



Review

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



Ashu Dastoor^{a,*}, Andrew Ryzhkov^a, Dorothy Durnford^b, Igor Lehnerr^c,
Alexandra Steffen^d, Heather Morrison^d

^a Air Quality Research Division, Environment Canada, 2121 TransCanada Highway, Dorval, QC H9P 1J3, Canada

^b Meteorological Service of Canada, Environment Canada, 2121 TransCanada Highway, Dorval, QC H9P 1J3, Canada

^c University of Waterloo, Department of Earth and Environmental Sciences, Waterloo, Ontario N2L 3G1, Canada

^d Environment Canada, Air Quality Research Division, Toronto, Ontario M3H 5T4, Canada

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 $15.5\text{--}31.0 \text{ Mg year}^{-1}$, 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 $\sim 12\%$ 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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Contents

1. Introduction	17
2. Factors included in the Global/Regional Atmospheric Heavy Metals Model (GRAHM)	17

* Corresponding author. Tel.: +1 514 421 4766; fax: +1 514 421 2106.
E-mail address: Ashu.Dastoor@ec.gc.ca (A. Dastoor).

3. Model simulations of Hg deposition in the Arctic and comparison with field measurements	18
4. Model simulations of Hg concentrations in surface air	20
5. Long-range transport and source attribution of Hg deposition in the Canadian Arctic	20
6. Temporal trends of ambient concentration and deposition of mercury	22
7. Future projections of Hg deposition under various Hg emission scenarios	25
8. Uncertainties in model estimates of deposition in the Arctic	25
9. Summary	26
Acknowledgments	26
References	26

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 period 1990–2005. 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

based on the global Hg budgets by Gbor et al. (2007), Shetty et al. (2008) and Mason (2009). Land-based natural emissions are spatially distributed according to the natural enrichment of Hg. Land re-emissions are spatially distributed according to the historic deposition of Hg and land-use type. Oceanic emissions depend on the distributions of primary production and atmospheric deposition. Both land and oceanic emissions temporally depend on the surface temperature. Scavenging of gaseous and particulate Hg species into atmospheric precipitation (wet deposition) and direct deposition to the earth's surface (dry deposition) are the removal mechanisms employed in the model. The dry deposition velocity is calculated using the resistance analogy (aerodynamic resistance, quasi-laminar sub-layer resistance, and surface resistance) for GEM and RGM, while the dry deposition velocity of PHg is calculated as fine particulate matter (Zhang et al., 2001, 2003). Durnford et al. (2012a, 2012b) developed and implemented a dynamic multi-layer snowpack/meltwater parameterization for mercury in GRAHM that interacts with mercury in the atmospheric boundary layer allowing the representation of bidirectional fluxes of mercury between air and snowpack/meltwater. Redox and diffusion processes of mercury species are represented in snowpack layers and meltwater.

Gas phase oxidation with O₃, OH radical and halogens (mainly Br) have been suggested as potential oxidants of GEM in the atmosphere. Although, the significance of GEM oxidation by O₃ and OH in the atmosphere is controversial based on several theoretical studies (Goodsite et al., 2004; Cremer et al., 2008), Subir et al. (2012) and Rutter et al. (2012) recommend including GEM oxidation with O₃ and OH in Hg models. Determined reaction rate constants for the oxidation of GEM by O₃ and OH in the atmosphere suggest a significantly shorter life time of GEM in the atmosphere compared to the ~1 year life time suggested by the observations. Calvert and Lindberg (2005) and Subir et al. (2012) suggest that GEM oxidation by O₃ and OH may be occurring in the atmosphere through complex reaction mechanism possibly involving surfaces and that the effective oxidation rate of GEM involving O₃/OH is likely much slower than determined by the experiments. Another possibility is that there are important reduction processes in the atmosphere that are currently unknown. Oxidation of GEM by Bromine radical is widely accepted as an important oxidation pathway in the polar and marine boundary layers, however very little data exists with respect to its significance and mechanism in the global atmosphere (Dibble et al., 2012). Only a limited number of reduction pathways for Hg in the aqueous phase have been identified and the mechanisms is not clear (Xiao et al., 1995; Van Loon et al., 2000; Si and Ariya, 2008). Hynes et al. (2009) concluded that the atmospheric importance of Hg reduction processes has not been established for any of the suggested reductants for Hg²⁺ so far and the role of reduction processes in the global atmosphere remains conjectural. In the current version of GRAHM, gaseous oxidation of mercury by O₃ and OH are included in the model with a rate constant for O₃ oxidation following Hall (1995) and one third of the reaction rate constant determined by Pal and Ariya (2004) for OH. Our choice of the reduced oxidation reaction rate for OH is constrained by the life time of GEM in the atmosphere of ~1 year. In the polar and marine boundary layers, the model includes gaseous oxidation of mercury by halogens, including atomic and molecular chlorine and bromine as well as bromine oxide (Ariya et al., 2002; Raofie and Ariya, 2003; Donohoue et al., 2006; Goodsite et al., 2004). Mercury is reduced in the aqueous phase photochemically and by the sulfite anion using rate constants from Xiao et al. (1995) and Van Loon et al. (2000) in GRAHM; however, the impact of these reduction processes is insignificant on Hg cycling in the model. Oxidation of Hg by Bromine species alone in all environments is currently being investigated in GRAHM. The GRAHM model simulations reported in this study were carried out at 1° × 1° latitude-longitude horizontal resolution and 28 vertical levels ending at 10 mb pressure level in the atmosphere.

3. Model simulations of Hg deposition in the Arctic and comparison with field measurements

In recent years, several models have provided estimates of Hg deposition to the Arctic. Early estimates tended to be higher as most models did not initially include post-AMDE Hg reemission or more comprehensive treatments of snow-phase redox chemistry. For example, using DEHM, which does not account for reemission from snow, Skov et al. (2004) estimated a deposition of 208 Mg year⁻¹ of Hg to the Arctic area north of the polar circle (66.56° N), of which 120 Mg year⁻¹ was attributed to AMDEs. Using an early version of GRAHM that did not incorporate post-AMDE re-emission, Ariya et al. (2004) estimated that 325 Mg year⁻¹ of Hg is deposited in the Arctic north of 60° including 100 Mg year⁻¹ deposited as a result of AMDEs. By incorporating bidirectional exchange of Hg fluxes in the Arctic in GRAHM to account for the fast re-emission of Hg from snowpack following AMDEs, Dastoor et al. (2008) obtained a lower estimate of 174 Mg year⁻¹ for the net deposition of Hg in snow north of 66.5° latitude. Net deposition is defined here as yearly gross deposition minus re-emission of seasonally deposited Hg to snowpacks. Brooks et al. (2006) measured Hg deposition, re-emission, and net surface gain fluxes of Hg at Barrow (Alaska) between March 25 and April 7, 2003, as 1.7 μg m⁻², 1.0 ± 0.2 μg m⁻² and 0.7 ± 0.2 μg m⁻², respectively. Dastoor et al. (2008) found excellent agreement between model-derived fluxes and measurements during the Barrow study with rates of 1.8 μg m⁻² deposition, 1.0 μg m⁻² re-emission and 0.8 μg m⁻² net surface gain of Hg. The most recent version of GRAHM yields a net deposition rate of 153 Mg year⁻¹ and a net surface gain of mercury of 117 Mg year⁻¹ above the Arctic Circle (66.5° N) (Durnford et al., 2012b). Net surface gain of mercury is estimated by subtracting all mercury emissions (including oceanic evasion) from gross deposition in the Arctic. The net gain of mercury from the atmosphere to the Arctic Ocean is estimated as 75 Mg year⁻¹ (58 Mg year⁻¹ direct deposition, 50 Mg year⁻¹ deposition via snow melt and 33 Mg year⁻¹ evasion) and the net gain to the terrestrial ecosystems north of 66.5° is estimated at 42 Mg year⁻¹ (29 Mg year⁻¹ direct deposition, 16 Mg year⁻¹ deposition via snow melt and 3 Mg year⁻¹ emissions from soils). In comparison, the yearly net deposition flux estimated by GLEMOS, DEHM and GEOS-Chem (two estimates) in the Arctic are 131 Mg year⁻¹, 110 Mg year⁻¹, and 80 Mg year⁻¹, -35 Mg year⁻¹ respectively (Travnikov and Ilyin, 2009; Christensen et al., 2004; Holmes et al., 2010; Fisher et al., 2012). Fig. 1 shows the annual net deposition of Hg (left) and annual average surface air concentrations (right) in the Arctic simulated by GRAHM. The model, in general, simulates a north to south deposition gradient with increasing deposition that could be attributed to Br concentrations (which are higher in coastal regions), proximity to emission sources, and increasing precipitation amounts. The eastern sub-Arctic displays higher Hg deposition and lower GEM concentrations compared to the western sub-Arctic. The eastern sub-Arctic appears to be directly influenced by North American sources.

