2176. https://doi.org/10.1126/science.1074153.

Meng, B., Feng, X., Qiu, G., Cai, Y., Wang, D., Li, P., et al., 2010. Distribution patterns of inorganic mercury and methylmercury in tissues of rice (*Oryza sativa L.*) plants and possible bioaccumulation pathways. J. Agric. Food Chem. 58 (8), 4951–4958. https://doi.org/10.1021/jf904557x.

Meng, B., Feng, X., Qiu, G., Liang, P., Li, P., Chen, C., et al., 2011. The process of methylmercury accumulation in rice (*Oryza sativa L.*). Environ. Sci. Technol. 45 (7), 2711–2717. https://doi.org/10.1021/es103384v.

Meng, B., Feng, X., Qiu, G., Anderson, C.W., Wang, J., Zhao, L., 2014a. Localization and speciation of mercury in brown rice with implications for pan-Asian public health. Environ. Sci. Technol. 48 (14), 7974–7981. https://doi.org/10.1021/es502000d.

Meng, M., Li, B., Shao, J.J., Wang, T., He, B., Shi, J.B., et al., 2014b. Accumulation of total mercury and methylmercury in rice plants collected from different mining areas in China. Environ. Pollut. 184, 179–186. https://doi.org/10.1016/j.envpol.2013.08.030.

Millard, G.D., Driscoll, C.T., Burns, D.A., Montesdeoca, M.R., Riva-Murray, K., 2018. Response of mercury in an Adirondack (NY, USA) forest stream to watershed lime application. Environ. Sci. Process. Impacts. 20 (4), 607–620. https://doi.org/10.1039/c7em00520b.

Millhollen, A.G., Gustin, M.S., Obrist, D., 2006. Foliar mercury accumulation and exchange for three tree species. Environ. Sci. Technol. 40 (19), 6001–6006. https://doi.org/10.1021/es0609194.

Mitchard, E.T.A., 2018. The tropical forest carbon cycle and climate change. Nature 559 (7715), 527–534. https://doi.org/10.1038/s41586-018-0300-2.

Mitchell, C.P.J., Branfireun, B.A., Kolka, R.K., 2008. Total mercury and methylmercury dynamics in upland–peatland watersheds during snowmelt. Biogeochemistry 90 (3), 225–241. https://doi.org/10.1007/s10533-008-9246-z.

Monteith, D.T., Stoddard, J.L., Evans, C.D., de Wit, H.A., Forsius, M., Hogasen, T., et al., 2007. Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. Nature 450 (7169), 537–540. https://doi.org/10.1038/nature06316.

Moreno, C.E., Fjeld, E., Lydersen, E., 2016. The effects of wildfire on mercury and stable isotopes (delta^{15}N, delta^{13}C) in water and biota of small boreal, acidic lakes in southern Norway. Environ. Monit. Assess. 188 (3), 178. https://doi.org/10.1007/s10661-016-5148-z.

Munthe, J., Hultberg, H., 2004. Mercury and methylmercury in runoff from a forested catchment - concentrations, fluxes, and their response to manipulations. Water Air Soil Pollut. Focus. 4, 607–618.

Munthe, J., Bodaly, R.A., Branfireun, B.A., Driscoll, C.T., Gilmour, C.C., Harris, R., et al., 2007. Recovery of mercury-contaminated fisheries. Ambio 36 (1), 33–44. https://doi.org/10.1579/0044-7447(2007)36[33:Romf]2.0.Co;2.

Murray, C.D., Buttle, J.M., 2003. Impacts of clearcut harvesting on snow accumulation and melt in a northern hardwood forest. J. Hydrol. 271 (1–4), 197–212 (doi:Pii S0022-1694(02)00352-9).

Myrbo, A., Swain, E.B., Johnson, N.W., Engstrom, D.R., Pastor, J., Dewey, B., et al., 2017. Increase in nutrients, mercury, and methylmercury as a consequence of elevated sulfate reduction to sulfide in experimental wetland mesocosms. J. Geophys. Res. Biogeosci. 122 (11), 2769–2785. https://doi.org/10.1002/2017jg003788.

Nagorski, S.A., Engstrom, D.R., Hudson, J.P., Krabbenhoft, D.P., Hood, E., DeWild, J.F., et al., 2014. Spatial distribution of mercury in southeastern Alaskan streams influenced by glaciers, wetlands, and salmon. Environ. Pollut. 184, 62–72. https://doi.org/10.1016/j.envpol.2013.07.040.

Nagy, K.L., Manceau, A., Gasper, J.D., Ryan, J.N., Aiken, G.R., 2011. Metallothionein-like multinuclear clusters of mercury(ii) and sulfur in peat. Environ. Sci. Technol. 45 (17), 7298–7306. https://doi.org/10.1021/es201025v.

Nartey, V.K., Klake, R.K., Hayford, E.K., Doamekpo, R.L.K., Appoh, R.K., 2011. Assessment of mercury pollution in rivers and streams around artisanal gold mining areas of the Birim north district of Ghana. J. Environ. Protect. 2, 1227–1239. https://doi.org/10.4236/jep.2011.29141.

Navratil, T., Shanley, J., Rohovec, J., Oulehle, F., Kram, P., Matouskova, S., et al., 2015. Mercury in stream water at five Czech catchments across a Hg and S deposition gradient. J. Geochem. Explor. 158, 201–211. https://doi.org/10.1016/j.gexplo.2015.07.016.

Nelson, S.J., Johnson, K.B., Kahl, J.S., Haines, T.A., Fernandez, I.J., 2007. Mass balances of mercury and nitrogen in burned and unburned forested watersheds at Acadia National Park, Maine, USA. Environ. Monit. Assess. 126 (1–3), 69–80. https://doi.org/10.1007/s10661-006-9332-4.

Obrist, D., 2007. Atmospheric mercury pollution due to losses of terrestrial carbon pools? Biogeochemistry 85 (2), 119–123. https://doi.org/10.1007/s10533-007-9108-0.

Obrist, D., Agnan, Y., Jiskra, M., Olson, C.L., Colegrove, D.P., Hueber, J., et al., 2017. Tundra uptake of atmospheric elemental mercury drives arctic mercury pollution. Nature 547 (7662). https://doi.org/10.1038/nature22997.

