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How relevant is the deposition of mercury onto snowpacks? – Part 1: A statistical study on the impact of environmental factors

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Abstract

A portion of the highly toxic methylmercury that bioaccumulates in aquatic life is created from mercury entering bodies of water with snowpack meltwater. To determine the importance of meltwater as a source of aquatic mercury, it is necessary to understand the environmental processes that govern the behavior of snowpack-related mercury. In this study we investigate relationships among 5 types of snowpack-related mercury observations and 20 model environmental variables. The observation types are the 24-h fractional loss of mercury from surface snow, and the concentrations of mercury in surface snow, seasonal snowpacks, the snowpack meltwater's ionic pulse, and long-term snowpack-related records. The model environmental variables include those related to atmospheric mercury, insolation, wind, atmospheric stability, snowpack physical characteristics, atmospheric pressure, and solid precipitation. Correlation coefficients and multiple linear regressions were calculated twice: once with all observations, and once with observations from locations presumably affected by oxidizing and stabilizing snowpack-related halogens excluded. We find that the presence of snowpack-related halogens has a significant impact on the behavior of snowpack-related mercury. Physically, snowpack-related mercury observations are most strongly controlled by the dry and wet depositions of oxidized mercury. The burial of mercury by fresh snowfalls and the wind driven ventilation of snowpacks are important processes. Indeed, in the absence of snowpack-related halogens, the 24-h fractional loss of mercury from surface snow is fully controlled by mercury deposition and surface-level atmospheric wind speed, stability, and surface pressure. The concentration of mercury in long-term records is affected by latitude, ventilation and surface pressure.

1 Introduction

In aquatic environments, given the presence of bacteria, mercury may be methylated. Methylation occurs in freshwater wetlands (Loseto et al., 2004; Goulet et al., 2007), peatlands (Mitchell et al., 2008a), lakes (Gilmour and Henry, 1991) and oceans

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(Mason and Fitzgerald, 1993; Monperrus et al., 2007; Sunderland et al., 2009). Since methylmercury bioaccumulates in fish and marine mammals and is a potent neurotoxin, it poses a serious health risk to humans. In the Arctic, where country foods include large marine mammals and fish, this issue is of great concern (Van Oostdam et al., 2005).

The source of the mercury found in the Arctic Ocean has been debated (Outridge et al., 2008). Although riverine outflow may be the dominant source of marine mercury locally (Leitch et al., 2007), it has been estimated that atmospheric deposition is the largest source of mercury in the Arctic Ocean as a whole (Outridge et al., 2008). This result is considered highly uncertain. An important source of the uncertainty is the lack of knowledge concerning the fate of mercury deposited onto snow-covered surfaces. The deposition of mercury onto snowpacks can be significant at high latitudes in spring as a result of Atmospheric Mercury Depletion Events (AMDEs; Schroeder et al., 1998; Lu et al., 2001; Berg et al., 2003; Ariya et al., 2004; Christensen et al., 2004; Heidam et al., 2004; Skov et al., 2004; Ferrari et al., 2005; Travnikov, 2005; Brooks et al., 2006; Kirk et al., 2006; Constant et al., 2007; Sommar et al., 2007; Johnson et al., 2008; Steffen et al., 2008). The mercury that is not revolatilized may enter the Arctic Ocean with the snowpack meltwater. To date, it is unknown what fraction of mercury is revolatilized from snowpacks.

The amount of mercury that is revolatilized from snowpacks can be closely related to the amount of mercury that is deposited, as is demonstrated by Fig. 1. This figure presents observed net GEM emission at Ny-Ålesund in 2008 and the simulated deposition of oxidized mercury through both wet and dry processes. The observations are presented in Steen et al. (2009). The mercury model involved is described in Sect. 2.2.1. Figure 1 provides the clearest evidence to date that the emission of mercury from snowpacks can be directly linked to previously-deposited mercury. As this figure demonstrates, the relationship between mercury deposition and emission is often very strong both in terms of magnitude and timing, with deposition preceding emission slightly. However, this figure also demonstrates that the mercury deposition/emission

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relationship is not simple. In the first half of the time series, total emission exceeds total deposition. The reverse appears to be true for the second half of the time series; the observation gap precludes a definitive statement. Furthermore, during other periods the deposition/emission relationship is less evident.

The close relationship of Fig. 1 between the simulated deposition of oxidized mercury and the observed net emission of GEM motivated us to attempt to decipher the environmental controls that govern the fate of mercury deposited onto snowpacks. If these controls can be determined, observations of snowpack-related mercury will provide an additional constraint on atmospheric mercury models. Additional constraints are required to improve the accuracy of these models.

The project of deciphering the environmental controls that determine the fate of mercury deposited onto snowpacks was divided into three parts. The first part reviewed the relevant literature, compiled datasets of observed concentrations of snowpack-related mercury, and theoretically determined the physical and chemical processes that govern the behavior of snowpack-related mercury. The results of this work are described in Durnford and Dastoor (2011).

The second part of the project consists of the present study. This study further investigates the theoretically-derived physico-chemical controls on snowpack-related mercury by statistically exploring the relationships between environmental variables and observations of mercury in snow-related media. The observations of mercury related to snowpacks were gathered from the literature and were presented in Durnford and Dastoor (2011) (Sect. 2.1); none of the snowpack-related mercury observations used in this statistical study are simulated. No new model development is described in the present study. However, we have used an established version of an atmospheric mercury model (Sect. 2.2.1) to provide the values of the environmental variables (Sect. 2.2.2). The environmental variables include meteorological fields, such as wind speed and precipitation, as well as mercury deposition fluxes (Sect. 2.2.2). This statistical study further supports the development of a parameterization for the fate of mercury deposited onto snowpacks.

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The third component of the project consists of the development of a snowpack/meltwater model for mercury for inclusion in large-scale atmospheric mercury models. The snowpack/meltwater model predicts the fate of mercury in snowpacks and snowpack meltwater based on mercury deposition and the local physical and chemical environments. It is based on the results of the first part of the project and the present statistical study. The snowpack/meltwater is described and its performance evaluated in Durnford et al. (2011).

Concerning processes involving mercury within the snowpack, gaseous elemental mercury (GEM) deposited onto snowpacks is likely emitted immediately given that it is highly labile (Steen et al., 2009). Deposited particulate mercury (PHg) is likely retained by the snowpack given that high concentrations of mercury and particles or their proxies are often collocated (Balogh et al., 2000; Schuster et al., 2002; St. Louis et al., 2005; Cobbett et al., 2007; Loewen et al., 2007; Poulain et al., 2007a, b; Witherow and Lyons, 2008; Jitaru et al., 2009). In contrast, deposited reactive gaseous mercury (RGM) may be reduced to GEM, primarily through photoreduction by UV-B radiation in the 305–320 nm wavelength range (Lalonde et al., 2003; Poulain et al., 2004; St. Louis et al., 2005; Dommergue et al., 2007; Faïn et al., 2007; Johnson et al., 2008; Sherman et al., 2010). GEM is likely the only mercuric species that is emitted. Prior to emission, a fraction may be reoxidized and, consequently, retained by the snowpack (Lalonde et al., 2003; Ferrari et al., 2004; Poulain et al., 2004, 2007b; Mann et al., 2005; Faïn et al., 2006, 2007, 2008; Lahoutifard et al., 2006; Lin et al., 2006; Dommergue et al., 2007).

GEM is apparently emitted from the top ~2 cm of the snowpack (Dommergue et al., 2007; Faïn et al., 2007; Brooks et al., 2008a; Johnson et al., 2008). Prior to emission, GEM molecules must be transported to the snowpack's surface. The transport is effected by molecular and turbulent diffusions (Albert and Shultz, 2002). Molecular diffusion is ubiquitous but slow (Albert and Shultz, 2002). Turbulent diffusion, or snowpack ventilation, is forced by atmospheric surface-level turbulence. This turbulence may be induced locally by wind interacting with a rough surface or by radiationally-forced thermal instability, or it may be generated elsewhere and imported (Kuhn, 2001;

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Albert and Shultz, 2002; Anderson and Neff, 2008).

The rates of photoreduction and revolatilization of GEM to the atmosphere increase significantly at the onset of snowmelt (Dommergue et al., 2003; Faïn et al., 2007; Sommar et al., 2007; Brooks et al., 2008b; Douglas et al., 2008). This causes a surge in the concentration of atmospheric surface-level GEM. Simultaneously, a considerable fraction of the snowpack's oxidized mercury content exits the snowpack in the meltwater's ionic pulse (Bales et al., 1990; Bishop et al., 1995; Allan et al., 2001; Kuhn, 2001; Lindberg et al., 2002; Dommergue et al., 2003, 2010). The ionic pulse lasts a few days (Bales et al., 1990; Bishop et al., 1995; Dommergue et al., 2003). It contains ionic concentrations that are higher than in the snowpack and that are 5–10 times higher than average meltwater concentrations (Bales et al., 1989, 1990).

The physico-chemical processes described above determine the fate of mercury in snowpacks, firn and ice (Durnford and Dastoor, 2011). The processes determining the fate of mercury in snowpack meltwater are similar. Regional variations in the behavior of snowpack-related mercury are produced by differing local environmental conditions (Durnford and Dastoor, 2011). For instance, snowpack-related halogens oxidize mercury while halides stabilize snowpack-related oxidized mercury (Lalonde et al., 2003; Ferrari et al., 2004; Faïn et al., 2006, 2008). Both the oxidation and the stabilization processes promote the retention of snowpack-related mercury. Thus, a smaller fraction of deposited mercury will be revolatilized at locations with elevated concentrations of snowpack-related halogen species.

A smaller fraction of deposited mercury will also be revolatilized at locations where the snowpack ventilation is weaker. This includes locations that experience weaker winds and/or less radiationally-induced atmospheric thermal instability (Albert and Shultz, 2002; Steffen et al., 2002; Lahoutifard et al., 2005; Steen et al., 2009; Durnford and Dastoor, 2011). Furthermore, for a given amount of atmospheric surface-level turbulence, which drives snowpack ventilation, the ventilation decreases with increasing snowpack density (Kuhn, 2001; Albert and Shultz, 2002; Domine et al., 2008).

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The revolatilization of mercury from snowpacks is also seemingly reduced by the reception of fresh snow. It has been hypothesized that new snowfalls can render mercury unavailable for emission by burying the previous surface layer along with its mercury content (Witherow and Lyons, 2008; Dommergue et al., 2010). As mentioned above, several studies reported that emitted GEM is sourced from only the top ~2 cm of the snowpack. It is possible that either the photoreduction that converts RGM to GEM within the snowpack (see above) is too weak at the former surface layer's new depth, or that the transport to the snowpack's surface of the GEM produced is too inefficient. As mentioned above, GEM is the only mercuric species emitted.

A smaller fraction of mercury is revolatilized from snowpacks under canopies than in adjacent open areas (Poulain et al., 2007b; Nelson et al., 2008). Although multiple processes contribute to this differential behavior (Fatnassi et al., 2006; Poulain et al., 2007b; Yue et al., 2008), the primary mechanism responsible for the higher retention rate in snowpacks under canopies is likely the shadowing effect (Poulain et al., 2007b). This effect reduces the amount of solar insolation reaching the surface of the snowpack. Consequently, since RGM is converted to GEM primarily through photoreduction (see above), less GEM is produced within the snowpack. As mentioned previously, only GEM is emitted from snowpacks.

At all locations, whether at mid or high latitudes, the revolatilization of mercury from snowpacks to the atmosphere depends on the difference of the GEM concentrations in the two media (Loux, 2001; Hansen et al., 2006). However, the extent of the impact of the concentration of atmospheric surface-layer GEM on this process is unknown. Furthermore, given the occurrence of high latitude AMDEs with their extremely low atmospheric surface-layer GEM concentrations (Schroeder et al., 1998; Lu et al., 2001; Bottenheim et al., 2002; Lindberg et al., 2002; Ferrari et al., 2005; Brooks et al., 2006; Kirk et al., 2006; Constant et al., 2007; Sommar et al., 2007; Steffen et al., 2008), it may be difficult to discern any general relationship between this concentration and the revolatilization. In contrast, it seems highly likely, given Fig. 1 and the discussion above, that revolatilization of mercury from snowpacks increases with mercury deposition.

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Concerning prior work related to the fate of mercury deposited onto snowpacks, Dastoor et al. (2008) described a representation of AMDEs and their associated deposition and revolatilization in a global three-dimensional model. The representation of the processes involved was simplistic. Holmes et al. (2010), who also used a global model, constructed a snowpack reservoir to accumulate deposited mercury. The revolatilization of this mercury was based on a 180 day lifetime, which decreased to 21 days when atmospheric temperatures exceeded 270 K. In addition to these global models, several small-scale process models that represent the behavior of snowpack-related mercury have been described in the literature. Both Ferrari et al. (2004) and Faïn et al. (2008) modeled the diffusion of GEM in the interstitial air of snow. Faïn et al. (2009) used a diffusion model to deduce historic atmospheric surface-level GEM concentrations from concentrations of GEM in firn air. Poulain et al. (2007b) constructed a mass balance for mercury in snowpacks where wet deposition, dry deposition and throughfall constituted mercury sources, while revolatilization and snowmelt constituted sinks. Values of wet deposition, revolatilization and snowmelt were calculated from observations of wet deposition and the concentration of mercury in the springtime snowpack, along with calculated rates of reduction under different canopies. The sum of dry deposition and throughfall was derived from these values and the observed snowpack mercury concentration.

