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Review

Atmospheric mercury in the Canadian Arctic. Part II: Insight from modeling

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HIGHLIGHTS

- Arctic is a sink of atmospheric mercury transported from lower latitudes.
- Deposition of Hg shows an increasing trend along a North–South gradient.
- Asian outflow of Hg contributes most to anthropogenic Hg deposition in the Arctic.
- Model simulates decreasing Hg in air but increasing Hg deposition in the Arctic.
- Emission controls worldwide can reduce Hg deposition in the Arctic.

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ABSTRACT

A review of mercury in the Canadian Arctic with a focus on field measurements is presented in part I (see Steffen et al., this issue). Here we provide insights into the dynamics of mercury in the Canadian Arctic from new and published mercury modeling studies using Environment Canada's mercury model. The model simulations presented in this study use global anthropogenic emissions of mercury for the period 1995–2005. The most recent modeling estimate of the net gain of mercury from the atmosphere to the Arctic Ocean is 75 Mg year^{-1} and the net gain to the terrestrial ecosystems north of 66.5° is 42 Mg year^{-1}. Model based annual export of riverine mercury from North American, Russian and all Arctic watersheds to the Arctic Ocean are in the range of 2.8–5.6, 12.7–25.4 and 15.5–31.0 Mg year^{-1}, respectively. Analysis of long-range transport events of Hg at Alert and Little Fox Lake monitoring sites indicates that Asia contributes the most ambient Hg to the Canadian Arctic followed by contributions from North America, Russia, and Europe. The largest anthropogenic Hg deposition to the Canadian Arctic is from East Asia followed by Europe (and Russia), North America, and South Asia. An examination of temporal trends of Hg using the model suggests that changes in meteorology and changes in anthropogenic emissions equally contribute to the decrease in surface air elemental mercury concentrations in the Canadian Arctic with an overall decline of ~12% from 1990 to 2005. A slow increase in net deposition of Hg is found in the Canadian Arctic in response to changes in meteorology. Changes in snowpack and sea-ice characteristics and increase in precipitation in the Arctic related with climate change are found to be primary causes for the meteorology-related changes in air concentrations and deposition of Hg in the region. The model estimates that under the emissions reduction scenario of worldwide implementation of the best emission control technologies by 2020, mercury deposition could potentially be reduced by 18–20% in the Canadian Arctic.

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1. Introduction

Inhabitants of northern Canada have among the highest rates of exposure to mercury in the world because traditional northern diets often include marine mammals and fish that contain high levels of mercury (Hg) (Braune et al., 2005; AMAP, 2011). Hg levels in many marine species are up to an order of magnitude higher now than in pre-industrial times (Outridge et al., 2002 & 2005). The Minamata Convention on Mercury (led by the United Nations Environment Programme; 2013) noted that the Arctic ecosystems and indigenous communities are particularly vulnerable to mercury pollution because of the biomagnification of mercury and contamination of traditional foods. Consequently, understanding the transport and fate of mercury in the Arctic is an important scientific and policy issue. A review of the understanding of atmospheric mercury transport and processes in the Canadian Arctic from the recent field measurements is presented in part 1 of the study (see Steffen et al. in this issue). This study explains pathways, sources, trends and budgets of mercury in the Canadian Arctic from new and published modeling studies using Environment Canada's mercury model (GRAHM).

Atmospheric mercury models simulate the cycling of Hg in the atmosphere starting from emissions to the air and ending with deposition to terrestrial and aquatic surfaces. Emissions to the air include anthropogenic emissions, natural emissions, and re-emissions of previously deposited Hg of both anthropogenic and natural origins. These models are used to estimate ambient Hg concentrations and deposition fluxes, to understand atmospheric transport pathways, to estimate source attribution, to explain long-term trends, and to predict future levels of Hg pollution. The models complement direct measurements by providing spatial coverage and detailed information on Hg budgets in the Arctic environment.