Obrist, D., Kirk, J.L., Zhang, L., Sunderland, E.M., Jiskra, M., Selin, N.E., 2018. A review of global environmental mercury processes in response to human and natural perturbations: changes of emissions, climate, and land use. Ambio 47 (2), 116–140. https://doi.org/10.1007/s13280-017-1004-9.

Olson, C., Jiskra, M., Biester, H., Chow, J., Obrist, D., 2018. Mercury in active-layer tundra soils of Alaska: concentrations, pools, origins, and spatial distribution. Glob. Biogeochem. Cy. 32 (7), 1058–1073. https://doi.org/10.1029/2017gb005840.

Orem, W., Gilmour, C., Axelrad, D., Krabbenhoft, D., Scheidt, D., Kalla, P., et al., 2011. Sulfur in the south Florida ecosystem: distribution, sources, biogeochemistry, impacts, and management for restoration. Crit. Rev. Environ. Sci. Technol. 41 (sup1), 249–288. https://doi.org/10.1080/10643389.2010.531201.

Oremland, R.S., Culbertson, C.W., Winfrey, M.R., 1991. Methylmercury decomposition in sediments and bacterial cultures - involvement of methanogens and sulfate reducers in oxidative demethylation. Appl. Environ. Microb. 57 (1), 130–137.

Osterwalder, S., Bishop, K., Alewell, C., Fritsche, J., Laudon, H., Åkerblom, S., et al., 2017. Mercury evasion from a boreal peatland shortens the timeline for recovery from legacy pollution. Sci. Rep. 7, 16022. https://doi.org/10.1038/s41598-017-16141-7.

Osterwalder, S., Sommar, J., Åkerblom, S., Jocher, G., Fritsche, J., Nilsson, M.B., ... Alewell, C., 2018. Comparative study of elemental mercury flux measurement techniques over a Fennoscandian boreal peatland. Atmos. Environ. 172, 16–25.

Osterwalder, S., Eugster, W., Feigenwinter, I., Jiskra, M., 2019. First eddy covariance flux measurements of gaseous elemental mercury (Hg^0) over a grassland. Atmos. Meas. Tech. Discuss. https://doi.org/10.5194/amt-2019-278.

Oswald, C.J., Branfireun, B.A., 2014. Antecedent moisture conditions control mercury and dissolved organic carbon concentration dynamics in a boreal headwater catchment. Water Resour. Res. 50 (8), 6610–6627. https://doi.org/10.1002/2013wr014736.

Oswald, C.J., Carey, S.K., 2016. Total and methyl mercury concentrations in sediment and water of a constructed wetland in the athabasca oil sands region. Environ. Pollut. 213, 628–637. https://doi.org/10.1016/j.envpol.2016.03.002.

Oswald, C.J., Heyes, A., Branfireun, B.A., 2014. Fate and transport of ambient mercury and applied mercury isotope in terrestrial upland soils: insights from the METAALICUS watershed. Environ. Sci. Technol. 48 (2), 1023–1031. https://doi.org/10.1021/es404260f.

Outridge, P.M., Mason, R.P., Wang, F., Guerrero, S., Heimbürger-Boavida, L.E., 2018. Updated global and oceanic mercury budgets for the United Nations Global Mercury Assessment. Environ. Sci. Technol. 52, 11466–11477. https://doi.org/10.1021/acs.est.8b01246.

Patel, K.F., Jakubowski, M.D., Fernandez, I.J., Nelson, S.J., Gawley, W., 2019. Soil nitrogen and mercury dynamics seven decades after a fire disturbance: a case study at Acadia National Park. Water Air Soil Pollut. 230, 29. https://doi.org/10.1007/s11270-019-4085-1.

Pearson, C., Howard, D., Moore, C., Obrist, D., 2019. Mercury and trace metal wet deposition across five stations in Alaska: controlling factors, spatial patterns, and source regions. Atmos. Chem. Phys. 19, 6913–6929. https://doi.org/10.5194/acp-19-6913-2019.

Peng, X.Y., Liu, F.J., Wang, W.X., Ye, Z.H., 2012. Reducing total mercury and methylmercury accumulation in rice grains through water management and deliberate selection of rice cultivars. Environ. Pollut. 162, 202–208. https://doi.org/10.1016/j.envpol.2011.11.024.

Pérez-Rodríguez, M., Horák-Terra, I., Rodríguez-Lado, L., Aboal, J.R., Martínez Cortizas, A., 2015. Long-term (~57 ka) controls on mercury accumulation in the southern hemisphere reconstructed using a peat record from Pinheiro Mire (Minas Gerais, Brazil). Environ. Sci. Technol. 49, 1356–1364. https://doi.org/10.1021/es504826d.

Pettersson, C., Bishop, K., Lee, Y.H., Allard, B., 1995. Relations between organic-carbon and methylmercury in humic rich surface waters from Svartberget catchment in northern Sweden. Water Air Soil Pollut. 80 (1–4), 971–979. https://doi.org/10.1007/Bf01189751.