Thus, to date, no-one has simulated the behavior of mercury in snowpacks in anywhere near its full complexity. Nor has a detailed study on the interaction between mercury in snowpack-related media and the local environment ever been performed; the current statistical study is unprecedented. As mentioned above, the results of the current study have been used to support the development of a snowpack/meltwater model for mercury. This model, which is described by Durnford et al. (2011), represents the primary physical and chemical processes that determine the fate of mercury deposited onto snowpacks. In the current statistical study, we employ 20 environmental variables and 5 types of mercury observations: the 24-h fractional loss of mercury from surface snow, and concentrations of mercury in surface snow, seasonal snowpacks,

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the snowpack meltwater's ionic pulse, and long-term snowpack-related records.

In the remainder of this article, Sect. 2 describes the snowpack-related mercury observations and the simulated environmental variables used in the study. The calculations performed are also described. In Sect. 3, the results are presented and discussed. Finally, Sect. 4 provides a summary and our conclusions.

2 Methodology

2.1 Observations

Durnford and Dastoor (2011) presented observations of five types of snowpack-related mercury: the 24-h fractional loss of mercury from surface snow, and concentrations of mercury in surface snow, seasonal snowpacks, the snowpack meltwater's ionic pulse, and long-term snowpack-related records. Seasonal snowpacks were defined as having existed for no more than two years; it seemed more appropriate to group second year snowpacks with seasonal snowpacks than long-term records. Consequently, the long-term snowpack-related records consist of glaciers, firn and snowpacks existing for more than two years.

The datasets used in this study were compiled by Durnford and Dastoor (2011). They are based on observations from numerous field studies performed in all regions of the globe. The reports of the field studies were published from 1971 to 2010. Thus, the datasets of snowpack-related mercury observations used in this study are not simulated. For readers wanting more information on the observations that contributed to these datasets, Durnford and Dastoor (2011) provided tables listing the time period covered by each contributing study, the environmental conditions during sampling, the sample size, and the mean, maximum and minimum data values.

At each location, a mean mercury data value was calculated from the means provided by the individual field studies gathered in Durnford and Dastoor (2011), weighted by sample size. Unspecified sample sizes were arbitrarily set to five. Only a small

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minority of sample sizes were unspecified. At many locations, the study where the sample size was unknown was the only study performed at that location; the assigning of an arbitrary sample size at these locations had no effect. In almost all other instances where the sample size was unspecified, numerous studies contributed to the location's mean. At these locations, it is extremely unlikely that the data value accompanying the unspecified sample size had an overly large impact on the location's mean. However, since a given location's mean mercury value may represent a single observed data value or the average of mean values provided by several different field studies, with each individual mean based on multiple observations, care must be taken not to overinterpret the study's results; some locations' observed means may be more reliable than others given the disparity in the number of contributing observations. Furthermore, sampling and analysis techniques varied between the contributing field studies. The mean snowpack-related mercury variable values calculated by Durnford and Dastoor (2011) are presented in Tables 1 through 5. The geographic distribution of these mean values is presented in Durnford and Dastoor (2011). Three long-term snowpack-related observations are included in Table 5 but not in Durnford and Dastoor (2011). Details concerning these additional observations are presented in Table 6. In Table 6, an ice core from the ice near Mizuho Station, Antarctica was sampled by Murozumi et al. (1978), a firn core from the Dasuopu glacier in Tibet was sampled by Wang et al. (2008), and an ice core from a glacier located at 4062 m a.s.l. on a saddle between the two summits of Belukha in the Siberian Altai (Olivier et al., 2003) was studied by Eyrikh et al. (2003).

In all, there are 9 mean values of the 24-h fractional loss of mercury from surface snow, 20 mean concentrations of mercury in surface snow, 23 mean concentrations of mercury in seasonal snowpacks, 8 mean concentrations of mercury in the snowpack meltwater's ionic pulse, and 13 mean concentrations of mercury in long-term snowpack-related records. Since the observations are not all from a uniform set of locations, the direct comparison of results for the different types of snowpack-related mercury observations is not applicable.

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2.2 Model environmental variables

2.2.1 The model

This study uses environmental variables simulated by Environment Canada's Global/Regional Atmospheric Heavy Metals model (GRAHM) (Dastoor and Larocque, 2004; Dastoor et al., 2008). GRAHM is an Eulerian chemical transport model built on top of EC's Global Environmental Multiscale-Global Deterministic Prediction System (GEM-GDPS) weather forecasting general circulation model (Côté et al., 1998a, b). The GEM-GDPS provides a single versus probabilistic ensemble forecast on a global versus regional domain. Meteorological and mercury processes are fully integrated in GRAHM: at each timestep: (1) mercury emissions are added to the model mercury concentrations, (2) the meteorological processes and mercury atmospheric physico-chemical processes are simulated, (3) the mercury species are transported, and (4) mercury is deposited. The simulations of the mercuric chemical transformations and depositional processes use information calculated by the meteorological component of the model during the same timestep, including boundary layer stability and the behavior of cloud water/ice. GRAHM has been shown to perform well compared to observations in past studies (Ryaboshapko et al., 2007a, b; Dastoor et al., 2008; Durnford et al., 2010).

The GEM-GDPS uses a semi-Lagrangian advection scheme to promote stability, and an implicit time scheme to control high frequency oscillations. The radiative transfer scheme is based on Fouquart and Bonnel (1980) and Garand and Mailhot (1990). Stratiform precipitation is calculated by a Sundqvist-based scheme (Sundqvist, 1978). A Kain-Fritsch scheme calculates deep convective precipitation (Kain and Fritsch, 1990). The turbulent mixing of meteorological and mercury species in the boundary layer is based on turbulent kinetic energy. Both turbulent kinetic energy and cloud liquid water/ice content are prognostic model variables.

In GRAHM, ozone and halogens oxidise mercury in the gas phase, while photochemistry and agents such as sulfur dioxide reduce mercury in the aqueous phase.

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Global three-dimensional monthly climatological concentrations of oxidants and reductants are used. Dry deposition is based on the resistance approach. Atmospheric Mercury Depletion Events (AMDEs) are simulated in springtime at high latitudes (Dastoor et al., 2008). Simulating AMDEs involves three distinct processes: (1) mercury oxidation, which requires the simulation of spatially and temporally localized releases of oxidizing bromine species to the atmosphere during spring; (2) the transport of mercury-depleted air masses; and (3) the representation of complex, heterogeneous atmosphere/snowpack mercury fluxes. It is likely that the relative importance of these three processes varies by location.

We use the global anthropogenic mercury emission fields produced by the Arctic Monitoring and Assessment Program (AMAP) for 2005 (Pacyna et al., 2010). Non-anthropogenic oceanic and terrestrial emissions of gaseous elemental mercury are based on the global mercury budget of Mason (2009). Terrestrial non-anthropogenic emissions are divided into direct natural emissions, and emissions of previously-deposited mercury. The former are distributed according to the natural geological enrichment of mercury. The latter are allocated according to the distribution of total deposition of mercury for historic years. The ratios of nonanthropogenic to anthropogenic emissions agree with published estimates for North America (Gbor et al., 2007) and East Asia (Shetty et al., 2008). The seasonal and diurnal variations of terrestrial emissions are based on the leaf area index and incoming direct solar radiation following Shetty et al. (2008). Oceanic emissions are modulated by the sea surface temperature.

GRAHM was run over a global domain at a 1 degree horizontal resolution with 28 and 58 vertical levels before and after 31 October 2006, respectively; GRAHM's vertical resolution follows that of the host model, GEM-GDPS. The vertical resolution of GEM-GDPS was increased in 2006 as part of a technical update of the model. We performed a series of interconnecting two-day simulations, where each simulation was initialised using observed meteorological analyses from the Canadian Meteorological Centre. Mercury concentrations were passed from one simulation to the next.

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2.2.2 The environmental variables

The 20 model environmental variables used in this study are presented in Table 7. These variables were chosen following Durnford and Dastoor (2011), who determined the physical and chemical processes that govern the behavior of snowpack-related mercury. It is hoped that these 20 variables together provide all the controls for the physical processes. Since we have multiple model variables describing different aspects of a given physical environmental characteristic, this set of 20 variables has some degree of redundancy built in. We choose to retain all 20 variables, nonetheless, in order to determine which aspect of each physical environmental characteristic is most relevant to the behavior of snowpack-related mercury. For instance, is an average wind speed of 4 m s^{-1} more effective at driving snowpack ventilation than an 8 % frequency of wind speeds of at least 6 m s^{-1}?

The environmental variables listed in Table 7 were calculated from 6-hourly model data from 2005 through 2009. The 5-yr averaging period filters out the intra-annual variability of the environment. Accumulated variables (dry and wet depositions of oxidized mercury, and total solid precipitation) represent the sum of the monthly values using the months of interest, averaged over the 5-yr period. Average variables (all variables other than accumulated variables and snow depth) are the average value of the months of interest over the 5-yr period. For all variables excluding snow depth, the months used are November through May in the Northern Hemisphere, and the corresponding months, May through November, in the Southern Hemisphere. The maximum snowpack depth within a single season represents, in the Northern Hemisphere, the five-year average of the difference between the maximum pack depth from February through June and January's depth. In the Southern Hemisphere, the difference between the maximum depth from August through December and July's depth is used. This process isolates a single season's contribution to multi-year snowpacks. For all variables, values are calculated separately for each hemisphere and then combined into a single global field.

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2.3 Procedure

In this study, we are relating observations of snowpack-related mercury to model environmental variables. Observations are valid at a single point. Model fields, however, provide average values for an entire grid cell. We interpolated the model fields to the observation's location using the inverse Cressman method (Cressman, 1959). Nonetheless, comparing the observation to the interpolated model value remains imperfect; observations are expected to exhibit significantly more fine-scale variability than the interpolated model values.

Determining relationships between the observations of snowpack-related mercury and the model environmental variables is also complicated by the fact that the observations were published anywhere between 1971 and 2010 (Sect. 2.1, Tables 1–5), while the model variables are based on simulations from 2005 through 2009 (Sect. 2.2.2). However, the observation at a given location represents a mean value, which is often based on means from multiple studies (Sect. 2.1, Tables 1–5). Similarly, the model values represent a 5-yr average. Since we are using mean values in both cases, we expect strong, low-frequency relationships between snowpack-related mercury observations and model environmental variables to be revealed, and not the high-frequency relationships.

To detect relationships between observations of snowpack-related mercury and model environmental variables, we performed two set of calculations. Each set of calculations used all model environmental variables. The two sets of mercury observations used are: Set1, which contains all observed mean values available for each of the five types of snowpack-related mercury observations; and Set2, which contains a subset of the observed mean values available for each observation type. Tables 1 to 5 indicate which of the snowpack-related mercury observations included in Set1 were also included in Set2.

In the creation of Set2, we subjectively removed mean values from locations where it is reasonable to believe that the snowpack-related media contains important levels

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of oxidizing and stabilizing halogen species. In some instances, information on the presence of snowpack-related halogen species was provided in the literature. Given that the distribution of snowpack-related halogens is highly heterogeneous (Garbarino et al., 2002; St. Louis et al., 2005, 2007; Constant et al., 2007; Poulain et al., 2007b), and that the sites of field studies contributing to a location's observed mean can be widely distributed, and that different types of observations are not always provided from the same sites, a location may be included in Set2 for some but not all observation types. If no information on the presence of snowpack-related halogen species was available in the articles describing the contributing field studies, we removed locations in coastal areas where elevated concentrations of these species are likely.

It is important to realize that snowpacks in locations experiencing AMDEs are not necessarily characterized by the high halogen contents that promote the retention of mercury within the snowpack. It is true that AMDEs are generated by halogens in the atmosphere and that AMDEs can lead to significant deposition of atmospheric mercury (Lu et al., 2001; Berg et al., 2003; Ariya et al., 2004; Christensen et al., 2004; Heidam et al., 2004; Skov et al., 2004; Ferrari et al., 2005; Travnikov, 2005; Brooks et al., 2006; Kirk et al., 2006; Constant et al., 2007; Sommar et al., 2007; Johnson et al., 2008; Steffen et al., 2008). However, the oxidized mercury that is produced during AMDEs may be transported prior to deposition. There is no guarantee that the triggering atmospheric halogen species and the oxidized mercury produced be transported in an identical manner, given their varying atmospheric lifetimes. Similarly, a location may experience AMDEs but little or no associated deposition of oxidized mercury, given that the atmospheric lifetime of GEM is far greater than that of oxidized mercury (Constant et al., 2007; Ferrari et al., 2008). Thus, halogens and mercury associated with AMDEs are not necessarily deposited in the same locations. Since it is the halogen content of the snowpacks that is relevant to the retention of mercury within the snowpacks, this differential transport and deposition is important.

When creating Set2, we also removed the five Greenlandic mean mercury concentrations that were reported in the 1970's. At some of these locations, stabilizing

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chlorides were observed in the snowpack (Weiss et al., 1975; Herron et al., 1977). Furthermore, the validity of these studies, with their tendency to report excessively high mercury concentrations, has been questioned (Jackson, 1997); both sampling procedures and analyzing techniques have improved since the 1970's. Thus, although the creation of Set2 is a subjective process, it was conducted in as objective a manner as possible. It was based on the thorough literature review described in Durnford and Dastoor (2011).

For both Set1 and Set2 we calculated the correlation coefficient (ρ) between each type of mercury observation and each environmental variable. For each type of mercury observation, we performed a multiple linear regression that involved all environmental variables for which the absolute value of the correlation coefficient was at least 0.35 (i.e. $|\rho| \geq 0.35$). Values for the snowpack-related mercury variables were calculated using the developed regression relationships. Finally, the correlation coefficient between the regression-derived and observed snowpack-related mercury variables' values was calculated. Thus, the difference in the results of these two sets of calculations should indicate the importance of chemical processes in determining the behavior of snowpack-related mercury.