Recent global mercury assessment report published by AMAP/UNEP provides an excellent review of global and regional models that have been applied to study the problem of mercury contamination (AMAP/UNEP, 2013). Atmospheric Hg models incorporate emissions of gaseous elemental mercury (GEM) and oxidized mercury as gas (Reactive Gaseous Mercury (RGM)) and on particles (Particulate Mercury (PHg)), and their parameterized gas and aqueous phase chemistry, phase exchange processes between air, aerosol and clouds, wet and dry deposition, boundary layer and cumulus cloud mixing, and transport. Atmospheric chemistry models require spatially and temporally resolved meteorological information to represent transport and physical–chemical processes. Some models use meteorological variables from external sources such as meteorological observational systems or meteorological model simulations; whereas, other models simulate meteorological variables along with chemical variables in the atmosphere. Owing to the global transport of Hg, hemispheric or global scale atmospheric models have been developed to estimate the atmospheric Hg contribution to the Arctic. The global models are typically integrated (advanced temporally) for a few years to establish a balance between the emissions, atmospheric Hg concentrations, and deposition. Current Hg models are primarily constrained by measurements of surface-level atmospheric GEM concentrations and wet deposition fluxes from North America and Europe where long-term observations with sufficient spatial coverage are available (AMAP/UNEP, 2013). A limited number of vertical profiles of GEM and fluxes of Hg from terrestrial and oceanic surfaces are also used to constrain the models. Current measurements of RGM and PHg concentrations in the atmosphere are very limited and have a high degree of uncertainty for use in model development (Kos et al., 2013; Gustin and Jaffe, 2010; Lyman et al., 2010). Since wet deposition of Hg occurs through the scavenging of oxidized Hg species in the atmosphere, it is possible to constrain the total oxidized Hg concentration in the lower troposphere using observed Hg concentrations in precipitation. Additionally, Hg modeling in the Arctic is further complicated by AMDE (Atmospheric Mercury Depletion Event) processes.

Environment Canada has developed the Global/Regional Atmospheric Heavy Metals Model (GRAHM) which includes a complete set of meteorological processes and Hg physical–chemical processes (Dastoor and Durnford, 2014; Durnford et al., 2012a,2012b; Dastoor et al., 2008; Dastoor and Larocque, 2004). Three other atmospheric models have been applied to model mercury in the Arctic are as follows: 1) Danish Eulerian Hemispheric Model (DEHM) (Christensen et al., 2004); 2) Global EMEP Multi-media Modelling System (GLEMOS) (Travnikov and Ilyin, 2009); and, 3) a global model driven by data from the Goddard Earth Observing System (GEOS-Chem) (Fisher et al., 2012; Holmes et al., 2010). All four models have incorporated AMDEs as part of the chemistry. The largest differences among models are found in the values and spatial distribution of natural emissions and re-emissions, the major oxidants of GEM, the reaction products represented and, in the Arctic, the GEM-Br oxidation rates, Br concentrations and parameterization of re-emission of GEM from the snowpack (Munthe et al., 2011).

2. Factors included in the Global/Regional Atmospheric Heavy Metals Model (GRAHM)

AMAP/UNEP (2011) provides a comparable set of historical global anthropogenic emissions of mercury for the period 1990 to 2005 which uses a standardized methodology and consistent information for estimating emissions from various sectors. Recently, AMAP/UNEP (2013) released a global mercury emissions inventory for 2010 which uses a revised methodology for the specification of emissions. The notable difference between the mercury emissions in 2005 and 2010 is a sharp increase in contribution from Artisanal and Small scale Gold Mining (ASGM) sector in 2010 (i.e. ~727 Mg in 2010 vs. ~350 Mg in 2005). Currently, a consistent set of historical mercury emissions (using same methodology) from 1990 to 2010 is unavailable; therefore, GRAHM simulations presented in this study use the anthropogenic emissions of mercury from AMAP/UNEP (2011) for the period1990–2005. The increases in Hg emissions from ASGM sector (in 2010) are primarily located in the southern hemisphere; thus, the impact of increased emissions from ASGM sector is expected to be minor in the Canadian Arctic. A full assessment of the impact of global anthropogenic mercury emissions in 2010 in the Canadian Arctic will be conducted in future modeling studies.

Total global emissions from natural sources and re-emissions of previously deposited Hg (from land and oceans) in GRAHM are