Phoenix, G.K., Bjerke, J.W., 2016. Arctic browning: extreme events and trends reversing arctic greening. Glob. Change Biol. 22 (9), 2960–2962. https://doi.org/10.1111/gcb.13261.
Podar, M., Gilmour, C.C., Brandt, C.C., Soren, A., Brown, S.D., Crable, B.R., et al., 2015. Global prevalence and distribution of genes and microorganisms involved in mercury methylation. Sci. Adv. 1 (9), e1500675. https://doi.org/10.1126/sciadv.1500675.
Poissant, L., Pilote, M., Yumvihoze, E., Lean, D., 2008. Mercury concentrations and foliage/atmosphere fluxes in a maple forest ecosystem in Quebec. Canada. J. Geophys. Res.-Atmos. 113, D10307. https://doi.org/10.1029/2007JD009510.
Porvari, P., Verta, M., Munthe, J., Haapanen, M., 2003. Forestry practices increase mercury and methyl mercury output from boreal forest catchments. Environ. Sci. Technol. 37 (11), 2389–2393. https://doi.org/10.1021/es0340174.
Poulin, B.A., Aiken, G.R., Nagy, K.L., Manceau, A., Krabbenhoft, D.P., Ryan, J.N., 2016. Mercury transformation and release differs with depth and time in a contaminated riparian soil during simulated flooding. Geochim. Cosmochim. Acta 176, 118–138. https://doi.org/10.1016/j.gca.2015.12.024.
Poulin, B.A., Ryan, J.N., Tate, M.T., Krabbenhoft, D.P., Hines, M.E., Barkay, T., et al., 2019. Geochemical factors controlling dissolved elemental mercury and methylmercury formation in Alaskan wetlands of varying trophic status. Environ. Sci. Technol. 53 (11), 6203–6213. https://doi.org/10.1021/acs.est.8b06041.
Qiu, G.L., Feng, X.B., Li, P., Wang, S.F., Li, G.H., Shang, L.H., Fu, X.W., 2008. Methylmercury accumulation in rice (*Oryza sativa L.*) grown at abandoned mercury mines in Guizhou, China. J. Agricul. Food Chem. 56, 2465–2468. https://doi.org/10.1021/jf073391a.
Rask, M., Nyberg, K., Markkanen, S.-L., Ojala, A., 1998. Forestry in catchments: effects on water quality, plankton, zoobenthos and fish in small lakes. Boreal Environ. Res. 3 (1), 75–86.
Rasmussen, P.E., Mierle, G., Nriagu, J.O., 1991. The analysis of vegetation for total mercury. Water Air Soil Pollut. 56 (1), 379–390.
Ravichandran, M., 2004. Interactions between mercury and dissolved organic matter - a review. Chemosphere 55 (3), 319–331. https://doi.org/10.1016/j.chemosphere.2003.11.011.
Rea, A.W., Lindberg, S.E., Scherbatskoy, T., Keeler, G.J., 2002. Mercury accumulation in foliage over time in two northern mixed-hardwood forests. Water Air Soil Pollut. 133 (1–4), 49–67. https://doi.org/10.1023/A:1012919731598.
Riggs, C.E., Kolka, R.K., Nater, E.A., Witt, E.L., Wickman, T.R., Woodruff, L.G., Butcher, J.T., 2017. Yellow perch (*Perca flavescens*) mercury unaffected by wildland fires in northern Minnesota. J. Environ. Qual. 46, 623–631. https://doi.org/10.2134/jeq2016.10.0418.
Riscassi, A.L., Scanlon, T.M., 2011. Controls on stream water dissolved mercury in three mid-Appalachian forested headwater catchments. Water Resour. Res. 47. https://doi.org/10.1029/2011wr010977.
Riscassi, A.L., Scanlon, T.M., 2013. Particulate and dissolved mercury export in streamwater within three mid-Appalachian forested watersheds in the US. J. Hydrol. 501, 92–100. https://doi.org/10.1016/j.jhydrol.2013.07.041.
Riscassi, A., Miller, C., Brooks, S., 2016. Seasonal and flow-driven dynamics of particulate and dissolved mercury and methylmercury in a stream impacted by an industrial mercury source. Environ. Toxicol. Chem. 35 (6), 1386–1400. https://doi.org/10.1002/etc.3310.
Risch, M.R., DeWild, J.F., Krabbenhoft, D.P., Kolka, R.K., Zhang, L.M., 2012. Litterfall mercury dry deposition in the eastern USA. Environ. Pollut. 161, 284–290. https://doi.org/10.1016/j.envpol.2011.06.005.
Risch, M.R., DeWild, J.F., Gay, D.A., Zhang, L., Boyer, E.W., Krabbenhoft, D.P., 2017. Atmospheric mercury deposition to forests in the eastern USA. Environ. Pollut. 228, 8–18. https://doi.org/10.1016/j.envpol.2017.05.004.
Rodenhouse, N.L., Lowe, W.H., Gebauer, R.L.E., McFarland, K.P., Bank, M.S., 2019. Mercury bioaccumulation in temperate forest food webs associated with headwater streams. Sci. Total Environ. 665, 1125–1134. https://doi.org/10.1016/j.scitotenv.2019.02.151.
Rothenberg, S.E., Feng, X.B., 2012. Mercury cycling in a flooded rice paddy. J. Geophys. Res-Biogeo. 117. https://doi.org/10.1029/2011JG001800.
Rothenberg, S.E., Kirby, M.E., Bird, B.W., DeRose, M.B., Lin, C.C., Feng, X., et al., 2010. The impact of over 100 years of wildfires on mercury levels and accumulation rates in two lakes in southern California, USA. Environ. Earth Sci. 60 (5), 993–1005. https://doi.org/10.1007/s12665-009-0238-7.
Rothenberg, S.E., Feng, X.B., Dong, B., Shang, L.H., Yin, R.S., Yuan, X.B., 2011. Characterization of mercury species in brown and white rice (*Oryza sativa L.*) grown in water-saving paddies. Environ. Pollut. 159, 1283–1289. https://doi.org/10.1016/j.scitotenv.2012.03.024.
Roy, V., Amyot, M., Carignan, R., 2009. Beaver ponds increase methylmercury concentrations in Canadian shield streams along vegetation and pond-age gradients. Environ. Sci. Technol. 43 (15), 5605–5611. https://doi.org/10.1021/es901193x.
Rudd, J.W., 1995. Sources of methyl mercury to freshwater ecosystems: a review. Water Air Soil Pollut. 80 (1–4), 697–713.
Saniewska, D., Beldowska, M., Beldowski, J., Saniewski, M., Gebka, K., Szubska, M., et al., 2018. Impact of intense rains and flooding on mercury riverine input to the coastal zone. Mar. Pollut. Bull. 127, 593–602. https://doi.org/10.1016/j.marpolbul.2017.12.058.
Schaefer, J.K., Morel, F.M.M., 2009. High methylation rates of mercury bound to cysteine by *Geobacter sulfurreducens*. Nat. Geosci. 2 (2), 123–126. https://doi.org/10.1038/Ngeo412.
Schaefer, J.K., Kronberg, R.M., Morel, F.M., Skyllberg, U., 2014. Detection of a key Hg methylation gene, hgca, in wetland soils. Environ. Microbiol. Rep. 6 (5), 441–447. https://doi.org/10.1111/1758-2229.12136.
Schelker, J., Burns, D.A., Weiler, M., Laudon, H., 2011. Hydrological mobilization of mercury and dissolved organic carbon in a snow-dominated, forested watershed: conceptualization and modeling. J. Geophys. Res-Biogeo. 116. https://doi.org/10.1029/2010jg001330.
Schroeder, W.H., Munthe, J., 1998. Atmospheric mercury - an overview. Atmos. Environ. 32 (5), 809–822. https://doi.org/10.1016/S1352-2310(97)00293-8.