In the Canadian Arctic, the average deposition flux using GRAHM (from all global sources) is simulated to be ~4.5 μg m⁻² year⁻¹ in the High Arctic (north of 66.5°), ~7 μg m⁻² year⁻¹ in Yukon sub-Arctic, ~7.5 μg m⁻² year⁻¹ in North West Territories sub-Arctic and ~6.5 μg m⁻² year⁻¹ in Nunavut sub-Arctic. Atmospheric Hg deposition has also been estimated using various field approaches, including dated lake sediment cores, snow measurements and wet deposition monitoring. Using dated sediment cores from 32 lakes across the Canadian Arctic, Muir et al. (2009) obtained net atmospheric Hg deposition fluxes of 2.8 μg m⁻² year⁻¹ (High Arctic) and 7.5 μg m⁻² year⁻¹ (subarctic), which is in good agreement with values obtained from GRAHM. Fluxes inferred from sediment cores exhibited a negative relationship between latitude and Hg deposition, consistent with the GRAHM model results (Muir et al., 2009). By comparison, Hg deposition fluxes in lake sediments from Northern Alaska were 2.8 ± 0.7 μg m⁻² year⁻¹ (Fitzgerald et al., 2005), which also agrees well with net atmospheric

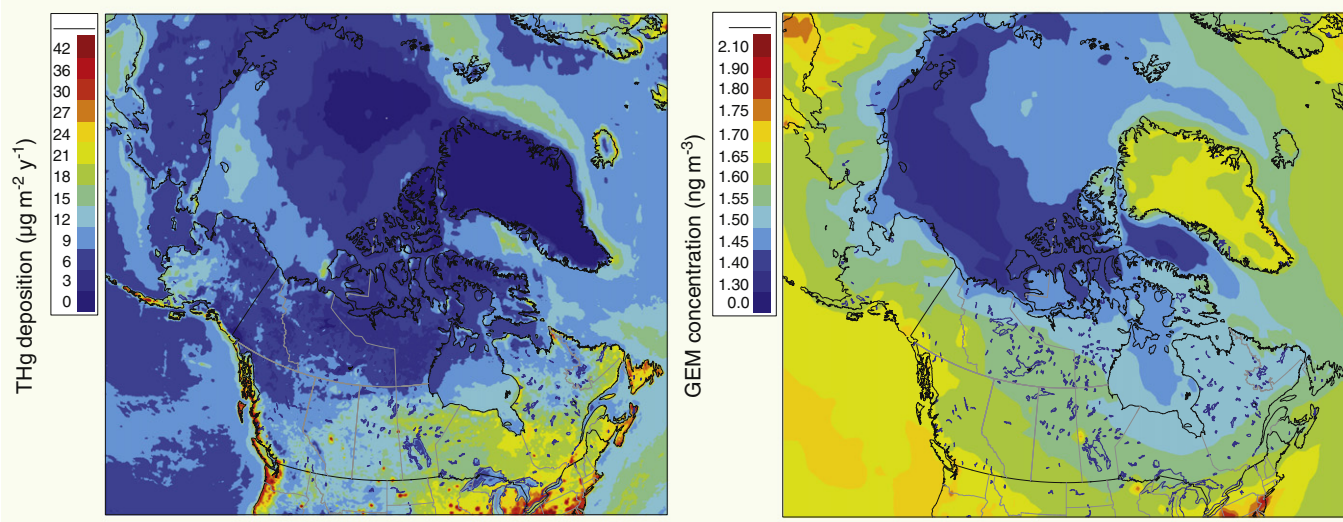


Fig. 1. Spatial distribution of THg deposition ($\mu\text{g m}^{-2} \text{year}^{-1}$; left), and surface air concentration of GEM (ng m^{-3} ; right) in the Canadian Arctic simulated by GRAHM in the year 2005.

deposition fluxes of $3.0 \mu\text{g m}^{-2} \text{year}^{-1}$ predicted by GEOS-Chem (Holmes et al., 2010). Flux values obtained from snowpack measurements in the Canadian Arctic are somewhat lower ($0.5\text{--}3.5 \mu\text{g m}^{-2}$; St. Louis et al., 2007) than the GRAHM-derived estimates, likely because they only reflect net Hg deposition taking place during the winter/spring season. Dastoor and Durnford (2014) found median concentrations of mercury in seasonal snowpacks simulated by the model within the measured range at Alert (Canada), Ny-Ålesund (Norway) and Barrow (USA) (e.g. 6.0 ng L^{-1} modeled and $4.9\text{--}6.0 \text{ ng L}^{-1}$ measured at Alert in spring 2005).

A few datasets of measured wet Hg flux are available from sub-Arctic precipitation collection stations at Churchill and Fort Vermilion (Alberta) in Canada (Sanei et al., 2010), and Kodiak, on the Pacific coast of Alaska (through the MDN, <http://nadp.sws.uiuc.edu/mdn>). Gross wet deposition fluxes from these stations were compared to model estimates from GRAHM for these sites (Sanei et al., 2010). The modeled wet Hg fluxes for the three sites ranged from 2.1 to $4.1 \mu\text{g m}^{-2} \text{year}^{-1}$ compared to measurements of 0.54 to $5.3 \mu\text{g m}^{-2} \text{year}^{-1}$. The model estimates were closest to the measured fluxes at Kodiak (GRAHM = +23%). The largest discrepancy between observations and model estimates was found at Churchill. Kirk et al. (2006) measured high concentrations of Hg in snow during AMDEs (over 100 ng L^{-1}). However, Sanei et al. (2010) did not measure high wet deposition fluxes ($0.5\text{--}2 \mu\text{g m}^{-2} \text{year}^{-1}$) at Churchill during spring using their method (see also Section 3), which perhaps is more consistent with the low snowpack Hg loads measured following reemission of Hg deposited during AMDEs ($0.04\text{--}0.4 \mu\text{g m}^{-2}$; Kirk et al., 2006). The relative contribution of wet and dry deposition at Arctic sites, including Churchill, is unknown. There is a discrepancy between measured and modeled partitioning of RGM and PHg at Alert (Fig. 3) that can give rise to inaccurate proportion of Hg deposition as dry and wet deposition. Dry deposition flux measurements and additional wet deposition measurements are required to fully evaluate the models and the impact of AMDEs in the Arctic.

Median mercury concentrations in snow collected on tables during precipitation events in the springtime (AMDEs season) are significantly higher at Barrow (47 ng L^{-1} in April–May, 2005) than at Alert (7.5 ng L^{-1} in March–May, 2005) which is well simulated by the model (37 ng L^{-1} at Barrow and 8.4 ng L^{-1} at Alert; Dastoor and Durnford, 2014). Bettles, Alaska and Fort Vermillion, Canada are in the Arctic Ocean watershed. The model simulates annual median mercury concentration in precipitation at Fort Vermillion accurately (3.8 ng L^{-1} modeled; 3.9 ng L^{-1} measured in 2007) but over-

estimates at Bettles (3.8 ng L^{-1} modeled; 2.1 ng L^{-1} measured in 2009) likely due to insufficient model resolution to resolve precipitation at this site. However, the mean model (4.8 ng L^{-1}) and observed (5.6 ng L^{-1}) mercury concentrations in precipitation at Bettles are closer (Dastoor and Durnford, 2014).

