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Review

Mercury in the Canadian Arctic Terrestrial Environment: An Update

Mary Gamberg [a,*], John Chételat [b], Alexandre J. Poulain [c], Christian Zdanowicz [d], Jiancheng Zheng [e]

[a] Gamberg Consulting, Box 30130, Whitehorse, Yukon, Canada Y1A 5M2
[b] Environment Canada, Ottawa, Ontario, Canada K1A 0H3
[c] University of Ottawa, Ottawa, Ontario, Canada K1N 6N5
[d] Uppsala University, Uppsala, Sweden
[e] Natural Resources Canada, Ottawa, Ontario, Canada K1A 0E4

HIGHLIGHTS

- THg and MeHg levels in Arctic snow on land are low, except in coastal areas.
- Hg levels in Canadian Arctic glaciers were stable for most of the past century.
- THg levels in glaciers show an increasing trend along a north-south gradient.
- Water-logged Arctic soils have the potential to methylate mercury.
- An Arctic caribou population shows no recent temporal trend in Hg levels.

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ABSTRACT

Contaminants in the Canadian Arctic have been studied over the last twenty years under the guidance of the Northern Contaminants Program. This paper provides the current state of knowledge on mercury (Hg) in the Canadian Arctic terrestrial environment. Snow, ice, and soils on land are key reservoirs for atmospheric deposition and can become sources of Hg through the melting of terrestrial ice and snow and via soil erosion. In the Canadian Arctic, new data have been collected for snow and ice that provide more information on the net accumulation and storage of Hg in the cryosphere. Concentrations of total Hg (THg) in terrestrial snow are highly variable but on average, relatively low (<5 ng L^{-1}), and methylmercury (MeHg) levels in terrestrial snow are also generally low (<0.1 ng L^{-1}). On average, THg concentrations in snow on Canadian Arctic glaciers are much lower than those reported on terrestrial lowlands or sea ice. Hg in snow may be affected by photochemical exchanges with the atmosphere mediated by marine aerosols and halogens, and by post-depositional redistribution within the snow pack. Regional accumulation rates of THg in Canadian Arctic glaciers varied little during the past century but show evidence of an increasing north-to-south gradient. Temporal trends of THg in glacier cores indicate an abrupt increase in the early 1990s, possibly due to volcanic emissions, followed by more stable, but relatively elevated levels. Little information is available on Hg concentrations and processes in Arctic soils. Terrestrial Arctic wildlife typically have low levels of THg (<5 μg g^{-1} dry weight) in their tissues, although caribou (*Rangifer tarandus*) can have higher Hg because they consume large amounts of lichen. THg concentrations in the Yukon's Porcupine caribou herd vary among years but there has been no significant increase or decrease over the last two decades.

Contents

* Corresponding author. Tel.: +1 867 334 3360.
E-mail address: mary.gamberg@gmail.com (M. Gamberg).

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1. Introduction

Arctic terrestrial ecosystems support a variety of wildlife including populations of mammals and birds that are important to the culture and traditional diet of northern Aboriginal peoples. Considerable information was collected on Hg in terrestrial mammals and birds from 1991 to 2003 during Phases I and II of the Northern Contaminants Program (NCP), with broad geographic coverage across the Canadian Arctic. In general, Hg levels are below levels of concern for wildlife health in large mammals such as caribou, moose, wolf and wolverine (Fisk et al., 2003). Terrestrial environments are also important in the regional Hg cycle because watersheds supply Hg to freshwater and marine ecosystems. Snow, ice, and soils on land are key reservoirs for atmospheric deposition and, in turn, can become sources of Hg through the melting of the cryosphere (terrestrial ice and snow) and via soil erosion (Douglas et al., 2012; Stern et al., 2012). A major knowledge gap identified in the second Canadian Arctic Contaminants Assessment Report (CACAR II) of the NCP was the lack of information on the fate of Hg following atmospheric deposition (Bidleman et al., 2003). Further study was recommended in order to understand the relationship between Hg deposition in snow and its incorporation into marine, freshwater, and terrestrial food webs (Bidleman et al., 2003). Since the CACAR II, several important advances have been made regarding the fate of Hg in the terrestrial environment. New data have been collected for snow and ice that provide more information on the net accumulation (flux) of Hg in the cryosphere, its storage and post-depositional fate. The monitoring of caribou and moose continued during Phase III of the NCP and included more frequent sampling of caribou to increase the statistical power of temporal trend monitoring for this key terrestrial indicator species. Vegetation (mostly lichen) was also measured for Hg levels to provide information on uptake at the base of Arctic terrestrial food webs.