Schroeder, W., Anlauf, K., Barrie, L., Lu, J., Steffen, A., Schneeberger, D., et al., 1998. Arctic springtime depletion of mercury. Nature 394, 331–332.
Schuster, P.F., Krabbenhoft, D.P., Naftz, D.L., Cecil, L.D., Olson, M.L., Dewild, J.F., et al., 2002. Atmospheric mercury deposition during the last 270 years: a glacial ice core record of natural and anthropogenic sources. Environ. Sci. Technol. 36 (11), 2303–2310. https://doi.org/10.1021/es0157503.
Schuster, P.F., Striegl, R.G., Aiken, G.R., Krabbenhoft, D.P., Dewild, J.F., Butler, K., et al., 2011. Mercury export from the Yukon River basin and potential response to a changing climate. Environ. Sci. Technol. 45 (21), 9262–9267. https://doi.org/10.1021/es202068b.
Schuster, P.F., Schaefer, K.M., Aiken, G.R., Antweiler, R.C., Dewild, J.F., Gryziec, J.D., et al., 2018. Permafrost stores a globally significant amount of mercury. Geophys. Res. Lett. 45 (3), 1463–1471. https://doi.org/10.1002/2017gl075571.
Schwesig, D., Krebs, O., 2003. The role of ground vegetation in the uptake of mercury and methylmercury in a forest ecosystem. Plant Soil 253 (2), 445–455. https://doi.org/10.1023/A:1024891014028.
Selin, N.E., Jacob, D.J., Yantosca, R.M., Strode, S., Jaegle, L., Sunderland, E.M., 2008. Global 3-D land-ocean-atmosphere model for mercury: present-day versus preindustrial cycles and anthropogenic enrichment factors for deposition. Glob. Biogeochem. Cy. 22 (3), GB3099. https://doi.org/10.1029/2008gb003282.
Shanley, J.B., Bishop, K., 2012. Mercury cycling in terrestrial watersheds. In: Banks, M.S. (Ed.), Mercury in the Environment: Pattern and Process, pp. 119–141.
Shanley, J.B., Mast, M.A., Campbell, D.H., Aiken, G.R., Krabbenhoft, D.P., Hunt, R.J., et al., 2008. Comparison of total mercury and methylmercury cycling at five sites using the small watershed approach. Environ. Pollut. 154 (1), 143–154. https://doi.org/10.1016/j.envpol.2007.12.031.
Shanley, J.B., Engle, M.A., Scholl, M., Krabbenhoft, D.P., Brunette, R., Olson, M.L., et al., 2015. High mercury wet deposition at a "clean air" site in Puerto Rico. Environ. Sci. Technol. 49, 12474–12482. https://doi.org/10.1021/acs.est.5b02430.
Shanley, J.B., Marvin-DiPasquale, M., Lane, O., Arendt, W., Hall, S., McDowell, W.H., 2019. Resolving a paradox-high mercury deposition, but low bioaccumulation in northeastern Puerto Rico. Ecotoxicology https://doi.org/10.1007/s10646-019-02108-z.
Sherman, L.S., Blum, J.D., Johnson, K.P., Keeler, G.J., Barres, J.A., Douglas, T.A., 2010. Mass-independent fractionation of mercury isotopes in arctic snow driven by sunlight. Nat. Geosci. 3 (3), 173–177. https://doi.org/10.1038/ngeo758.
Shu, R., Wang, Y.J., Zhong, H., 2016. Biochar amendment reduced methylHg accumulation in rice plants. J. Hazard. Mater. 313, 1–8. https://doi.org/10.1016/j.jhazmat.2016.03.080.
Simone, F.D., Artaxo, P., Bencardino, M., Cinnirella, S., Carbone, F., D'Amore, F., Dommergue, A., Feng, X.B., Gencarelli, C.N., Hedgecock, I.M., Landis, M.S., 2017. Particulate-phase mercury emissions from biomass burning and impact on resulting deposition: a modelling assessment. Atmos. Chem. Phys. 17 (3), 1881–1899. https://doi.org/10.5194/acp-17-1881-2017.
Siwik, E.I.H., Campbell, L.M., Mierle, G., 2009. Fine-scale mercury trends in temperate deciduous tree leaves from Ontario, Canada. Sci. Total Environ. 407 (24), 6275–6279. https://doi.org/10.1016/j.scitotenv.2009.08.044.
Skyllberg, U., Qian, J., Frech, W., Xia, K., Bleam, W.F., 2003. Distribution of mercury, methyl mercury and organic sulphur species in soil, soil solution and stream of a boreal forest catchment. Biogeochemistry 64 (1), 53–76. https://doi.org/10.1023/A:1024904502633.
Skyllberg, U., Drott, A., Lambertsson, L., Bjorn, E., Karlsson, T., Johnson, T., et al., 2007. Net methylmercury production as a basis for improved risk assessment of mercury-contaminated sediments. Ambio 36 (6), 437–442. https://doi.org/10.1579/0044-7447(2007)36[437:Nmpaab]2.0.Co;2.
Skyllberg, U., Westin, M.B.R., Meili, M., Björn, E., 2009. Elevated concentrations of methyl mercury in streams after forest clear-cut: a consequence of mobilization from soil or new methylation? Environ. Sci. Technol. 43 (22), 8535–8541. https://doi.org/10.1021/es900996z.
Smith, S.L., Romanovsky, V.E., Lewkowicz, A.G., Burn, C.R., Mallard, M., Clow, G.D., et al., 2010. Thermal state of permafrost in North America: a contribution to the international polar year. Perma. Periglac. Proc. 21 (2), 117–135. https://doi.org/10.1002/ppp.690.
Smith-Downey, N.V., Sunderland, E.M., Jacob, D.J., 2010. Anthropogenic impacts on global storage and emissions of mercury from terrestrial soils: insights from a new global model. J. Geophys. Res-Biogeo 115 (G03008). https://doi.org/10.1029/2009JG001124.
Soerensen, A.L., Jacob, D.J., Schartup, A.T., Fisher, J.A., Lehnherr, I., St Louis, V.L., et al., 2016. A mass budget for mercury and methylmercury in the Arctic Ocean. Glob. Biogeochem. Cy. 30 (4), 560–575. https://doi.org/10.1002/2015gb005280.
Somenahally, A.C., Hollister, E.B., Loeppert, R.H., Yan, W.G., Gentry, T.J., 2011. Microbial communities in rice rhizosphere altered by intermittent and continuous flooding in fields with long-term arsenic application. Soil Biol. Biochem. 43, 1220–1228. https://doi.org/10.1016/j.soilbio.2011.02.011.
Sommar, J., Zhu, W., Lin, C.-J., Feng, X., 2013. Field approaches to measure Hg exchange between natural surfaces and the atmosphere - a review. Crit. Rev. Environ. Sci. Technol. 43, 1657–1739. https://doi.org/10.1080/10643389.2012.671733.
Sondergaard, J., Tamstorf, M., Elberling, B., Larsen, M.M., Mylius, M.R., Lund, M., et al., 2015. Mercury exports from a high-arctic river basin in northeast Greenland (74 degrees N) largely controlled by glacial lake outburst floods. Sci. Total Environ. 514, 83–91. https://doi.org/10.1016/j.scitotenv.2015.01.097.
Song, S., Selin, N.E., Soerensen, A.L., Angot, H., Artz, R., Brooks, S., et al., 2015. Top-down constraints on atmospheric mercury emissions and implications for global biogeochemical cycling. Atmos. Chem. Phys. 15, 7103–7125. https://doi.org/10.5194/acp-15-7103-2015.