The calculation of the final correlation coefficient between the regression-derived and observed values of the snowpack-related mercury variable constitutes a test of the regression's performance. It evaluates the extent to which the ensemble of environmental variables that participated in the regression controls the processes that determine the observed values of the snowpack-related mercury variable. Lower coefficients suggest that the environmental control of at least one process that is important in determining the value of the mercury variable was not included in the regression. In contrast, a unity-valued correlation coefficient indicates that the multiple linear regression was successful: the ensemble of the environmental variables that participated in the regression represents the controls of all the processes that determine the value of the mercury variable; the ensemble of environmental variables fully, albeit indirectly, controls the mercury variable's value.

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3 Results and discussion

Figure 2 presents individual correlation coefficients between the snowpack-related mercury observations from Set1 and the model environmental variables. Set1 includes all observations (Sect. 2.3). Only coefficients for which the absolute value is at least 0.35 ($|\rho| \geq 0.35$) are presented; it is these environmental variables that participate in the multiple linear regressions.

In Fig. 2, the average of the absolute values of the individual correlation coefficients for each observation type is provided at the bottom of the panel. This average is no higher than 0.45 except in Fig. 2d. The weakness of the average correlation is surprising given that, theoretically, a single set of physical and chemical processes governs the behavior of snowpack-related mercury at all locations (Durnford and Dastoor, 2011); one would expect the general nature of these processes to yield stronger correlations. Furthermore, the collection of model environmental variables shown is unrealistic. For instance, the 24-h fractional loss of mercury from surface snow is correlated only with the dry deposition of oxidized mercury (DOxDp). Similarly, over half of the environmental variables presented for the concentration of mercury in long-term snowpack-related records are related to insolation (i.e. variables 4 to 6: Alb, SW, LAI).

The only type of mercury observation to show somewhat reasonable results in Fig. 2 is the concentration of mercury in the snowpack meltwater's ionic pulse (Fig. 2d). For this observation type, the average of the absolute value of the individual correlation coefficients is a much stronger 0.58. Furthermore, the correlated environmental variables represent a more realistic range of the relevant physical processes (Durnford and Dastoor, 2011). In view of the generally weak correlation coefficients calculated for Set1, the correlated variables of Fig. 2d will not be discussed further.

Figure 3 presents scatter plots of snowpack-related mercury variables as observed and as calculated using Set1's regression relationships. The correlation coefficient between the regression-derived and observed snowpack-related mercury variable values is provided in each panel's lower right corner. Given Fig. 2's generally low

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correlations, the unsatisfactory correlation coefficients between the observed and calculated snowpack-related mercury variables of Fig. 3 are not unexpected. As discussed in Sect. 2.3, a low coefficient suggests that the environmental control of at least one process that is important in determining the value of the mercury variable was not included in the regression.

Although the overall results of Set1's Fig. 3 are disappointing, there are interesting aspects to this figure. For the 24-h fractional loss of mercury from surface snow (Fig. 3a), most of the observations are aligned, apart from one outlying black point, which represents Barrow. At some locations in the vicinity of Barrow, the halogen content of the snowpacks is extremely high (Garbarino et al., 2002). Given that halogens oxidize and stabilize snowpack-related mercury, the irregular behavior observed at Barrow in Fig. 3a is not surprising. Following the procedures used to create Set2 (Sect. 2.3), Barrow, and only Barrow, is excluded from Set2's observations of the 24-h fractional loss of mercury from surface snow (Table 1); it is not Barrow's apparent status as an outlier that caused it to be the only data point excluded.

Similarly, a subjective examination of Fig. 3e, which pertains to the concentration of mercury in long-term snowpack-related records, suggests that either the smallest of the four observed concentrations represented by a red circle is an outlier, or the four greatest observed concentrations represented by green circles. The smallest observation represented by a red circle is from the Belukha glacier in Siberia (Eyrikh et al., 2003; Tables 5, 6) while the observations represented by the green circles are from Greenlandic locations that were sampled in the 1970's (Weiss et al., 1971, 1975; Carr and Wilkniss, 1973; Herron et al., 1977; Appelquist et al., 1978; Table 5). The second lowest observed concentration represented by a red circle in Fig. 3e is from the Col du Dôme glacier in the Alps (Jitaru et al., 2003; Table 5), which is at an elevation of 4250 m a.s.l. The concentration at this elevated site represents low free-tropospheric mercury concentrations (Durnford and Dastoor, 2011). Since the sampling site on the Belukha glacier was also at an elevation of over 4000 m a.s.l (Sect. 2.1), the observation from the Belukha glacier likely also reflects low free-tropospheric mercury

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concentrations. Thus, the concentration at the Belukha glacier is reasonable. In contrast, chlorides were observed in some of the Greenlandic snowpack samples (Weiss et al., 1975; Herron et al., 1977). Furthermore, the validity of these early Greenlandic observations has been questioned (Jackson, 1997). Therefore, the five observations represented by green circles are excluded from Set2, while the observation from the Belukha glacier is retained (Tables 3, 5).

In Fig. 3b, which pertains to the concentration of mercury in surface snow, each group of colored circles tends to form a somewhat horizontal line. This suggests a latitudinal characteristic. However, since the actual observations are not at all latitudinally organized, the regression has produced unrealistic results.

Given that the results pertaining to the concentration of mercury in the snowpack meltwater's ionic pulse were the most realistic of Fig. 2, it is not surprising that the results associated with this type of mercury observation are the best of Fig. 3. Indeed, the unity-valued correlation coefficient of Fig. 3d indicates that this type of mercury observation is fully controlled by the ensemble of correlated environmental variables.

The results from Set2's multiple linear regressions are presented in Fig. 4. The correlation coefficients between the observed and regression-derived snowpack-related mercury variables have increased significantly: the average coefficient of 0.59 in Fig. 3 has jumped to 0.95 in Fig. 4. Although some of this gain in performance may simply reflect the reduced number of points processed, this is not the sole explanation; there are no longer any outliers in Fig. 4a nor any horizontally-aligned latitudinal bands in Fig. 4b, while the distribution of the remaining points in Fig. 4e has been completely reconfigured. Furthermore, Set2's collections of correlated model environmental variables (Fig. 5) have changed completely and are now far more closely aligned with theoretical expectations (see below). Thus, the success of Set2's results is primarily caused by our exclusion of observations based on the halogen content of the snowpack-related media. Our results indicate conclusively that the oxidation and stabilization of snowpack-related mercury by halogen species have a significant impact on the behavior of snowpack-related mercury.

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The demonstrated significant impact of halogen species on the fate of snowpack-related mercury indicates a potentially important consequence of climate change. Oxidizing halogen species are released to the atmosphere in association with refreezing sea ice leads (Grenfell and Maykut, 1977; Simpson et al., 2007a, b; Zhao et al., 2008). Oxidation by the reactive bromine species released causes AMDEs (Schroeder et al., 1998; Lu et al., 2001; Bottenheim et al., 2002; Brooks et al., 2006; Sommar et al., 2007), which are accompanied by an important deposition of oxidized mercury (Schroeder et al., 1998; Lu et al., 2001; Berg et al., 2003; Ariya et al., 2004; Christensen et al., 2004; Heidam et al., 2004; Skov et al., 2004; Ferrari et al., 2005; Travnikov, 2005; Brooks et al., 2006; Kirk et al., 2006; Constant et al., 2007; Sommar et al., 2007; Johnson et al., 2008; Steffen et al., 2008). Since some compounds formed within the snowpack between mercury and halides are stable (Lalonde et al., 2003; Ferrari et al., 2004b; Faïn et al., 2006, 2008), the revolatilization of the deposited mercury is reduced from snowpacks containing high concentrations of halides. Thus, if, in a warmer climate, sea ice becomes more dynamic such that the formation and refreezing of sea ice leads is more frequent, concentrations of mercury in snowpacks over sea ice may increase significantly.

A second important result from Fig. 4 relates to the interpretation of the high correlation coefficients that were obtained for Set2 between the regression-derived and observed mercury values. A unity-valued correlation coefficient suggests that the ensemble of environmental variables that participated in the regression related to a given mercury observation type provides the controls for all the processes that govern the value of that observation type (Sect. 2.3); the ensemble of participating environmental variables fully, but indirectly, controls the mercury variable's value. Thus, Fig. 4 indicates that the 24-h fractional loss of mercury in surface snow, and the concentrations of mercury in surface snow and the snowpack meltwater's ionic pulse are entirely or virtually entirely controlled by the ensemble of environmental variables that participated in their regressions. Similarly, the concentration of mercury in seasonal snowpacks is also very well controlled by its ensemble of correlated model environmental variables.

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However, the concentration of mercury in long-term snowpack-related records is less well controlled by its ensemble of environmental variables. It is possible that at least one process that is relevant to the concentration of mercury in long-term snowpack-related records is not represented by the 20 model environmental variables provided. Alternatively, it is possible that observations from more locations are required to improve the results of this mercury observation type's multiple linear regression.

Figure 5 demonstrates that, for each type of snowpack-related mercury observation, the correlated model environmental variables are more strongly correlated in Set2 than in Set1; the average of the absolute value of the individual correlation coefficients is greater for each type of mercury observation in Set2 than Set1 (Figs. 5 versus 2). The greatest increase, of 0.40 to 0.71, occurs for the concentration of mercury in surface snow. The next greatest increase is from 0.45 to 0.69 for the 24-h fractional loss of mercury in surface snow.

Figure 5 also demonstrates that the number of model environmental variables that are sufficiently strongly correlated to be presented (i.e. $|\rho| \geq 0.35$) increased for Set2 over Set1 for all short-term snowpack-related mercury observation types considered. The concentration of mercury in the snowpack meltwater's ionic pulse gained 7 variables, yielding a total of 14 correlated model environmental variables for Set2, while the concentration of mercury in seasonal snowpacks gained 6 variables, yielding a total of 10 correlated model environmental variables for Set2. Although fewer model environmental variables are presented in Fig. 5e than Fig. 2e for the concentration of mercury in long-term snowpack-related records, the collection of variables shown for Set2 is more realistic.

Considering the collection of model environmental variables that are correlated with the 24-h fractional loss of mercury from surface snow, Fig. 5a reveals that the fractional loss increases with the deposition of oxidized mercury through both dry (DOxDp) and wet (WOxDp) processes. This is as one might expect. As the frequency of wind speeds of at least $6\,m\,s^{-1}$ (WdSpF6) increases, so does the loss. This agrees with Albert and Shultz (2002), Steffen et al. (2002), Lahoutifard et al. (2005) and Steen et al. (2009)

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who found that wind speed significantly affects mercury revolatilization. Moreover, the loss increases with decreasing surface-layer atmospheric stability (SfcSFn). Since a lower stability promotes the formation of snowpack-ventilating turbulence, these two relationships indicate the importance of snowpack ventilation in driving emission. It is probable that the positive correlation between surface pressure (SfcP) and the 24-h fractional loss of mercury from surface snow reflects the fact that higher atmospheric pressures are accompanied by sunnier conditions. Sunnier conditions presumably promote the photoreduction of the snowpack's oxidized mercury content. It is also possible that this positive correlation reflects the likelihood that the sunnier conditions associated with increasing surface pressures enhance atmospheric radiationally-driven thermal instability. Atmospheric instability likely promotes snowpack ventilation (Durnford and Dastoor, 2011).

The relationship between the concentration of mercury in surface snow and the model environmental variables of Fig. 5b is more difficult to interpret; the only transparent relationship is the increase in this type of mercury concentration with increasing dry (DOxDp) and wet (WOxDp) depositions of oxidized mercury. The unexpected positive correlation of Fig. 5b between the frequency of wind speeds of at least $6\,m\,s^{-1}$ (WdSpF6) and the mercury concentration may indicate the importance of blowing snow. It is possible that it is the deposition of the blowing snow that is important: the addition of a new surface layer of snow would decrease the photoreduction of oxidized mercury at a given snowpack depth, and, consequently, promote mercury retention. This correlation may also reflect the fact that strong winds increase the density of the surface layer of snow, thereby reducing snowpack ventilation and promoting mercury retention (Kuhn, 2001; Albert and Shultz, 2002; Domine et al., 2008). The fact that the correlations of the two pressure variables (SfcP, SLP) are of opposite signs indicates the importance of altitude. The positive correlation with surface pressure (SfcP) suggests that the mercury concentration decreases with altitude. This may reflect the fact that anthropogenic mercury emissions are more concentrated at lower elevations. The negative correlation with sea level pressure (SLP) may indicate that the sunny

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conditions typical of high pressure systems promote both photoreduction within the snowpack, which would increase the pool of GEM available for revolatilization (Lalonde et al., 2003; Poulain et al., 2004; St. Louis et al., 2005; Dommergue et al., 2007; Faïn et al., 2007; Johnson et al., 2008; Sherman et al., 2010), and radiationally-driven atmospheric turbulence, which drives snowpack ventilation (Durnford and Dastoor, 2011). Both processes would act to reduce the concentration of mercury in surface snow. The correlation of this mercury concentration with the concentration of surface-level atmospheric GEM (GEM) has probably been strongly affected by high-latitude springtime AMDEs. Since AMDEs depress the high-latitude atmospheric GEM concentration throughout the season, creating a strong latitudinal gradient in this concentration, this relationship is likely meaningless.