Durnford et al. (2012b) and Fisher et al. (2012) estimated depositional and emission fluxes of mercury in the terrestrial and marine ecosystems in the Arctic. Differences in the simulated atmosphere/ocean mercury fluxes by GEOS-Chem (Fisher et al., 2012) and GRAHM (Durnford et al., 2012b) were discussed by Dastoor and Durnford (2014). Both models estimated very similar total inputs of mercury to the Arctic Ocean from the atmosphere and rivers combined ($125/124\text{--}139 \text{ Mg year}^{-1}$ for GEOS-Chem/GRAHM). However, the riverine source ($80/15.5\text{--}31 \text{ Mg year}^{-1}$ for GEOS-Chem/GRAHM) was dominant in GEOS-Chem while the atmosphere was the dominant source ($45/108 \text{ Mg year}^{-1}$ for GEOS-Chem/GRAHM) of mercury to the Arctic Ocean in GRAHM. Comparison of modeled to observed Hg concentrations in air, precipitation and snowpacks suggest that GRAHM does not overpredict mercury deposition in the Arctic (Dastoor and Durnford, 2014). The riverine delivery of mercury to the Arctic Ocean is poorly constrained by observations (Kirk et al., 2012). Estimates of the riverine source (in Mg year^{-1}) include 12.5 (max = 39 ; Outridge et al., 2008), $15.5\text{--}31$ (based on GRAHM simulated snowpack meltwater mercury concentrations; Dastoor and Durnford, 2014), 50 (based on observed Dissolved Organic Carbon (DOC) to mercury ratios from multiple measurements; Dastoor and Durnford, 2014), 80 (inferred with GEOS-Chem simulation of the Arctic Ocean mercury budgets; Fisher et al., 2012), and 108 (deduced from observed DOC to mercury ratios from Yukon River; Kirk et al., 2012). Dastoor and Durnford (2014) also provided separate estimates of riverine Hg exports from the North American and Russian watersheds to the Arctic Ocean at 8.6 and $41.6 \text{ Mg year}^{-1}$, respectively, based on observed DOC/Hg ratios and in the range of $2.8\text{--}5.6$ and $12.7\text{--}25.4 \text{ Mg year}^{-1}$, respectively, based on modeled mercury in meltwater.

GEOS-Chem and GRAHM provide very different estimates of evasion from the Arctic Ocean ($90/33 \text{ Mg year}^{-1}$ for GEOS-Chem/GRAHM). The limited observations of evasion from the Arctic Ocean indicate great variability both temporally and spatially. GEOS-Chem's stronger simulated ocean evasion of $44 \text{ ng m}^{-2} \text{day}^{-1}$ in August/September is in line with the limited observations in Canadian archipelago (Kirk et al., 2012). GRAHM's evasion over the central Arctic Ocean is stronger than in GEOS-Chem. Strong evasion polewards of 80° N is supported by the observation-based study of Hirdman et al. (2009). Despite their different estimates of total evasion, both GEOS-Chem and GRAHM agree

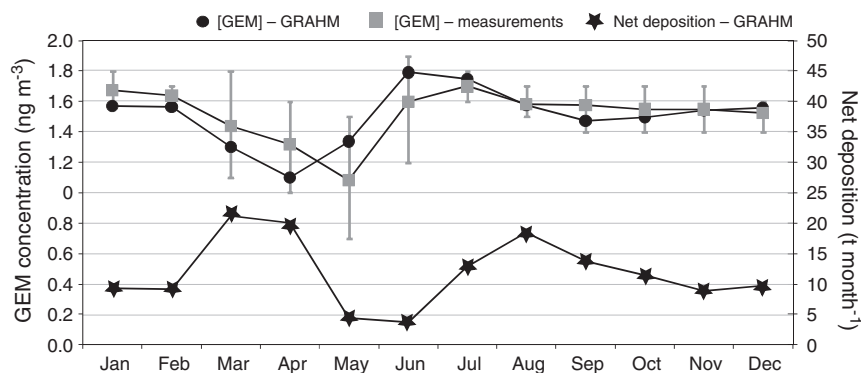


Fig. 2. Monthly mean ambient GEM concentrations (north of 66.5°) simulated by GRAHM and observed concentrations averaged over six Arctic sites: Barrow (USA), Alert (Canada), Station Nord (Greenland), Ny-Ålesund (Norway), Andoya (Norway), and Amderma (Russia) (ng m^{-3} ; left scale). Total monthly deposition was simulated by GRAHM (Mg month^{-1} ; right scale).

that ocean evasion supports the elevated concentrations of atmospheric mercury observed during summer in the Arctic. The disparity in the two model's mercury flux estimates reflects the current uncertainties in mercury modeling in the Arctic. To close the gap between modeling estimates, comprehensive observations in all compartments of the Arctic ecosystem are needed.

4. Model simulations of Hg concentrations in surface air

Dastoor and Durnford (2014) compared simulated concentrations of GEM and RGM in air with observed data at Alert (Canada), Ny-Ålesund (Norway), Amderma (Russia), and Barrow (USA) in all seasons for 2005–2009. The model median concentrations of GEM and RGM were found within the range of observed medians at all locations (e.g., 1.22 ng/m^3 and 0.1 ng/m^3 modeled GEM and RGM concentrations in spring 2005, respectively, and 1.11 ng/m^3 and 0.17 ng/m^3 measured GEM and RGM concentrations, respectively, at Alert in spring 2005).

Monthly mean GEM concentrations and deposition fluxes derived from GRAHM were compared to measured monthly GEM means from combined years at six Arctic monitoring sites: Barrow (USA), Alert (Canada), Station Nord (Greenland), Ny-Ålesund (Norway), Andoya (Norway), and Amderma (Russia) (Fig. 2) in this study. The GRAHM-derived and the measured yearly average GEM concentrations at the six sites are 1.47 and 1.52 ng m^{-3} , respectively. Close agreement between the two averages suggests that very similar physical-chemical processes are occurring throughout coastal circumpolar Arctic regions because the model employs a common set of mercury process parameterizations at all sites. Dastoor and Durnford (2014) show that the surface air concentrations of GEM in the Arctic tend to peak during the snowmelt season (end of May to mid-June at Alert) which is supported primarily by revolatilisation of Hg from snowpacks/meltwater; some of the highest GEM concentrations are observed (also shown by model) around this time. GEM concentrations are also elevated later in summer (mid July to early August at Alert). The summer maximum in Arctic GEM air concentrations is supported by oceanic evasion (Dastoor and Durnford, 2014; Fisher et al., 2012).

The model chemical parameterization is capable of simulating the seasonal pattern of oxidized Hg (RGM and PHg) at Alert (Fig. 3). The magnitude of oxidation in the springtime is simulated within the measured range, whereas there is notable discrepancy in estimating the partitioning between gas and aerosol phases. Currently, partitioning between RGM and PHg in the model is uncertain because the heterogeneous chemistry is a knowledge gap.