Song, S., Selin, N.E., Gratz, L.E., Ambrose, J.L., Jaffe, D.A., Shah, V., et al., 2016. Constraints from observations and modeling on atmosphere–surface exchange of mercury in eastern North America. Elem. Sci. Anth. 4, 000100. https://doi.org/10.12952/journal.elementa.000100.

Sonke, J.E., Heimbürger, L.E., 2012. Mercury in flux. Nat. Geosci. 5 (7), 447–448. https://doi.org/10.1038/ngeo1508.

Sonke, J.E., Teisserenc, R., Heimburger-Boavida, L.-E., Petrova, M.V., Marusczak, N., Le Dantec, T., et al., 2018. Eurasian river spring flood observations support net Arctic Ocean mercury export to the atmosphere and Atlantic Ocean. Proc. Natl. Acad. Sci. U. S. A. 115 (50), E11586-E11594. https://doi.org/10.1073/pnas.1811957115.

Sørensen, R., Meili, M., Lambertsson, L., Brömssen, C.v., Bishop, K., 2009. The effect of forest harvest operations on mercury and methylmercury in two boreal streams: relatively small changes in the first two years prior to site preparation. Ambio 38 (7), 364–372. https://doi.org/10.1579/0044-7447-38.7.364.

Sprovieri, F., Pirrone, N., Bencardino, M., D'Amore, F., Angot, H., Barbante, C., et al., 2017. Five-year records of mercury wet deposition flux at GMOS sites in the northern and southern hemispheres. Atmos. Chem. Phys. 17, 2689–2708. https://doi.org/10.5194/acp-17-2689-2017.

St Pierre, K.A., Zolkos, S., Shakil, S., Tank, S.E., St Louis, V.L., Kokelj, S.V., 2018. Unprecedented increases in total and methyl mercury concentrations downstream of retrogressive thaw slumps in the western Canadian arctic. Environ. Sci. Technol. 52 (24), 14099–14109. https://doi.org/10.1021/acs.est.8b05348.

Steffen, A., Douglas, T., Amyot, M., Ariya, P., Aspmo, K., Berg, T., et al., 2008. A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and snow. Atmos. Chem. Phys. 8, 1445–1482. https://doi.org/10.5194/acp-8-1445-2008.

Stern, G.A., Macdonald, R.W., Outridge, P.M., Wilson, S., Chetelat, J., Cole, A., et al., 2012. How does climate change influence arctic mercury? Sci. Total Environ. 414, 22–42. https://doi.org/10.1016/j.scitotenv.2011.10.039.

Stoken, O.M., Riscassi, A.L., Scanlon, T.M., 2016. Association of dissolved mercury with dissolved organic carbon in us rivers and streams: the role of watershed soil organic carbon. Water Resour. Res. 52 (4), 3040–3051. https://doi.org/10.1002/2015wr017849.

Streets, D.G., Horowitz, H.M., Lu, Z., Levin, L., Thackray, C.P., Sunderland, E.M., 2019. Five hundred years of anthropogenic mercury: spatial and temporal release profiles. Environ. Res. Lett. 14, 084004. https://doi.org/10.1088/1748-9326/ab281f.

Strickman, R.J., Mitchell, C.P.J., 2017. Accumulation and translocation of methylmercury and inorganic mercury in *Oryza sativa*: an enriched isotope tracer study. Sci. Total Environ. 574, 1415–1423. https://doi.org/10.1016/j.scitotenv.2016.08.068.

Strickman, R.J.S., Fulthorpe, R.R., Coleman Wasik, J.K., Engstrom, D.R., Mitchell, C.P.J., 2016. Experimental sulfate amendment alters peatland bacterial community structure. Sci. Total Environ. 566-567, 1289–1296. https://doi.org/10.1016/j.scitotenv.2016.05.189.

Sturm, M., Racine, C., Tape, K., 2001. Increasing shrub abundance in the Arctic. Nature 411, 546–547. https://doi.org/10.1038/35079180.