The intent of this paper is to evaluate the state of science on Hg in terrestrial environments of the Canadian Arctic, providing a more geographically-focussed approach than the recent general circumpolar review of Douglas et al. (2012). The discovery of Atmospheric Depletion Events (AMDEs) (Steffen et al., 2005) has influenced much of the abiotic research in the Canadian North over the last decade, resulting in the major focus of this review on terrestrial snow and ice as repositories of atmospheric Hg. However, the terrestrial environment is generally less studied than the freshwater and marine environments, and this review also serves to identify important knowledge gaps that should be addressed because of their potential role in the cycling of Hg in the Arctic environment.

2. Terrestrial Hg cycle

2.1. General overview

Mercury enters the Arctic terrestrial environment primarily as divalent inorganic Hg [Hg(II)] via two main pathways: atmospheric deposition and geological weathering (Fig. 1). MeHg is also deposited from the atmosphere although this flux is probably small relative to inorganic Hg (Lehnerr, 2012). Hg(II) in snow, ice, and soil can have several fates: 1) photochemical reduction to gaseous elemental Hg (GEM) and subsequent evasion to the atmosphere, contributing to reemission of newly deposited Hg (II); 2) transient storage in the snowpack or glacier ice; 3) microbial methylation in soils and possibly also in snow (Larose et al., 2010); 4) transport to aquatic ecosystems via snowmelt, surface runoff, and erosion; 5) uptake by vegetation, with a subsequent return to snow or soils (via throughfall and litterfall) or entering the terrestrial food chain through herbivores and inevitably returning to soils through decomposition.

2.2. Photochemical Hg (II) reduction and emission

Photoreduction and subsequent evasion of volatile Hg^0, which is the main mechanism thought to drive Hg emissions from snow and ice, was discussed thoroughly by Steffen et al. (2013a) in an atmospheric context because of its importance following AMDEs. In soil, Hg (II) originating from atmospheric deposition and geological weathering may also undergo photochemical reduction and subsequent evasion/ emission to the atmosphere. Sunlight intensity, temperature and soil moisture may affect the rate of GEM emission from soil (Carpi and Lindberg, 1998; Schlüter, 2000), while oxidation of GEM by abiotic or microbial processes may reduce net emission from soils. In temperate regions, Hg(II) reduction may be driven by sunlight penetrating into the top few millimetres of soil, by the presence of abiotic reductants such as humic and fulvic acids, or by reactions mediated by microbes (Gabriel and Williamson, 2004). Low emission rates might be expected from Arctic soils because of lower temperatures, lower annual solar radiation, and lower microbial activity compared to temperate regions. However, climate change may impact many of the yet unidentified environmental conditions that influence soil Hg (II) reduction and evasion in the Arctic. Measurements made at various sites across Canada, mostly south of 60° N latitude, revealed a positive correlation between the flux of gaseous Hg to the atmosphere and the THg concentration of the mineral substrate (Schroeder et al., 2005). Emissions of gaseous Hg from moss-covered mercuriferous shale at MacMillan Pass in Yukon (mean: 1.5 ng m^{-2} h^{-1}, range: -0.60–10.63 ng m^{-2} h^{-1}) were considerably higher than from moss-covered substrate having background levels of geological Hg near Kuujjuarapik, Quebec (mean: 0.08 ng m^{-2} h^{-1}, range: -0.58–1.3 ng m^{-2} h^{-1}) (Schroeder et al., 2005). Mechanisms of Hg reduction and estimates of emission from terrestrial substrates are significant knowledge gaps in the Arctic Hg cycle.

2.3. MeHg deposition and production

Although pathways for Hg methylation in Arctic terrestrial ecosystems remain poorly characterized, several novel processes have been

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