5. Long-range transport and source attribution of Hg deposition in the Canadian Arctic

The origin of atmospherically deposited Hg in the Arctic was analyzed by estimating the contribution from anthropogenic emissions in the source regions shown in Fig. 4 (in this study). Total anthropogenic emissions of Hg (in Mg y^{-1}) in these regions and the percent contribution to total global anthropogenic emissions are provided. The combined contributions to deposition of natural emissions and re-

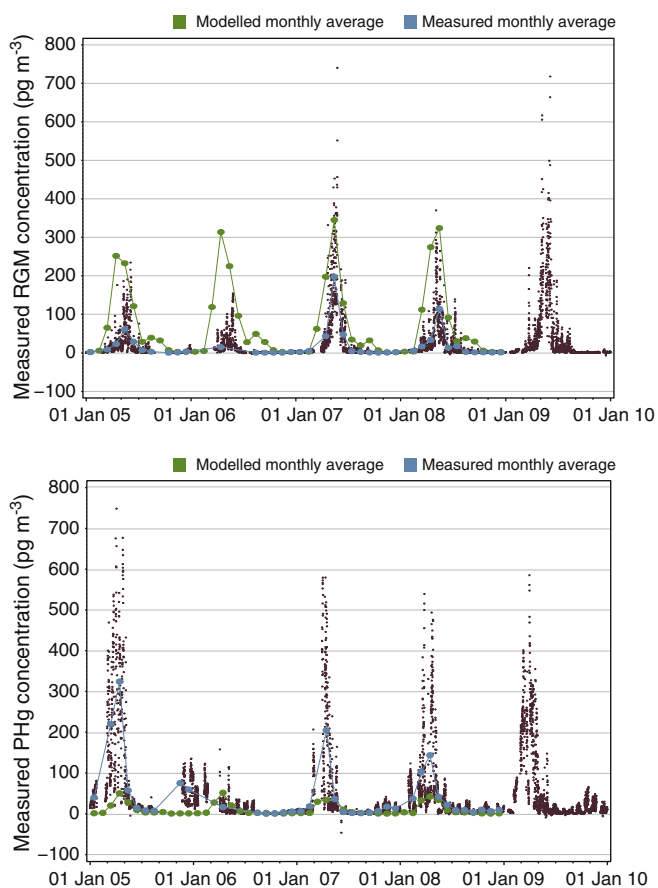


Fig. 3. Surface air concentrations of RGM (top panel) and PHg (bottom panel) simulated by GRAHM and measured at Alert (Nunavut) in pg m^{-3} . Dark purple dots represent the concentrations from measured samples, blue dots represent the monthly average of those measured samples, and green dots represent the monthly average concentration predicted by the model.

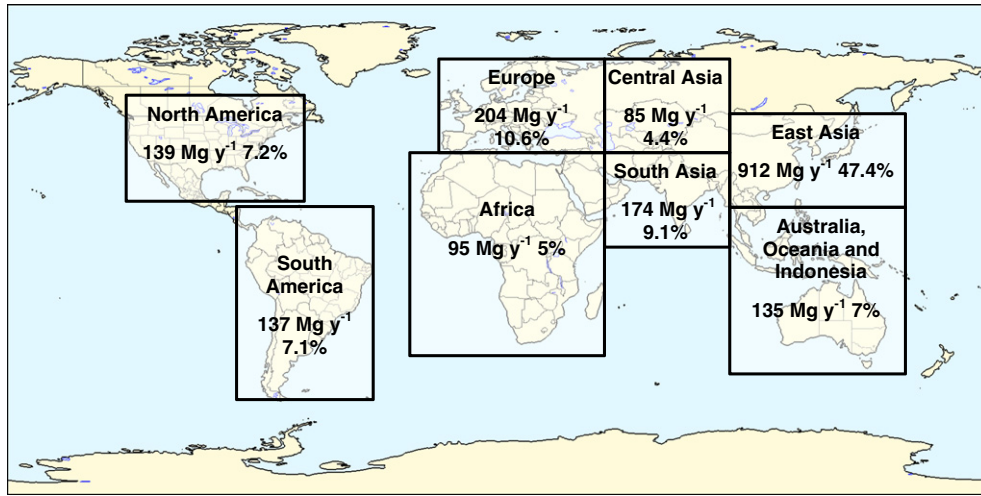


Fig. 4. Location of source regions considered in the source attribution study. Total anthropogenic emissions (Mg year^{-1}) in these regions and the percent contributions to global anthropogenic emissions of Hg for 2005 are shown on the map. Receptor regions in the study are the same as the source regions.

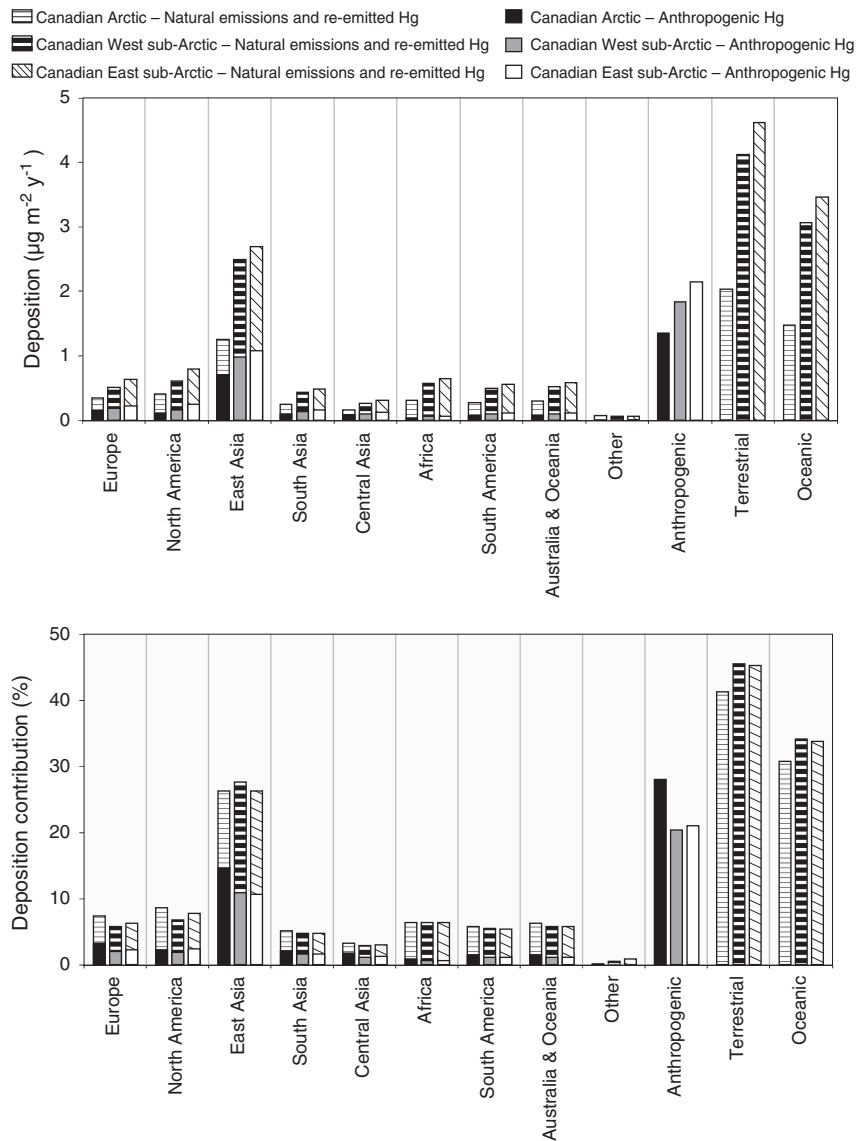


Fig. 5. Contributions of major Hg emission source regions to THg deposition to the Canadian High Arctic, western sub-Arctic and eastern sub-Arctic simulated by GRAHM in 2005. Contributions are presented as absolute values ($\mu\text{g m}^{-2} \text{ year}^{-1}$; top panel) and as percentages (bottom panel).

emissions, anthropogenic emissions, and oceanic emissions were simulated using the GRAHM model.

Fig. 5 presents estimates of contributions to the 2005 deposition flux from each source region as well as total contributions from all anthropogenic, terrestrial, and oceanic emissions as absolute values (top panel) and as a percentage of total deposition to Canadian High Arctic and sub-Arctic regions (bottom panel). The eastern sub-Arctic region showed the highest deposition fluxes of all the Arctic regions, possibly because of higher precipitation rates, favorable land-use types for dry deposition, and the presence of AMDEs around Hudson Bay.