Tang, J.Y., Riley, W.J., 2015. Weaker soil carbon-climate feedbacks resulting from microbial and abiotic interactions. Nat. Clim. Chang. 5 (1), 56–60. https://doi.org/10.1038/nclimate2438.

Tang, W., Cheng, J.P., Zhao, W.C., Wang, W.H., 2015. Hg levels and estimated total daily intakes for children and adults from an electronic waste recycling area in Taizhou, China: key role of rice and fish consumption. J. Environ. Sci. 34, 107–115. https://doi.org/10.1016/j.jes.2015.01.029.

Tang, W.L., Dang, F., Evans, D., Zhong, H., Xiao, L., 2017. Understanding reduced inorganic Hg accumulation in rice following selenium application: selenium application routes, speciation and doses. Chemosphere 169, 369–376. https://doi.org/10.1016/j.chemosphere.2016.11.087.

Taylor, H., Appleton, J.D., Lister, R., Smith, B., Chitamweba, D., Mkumbo, O., et al., 2005. Environmental assessment of mercury contamination from the Rwamagasa artisanal gold mining centre, Geita District, Tanzania. Sci. Total Environ. 343 (1–3), 111–133. https://doi.org/10.1016/j.scitotenv.2004.09.042.

Telmer, K.M., Veiga, M.M., 2009. World emissions of mercury from artisanal and small scale gold mining. In: Mason, R., Pirrone, N. (Eds.), Mercury Fate and Transport in the Global Atmosphere, pp. 131–172. https://doi.org/10.1007/978-0-387-93958-2_6.

Tjerngren, I., Meili, M., Bjorn, E., Skyllberg, U., 2012. Eight boreal wetlands as sources and sinks for methyl mercury in relation to soil acidity, C/N ratio, and small-scale flooding. Environ. Sci. Technol. 46 (15), 8052–8060. https://doi.org/10.1021/es300845x.

Treharne, R., Bjerke, J.W., Tømmervik, H., Stendardi, L., Phoenix, G.K., 2019. Arctic browning: impacts of extreme climatic events on heathland ecosystem CO_2 fluxes. Glob. Change Biol. 25, 489–503. https://doi.org/10.1111/gcb.14500.

Tremblay, A., Cloutier, L., Lucotte, M., 1998. Total mercury and methylmercury fluxes via emerging insects in recently flooded hydroelectric reservoirs and a natural lake. Sci. Total Environ. 219 (2–3), 209–221. https://doi.org/10.1016/S0048-9697(98)00227-7.

Ukonmaanaho, L., Starr, M., Kantola, M., Laurén, A., Piispanen, J., Pietilä, H., et al., 2016. Impacts of forest harvesting on mobilization of Hg and MeHg in drained peatland forests on black schist or felsic bedrock. Environ. Monit. Assess. 188 (4), 228. https://doi.org/10.1007/s10661-016-5210-x.

Ulrich, P.D., Sedlak, D.L., 2010. Impact of iron amendment on net methylmercury export from tidal wetland microcosms. Environ. Sci. Technol. 44 (19), 7659–7665. https://doi.org/10.1021/es1018256.

UNEP, 2013a. Global Mercury Assessment. Sources, Emissions, Releases and Environmental Transport. UNEP Chemicals Branch, Geneva, Switzerland, p. 2013.

UNEP, 2013b. Minamata Convention on Mercury: Text and Annexes. UNEP Chemicals Branch, Geneva, Switzerland.

UNEP, 2019. Global 3ercury Assessment. Sources, emissions, releases and environmental transport. UNEP Chemicals and Health Branch, Geneva, Switzerland, p. 2018 (ISBN: 978-92-807-3744-8).

Veiga, M.M., Maxson, P.A., Hylander, L.D., 2006. Origin and consumption of mercury in small-scale gold mining. J. Clean. Prod. 14 (3–4), 436–447. https://doi.org/10.1016/j.jclepro.2004.08.010.

Vermilyea, A.W., Nagorski, S.A., Lamborg, C.H., Hood, E.W., Scott, D., Swarr, G.J., 2017. Continuous proxy measurements reveal large mercury fluxes from glacial and forested watersheds in Alaska. Sci. Total Environ. 599, 145–155. https://doi.org/10.1016/j.scitotenv.2017.03.297.

Vishnivetskaya, T.A., Hu, H.Y., Van Nostrand, J.D., Wymore, A.M., Xu, X.H., Qiu, G.L., Feng, X.B., Zhou, J.Z., Brown, S.D., Brandt, C.C., Podar, M., Gu, B.H., Elias, D.A., 2018. Microbial community structure with trends in methylation gene diversity and abundance in mercury-contaminated rice paddy soils in Guizhou, China. Environ. Sci-Proc. Imp. 20, 673–685. https://doi.org/10.1039/c7em00558j.

Voss, B.M., Peucker-Ehrenbrink, B., Eglinton, T.I., Spencer, R.G.M., Bulygina, E., Galy, V., et al., 2015. Seasonal hydrology drives rapid shifts in the flux and composition of dissolved and particulate organic carbon and major and trace ions in the Fraser River. Canada. Biogeosciences. 12 (19), 5597–5618. https://doi.org/10.5194/bg-12-5597-2015.

Walmsley, J.D., Godbold, D.L., 2010. Stump harvesting for bioenergy – a review of the environmental impacts. Forestry 83 (1), 17–38. https://doi.org/10.1093/forestry/cpp028.

Wang, X., Li, B., Tam, N.F.Y., Huang, L., Qi, X.L., Wang, H.B., Ye, Z.H., Meng, M., Shi, J.B., 2014a. Radial oxygen loss has different effects on the accumulation of total Hg and methylHg in rice. Plant Soil 385, 343–355. https://doi.org/10.1007/s11104-014-2239-x.

Wang, X., Lin, C.J., Feng, X., 2014b. Sensitivity analysis of an updated bidirectional air–surface exchange model for elemental mercury vapor. Atmos. Chem. Phys. 14, 6273–6287. https://doi.org/10.5194/acp-14-6273-2014.