The dry (DOxDp) and wet (WOxDp) depositions of oxidized mercury are also, as one might expect, positively correlated with the concentration of mercury in seasonal snowpacks (Fig. 5c). Interestingly, the correlation with wet deposition is significantly stronger than that of dry deposition. This indicates the importance of burial in increasing the mercury concentration (Dommergue et al., 2010; Witherow and Lyons, 2008). The negative correlation with average wind speed (WdSpAv) is also expected, given that wind-induced snowpack ventilation promotes mercury emission (Kuhn, 2001; Albert and Shultz, 2002; Steffen et al., 2002; Lahoutifard et al., 2005; Anderson and Neff, 2008; Steen et al., 2009). The positive correlations with all three solid precipitation variables (PrTot, PrF24h, PrF6h) likely indicate the importance of both wet deposition and the burial of previously-deposited mercury by fresh snowfalls. The positive correlation with surface-level relative humidity (RH) may also indicate the importance of the burial of mercury by fresh snowfalls. However, the strong latitudinal variation of atmospheric surface-level temperature (SfcT), along with the positive correlation between this temperature and the concentration of mercury in seasonal snowpacks, indicates that this mercury concentration tends to increase with latitude; either not all observations affected by snowpack-related oxidizing and stabilizing halogens were excluded from Set2, or AMDE-related mercury deposition has a lasting impact on snowpack

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mercury concentrations. In contrast, it is likely that the correlation with sea level pressure (SLP) is meaningless; the correlation disappears if the three outlying antarctic observations are excluded (not shown). Also meaningless is the correlation between the concentrations of surface-level atmospheric GEM (GEM) and mercury in seasonal snowpacks; high-latitude AMDEs have an overly strong impact (not shown).

Of the 14 model environmental variables correlated with the snowpack meltwater's ionic pulse, 8 of the variables were assigned zero-valued coefficients by the multiple linear regression. This indicates that these variables were determined to be linearly dependent on another variable included in the regression. Since these 8 variables (the concentration of surface-level atmospheric GEM (GEM), albedo (Alb), both wind speed variables (WdSpAv, WdSpF6), turbulent kinetic energy (TKE) and all three solid precipitation variables (PrTot, PrF24h, PrF6h)) were excluded from the regression, we will not discuss them further.

The dry (DOxDp) and wet (WOxDp) depositions of oxidized mercury are, as one might expect, positively correlated with the concentration of mercury in the snowpack meltwater's ionic pulse (Fig. 5d). The negative correlation between the leaf area index (LAI) and the meltwater mercury concentration likely indicates the presences of a latitudinal gradient in this concentration; since most observations of snowpack-related mercury are made in the open, the impact of a canopy's shadowing effect on snowpack-related mercury concentrations is not truly being examined. The negative correlation between surface-level atmospheric temperature (SfcT) and the meltwater mercury concentration also suggests that this mercury concentration increases with latitude. The tendency of the concentration of mercury in the snowpack meltwater's ionic pulse to increase latitudinally suggests that either some of the remaining arctic observations of meltwater mercury were affected by oxidation by snowpack-related halogen species and stabilization by snowpack-related halides and should have been excluded from Set2, or that the important springtime high-latitude AMDE-associated mercury deposition has an impact on the concentration of mercury in the snowpack meltwater's ionic pulse.

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The negative correlation between the concentration of mercury in the snowpack meltwater's ionic pulse and snow depth (SnoDp) suggests that the snowpack's mercury content was diluted by the greater water content. Likewise, the positive correlation with sea level pressure (SLP) suggests that the mercury concentration is depressed through dilution; decreasing pressures are accompanied by a greater likelihood of precipitation. However, as discussed above, the concentration of mercury in seasonal snowpacks increases with increasing precipitation, likely as a result of the importance of both wet deposition and the burial of previously-deposited mercury by fresh snowfalls. This apparent contradiction may indicate that the generation of the ionic pulse (Bales et al., 1989, 1990; Bishop et al., 1995; Allan et al., 2001; Kuhn, 2001; Lindberg et al., 2002; Dommergue et al., 2003, 2010) is variable; the snowpack meltwater's ionic pulse may be weaker, i.e. more dilute, in a deeper snowpack. No evidence is available to prove or disprove this hypothesis. However, it is known that multiple melt-freeze cycles enhance concentrations in the ionic pulse (Kuhn, 2001); deeper snowpacks may tend to undergo fewer melt-freeze cycles.

The concentration of mercury in long-term snowpack-related records is correlated with the fewest model environmental variables of the five types of mercury observations. The positive correlation with short-wave insolation absorbed at the ground (SW) is not related to photoreduction; such a relationship would produce anticorrelation. Instead, the positive correlation suggests a latitudinal gradient in these mercury concentrations. It may be that none of the high-latitude glaciers are in locations affected by AMDEs and their important mercury deposition, while midlatitude glaciers are more affected by the predominantly midlatitude sources of anthropogenic mercury. Even natural sources of mercury, given the presence of the strong midlatitude upper-level zonal winds, may be more strongly represented in mid-latitude long-term snowpack-related records. Similarly, the anticorrelation of the concentration of mercury in long-term snowpack-related records with average surface-level atmospheric wind speed (WdSpAv) may reflect the fact that surface-level wind speeds are stronger at higher latitudes where anthropogenic sources of mercury may have less impact,

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and/or may indicate that ventilation can penetrate into snowpacks that are more than two season's old, and/or may indicate that the ventilation of snowpacks less than two years old impacts the concentration of mercury in long-term snowpack-related records. The anticorrelation between surface pressure (SfcP) and this mercury concentration may support the importance of ventilation for the concentration of mercury in long-term snowpack-related records since the sunnier conditions that accompany increasing surface pressures promote radiationally-driven surface-level atmospheric turbulence; turbulence drives snowpack ventilation, which increases revolatilization. However, this anticorrelation likely also reflects the fact that the sunnier, drier conditions that accompany higher pressures promote photoreduction and, consequently, revolatilization, as well as reduce the likelihood of mercury being buried.

4 Summary and conclusions

In this statistical study we used the mean values of 5 types of snowpack-related mercury observations and the five-year average of 20 model environmental variables. The snowpack-related mercury observations were gathered from published reports of field studies. The model environmental variables represent the controls on the physical and chemical processes that govern the behavior of snowpack-related mercury.

We performed two sets of calculations for this study. We first calculated individual correlation coefficients between each type of snowpack-related mercury observation and each of the model environmental variables. All sufficiently strongly correlated environmental variables, where the absolute value of the correlation coefficient was at least 0.35 (i.e. $|\rho| \geq 0.35$), subsequently participated in a multiple linear regression for the given observation type. The first set of calculations, Set1, involved all available observations. Locations that were assumed to be strongly affected by the oxidation and/or stabilization of snowpack-related mercury by halogen species were excluded from the second set of calculations, Set2. Also excluded were the mean mercury concentrations based on studies performed in Greenland in the 1970's; chloride concentrations

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are elevated at some of these sites. Furthermore, the validity of these concentrations has been questioned.

For each of the five types of snowpack-related mercury observations, both the average correlation and the number of correlated model environmental variables increased for Set2 over Set1. Furthermore, the ability to estimate snowpack-related mercury variable values using the multiple linear regression relationships was considerably improved for Set2 over Set1. Indeed, the 24-h fractional loss of mercury in surface snow, and the concentrations of mercury in surface snow and the snowpack meltwater's ionic pulse are fully controlled by Set2's correlated model environmental variables. These results indicate clearly that the oxidation and stabilization of snowpack-related mercury by halogen species has a significant impact on the behavior of snowpack-related mercury. Furthermore, the ability of Set2's model environmental variables to control the snowpack-related mercury observations indicates that these observations do, indeed, have the potential to constrain atmospheric mercury models; additional constraints will help to improve the accuracy of these models.

The group of physical environmental variables that governs the 24-h fractional loss of mercury from surface snow is the most transparent; the fractional loss increases with increasing depositions of oxidized mercury through dry and wet processes, an increasing frequency of wind speeds of at least $6\,m\,s^{-1}$, decreasing surface-level atmospheric stability, and increasing surface pressure. Increasing surface pressures are accompanied by sunnier conditions, which promote photoreduction. The concentration of mercury in seasonal snowpacks is clearly strongly affected by the burial of mercury by fresh snowfalls. These results are all expected. Somewhat less expected is the fact that the concentration of mercury in long-term snowpack-related records is determined to a certain extent by latitude. This latitudinal dependence may reflect the importance either of midlatitude anthropogenic sources of mercury or of midlatitude upper-level zonal winds.

Considering the overall importance of the individual model environmental variables for Set2, we find that wet and dry depositions of oxidized mercury have the strongest

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impact on snowpack-related mercury variables. The fact that wet deposition is more strongly correlated than dry deposition for concentrations of mercury in surface snow, seasonal snowpacks and the snowpack meltwater's ionic pulse, while the reverse is true for the 24-h fractional loss of mercury in surface snow, suggests that some oxidized mercury deposited through wet processes is buried by the precipitation, which renders it unavailable for emission. The positive correlations between the three solid precipitation variables and the concentration of mercury in seasonal snowpacks also indicate the importance of burial.

It is possible that the burial of mercury deposited through wet processes is caused by the ability of a new layer of snow to reduce the penetration of solar radiation within the snowpack. This would diminish photoreduction within the snowpack and, consequently, revolatilization. It is also possible that mercury deposited through wet processes is less easily photoreduced than mercury deposited through dry processes and/or that the GEM produced from the photoreduction of oxidized mercury is less easily revolatilized when wet rather than dry deposition processes are involved. If so, this suggests that the mercury contained in falling snowflakes tends to be centrally located. This, in turn, suggests that mercuric compounds tend to constitute the condensation/nucleation sites of snowflakes rather than adsorbing to falling snowflakes. In dry deposition, the mercury is sorbed onto aerosol surfaces.

For the remaining correlated model environmental variables for Set2, both wind speed variables are important for determining the snowpack-related mercury variable value. The two pressure variables, surface pressure and sea level pressure, are also important, although the interpretation of their relationship with snowpack-related mercury has proved to be somewhat problematic. It is possible that their greatest importance lies in the fact that higher pressures are accompanied by sunnier, photoreduction-promoting conditions. The environmental variables that are linked to snowpack characteristics are important only in relation to the concentrations of mercury in seasonal snowpacks and the snowpack meltwater's ionic pulse. Surprisingly, snowpack density is uncorrelated with all snowpack-related mercury observation types;

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model density values may be insufficiently accurate for any actual relationships to be revealed. Surface-level stability is important only for the 24-h fractional loss of mercury from surface snow. The concentration of surface-level atmospheric GEM and the three model environmental variables that are directly related to insolation are of less importance than expected.

In the future, the nature of the relationship between snowpack-related mercury and the environment could be further elucidated if the concentrations of halogens and other oxidants and stabilizing agents were included in the calculations. The reliability of the calculations might also improve if the value of the environmental variables were measured versus simulated. Having observations of snowpack-related mercury from a wider variety of locations would further improve the accuracy of the calculations.

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References

Albert, M. R. and Shultz, E. F.: Snow and firn properties and air-snow transport processes at Summit, Greenland, Atmos. Environ., 36, 2789–2797, 2002.

Allan, C. J., Heyes, A., Roulet, N. T., St. Louis, V. L., and Rudd, J. W. M.: Spatial and temporal dynamics of mercury in Precambrian Shield upland runoff, Biogeochemistry, 52, 13–40, 2001.

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Anderson, P. S. and Neff, W. D.: Boundary layer physics over snow and ice, Atmos. Chem. Phys., 8, 3563–3582, doi:10.5194/acp-8-3563-2008, 2008.
Appelquist, H., Jensen, K. O., Sevel, T., and Hammer, C.: Mercury in the Greenland ice sheet, Nature, 273, 657–659, 1978.
Ariya, P. A., Dastoor, A. P., Amyot, M., Schroeder, W. H., Barrie, L., Anlauf, K., Raofie, F., Ryzhkov, A., Davignon, D., Lalonde, J., and Steffen, A.: The Arctic: a sink for mercury, Tellus B, 56, 397–403, 2004.
Aspmo, K., Temme, C., Berg, T., Ferrari, C., Gauchard, P.-A., Fain, X., and Wibetoe, G.: Mercury in the atmosphere, snow and melt water ponds in the North Atlantic Ocean during Arctic summer, Environ. Sci. Technol., 40, 4083—4089, 2006.
Bales, R. C., Davis, R. E., and Stanley, D. A.: Ion elution through shallow homogeneous snow, Water Resour. Res., 25, 1869–1877, 1989.
Bales, R. C., Sommerfeld, R. A., and Kebler, D. G.: Ionic tracer movement through a Wyoming snowpack, Atmos. Environ., 24, 2749–2758, 1990.
Balogh, S. J., Meyer, M. L., Hansen, N. C., Moncrief, J. F., and Gupta, S. C.: Transport of mercury from a cultivated field during snowmelt, J. Environ. Qual., 29, 871–874, 2000.
Berg, T., Bartnicki, J., Munthe, J., Lattila, H., Hrehoruk, J., and Mazur, A.: Atmospheric mercury species in the European Arctic: measurements and modeling, Atmos. Environ., 35, 2569–2582, 2001.
Berg, T., Sekkesæter, S., Steinnes, E., Valdal, A.-K., and Wibetoe, G.: Springtime depletion of mercury in the European Arctic as observed at Svalbard, Sci. Total Environ., 304, 43–51, 2003.
Bishop, K., Lee, Y.-H., Pettersson, C., and Allard, B.: Methylmercury output from the Svartberget catchment in Northern Sweden during spring flood, Water Air Soil Poll., 80, 445–454, 1995.
Bloom, N. S. and Watras, C. J.: Observations of methylmercury in precipitation, Sci. Total Environ., 87/88, 199–207, 1989.
Bottenheim, J. W., Fuentes, J. D., Tarasick, D. W., and Anlauf, K. G.: Ozone in the Arctic lower troposphere during winter and spring 2000 (ALERT2000), Atmos. Environ., 36, 2535–2544, 2002.
Brooks, S., Arimoto, R., Lindberg, S., and Southworth, G.: Antarctic polar plateau snow surface conversion of deposited oxidized mercury to gaseous elemental mercury with fractional long-term burial, Atmos. Environ., 42, 2877–2884, 2008a.