Fig. 5 (bottom panel) shows the percent contribution from various source regions to deposition in the Canadian Arctic. The anthropogenic contribution to Hg deposition is higher (28%) in the Canadian High Arctic than the sub-Arctic (20–21%) due to higher contributions from East Asia (15%, 11% in High Arctic and sub-Arctic, respectively) and Europe (3%, 2% in High Arctic and sub-Arctic, respectively). East Asian emissions contribute the largest proportion of anthropogenic Hg deposition to the Canadian Arctic (10–15%) followed by Europe (2–3%), North America (2–2.5%), and South Asia (1.5–2%). Overall in the Canadian Arctic, the total contribution from all emissions in East Asia (26 to 28%) to annual Hg deposition is more than twice that of the next biggest contributors, the U.S. (7 to 9%) followed by Europe (6 to 7%). These results suggest that increasing emissions in East Asia could increase deposition of Hg to the Canadian Arctic and offset the decrease in deposition that has resulted from reductions of North American, European and Russian emissions. An estimated two thirds of deposition to the Canadian Arctic comes from natural emissions and re-emitted Hg from land and oceans. The re-emitted mercury is now the dominant portion of terrestrial and marine emissions as a result of anthropogenic depositions since industrialization. Accurate representation of re-emission of mercury from various surfaces is essential for applying the models to estimate future changes in deposition.

Durnford et al. (2010) analyzed Hg transport pathways and estimated source attribution of air concentrations of GEM from Asia, North America, Russia and Europe at six Arctic measurement sites, as well as three subarctic and eight midlatitude sites in Canada. Despite less efficient transport mechanism than those of North America and Russia, Asia was found to be the dominant source of GEM at all verification sites; it contributed the most mercury (29–37% at all stations, seasons and vertical heights). Presence of roughly half of the global anthropogenic emissions of Hg in Asia results in its contribution being dominant in Canada including the Arctic. Asian outflow of Hg generated the most long range transport events, causing 43%, 67% and 75% of the high Hg events at the Arctic, sub-Arctic and midlatitude sites, respectively. Similar to the results shown in Fig. 6, results from this analysis indicated that Asia contributed by far the most Hg from all types of emissions at both Alert in 2000 (~30%) and at Little Fox Lake (Yukon) (~35%) in 2008. At both sites, the next largest source regions contributing to atmospheric Hg levels were North America (~10%), Russia (~6%), and Europe (~5%). Little Fox Lake has been assessed as a good location for monitoring Asian long-range transport (Durnford et al., 2010). Typically Hg plumes from East Asia that arrive western Canada reach the Yukon first then travel south on the lee side of the Rockies.

6. Temporal trends of ambient concentration and deposition of mercury

Long-term temporal changes in atmospheric Hg levels in the Arctic can be used to monitor the effect of changes in anthropogenic Hg emissions worldwide. However, it is challenging to relate these observed trends to the spatially and temporally heterogeneous changes in global anthropogenic Hg emissions. Moreover, the impact of concurrent changes in weather patterns, land and oceanic emissions, atmospheric chemical composition, land use, and other climate change related factors on trends in Hg levels in the environment make it difficult to detect the changes due to emissions alone. Modeling the impacts of changes in

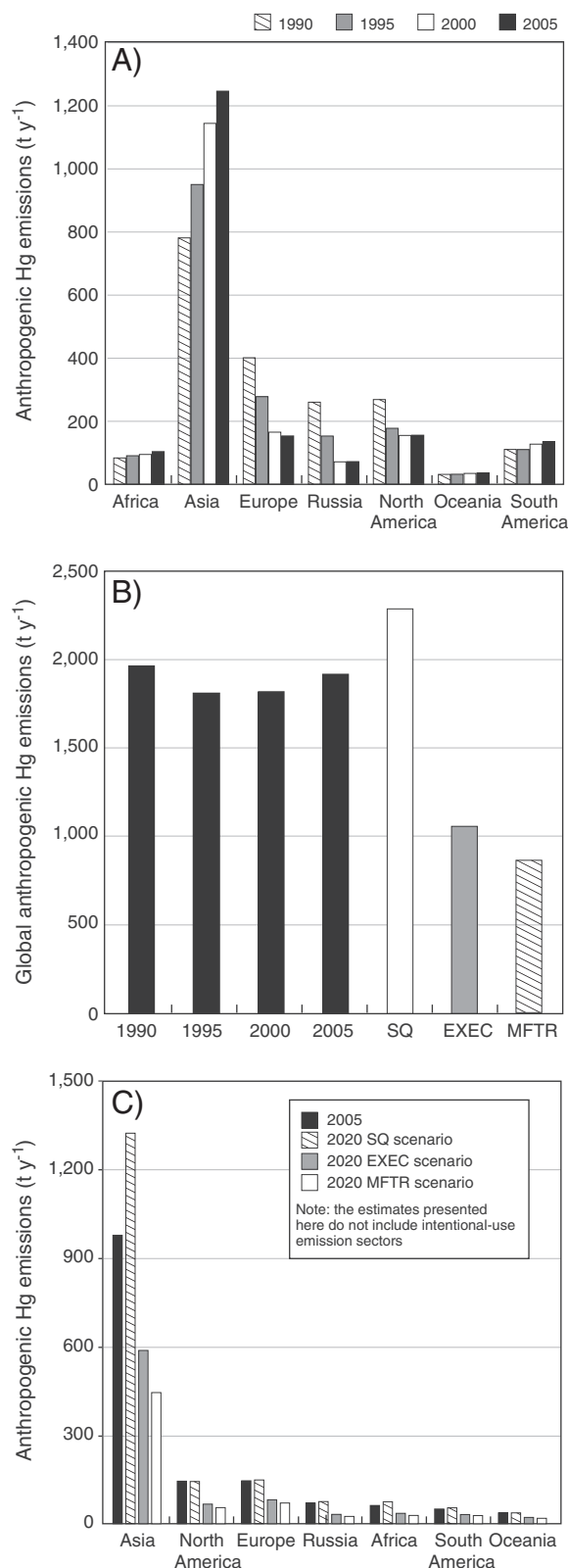


Fig. 6. A) Revised estimates of yearly anthropogenic Hg emissions to air in 1990, 1995, 2000, and 2005 from different regions (Mg year^{-1}) (source: AMAP, 2011). B) Total yearly global estimates of Hg emissions in 1990, 1995, 2000, and 2005 and three future (2020) scenarios (Mg year^{-1}). C) Regional yearly anthropogenic emissions of Hg under three future scenarios for the year 2020: Status Quo (SQ), Extended Emissions Control (EXEC), and Maximum Feasible Technological Reduction (MFTR) (Mg year^{-1}) (source: AMAP, 2011).

emissions and environmental factors on the Hg trends, both independently and in combination, can help explain the observed Hg trends in the Arctic and predict future Hg levels based on projected future emissions. Thus, GRAHM was applied to assess the impact of changing

anthropogenic emissions on Hg concentrations and deposition in the Arctic (in this study). Fig. 6 shows the changes in anthropogenic Hg emissions within major emission regions and globally for 1990, 1995, 2000, and 2005. Fig. 6 also shows the expected changes in global and