Wang, X., Ye, Z.H., Li, B., Huang, L.N., Meng, M., Shi, J.B., Jiang, G.B., 2014c. Growing rice aerobically markedly decreases mercury accumulation by reducing both Hg bioavailability and the production of MeHg. Environ. Sci. Technol. 48, 1878–1885. https://doi.org/10.1021/es4038929.

Wang, X., Bao, Z., Lin, C.-J., Yuan, W., Feng, X., 2016. Assessment of global mercury deposition through litterfall. Environ. Sci. Technol. 50, 8548–8557. https://doi.org/10.1021/acs.est.5b06351.

Wang, Y.J., Dang, F., Zhao, J.T., Zhong, H., 2016a. Selenium inhibits sulfate-mediated methylHg production in rice paddy soil. Environ. Pollut. 213, 232–239. https://doi.org/10.1016/j.envpol.2016.02.021.

Wang, Y.J., Wei, Z.B., Zeng, Q.L., Zhong, H., 2016b. Amendment of sulfate with Se into soils further reduces methylHg accumulation in rice. J. Soils Sediments 16, 2720–2727. https://doi.org/10.1007/s11368-016-1453-y.

Wang, X., Yuan, W., Lin, C.-J., Zhang, L., Zhang, H., Feng, X., 2019. Climate and vegetation as primary drivers for global mercury storage in surface soil. Environ. Sci. Technol. 53, 10665–10675. https://doi.org/10.1021/acs.est.9b02386.

Wiedinmyer, C., Friedli, H., 2007. Mercury emission estimates from fires: an initial inventory for the United States. Environ. Sci. Technol. 41 (23), 8092–8098. https://doi.org/10.1021/es071289o.

Wiener, J.G., Knights, B.C., Sandheinrich, M.B., Jeremiason, J.D., Brigham, M.E., Engstrom, D.R., et al., 2006. Mercury in soils, lakes, and fish in Voyageurs National Park (Minnesota): importance of atmospheric deposition and ecosystem factors. Environ. Sci. Technol. 40 (20), 6261–6268. https://doi.org/10.1021/es060822h.

Windham-Myers, L., Marvin-Dipasquale, M., Krabbenhoft, D.P., Agee, J.L., Cox, M.H., Heredia-Middleton, P., et al., 2009. Experimental removal of wetland emergent vegetation leads to decreased methylmercury production in surface sediment. J. Geophys. Res.-Biogeosci. 114 (G00C05). https://doi.org/10.1029/2008jg000815.

Windham-Myers, L., Fleck, J.A., Ackerman, J.T., Marvin-DiPasquale, M., Stricker, C.A., Heim, W.A., et al., 2014. Mercury cycling in agricultural and managed wetlands: a synthesis of methylmercury production, hydrologic export, and bioaccumulation from an integrated field study. Sci. Total Environ. 484, 221–231. https://doi.org/10.1016/j.scitotenv.2014.01.033.

Woerndle, G.E., Tsui, M.T.-K., Sebestyen, S.D., Blum, J.D., Nie, X., Kolka, R.K., 2018. New insights on ecosystem mercury cycling revealed by stable isotopes of mercury in water flowing from a headwater peatland catchment. Environ. Sci. Technol. 52 (4), 1854–1861. https://doi.org/10.1021/acs.est.7b04449.

Wright, L.P., Zhang, L., Marsik, F.J., 2016. Overview of mercury dry deposition, litterfall, and throughfall studies. Atmos. Chem. Phys. 16, 13399–13416. https://doi.org/10.5194/acp-16-13399-2016.

Wu, P., Bishop, K., von Brömssen, C., Eklöf, K., Futter, M., Hultberg, H., et al., 2018. Does forest harvest increase the mercury concentrations in fish? Evidence from Swedish lakes. Sci. Total Environ. 622-623, 1353–1362. https://doi.org/10.1016/j.scitotenv.2017.12.075.

Wu, P., Kainz, M.J., Bravo, A.G., Åkerblom, S., Sonesten, L., Bishop, K., 2019. The importance of bioconcentration into the pelagic food web base for methylmercury biomagnification: a meta-analysis. Sci. Total Environ. 646, 357–367.

Yang, Z.M., Fang, W., Lu, X., Sheng, G.P., Graham, D.E., Liang, L.Y., et al., 2016. Warming increases methylmercury production in an arctic soil. Environ. Pollut. 214, 504–509. https://doi.org/10.1016/j.envpol.2016.04.069.

Yang, Y., Meng, L.H., Yanai, R.D., Montesdeoca, M., Templer, P.H., Asbjornsen, H., Rustad, L.E., Driscoll, C.T., 2019. Climate change may alter mercury fluxes in northern hardwood forests. Biogeochemistry https://doi.org/10.1007/s10533-019-00605-1.

Yu, R.Q., Adatto, I., Montesdeoca, M.R., Driscoll, C.T., Hines, M.E., Barkay, T., 2010. Mercury methylation in sphagnum moss mats and its association with sulfate-reducing bacteria in an acidic adirondack forest lake wetland. FEMS Microbiol. Ecol. 74 (3), 655–668. https://doi.org/10.1111/j.1574-6941.2010.00978.x.

Yuan, W., Sommar, J., Lin, C.-J., Wang, X., Li, K., Liu, Y., et al., 2019a. Stable isotope evidence shows re-emission of elemental mercury vapor occurring after reductive loss from foliage. Environ. Sci. Technol. 53 (2), 651–660. https://doi.org/10.1021/acs.est.8b04865.

Yuan, W.P., Zheng, Y., Piao, S.L., Ciais, P., Lombardozzi, D., Wang, Y.P., et al., 2019b. Increased atmospheric vapor pressure deficit reduces global vegetation growth. Sci. Adv. 5 (8). https://doi.org/10.1126/sciadv.aax1396.

Zagar, D., Knap, A., Warwick, J.J., Rajar, R., Horvat, M., Cetina, M., 2006. Modelling of mercury transport and transformation processes in the Idrijca and Soca river system. Sci. Total Environ. 368, 149–163. https://doi.org/10.1016/j.scitotenv.2005.09.068.

Zhang, L.M., Wright, L.P., Blanchard, P., 2009. A review of current knowledge concerning dry deposition of atmospheric mercury. Atmos. Environ. 43 (37), 5853–5864. https://doi.org/10.1016/j.atmosenv.2009.08.019.