ACPD
12, 387–439, 2012

Brooks, S., Lindberg, S., Southworth, G., and Arimoto, R.: Springtime atmospheric mercury speciation in the McMurdo, Antarctica coastal region, Atmos. Environ., 42, 2885–2893, 2008b.

Brooks, S. B., Saiz-Lopez, A., Skov, H., Lindberg, S. E., Plane, J. M. C., and Goodsite, M. E.: The mass balance of mercury in the springtime arctic environment, Geophys. Res. Lett., 33, L13812, doi:10.1029/2005GL025525, 2006.

Boutron, C. F., Vandal, G. M., Fitzgerald, W. F., and Ferrari, C. P.: A forty year record of mercury in central Greenland snow, Geophys. Res. Lett., 25, 3315–3318, 1998.

Capelli, R., Minganti, V., Chiarini, C., and De Pellegrini, R.: Mercury in snow layers from the Antarctica, Int. J. Environ. An. Ch., 71, 289–296, 1998.

Carr, R. A. and Wilkniss, P. E.: Mercury in the Greenland ice sheet: Further data, Science, 181, 843–844, 1973.

Christensen, J. H., Brandt, J., Frohn, L. M., and Skov, H.: Modelling of Mercury in the Arctic with the Danish Eulerian Hemispheric Model, Atmos. Chem. Phys., 4, 2251–2257, doi:10.5194/acp-4-2251-2004, 2004.

Cobbett, F. D., Steffen, A., Lawson, G., and Van Heyst, B. J.: GEM fluxes and atmospheric mercury concentrations (GEM, RGM and Hg^p) in the Canadian Arctic at Alert, Nunavut, Canada (February-June 2005), Atmos. Environ., 41, 6527–6543, 2007.

Constant, P., Poissant, L., Villemur, R., Yumvihoze, E., and Lean, D.: Fate of inorganic mercury and methyl mercury within the snow cover in the low arctic tundra on the shore of Hudson Bay (Québec, Canada), J. Geophys. Res., 112, D08309, doi:10.1029/2006JD007961, 2007.

Côté, J., Desmarais, J.-G., Gravel, S., Méthot, A., Patoine, A., Roch, M., and Staniforth, A.: The operational CMC-MRB Global Environmental Multiscale (GEM) Model: Part II – Results, Mon. Weather Rev., 126, 1397–1418, 1998a.

Côté, J., Gravel, S., Méthot, A., Patoine, A., Roch, M., and Staniforth, A.: The operational CMC-MRB Global Environmental Multiscale (GEM) Model: Part I – Design considerations and formulation, Mon. Weather Rev., 126, 1373–1395, 1998b.

Cressman, G. P.: An operational objective analysis system, Mon. Weather Rev., 87, 367–374, 1959.

Dastoor, A. P. and Larocque, Y.: Global circulation of atmospheric mercury: A modeling study, Atmos. Environ., 38, 147–161, 2004.

Dastoor, A. P., Davignon, D., Theys, N., Van Roozendael, M., Steffen, A., and Ariya, P. A.: Modeling dynamic exchange of gaseous elemental mercury at polar sunrise, Environ. Sci.

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12, 387–439, 2012

Technol., 42, 5183–5188, doi:10.1021/es800291w, 2008.
Domine, F., Albert, M., Huthwelker, T., Jacobi, H.-W., Kokhanovsky, A. A., Lehning, M., Picard, G., and Simpson, W. R.: Snow physics as relevant to snow photochemistry, Atmos. Chem. Phys., 8, 171–208, doi:10.5194/acp-8-171-2008, 2008.
Dommergue, A., Ferrari, C. P., Gauchard, P.-A., and Boutron, C. F.: The fate of mercury species in a sub-arctic snowpack during snowmelt, Geophys. Res. Lett., 30, 1621, doi:10.1029/2003GL017308, 2003.
Dommergue, A., Bahlmann, E., Ebinghaus, R., Ferrari, C., and Boutron, C.: Laboratory simulation of Hg^0 emissions from a snowpack, Anal. Bioanal. Chem., 388, 319–327, 2007.
Dommergue, A., Larose, C., Faïn, X., Clarisse, O., Foucher, D., Hintelmann, H., Schneider, D., and Ferrari, C. P.: Deposition of mercury species in the Ny-Ålesund area (79° N) and their transfer during snowmelt, Environ. Sci. Technol., 44, 901–907, doi:10.1021/es902579m, 2010.
Douglas, T. A., Sturm, M., Simpson, W. R., Blum, J. D., Alvarez-Aviles, L., Keeler, G. J., Perovich, D. K., Biswas, A., and Johnson, K.: Influence of snow and ice crystal formation and accumulation on mercury deposition to the Arctic, Environ. Sci. Technol., 42, 1542–1551, 2008.
Durnford, D. and Dastoor, A.: The behavior of mercury in the cryosphere: A review of what we know from observations, J. Geophys. Res., 116, D06305, doi:10.1029/2010JD014809, 2011.
Durnford, D., Dastoor, A., Figueras-Nieto, D., and Ryjkov, A.: Long range transport of mercury to the Arctic and across Canada, Atmos. Chem. Phys., 10, 6063–6086, doi:10.5194/acp-10-6063-2010, 2010.
Durnford, D., Dastoor, A., Ryzhkov, A., Poissant, L., Pilote, M., Figueras-Nieto, D.: How relevant is the deposition of mercury onto snowpacks? – Part 2: A modeling study, Atmos. Chem. Phys. Discuss., submitted, 2011.
Eyrikh, S., Schwikowski, M., Gäggeler, H. W., Tobler, L., and Papina, T.: First mercury determination in snow and firn from high-mountain glaciers in the Siberian Altai by CV-ICP-MS, J. Phys. IV, 107, 431–434, 2003.
Faïn, X., Ferrari, C. P., Gauchard, P.-A., Magand, O., and Boutron, C.: Fast depletion of gaseous elemental mercury in the Kongsvegen Glacier snowpack in Svalbard, Geophys. Res. Lett., 33, L06826, doi:10.1029/2005GL025223, 2006.
Faïn, X., Grangeon, S., Bahlmann, E., Fritsche, J., Obrist, D., Dommergue, A., Ferrari, C. P.,

ACPD
12, 387–439, 2012

Cairns, W., Ebinghaus, R., Barbante, C., Cescon, P., and Boutron, C.: Diurnal production of gaseous mercury in the alpine snowpack before snowmelt, J. Geophys. Res., 112, D21311, doi:10.1029/2007JD008520, 2007.

Faïn, X., Ferrari, C. P., Dommergue, A., Albert, M., Battle, M., Arnaud, L., Barnola, J.-M., Cairns, W., Barbante, C., and Boutron, C.: 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, doi:10.5194/acp-8-3441-2008, 2008.

Faïn, X., Ferrari, C. P., Dommergue, A., Albert, M. R., Battle, M., Severinghaus, J., Arnaud, L., Barnola, J.-M., Cairns, W., Barbante, C., and Boutron, C.: Polar firn air reveals large-scale impact of anthropogenic mercury emissions during the 1970s, P. Natl. Acad. Sci. USA, 106, 16114–16119, doi:10.1073/pnas.0905117106, 2009.

Fatnassi, H., Boulard, T., Poncet, C., and Chave, M.: Optimisation of greenhouse insect screening with computational fluid dynamics, Biosyst. Eng., 93, 301–312, 2006.

Ferrari, C. P., Dommergue, A., Veysseyre, A., Planchon, F., and Boutron, C. F.: Mercury speciation in the French seasonal snow cover, Sci. Total Environ., 287, 61–69, 2002.

Ferrari, C. P., Dommergue, A., Boutron, C. F., Skov, H., Goodsite, M., and Jensen, B.: Nighttime production of elemental gaseous mercury in interstitial air of snow at Station Nord, Greenland, Atmos. Environ., 38, 2727–2735, 2004.

Ferrari, C. P., Gauchard, P.-A., Aspmo, K., Dommergue, A., Magand, O., Bahlmann, E., Nagorski, S., Temme, C., Ebinghaus, R., Steffen, A., Banic, C., Berg, T., Planchon, F., Barbante, C., Cescon, P., and Boutron, C. F.: Snow-to-air exchanges of mercury in an Arctic seasonal snow pack in Ny-Ålesund, Svalbard, Atmos. Environ., 39, 7633–7645, 2005.

Ferrari, C. P., Padova, C., Faïn, X., Gauchard, P.-A., Dommergue, A., Aspmo, K., Berg, T., Cairns, W., Barbante, C., Cescon, P., Kaleshke, L., Richter, A., Wittrock, F., and Boutron, C.: Atmospheric mercury depletion event study in Ny-Alesund (Svalbard) in spring 2005. Deposition and transformation of Hg in surface snow during springtime, Sci. Total Environ., 397, 167–177, 2008.

Fitzgerald, W. F., Mason, R. P., and Vandal, G. M.: Atmospheric cycling and air-water exchange of mercury over mid-continental lacustrine regions, Water Air Soil Poll., 56, 745–767, 1991.

Fouquart, Y. and Bonnel, B.: Computations of solar heating of the earth's atmosphere: a new parameterization, Contrib. Atmos. Phys., 53, 35–62, 1980.

Garand, L. and Mailhot, J.: The influence of infrared radiation on numerical weather forecasts, in Proceedings of the 7th Conference on Atmospheric Radiation, J146–J151, American

ACPD
12, 387–439, 2012

Meteorological Society, US, 1990.
Garbarino, J. R., Snyder-Conn, E., Leiker, T. J., and Hoffman, G. L.: Contaminants in Arctic snow collected over Northwest Alaskan sea ice, Water Air Soil Poll., 139, 183–214, 2002.
Gbor, P. K., Wen, D., Meng, F., Yang, F., and Sloan, J. J.: Sloan Modeling of mercury emission, transport and deposition in North America, Atmos. Environ., 41, 1135–1149, 2007.
Gilmour, C. C. and Henry, E. A.: Mercury methylation in aquatic systems affected by acid deposition, Environ. Pollut., 71, 131–169, 1991.
Goulet, R. R., Holmes, J., Page, B., Poissant, L., Siciliano, S. D., Lean, D. R. S., Wang, F., Amyot, M., and Tessier, A.: Mercury transformations and fluxes in sediments of a riverine wetland, Geochim. Cosmochim. Ac., 71, 3393–3406, 2007.
Grenfell, T. C. and Maykut, G. A.: The optical properties of ice and snow in the Arctic Basin, J. Glaciol., 18, 445–463, 1977.
Hansen, K. M., Halsall, C. J., and Christensen, J. H.: A dynamic model to study the exchange of gas-phase persistent organic pollutants between air and a seasonal snowpack, Environ. Sci. Technol., 40, 2644–2652, doi:10.1021/es051685b, 2006.
Heidam, N. Z., Christensen, J., Wåhlin, P., and Skov, H.: Arctic atmospheric contaminants in NE Greenland: levels, variations, origins, transport, transformations and trends 1990–2001, Sci. Total Environ., 331, 5–28, 2004.
Herron, M. M., Langway Jr., C. C., Weiss, H. V., Hurley, P., Kerr, R., and Cragin, J. H.: Vanadium and other elements in Greenland ice cores, in: Isotopes and Impurities in Snow and Ice, Proceedings of symposium held during the 16th International Union of Geodesy and Geophysics symposium, Grenoble, 1975, Int. Ass. Hydrol. Sci., 118, 98–102, available at: http://iahs.info/redbooks/118.htm, 1977.
Hicks, J., Marko, K., and Sahulka, D.: Metals in snow columns, in: Flin Flon soils study, Tech. Rep. Appendix R, Hudson Bay Mining and Smelting Co., Ltd., Flin Flon, Manitoba, Canada, available at: http://www.flinflonsoilsstudy.com/doclibrary.php, 2008.
Holmes, C. D., Jacob, D. J., Corbitt, E. S., Mao, J., Yang, X., Talbot, R., and Slemr, F.: Global atmospheric model for mercury including oxidation by bromine atoms, Atmos. Chem. Phys., 10, 12037–12057, doi:10.5194/acp-10-12037-2010, 2010.
Jackson, T. A.: Long-range atmospheric transport of mercury to ecosystems, and the importance of anthropogenic emissions – a critical review and evaluation of the published evidence, Environ. Rev., 5, 99–120, 1997.
Jitaru, P., Infante, H. G., Ferrari, C. P., Dommergue, A., Boutron, C. F., and Adams, F. C.:

ACPD
12, 387–439, 2012

Present century record of mercury species pollution in high altitude alpine snow and ice, J. Phys IV, 107, 683–686, doi:10.1051/jp4:20030395, 2003.
Jitaru, P., Gabrielli, P., Marteel, A., Plane, J. M. C., Planchon, F. A. M., Gauchard, P.-A., Ferrari, C. P., Boutron, C. F., Adams, F. C., Hong, S., Cescon, P., and Barbante, C.: Atmospheric depletion of mercury over Antarctica during glacial periods, Nat. Geosci., 2, 505–508, doi:10.1038/ngeo549, 2009.
Johnson, K. P., Blum, J. D., Keeler, G. J., and Douglas, T. A.: Investigation of the deposition and emission of mercury in arctic snow during an atmospheric mercury depletion event, J. Geophys. Res., 113, D17304, doi:10.1029/2008JD009893, 2008.
Kain, J. S. and Fritsch, J. M.: A one-dimensional entraining/detraining plume model and its application in convective parameterization, J. Atmos. Sci., 47, 2784–2802, 1990.
Kirk, J. L., St. Louis, V. L., and Sharp, M. J.: Rapid reduction and emission of mercury deposited into snowpacks during atmospheric mercury depletion events at Churchill, Manitoba, Canada, Environ. Sci. Technol., 40, 7590–7596, 2006.
Kuhn, M.: The nutrient cycle through snow and ice, a review, Aquat Sci., 63, 150–167, 2001.
Lahoutifard, N., Sparling, M., and Lean, D.: Total and methyl mercury patterns in Arctic snow during springtime at Resolute, Nunavut, Canada, Atmos. Environ., 39, 7597–7606, 2005.
Lahoutifard, N., Poissant, L., and Scott, S. L.: Scavenging of gaseous mercury by acidic snow at Kuujjuarapik, Northern Québec, Sci. Total Environ., 355, 118–126, 2006.
Lalonde, J. D., Poulain, A. J., and Amyot, M.: The role of mercury redox reactions in snow on snow-to-air mercury transfer, Environ. Sci. Technol., 36, 174–178, 2002.
Lalonde, J. D., Amyot, M., Doyon, M.-R., and Auclair, J.-C.: Photo-induced Hg(II) reduction in snow from the remote and temperate Experimental lakes Area (Ontario, Canada), J. Geophys. Res., 108, 4200, doi:10.1029/2001JD001534, 2003.
Lamborg, C. H., Fitzgerald, W. F., Vandal, G. M., and Rolfhus, K. R.: Atmospheric mercury in northern Wisconsin: sources and species, Water Air Soil Poll., 80, 189–198, 1995.
Larose, C., Dommergue, A., De Angelis, M., Cossa, D., Averty, B., Marusczak, N., Soumis, N., Schneider, D., and Ferrari, C.: Springtime changes in snow chemistry lead to new insights into mercury methylation in the Arctic, Geochim. Cosmochim. Ac., 74, 6263–6275, doi:10.1016/j.gca.2010.08.043, 2010.
Leitch, D. R., Carrie, J., Lean, D., Macdonald, R. W., Stern, G. A., and Wang, F.: The delivery of mercury to the Beaufort Sea of the Arctic Ocean by the Mackenzie River, Sci. Total Environ., 373, 178–195, 2007.

ACPD
12, 387–439, 2012

Lin, C.-J., Pongprueksa, P., Lindberg, S. E., Pehkonen, S. O., Byun, D., and Jang, C.: Scientific uncertainties in atmospheric mercury models I: Model science evaluation, Atmos. Environ., 40, 2911–2928, 2006.

Lindberg, S. E., Brooks, S., Lin, C-J., Scott, K., Meyers, T., Chambers, L., Landis, M., and Stevens, R.: Formation of reactive gaseous mercury in the Arctic: Evidence of oxidation of Hg^0 to gas-phase Hg-II compounds after Arctic sunrise, Water Air Soil Poll.: Focus, 1, 295–302, 2001.

Lindberg, S. E., Brooks, S., Lin, C-J., Scott, K. J., Landis, M. S., Stevens, R. K., Goodsite, M., and Richter, A.: Dynamic oxidation of gaseous mercury in the Arctic troposphere at Polar Sunrise, Environ. Sci. Technol., 36, 1245–1256, 2002.

Loewen, M., Kang, S., Armstrong, D., Zhang, Q., Tomy, G., and Wang, F.: Atmospheric transport of mercury to the Tibetan Plateau, Environ. Sci. Technol., 41, 7632–7638, 2007.

Loseto, L. L., Lean, D. R. S., and Siciliano, S. D.: Snowmelt sources of methylmercury to High Arctic ecosystems, Environ. Sci. Technol., 38, 3004–3010, 2004.

Loux, N. T.: Monitoring cyclical air/water elemental mercury exchange, J. Environ. Monit., 3, 43–48, doi:10.1039/b005545j, 2001.

Lu, J. Y., Schroeder, W. H., Barrie, L. A., Steffen, A., Welch, H. E., Martin, K., Lockhart, L., Hunt, R. V., Boila, G., and Richter, A.: Magnification of atmospheric mercury deposition to polar regions in springtime: the link to tropospheric ozone depletion chemistry, Geophys. Res. Lett., 28, 3219–3222, 2001.

Mann, J. L., Long, S. E., Shuman, C. A., and Kelly, W. R.: Determination of mercury content in a shallow firn core from Greenland by isotope dilution inductively coupled plasma mass spectrometry, Water Air Soil Poll., 163, 19–32, 2005.

Mason, R.: Mercury emissions from natural processes and their importance in the global mercury cycle, in Mercury fate and transport in the global atmosphere, Springer US, 173–191, 2009.

Mason, R. P. and Fitzgerald, W. F.: The distribution and biogeochemical cycling of mercury in the equatorial Pacific Ocean, Deep-Sea Res. Pt. 1, 40, 1897–1924, 1993.

Mitchell, C. P. J., Branfireun, B. A., and Kolka, R. K.: Assessing sulfate and carbon controls on net methylmercury production in peatlands: An in situ mesocosm approach, Appl. Geochem., 23, 503–518, 2008a.

Mitchell, C. P. J., Branfireun, B. A., and Kolka, R. K.: Total mercury and methylmercury dynamics in upland-peatland watersheds during snowmelt, Biogeochemistry, 90, 225–241,

ACPD
12, 387–439, 2012

doi:10.1007/s10533-008-9246-z, 2008b.
Monperrus, M., Tessier, E., Amouroux, D., Leynaert, A., Huonnic, P., and Donard, O. F. X.: Mercury methylation, demethylation and reduction rates in coastal and marine surface waters of the Mediterranean Sea, Mar. Chem., 107, 49–63, 2007.
Murozumi, M., Nakamura, S., and Yoshida, Y.: Compile data of chemical compositions in ice cores drilled at Mizuho Station, Memoirs of National Institute of Polar Research, 10, 167–168, 1978.
Nelson, S. J., Johnson, K. B., Weathers, K. C., Loftin, C. S., Fernandez, I. J., Kahl, J. S., and Krabbenhoft, D. P.: A comparison of winter mercury accumulation at forested and no-canopy sites measured with different snow sampling techniques, Appl. Geochem., 23, 384–398, 2008.
Olivier, S., Schwikowsk, M., Brütsch, S., Eyrikh, S., Gäggeler, H. W., Lüthi, M., Papina, T., Saurer, M., Schotterer, U., Tobler, L., and Vogel, E.: Glaciochemical investigation of a ice core from Belukha glacier, Siberian Altai, Geophys. Res. Lett., 30, 2019, doi:10.1029/2003GL018290, 2003.
Outridge, P. M., Macdonald, R. W., Wang, F., Stern, G. A., and Dastoor, A. P.: A mass balance inventory of mercury in the Arctic Ocean, Environ. Chem., 5, 89–111, doi:10.1071/EN08002, 2008.
Pacyna, E. G., Pacyna, J. M., Sundseth, K., Munthe, J., Kindbom, K., Wilson, S., Steenhuisen, F., and Maxson, P.: Global emission of mercury to the atmosphere from anthropogenic sources in 2005 and projections to 2020, Atmos. Environ., 44, 2487–2499, doi:10.1016/j.atmosenv.2009.06.009, 2010.
Poulain, A. J., Lalonde, J. D., Amyot, M., Shead, J. A., Raofie, F., and Ariya, P. A.: Redox transformations of mercury in an Arctic snowpack at springtime, Atmos. Environ., 38, 6763–6774, 2004.
Poulain, A. J., Garcia, E., Amyot, M., Campbell, P. G. C., and Ariya, P. A.: Mercury distribution, partitioning and speciation in coastal vs. inland High Arctic snow, Geochim. Cosmochim. Ac., 71, 3419–3431, 2007a.
Poulain, A. J., Roy, V., and Amyot, M.: Influence of temperate mixed and deciduous tree covers on Hg concentrations and photoredox transformations in snow, Geochim. Cosmochim. Ac., 71, 2448–2462, 2007b.
Ryaboshapko, A., Bullock, R., Christensen, J., Cohen, M., Dastoor, A., Ilyin, I., Petersen, G., Syrakov, D., Artz, R., Davignon, D., Draxler, R., and Munthe, J.: Intercomparison study of

ACPD
12, 387–439, 2012

atmospheric mercury models: 1. Comparison of models with short-term measurements, Sci. Total Environ., 376, 228–240, 2007a.
Ryaboshapko, A., Bullock, R., Christensen, J., Cohen, M., Dastoor, A., Ilyin, I., Petersen G., Syrakov, D., Travnikov, O., Artz, R., Davignon, D., Draxler, R., Munthe, J., and Pacyna, J.: Intercomparison Study of Atmospheric Mercury Models: 2. Modelling results vs. long-term observations and comparison of country deposition budgets, Sci. Total Environ., 377, 319–333, 2007b.
Schroeder, W. H., Anlauf, K. G., Barrie, L. A., Lu, J. Y., Steffen, A., Schneeberger, D. R., and Berg, T.: Arctic springtime depletion of mercury, Nature, 394, 331–332, 1998.
Schuster, P. F., Krabbenhoft, D. P., Naftz, D. L., Cecil, L. D., Olson, M. K., Dewild, J. F., Susong, D. D., Green, J. R., and Abbott, M. L.: Atmospheric mercury deposition during the last 270 years: A glacial ice core record of natural and anthropogenic sources, Environ. Sci. Technol., 36, 2303–2310, 2002.
Sheppard, D. S., Patterson, J. E., and McAdam, M. K.: Mercury content of Antarctic ice and snow: Further results, Atmos. Environ., 25A, 1657–1660, 1991.
Sherman, L. S., Blum, J. D., Johnson, K. P., Keeler, G. J., Barres, J. A., and Douglas, T. A.: Mass-independent fractionation of mercury isotopes in Arctic snow driven by sunlight, Nat. Geosci., 3, 173–177, doi:10.1038/ngeo758, 2010.
Shetty, S. K., Lin, C.-J., Streets, D. G., and Jang, C.: Model estimate of mercury emission from natural sources in East Asia, Atmos. Environ., 42, 8674–8685, 2008.
Simpson, W. R., Carlson, D., Hönninger, G., Douglas, T. A., Sturm, M., Perovich, D., and Platt, U.: First-year sea-ice contact predicts bromine monoxide (BrO) levels at Barrow, Alaska better than potential frost flower contact, Atmos. Chem. Phys., 7, 621–627, doi:10.5194/acp-7-621-2007, 2007a.
Simpson, W. R., von Glasow, R., Riedel, K., Anderson, P., Ariya, P., Bottenheim, J., Burrows, J., Carpenter, L. J., Frieß, U., Goodsite, M. E., Heard, D., Hutterli, M., Jacobi, H.-W., Kaleschke, L., Neff, B., Plane, J., Platt, U., Richter, A., Roscoe, H., Sander, R., Shepson, P., Sodeau, J., Steffen, A., Wagner, T., and Wolff, E.: Halogens and their role in polar boundary-layer ozone depletion, Atmos. Chem. Phys., 7, 4375–4418, doi:10.5194/acp-7-4375-2007, 2007b.
Skov, H., Christensen, J. H., Goodsite, M. E., Heidam, N. Z., Jensen, B., Wåhlin, P., and Geernaert, G.: Fate of elemental mercury in the Arctic during atmospheric mercury depletion episodes and the load of atmospheric mercury to the Arctic, Environ. Sci. Technol., 38, 2373–2382, 2004.