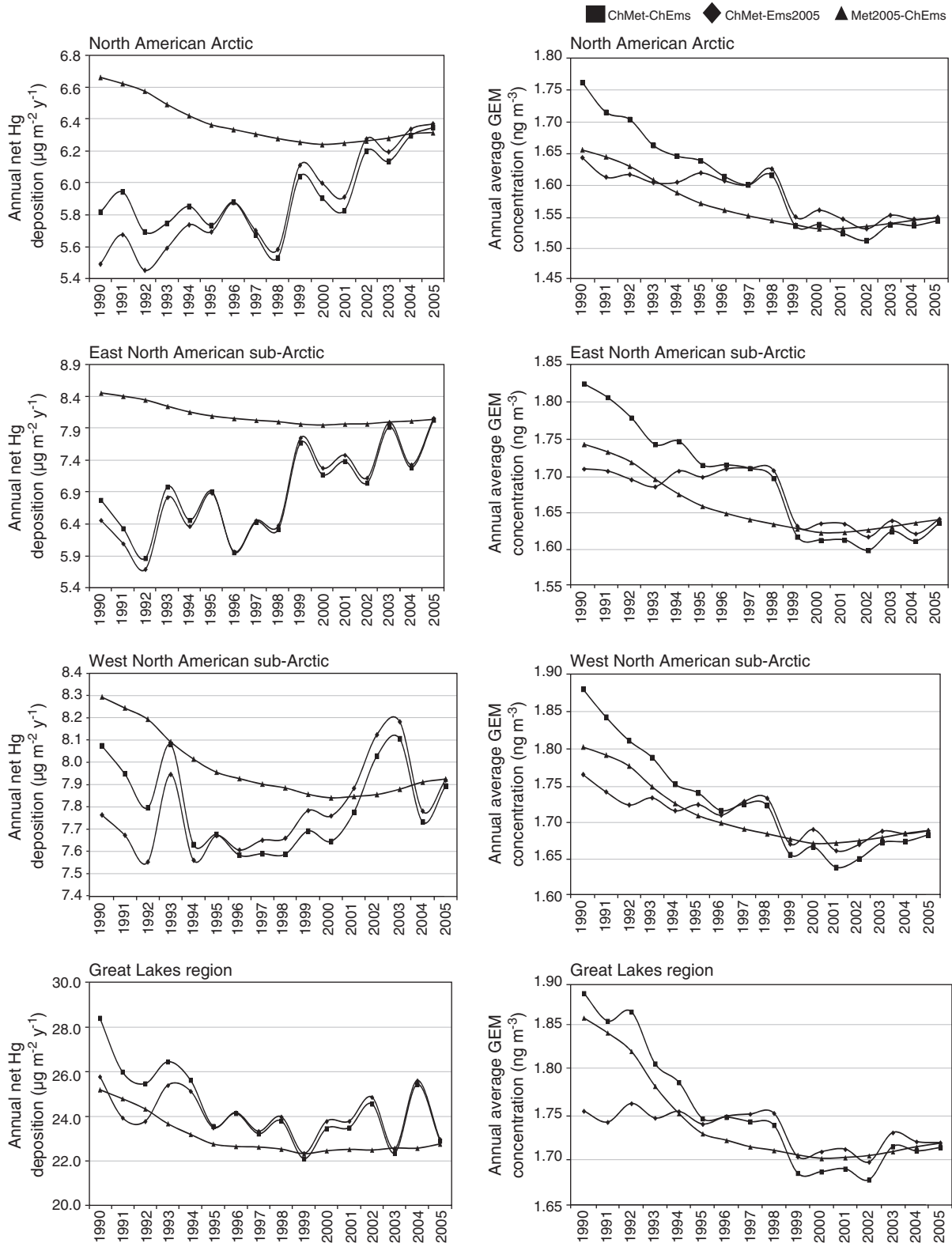


Fig. 7. Modeled yearly trends of net Hg deposition ($\mu\text{g m}^{-2} \text{year}^{-1}$; left panels) and yearly average concentration of surface air GEM (ng m^{-3} ; right panels) in the North American High Arctic ($66.6\text{--}90^\circ \text{N}$, $170\text{--}10^\circ \text{W}$), east and west sub-Arctic regions (east: $60\text{--}66.5^\circ \text{N}$, $100\text{--}10^\circ \text{W}$; west: $60\text{--}66.5^\circ \text{N}$, $170\text{--}100^\circ \text{W}$), and Great Lakes region ($40\text{--}50^\circ \text{N}$, $94\text{--}74^\circ \text{W}$). Three trend simulations were conducted: changing meteorology and changing emissions (ChMet–ChEms), changing meteorology and emissions constant at 2005 levels (ChMet–Ems2005), and changing emissions and meteorology constant at 2005 conditions (Met2005–ChEms).

regional anthropogenic emissions under three future scenarios for 2020 (described later in the section). A comparable set of historical global anthropogenic emissions were developed for the period 1990 to 2005 using a standardized methodology and consistent information for estimating certain emissions by AMAP (AMAP/UNEP, 2011). According to these reanalyzed emission inventories, the emissions in Europe and North America decreased most rapidly from 1990 to 2000, and decreased less from 2000 to 2005. The emissions from other continents, most notably from Asia, steadily increased from 1990 to 2005. Thus, global anthropogenic Hg emissions decreased from 1990 to 1995 by 7.8%, remained steady from 1995 to 2000 and increased by 5.6% from 2000 to 2005 according to AMAP inventories. It should be noted here that these inventories do not capture inter-annual changes in emissions which introduce a source of uncertainty to the modeling results, nor do they include natural emissions that may not be constant.

Atmospheric Hg concentrations and deposition were simulated from the year 1990 to 2005 based on changes in meteorology and anthropogenic emissions (Base Run). Two additional model simulations were performed to distinguish the impacts of changes in emissions and meteorology. In the first set of simulations, the emissions were kept constant at the year 2005 level and only meteorology was allowed to change from 1990 to 2005. In the second set, the meteorology was kept constant at year 2005 conditions and emissions were allowed to change from 1990 to 2005. Fig. 8 shows temporal trends of annual average net deposition and surface air GEM concentration from base and controlled simulations for the North American High Arctic (66.6–90° N, 170–10° W), and east and west sub-Arctic regions (east: 60–66.5° N, 100–10° W; west: 60–66.5° N, 170–100° W). Also shown for comparison are the temporal trends of annual average net deposition and surface air GEM concentrations for the Great Lakes region (40–50° N, 94–74° W).

Comparison of the simulated temporal trends from model simulations with and without changing meteorology shows that inter-annual variability in deposition and air concentration is driven by inter-annual variability in meteorology, linked with climate oscillations. These results suggest that temporal trends detected over short periods of time or from intermittent datasets may not be conclusive evidence of changes in emissions. Fig. 7 shows a decreasing trend in surface air concentrations from 1990 to 2005 for all three regions. In contrast, a constant or slow increasing deposition trend is found in High and sub-Arctic regions as opposed to the decreasing deposition trend found in the Great Lakes region. From 1990 to 2005, the model simulation shows a 15% increase in deposition in the High Arctic due to changes in meteorology, which is offset by decreases in North American and European emissions by 5%, resulting in an overall increase in the deposition by 10%. During this same period in the eastern sub-Arctic, simulations of deposition suggest an increase of 25% from meteorological

changes and a decrease of 5% from emission changes, leading to an overall increase of 20% in deposition. Almost no change in deposition is found in the western sub-Arctic where meteorological changes have led to a 2% increase and emission changes have led to a 4% decrease resulting in an overall 2% decrease in deposition over the 1990 to 2005 period. The differences in temporal trends of GEM concentrations between the High Arctic and sub-Arctic regions are less pronounced compared to the differences in trends of deposition fluxes between these regions; deposition processes are influenced more strongly by local meteorology compared to the concentrations of GEM. Decreases of 6% and 4% in surface air concentrations of GEM are simulated by the model due to changes in meteorology for High Arctic and sub-Arctic regions, respectively, from 1990 to 2005. Additional decrease of 6% in surface air concentrations of GEM is simulated by the model due to changes in emissions for all three regions. Total decrease of 12% (in 15 years) in surface air concentrations of GEM in the High Arctic is consistent with the measured decrease in air concentrations of GEM at Alert (see Steffen et al. in this issue). Modeled and observed temporal trends of average surface air GEM concentrations from October to December for Alert are shown in Fig. 8. A small decrease in observed surface air concentrations of GEM from 1995 to 2000 and the subsequent increase from 2000 to 2005 are found to be consistent with changes in global anthropogenic emissions during this period. These results indicate that Alert is a good location to detect changes in global anthropogenic Hg emissions. In contrast to High and sub-Arctic regions in North America, decreases in Hg deposition (–20% in ~15 years) as well as surface air GEM concentrations (–10% in ~15 years) are simulated by the model for the Great Lakes region. These decreases are related mainly to reductions in North American anthropogenic Hg emissions. Direct emissions of oxidized Hg from local sources contribute to the deposition fluxes in the Great Lakes region. Thus, a larger decrease in deposition flux was observed in the simulation compared to the decrease in GEM surface air concentration. The long-term wet deposition data from North American mid-latitudes that are available from the NADP (National Atmospheric Deposition Program) suggest a small decrease between 1997 and 2005 which is consistent with modeled wet deposition trends in the Great Lakes region. These results illustrate that Hg trends observed at temperate locations cannot be extrapolated to the Arctic because the changes in the Arctic are dominated by the changes in meteorology and global anthropogenic emissions whereas Hg trends in the temperate regions are more influenced by the regional changes in emissions.