Zhang, T., Kim, B., Levard, C., Reinsch, B.C., Lowry, G.V., Deshusses, M.A., et al., 2012b. Methylation of mercury by bacteria exposed to dissolved, nanoparticulate, and microparticulate mercuric sulfides. Environ. Sci. Technol. 46 (13), 6950–6958. https://doi.org/10.1021/es203181m.

Zhang, H., Feng, X.B., Jiang, C..X., Li, Q.H., Gu, C.H., Shang, L.H., Li, P., Lin, Y., Larssen, T., 2014. Understanding the paradox of selenium contamination in Hg mining areas: high soil content and low accumulation in rice. Environ. Pollut. 188, 2736. https://doi.org/10.1016/j.envpol.2014.01.012.

Zhang, H., Fu, X.W., Lin, C.J., Shang, L.H., Zhang, Y.P., Feng, X.B., et al., 2016c. Monsoon-facilitated characteristics and transport of atmospheric mercury at a high-altitude background site in southwestern China. Atmos. Chem. Phys. 16 (20), 13131–13148. https://doi.org/10.5194/acp-16-13131-2016.

Zhang, H., Holmes, C.D., Wu, S., 2016d. Impacts of changes in climate, land use and land cover on atmospheric mercury. Atmos. Environ. 141, 230–244. https://doi.org/10.1016/j.atmosenv.2016.06.056.

Zhang, L., Wu, Z., Cheng, I., Wright, L.P., Olson, M.L., Gay, D.A., et al., 2016a. The estimated six-year mercury dry deposition across North America. Environ. Sci. Technol. 50, 12864–12873. https://doi.org/10.1021/acs.est.6b04276.

Zhang, Y., Jacob, D.J., Horowitz, H.M., Chen, L., Amos, H.M., Krabbenhoft, D.P., et al., 2016b. Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions. Proc. Natl. Acad. Sci. U. S. A. 113, 526–531. https://doi.org/10.1073/pnas.1516312113.

Zhang, Y., Liu, Y.R., Lei, P., Wang, Y.J., Zhong, H., 2018. Biochar and nitrate reduce risk of methylHg in soils under straw amendment. Sci. Total Environ. 619-620, 384–390. https://doi.org/10.1016/j.scitotenv.2017.11.106.

Zhang, J., Wu, S..C., Xu, Z.T., Wang, M.Y., Man, Y.B., Christie, P., Liang, P., Shan, S.D., Wong, M.H., 2019. The role of sewage sludge biochar in methylHg formation and accumulation in rice. Chemosphere 2019 (218), 527–533. https://doi.org/10.1016/j.chemosphere.2018.11.090.

Zhao, M., Running, S.W., 2010. Drought-induced reduction in global terrestrial net primary production from 2000 through 2009. Science 329, 940–943. https://doi.org/10.1126/science.1192666.

Zhao, J.T., Li, Y.F., Li, Y.Y., Gao, Y.X., Li, B., Hu, Y., Zhao, Y.L., Chai, Z.F., 2014. Selenium modulates Hg uptake and distribution in rice (*Oryza sativa L.*), in correlation with Hg species and exposure level. Metallomics 6, 1951–1957. https://doi.org/10.1039/c4mt00170b.

Zhao, L., Anderson, C.W.N., Qiu, G.L., Meng, B., Wang, D.Y., Feng, X.B., 2016a. Mercury methylation in paddy soil: source and distribution of mercury species at a Hg mining area, Guizhou Province. China. Biogeosciences 13, 2429–2440. https://doi.org/10.5194/bg-13-2429-2016.

Zhao, L., Qiu, G.L., Anderson, C.W.N., Meng, B., Wang, D.Y., Shang, L.H., Yan, H.Y., Feng, X.B., 2016b. Mercury methylation in rice paddies and its possible controlling factors in the Hg mining area, Guizhou province, southwest China. Environ. Pollut. 215, 1–9. https://doi.org/10.1016/j.envpol.2016.05.001.

Zheng, S., Gu, B., Zhou, Q., Li, Y., 2013. Variations of mercury in the inflow and outflow of a constructed treatment wetland in south Florida, USA. Ecol. Eng. 61, 419–425. https://doi.org/10.1016/j.ecoleng.2013.10.015.

Zheng, W., Obrist, D., Weis, D., Bergquist, B.A., 2016. Mercury isotope compositions across North American forests. Glob. Biogeochem. Cy. 30, 1475–1492. https://doi.org/10.1002/2015GB005323.

Zhong, S.Q., Qiu, G.L., Feng, X.B., Lin, C., Bishop, K., 2018. Sulfur and iron influence the transformation and accumulation of mercury and methylmercury in the soil-rice system. J. Soils Sediments 18, 578–585. https://doi.org/10.1007/s11368-017-1786-1.

Zhou, J., Feng, X., Liu, H., Zhang, H., Fu, X., Bao, Z., et al., 2013. Examination of total mercury inputs by precipitation and litterfall in a remote upland forest of southwestern China. Atmos. Environ. 81, 364–372.

Zhu, W., Sommar, J., Lin, C.J., Feng, X., 2015a. Mercury vapor air–surface exchange measured by collocated micrometeorological and enclosure methods - part I: data comparability and method characteristics. Atmos. Chem. Phys. 15, 685–702.

Zhu, W., Sommar, J., Lin, C.J., Feng, X., 2015b. Mercury vapor air–surface exchange measured by collocated micrometeorological and enclosure methods – part II: Bias and uncertainty analysis. Atmos. Chem. Phys. 15, 5359–5376.

Zhu, W., Lin, C.J., Wang, X., Sommar, J., Fu, X., Feng, X., 2016. Global observations and modeling of atmosphere–surface exchange of elemental mercury: a critical review. Atmos. Chem. Phys. 16, 4451–4480.

Zhu, S., Zhang, Z., Zagar, D., 2018. Mercury transport and fate models in aquatic systems: a review and synthesis. Sci. Total Environ. 639, 538–549. https://doi.org/10.1016/j.scitotenv.2018.04.397.