ACPD
12, 387–439, 2012

Snyder-Conn, E., Garbarino, J. R., Hoffman, G. L., and Oelkers, A.: Soluble trace elements and total mercury in Arctic Alaskan snow, Arctic, 50, 201–215, 1997.
Sommar, J., Wängberg, I., Berg, T., Gårdfeldt, K., Munthe, J., Richter, A., Urba, A., Wittrock, F., and Schroeder, W. H.: Circumpolar transport and air-surface exchange of atmospheric mercury at Ny-Ålesund (79° N), Svalbard, spring 2002, Atmos. Chem. Phys., 7, 151–166, doi:10.5194/acp-7-151-2007, 2007.
Steen, A. O., Berg, T., Dastoor, A. P., Durnford, D. A., Hole, L. R., and Phaffhuber, K. A.: Dynamic exchange of gaseous elemental mercury during polar night and day, Atmos. Environ., 43, 5604–5610, 2009.
Steffen, A., Schroeder, W., Bottenheim, J., Narayan, J., and Fuentes, J. D.: Atmospheric mercury concentrations: measurements and profiles near snow and ice surfaces in the Canadian Arctic during Alert 2000, Atmos. Environ., 36, 2653–2661, 2002.
Steffen, A., Douglas, T., Amyot, M., Ariya, P., Aspmo, K., Berg, T., Bottenheim, J., Brooks, S., Cobbett, F., Dastoor, A., Dommergue, A., Ebinghaus, R., Ferrari, C., Gardfeldt, K., Goodsite, M. E., Lean, D., Poulain, A. J., Scherz, C., Skov, H., Sommar, J., and Temme, C.: A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and snow, Atmos. Chem. Phys., 8, 1445–1482, doi:10.5194/acp-8-1445-2008, 2008.
St. Louis, V. L., Rudd, J. W. M., Kelly, C. A., and Barrie, L. A.: Wet deposition of methyl mercury in northwestern Ontario compared to other geographic locations, Water Air Soil Poll., 80, 405–414, 1995.
St. Louis, V. L., Sharp, M. J., Steffen, A., May, A., Barker, J., Kirk, J. L., Kelly, D. J. A., Arnott, S. E., Keatley, B., and Smol, J. P.: Some sources and sinks of monomethyl and inorganic mercury on Ellesmere Island in the Canadian High Arctic, Environ. Sci. Technol., 39, 2686–2701, 2005.
St. Louis, V. L., Hintelmann, H., Graydon, J. A., Kirk, J. L., Barker, J., Dimock, B., Sharp, M. J., and Lehnherr, I.: Methylated mercury species in Canadian High Arctic marine surface waters and snowpacks, Environ. Sci. Technol., 41, 6433–6441, 2007.
Sunderland, E. M., Krabbenhoft, D. P., Moreau, J. W., Strode, S. A., and Landing, W. M.: Mercury sources, distribution, and bioavailability in the North Pacific Ocean: Insights from data and models, Global Biogeochem. Cy., 23, GB2010, doi:10.1029/2008GB003425, 2009.
Sundqvist, H.: A parameterization scheme for non-convective condensation including prediction of cloud water content, Q. J. Roy. Meteor. Soc., 104, 677–690, 1978.
Susong, D. D., Abbott, M. L., and Krabbenhoft, D. P.: Mercury accumulation in snow on the

ACPD
12, 387–439, 2012

Idaho National Engineering and Environmental laboratory and surrounding region, southeast Idaho, USA, Environ. Geol., 43, 357–363, 2003.
Travnikov, O.: Contribution of the intercontinental atmospheric transport to mercury pollution in the Northern Hemisphere, Atmos. Environ., 39, 7541–7548, 2005.
Vandal, G. M., Fitzgerald, W. F., Boutron, C. F., and Candelone, J.-P.: Variations in mercury deposition to Antarctica over the past 34,000 years, Nature, 362, 621–623, 1993.
Van Oostdam, J., Donaldson, S. G., Feeley, M., Arnold, D., Ayotte, P., Bondy, G., Chan, L., Dewaily, É., Furgal, C. M., Kuhnlein, H., Loring, E., Muckle, G., Myles, E., Receveur, O., Tracy, B., Gill, U., and Kalhok, S.: Human health implications of environmental contaminants in Arctic Canada: A review, Sci. Total Environ., 351–352, 165–246, 2005.
Wang, X.-p., Yao, T.-d., Wang, P.-l., Wei-Yang, and Tian, L.-d.: The recent deposition of persistent organic pollutants and mercury to the Dasuopu glacier, Mt. Xixiabangma, central Himalayas, Sci. Total. Environ., 394, 134–143, 2008.
Weiss, H., Bertine, K., Koide, M., and Goldberg, E. D.: The chemical composition of a Greenland glacier, Geochim. Cosmochim. Ac., 39, 1–10, 1975.
Weiss, H. V., Koide, M., and Goldberg, E. D.: Mercury in a Greenland ice sheet: Evidence of recent input by man, Science, 174, 692–694, 1971.
Weiss, H. V., Herron, M. M., and Langway Jr., C. C.: Natural enrichments of elements in snow, Nature, 274, 352–353, 1978.
Witherow, R. A. and Lyons, W. B.: Mercury deposition in a Polar Desert ecosystem, Environ. Sci. Technol., 42, 4710–4716, 2008.
Yue, W., Meneveau, C., Parlange, M. B., Zhu, W., Kang, H. S., and Katz, J.: Turbulent kinetic energy budgets in a model canopy: comparisons between LES and wind-tunnel experiments, Environ. Fluid Mech., 8, 73–95, 2008.
Zhao, T. L., Gong, S. L., Bottenheim, J. W., McConnell, J. C., Sander, R., Kaleschke, L., Richter, A., Kerkweg, A., Toyota, K., and Barrie, L. A.: A three-dimensional model study on the production of BrO and Arctic boundary layer ozone depletion, J. Geophys. Res., 113, D24304, doi:10.1029/2008JD010631, 2008.

ACPD
12, 387–439, 2012

Table 1. Mean 24-h losses of total mercury from surface snow.

Location	Latitude (° N)	Longitude (° E)	24-h loss (%)	Involved in 2nd set of calculations	References
Antarctic					
McMurdo	−77.5	159.8	34	yes	Brooks et al. (2008b)
Midlatitude					
Northwestern Ontario	49.7	−93.7	39	yes	Lalonde et al. (2003)
Ste Foy	47.3	−71.3	36	yes	Lalonde et al. (2002)
Subarctic					
Churchill	58.8	−94.1	51	yes	Kirk et al. (2006)
Kuujjuarapik/ Whapmagoostui	55.3	−77.8	47	yes	Dommergue et al. (2003); Constant et al. (2007)
Arctic					
Barrow	71.3	−156.6	20	no	Johnson et al. (2008)
Cornwallis Island	74.9	−95.0	48	yes	Poulain et al. (2004)
Ellesmere Island	82.0	−75.0	30	yes	St. Louis et al. (2005)
Ny-Ålesund	78.9	11.9	42	yes	Sommar et al. (2007); Dommergue et al. (2010)

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Table 2. Mean concentrations of total mercury in surface snow.

Location	Latitude (° N)	Longitude (° E)	Hg ($ng\,l^{-1}$)	Involved in 2nd set of calculations	References
Antarctic					
South Pole Station	−90.0	0.0	198.0	yes	Brooks et al. (2008a)
McMurdo	−77.5	159.8	101.7	no	Sheppard et al. (1991); Brooks et al. (2008b)
Midlatitude					
Northwestern Ontario	49.7	−93.7	1.6	yes	St. Louis et al. (1995); Lalonde et al. (2003)
Wisconsin	46.0	−89.7	4.1	yes	Bloom and Watras (1989); Fitzgerald et al. (1991); Lamborg et al. (1995)
Ste Foy	47.3	−71.3	3.3	yes	Lalonde et al. (2002)
Maine	44.4	−68.3	9.9	yes	Nelson et al. (2008)
Alps	45.3	5.8	67.6	no	Ferrari et al. (2002); Faïn et al. (2007)
Subarctic					
Churchill	58.8	−94.1	36.0	no	Kirk et al. (2006)
Kuujjuarapik/ Whapmagoostui	55.3	−77.8	10.0	yes	Dommergue et al. (2003); Lahoutifard et al. (2006); Constant et al. (2007)
Arctic					
Barrow	71.3	−156.6	50.6	no	Weiss et al. (1978); Lindberg et al. (2001, 2002); Brooks et al. (2006, 2008b); Douglas et al. (2008); Johnson et al. (2008); Sherman et al. (2010)
Ship: Arctic Ocean	79.0	−154.0	21.0	no	Lu et al. (2001)
Canadian Archipelago	76.0	−98.0	45.0	no	Lu et al. (2001)
Resolute Bay/Cornwallis Island	74.9	−95.0	3.8	no	Lu et al. (2001); Poulain et al. (2004); Lahoutifard et al. (2005); Poulain et al. (2007a)
Cornwallis, Ellesmere Islands	78.5	−85.0	30.3	no	St. Louis et al. (2007)
Ellesmere Island	82.0	−75.0	3.2	yes	St. Louis et al. (2005)
Hudson Bay/Baffin Bay/Davis Str	66.3	−69.7	55.0	no	Lu et al. (2001)
Labrador Sea	57.0	−53.0	38.0	no	Lu et al. (2001)
Summit	72.6	−38.5	0.9	yes	Mann et al. (2005)
Ship: N Atlantic, Arctic Oceans	83.5	0.0	3.3	no	Aspmo et al. (2006)
Ny-Ålesund	78.9	11.9	44.5	no	Berg et al. (2001, 2003); Sommar et al. (2007); Ferrari et al. (2005, 2008); Steen et al. (2009); Dommergue et al. (2010); Larose et al. (2010)

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Table 3. Mean concentrations of total mercury in seasonal snowpacks.

Location	Latitude (° N)	Longitude (° E)	Hg (ng l^{-1})	Involved in 2nd set of calculations	References
Antarctic					
South Pole Station	−90.0	0.0	10.0	yes	Brooks et al. (2008a)
McMurdo	−77.5	159.8	0.2	yes	Sheppard et al. (1991)
McCarthy Ridge	−74.6	163.1	0.5	yes	Capelli et al. (1998)
Hercules Névé	−73.1	165.5	0.2	yes	Capelli et al. (1998)
Midlatitude					
Tibetan Plateau	31.5	90.0	4.5	yes	Loewen et al. (2007)
Idaho	43.5	−112.5	5.7	yes	Susong et al. (2003)
Flin Flon	54.8	−101.9	520	no	Hicks et al. (2008)
Minnesota	46.0	−94.0	1.0	yes	Balogh et al. (2000)
Northwestern Ontario	49.7	−93.7	0.6	yes	Lalonde et al. (2003)
North-central Minnesota	47.5	−93.5	0.8	yes	Mitchell et al. (2008b)
Laurentians	46.0	−74.0	2.7	yes	Poulain et al. (2007b)
Maine	44.4	−68.3	14.0	yes	Nelson et al. (2008)
Alps	45.3	5.8	130.6	no	Faïn et al. (2007)
Subarctic					
Churchill	58.8	−94.1	15.7	no	Kirk et al. (2006)
Kuujjuarapik/ Whapmagoostui	55.3	−77.8	5.9	yes	Dommergue et al. (2003); Constant et al. (2007)
Arctic					
Barrow	71.3	−156.6	17.7	no	Snyder-Conn et al. (1997); Garbarino et al. (2002); Lindberg et al. (2002); Douglas et al. (2008); Johnson et al. (2008)
Cornwallis Island	74.9	−95.0	6.0	no	Poulain et al. (2004, 2007a)
Cornwallis, Ellesmere Islands	78.5	−85.0	28.0	no	St. Louis et al. (2007)
Ellesmere Island	82.0	−75.0	1.1	yes	St. Louis et al. (2005)
Alert	83.0	−62.6	10.1	yes	Cobbett et al. (2007)
Station Milcent	70.3	−44.6	494	no	Herron et al. (1977)
Dye-3	65.2	−43.8	46	no	Weiss et al. (1975)
Ny-Ålesund	78.9	11.9	8.5	yes	Ferrari et al. (2005); Dommergue et al. (2010); Larose et al. (2010)

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Table 4. Mean concentrations of total mercury in the snowpack meltwater's ionic pulse.

Location	Latitude (° N)	Longitude (° E)	Hg (ng l^{-1})	Involved in 2nd set of calculations	References
Midlatitude					
Northwestern Ontario	49.7	−93.7	7.9	yes	Allan et al. (2001)
North-central Minnesota	47.5	−93.5	13.0	yes	Mitchel et al. (2008b)
Alps	45.3	5.8	72	no	Faïn et al. (2007)
Subarctic					
Churchill	58.8	−94.1	4.4	no	Kirk et al. (2006)
Kuujjuarapik/ Whapmagoostui	55.3	−77.8	11.9	yes	Dommergue et al. (2003)
Svartberget Catchment	64.2	19.8	3.5	yes	Bishop et al. (1995)
Arctic					
Barrow	71.3	−156.6	21.3	yes	Lindberg et al. (2002); Douglas et al. (2008)
Ny-Ålesund	78.9	11.9	6.4	yes	Dommergue et al. (2010)

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Table 5. Mean concentrations of total mercury in long-term snowpack-related records.

Location	Latitude (° N)	Longitude (° E)	Hg (ng l^{-1})	Involved in 2nd set of calculations	References
Antarctic					
Commonwealth Glacier	−77.5	16.5	3.8	yes	Witherow and Lyons (2008)
Dome C	−76.0	124.0	3.7	yes	Vandal et al. (1993); Jitaru et al. (2009)
Mizuho Station	−70.7	44.3	1.5	yes	Murozumi et al. (1978)
Midlatitude					
Dasuopu Glacier	28.4	85.7	8.6	yes	Wang et al. (2008)
Upper Fremont Glacier	43.3	−109.4	7.5	yes	Schuster et al. (2002)
Col du Dome glacier	45.8	6.8	2.2	yes	Jitaru et al. (2003)
Belukha glacier	49.8	86.6	1.7	yes	Eyrikh et al. (2003)
Arctic					
Camp Century	77.2	−61.1	87.6	no	Weiss et al. (1971); Carr and Wilkniss (1973)
Site 2	77.0	−56.1	135	no	Carr and Wilkniss (1973)
Station Milcent	70.3	−44.6	513	no	Herron et al. (1977)
Dye-3	65.2	−43.8	40.4	no	Weiss et al. (1975); Appelquist et al. (1978)
Summit	72.6	−38.5	3.3	yes	Boutron et al. (1998); Mann et al. (2005); Faïn et al. (2008)
Crete	71.1	−37.3	8.3	no	Appelquist et al. (1978)

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Table 6. Mean concentrations of total mercury in long-term snowpack-related records that were not included in Durnford and Dastoor (2011).