An analysis of modeled meteorological trends from 1990 to 2005 shows a slowly increasing trend in precipitation amounts, decreasing snow cover extent and increasing regions of first year sea-ice in North American Arctic. These changes are consistent with the observational evidence in the Arctic and are attributed to

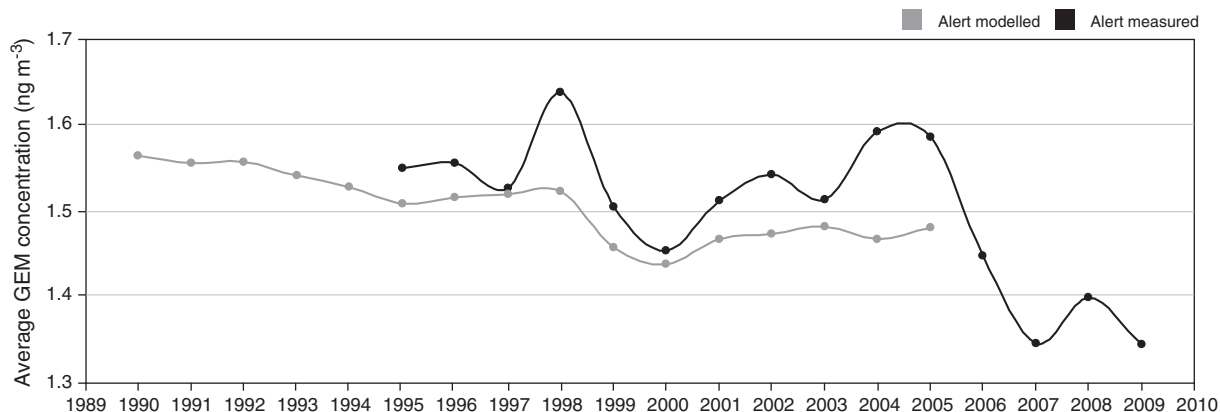


Fig. 8. Comparison of measured (dark purple) and modeled (green) average GEM concentrations (ng m^{-3}) in surface air from October to December at Alert (Nunavut).

climate change (Bekryaev et al., 2010; Cavalieri and Parkinson, 2012; Intergovernmental Panel on Climate Change (IPCC), Climate Change, 2013). Increasing precipitation rates result in a small increase in wet depositional fluxes of Hg in the model. In GRAHM, snowpacks over first year sea ice are considered enriched in halogens; high levels of halogens in these snowpacks suppress reduction and re-emission of deposited mercury (Durnford and Dastoor, 2011). Increasing regions of snowpacks over first year sea-ice and decreasing snow cover extent both lead to decreasing trend in mercury re-emission rates from snowpacks in the model.

In Arctic and sub-Arctic regions, there are no long-term records of direct measurements of wet or dry deposition. The relationship between the trends in emissions (or atmospheric deposition of Hg) and lake sediment Hg fluxes is not fully understood (Goodsite et al., 2013). For example, there has been a measurable decrease in atmospheric concentrations of Hg, by contrast, an increasing trend in Hg flux to Arctic lake sediments is observed in recent decades (Munthe et al., 2011), although there is some suggestion that Hg fluxes in subarctic lake sediments are also starting to decrease (Muir et al., 2009). Climate change has also been suggested as a possible explanation for increasing Hg fluxes to lake sediments (Kirk et al., 2011; Chételat et al., in this issue; Outridge et al., 2007; Munthe et al., 2011). In addition to the impact of changes in emissions, the decrease in atmospheric GEM concentrations can also occur by an increase in deposition rate of Hg to the surfaces through increased oxidation of GEM or by a reduction in re-emission of GEM from surfaces. The modeled trends (Fig. 7) show that changes in meteorology in recent years may have led to an increase in net Hg deposition in the North American Arctic and an associated decrease in air concentrations of GEM. This may partly explain the increasing Hg fluxes in Arctic lake sediments. It should be pointed out that the version of the GRAHM model employed here did not include the impact of changes in anthropogenic emissions on terrestrial re-emission rates other than for re-emissions from snow. Also, potential changes in rate of oceanic evasion of mercury as a result of changes in meteorology are not considered here.

Implications of climate change related factors to the mercury levels in the Arctic ecosystems are complex and multidirectional (Stern et al., 2012). Reductions in snow cover extent and depth in the Arctic may reduce extent of re-emission of mercury from cryosphere. Increased activity of freezing and re-freezing of leads and first year sea-ice regions may promote more active bromine chemistry leading to more frequent AMDEs. Also, increased regions of first year sea ice may lead to retention of more of the deposited mercury in overlying snowpacks because of the availability of oxidizing halogens in these snowpacks (as found in this study). On the other hand, high temperatures and shallow boundary layer may shift the atmospheric kinetics of AMDEs to reduced oxidation, higher surface temperature may enhance the photo-reduction and re-emission of Hg from the Arctic Ocean. Changes in primary productivity may result in biochemical changes in mercury that are not fully understood.

The modeled temporal trends of Hg in the Arctic reported in this study are in contrast with an analysis of the impact of changes in meteorological parameters on inter-annual variability (1979–2008) in atmospheric Hg and Hg deposition conducted using GEOS-Chem (Fisher et al., 2013). GEOS-Chem results suggests a decrease in Hg deposition in the Arctic with an increase in high solar radiation, due to increased photoreduction and re-emission of Hg from snowpacks, coupled with decreased deposition during AMDEs as a result of warmer temperatures. Currently, the representation of the interactions of climate change with Hg cycling in both models is inadequate to fully describe the complexity of these processes. Advanced representation of biogeochemical processes of mercury in air and surfaces as well as better representation of sea-ice dynamics are required to reduce the uncertainties in model estimates.

7. Future projections of Hg deposition under various Hg emission scenarios

Expected levels of Hg in deposition and surface air concentrations in 2020 were modeled (in this study) under three future global anthropogenic emissions scenarios briefly described as follows (for details see AMAP/UNEP, 2011). The *Status Quo* (SQ) scenario assumes that current patterns, practices, and uses that result in Hg emissions to air continue in the future. The *Extended Emissions Control* (EXEC) scenario assumes economic progress at a rate reflecting the future development of industrial technologies and emissions control technologies. Mercury-reducing technologies currently employed throughout Europe and North America would be implemented globally. The *Maximum Feasible Technological Reduction* (MFTR) scenario assumes all available solutions and measures are implemented globally, which will lead to the maximum degree of reduction of Hg emissions and Hg discharges to any environment.

The SQ emission scenario predicts a moderate increase of emissions in all regions except for North America (Fig. 6). The EXEC and MFTR scenarios both predict an emission decrease roughly by a factor of two and the difference between these scenarios is not significant. The largest reduction in Hg emissions is expected in Asia. Under the SQ scenario, the overall global anthropogenic emissions are estimated to increase by 19% between 2005 and 2020. Under the EXEC and MFTR scenarios, the emissions are expected to decrease in 2020 by up to 45% and up to 55%, respectively.