Location	Latitude (° N)	Longitude (° E)	Time period	sample size	Mean ($ng\,l^{-1}$)	Reference
Antarctic						
Mizuho Station, Antarctica	−70.7	44.3	–	26	1.48	Murozumi et al. (1978)
Midlatitudes						
Dasuopu Glacier, Tibet	28.4	85.7	1998–2005	41	8.59	Wang et al. (2008)
Belukha Glacier, Siberia	49.8	86.6	–	128	1.67	Eyrikh et al. (2003)

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Table 7. Model environmental variables.

Number	Variable	Description	Notes
atmospheric mercuric environment			
1	GEM	concentration of surface-level atmospheric GEM	–
2	DOxDp	deposition of oxidized mercury through dry processes	–
3	WOxDp	deposition of oxidized mercury through wet processes	–
variables impacting the photoreduction of oxidized mercury in snowpack-related media			
4	Alb	Albedo	varies monthly
5	SW	amount of short-wave radiation absorbed at the surface	–
6	LAI	leaf area index	varies monthly
variables describing atmospheric surface-level wind			
7	WdSpAv	average wind speed	snowpack ventilation driven by wind pumping is more effective with stronger winds
8	WdSpF6	frequency of wind speeds of at least 6 m s^{-1}	
variables describing the surface-layer atmospheric stability[a]			
9	TKE	surface-level turbulent kinetic energy	produced by wind shear and static instability, lost through viscous dissipation
10	HPBL	surface stability function	based on the bulk Richardson number[b]
11	SfcSFn	height of the planetary boundary layer	–
variables describing or related to the physical characteristics of the snowpack			
12	SnoDp	maximum snowpack depth within a single season	–
13	SnoDn	average snowpack density	–
14	RH	surface-level relative humidity	–
15	SfcT	surface-level temperature	–
variables describing surface-level atmospheric pressure			
16	SfcP	surface pressure[c]	as pressure increases locally, skies clear. This promotes photoreduction within the snowpack, and emission-augmenting radiationally-induced thermal instability. This also diminishes the likelihood that surface-level snowpack mercury will be buried by fresh snowfalls
17	SLP	sea level pressurec	
variables describing solid precipitation			
18	PrTot	total solid precipitation	surface-layer snowpack-related mercury can be rendered less available for emission by being buried by new solid precipitation
19	PrF24h	frequency of solid precipitation of at least 5 mm over 24 h	
20	PrF6h	frequency of solid precipitation of at least 0.5 mm over 6 h	

[a] snowpack ventilation increases with decreasing surface-level stability; [b] compares the strengths of turbulence produced thermally and by vertical shear; [c] increases with altitude

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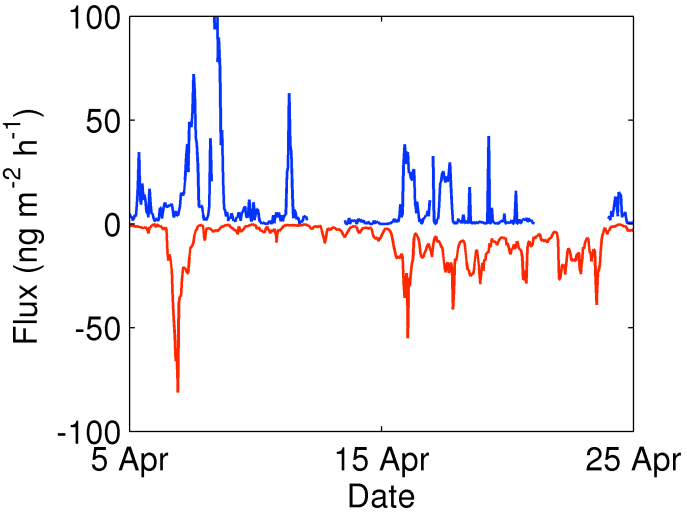

Fig. 1. Time series of observed net emission (blue) (Steen et al., 2009) and simulated deposition of oxidized mercury by both wet and dry processes (red) for Ny-Ålesund in 2008.

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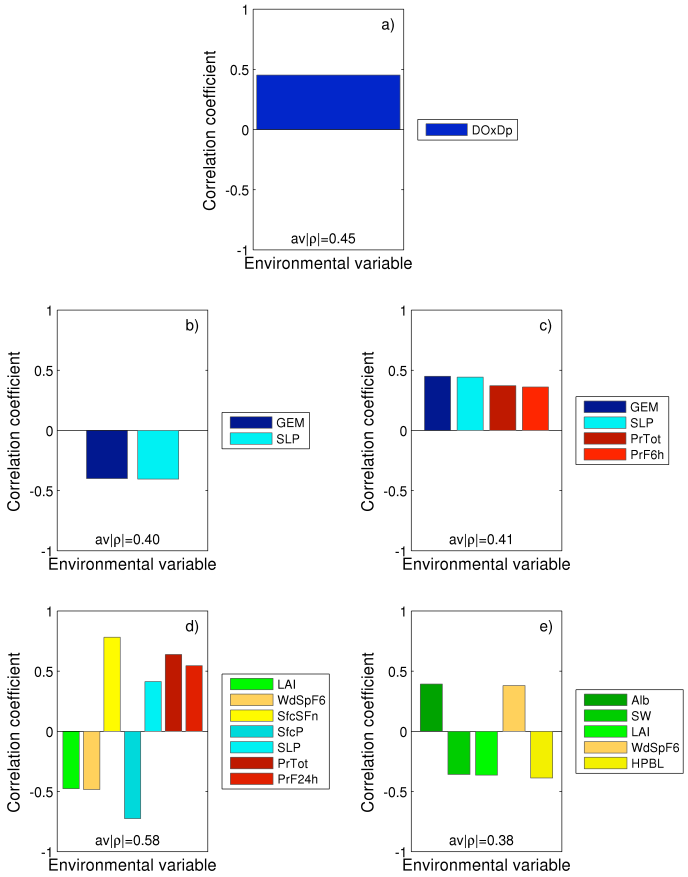

Fig. 2. Correlation coefficients of the model environmental variables that are correlated ($|\rho| \geq 0.35$) with observed **(a)** 24-h loss of mercury from surface snow, and concentration of mercury in **(b)** surface snow, **(c)** seasonal snowpacks, **(d)** the snowpack meltwater's ionic pulse, and **(e)** long-term snowpack-related records, for Set1 calculations. Each panel's legend lists the model environmental variables that are correlated with the panel's observed snowpack-related variable. Provided at the bottom of each panel is the average of the absolute value of the correlation coefficients presented in that panel.

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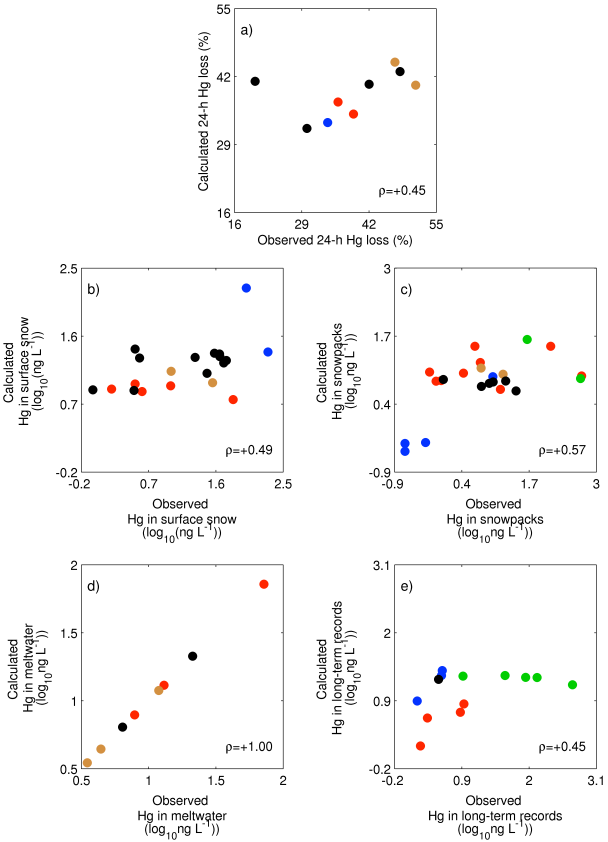

Fig. 3. Regression-derived, or calculated, versus observed snowpack-related mercury variables for Set1: **(a)** the 24-h loss of mercury from surface snow, and for the concentration of mercury in **(b)** surface snow, **(c)** seasonal snowpacks, **(d)** the snowpack meltwater's ionic pulse, and **(e)** long-term snowpack-related records. Calculated variable values are derived using the regression relationships (Sect. 2.3). Blue, red, brown, black and green circles represent observations from, respectively, Antarctica, midlatitudes, the subarctic, the Arctic, and Greenlandic locations discussed in studies from the 1970's. In each panel's lower right corner is the correlation coefficient between the calculated and observed sets of mercury values.

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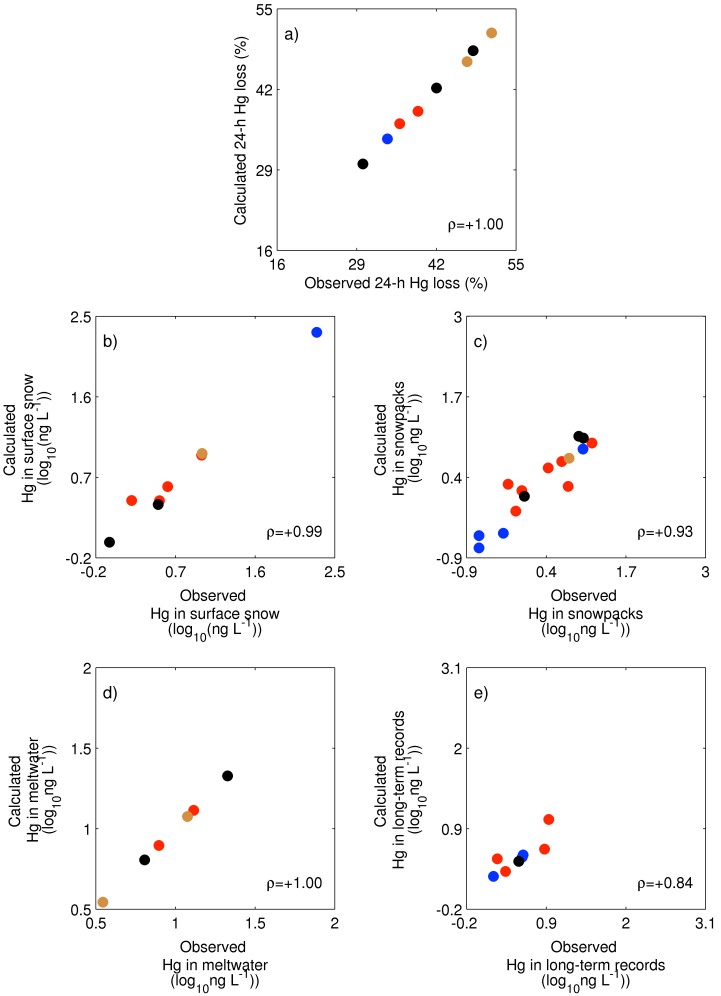

Fig. 4. As per Fig. 3 for Set2.

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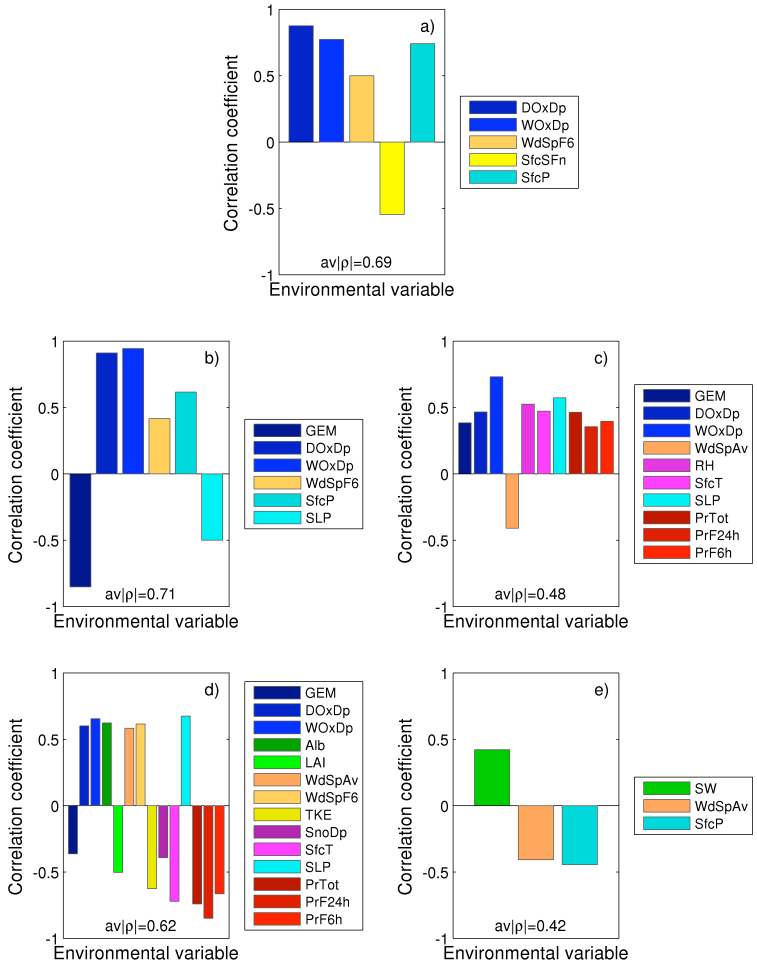

Fig. 5. As per Fig. 2 for Set2.

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