The model estimates were performed keeping the global meteorological conditions unchanged in all simulations for 2020 scenarios; thus, no impact of climate change is taken into account. In the Canadian High and sub-Arctic, average increases of 3% are expected for annual GEM concentrations in 2020 if emissions are to increase under the SQ scenario. In the Canadian High Arctic, average decreases of 9.1% and 10.9% can be expected if the emissions are to be reduced according to EXEC and MFTR scenarios, respectively. In the Canadian sub-Arctic, average decreases of 9.6% and 11.5% are expected for EXEC and MFTR scenarios, respectively. Differences in simulated changes in Hg concentrations between Arctic sectors are small. The largest estimated increase in Hg concentration is in the western sub-Arctic under the SQ scenario and the decrease is largest in the eastern European sub-Arctic and west Asian sub-Arctic (not shown here). In the Canadian High and sub-Arctic, average increases in deposition of 5.2% and 4.8%, respectively, are expected by 2020 under the SQ scenario. Average decreases of 14.9% and 18% are simulated under the EXEC and MFTR scenarios, respectively, in the Canadian High Arctic. In the Canadian sub-Arctic, average decreases of 16.5% and 19.9% are expected for EXEC and MFTR scenarios, respectively. These results suggest that the changes in anthropogenic oxidized Hg in these scenarios have an influence on deposition in the Arctic and sub-Arctic through direct transport of these species. Stronger variations in deposition are the result of regional differences in meteorology that influence dry and wet deposition. The impact of changes in remote emissions such as Asia is larger in the free troposphere compared to surface air. The increase in deposition estimates is found to be somewhat larger in the Arctic compared to the sub-Arctic, which is consistent with differences in source attribution for these two regions.

8. Uncertainties in model estimates of deposition in the Arctic

The two most fundamental parameters that determine the exchange of Hg between the earth's surface and atmosphere are air emissions of GEM and its overall oxidation rate. These parameters are important because of the dominance of GEM in emissions and its physical property of low solubility. Current estimates of anthropogenic emissions are more reliable than estimates of natural emissions and re-emissions of GEM which are estimated to account for approximately half to two-thirds of total global GEM emissions (Sunderland and Mason, 2007). Although GEM is the dominant species in anthropogenic Hg emissions,

current inventories also suggest significant emissions of oxidized Hg. The uncertainties in precise partitioning of anthropogenic Hg species at the stack or in-plume have an impact on the extent of the long-range transport of Hg simulated by the models. Current anthropogenic emission inventories lack information on temporal variations in emissions, leading to further modeling uncertainties.

The next major limitation of current atmospheric Hg models is uncertainty in Hg chemistry (Subir et al., 2011, 2012). The uncertainties in gaseous Hg chemistry are associated with the reported kinetic constants and the lack of identified products of Hg reactions. The existing Hg kinetic parameters for the gas and aqueous phases, obtained from both theoretical calculations and laboratory measurements, vary significantly. In addition, there is limited knowledge of the temperature dependence of Hg reactions for the wide range of temperatures in the atmosphere. Laboratory experimental data point to the importance of heterogeneous Hg chemistry on surfaces that could be relevant in the atmosphere. However, little is known about Hg reactions and equilibrium processes that take place on surfaces. As a result, it is difficult to quantify gas-particle partitioning of Hg in the atmosphere, which leads to uncertainty in model predictions of Hg deposition. Other knowledge gaps related to Hg processes are inadequate knowledge of scavenging characteristics of Hg in liquid and solid condensates, and dry deposition velocities. Uncertainties in model results are also caused by the uncertainties in meteorological parameters.

Given the current knowledge gaps, models rely on observational data to constrain the parameterization of non-anthropogenic emissions and Hg chemical mechanisms by evaluating the model outputs with measurements of Hg species in air and precipitation. Significant progress has been made in reducing the uncertainties in Hg models in the last decade as a result of the increase in measurement data. Finally, terrestrial and ocean models that include biogeochemical processes of Hg need to be linked to atmospheric Hg models to improve estimates of natural and revolatilized emissions of Hg to fully assess the impact of changing anthropogenic emissions and climate on Hg deposition in the Canadian Arctic.

9. Summary

Environment Canada's mercury model GRAHM model incorporates emissions and processes of Hg in the atmosphere and exchange of Hg between air, snow, vegetation and water. The model has been used to estimate regional to global scale Hg deposition, interpret Hg measurements, and understand past, present, and future trends and sources of Hg to the Arctic region. Detailed model simulations using GRAHM for the Alert and Little Fox Lake monitoring sites in Canadian High Arctic and sub-Arctic, respectively, indicated that Asia likely contributes the most Hg in air—from anthropogenic, natural, and re-emission sources—in the Canadian Arctic, followed by contributions from North America, Russia, and Europe. Most of the variability in model simulations for both locations was determined by long-range transport of Hg from Asia. However, transport of mercury from Russia is found to be most efficient compared with transport efficiencies from all other source regions.

Using GRAHM, it was estimated that there a net gain of 117 Mg of Hg annually at the surfaces in the Arctic (north of 66.5°), which indicates that the Arctic is a sink for atmospheric Hg. The net gain of mercury from the atmosphere to the Arctic Ocean was estimated at 75 Mg year⁻¹ (58 Mg year⁻¹ direct deposition, 50 Mg year⁻¹ deposition via snow melt and 33 Mg year⁻¹ evasion) and, in the terrestrial ecosystems North of 66.5°, the net gain was estimated at 42 Mg year⁻¹ (29 Mg year⁻¹ direct deposition, 16 Mg year⁻¹ deposition via snow melt and 3 Mg year⁻¹ emissions from soils). Using simulated mercury concentrations in meltwater, annual export of riverine mercury from North American, Russian and all Arctic watersheds to the Arctic Ocean were estimated in the range of 2.8–5.6, 12.7–25.4 and 15.5–31.0 Mg year⁻¹, respectively. The model illustrated that snowmelt revolatilization and the

oceanic evasion of mercury are the causes for high summertime concentrations of mercury in air at Alert (NU).

Both model simulations and estimates derived from lake sediments (Muir et al., 2009) show that the deposition of atmospheric Hg generally decreases with increasing latitude, and deposition of atmospheric Hg in Arctic and subarctic lakes is significantly lower than in mid-latitude North American lakes. Source attribution analyses for mercury deposition in the Arctic were performed with the model using Hg emissions data from 2005. In the Canadian Arctic, global anthropogenic emissions, terrestrial emissions, and oceanic emissions were predicted to have contributed to approximately 30%, 40% and 30% of Hg deposition, respectively. The anthropogenic contribution to Hg deposition was slightly higher in the High Arctic than in the sub-Arctic due to greater contributions from East Asia to the High Arctic. The largest anthropogenic source region with respect to Hg deposition to the Canadian Arctic is East Asia followed by Europe, North America, and South Asia. The absolute contribution from East Asia to Hg deposition is slightly higher in the western sub-Arctic region and deposition from North America is slightly higher in the High Arctic and eastern sub-Arctic.

According to modeling analyses in mid-latitude locations of North America, the decrease in regional Hg emissions was well reflected by a decrease in air concentrations and deposition. However, in the High Arctic, the GRAHM model results suggested that the Hg trends in air were equally related to changes in meteorology and global changes in anthropogenic emissions. In addition to inter-annual variability, the changes in meteorology reflected the impact of climate change between 1990 and 2005 in these simulations. Modeling results showed a slow increasing trend in Hg deposition in contrast to a decreasing trend in surface air GEM concentrations in the Arctic. The increase in net deposition in the Arctic was found to be primarily related to changes in snowpack and sea ice conditions and a small increase in precipitation rates. Results from the model and measured GEM concentrations illustrate that Hg trends observed at temperate locations cannot be extrapolated to the Arctic.

If emissions were to increase along a *status quo* scenario, model simulations projected an average increase in atmospheric Hg deposition of about 5% in 2020 for the Canadian Arctic. Maximum decreases in Hg deposition of 18–20% are expected under a scenario where the best control technologies are implemented.

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