Polar firn air reveals large-scale impact of anthropogenic mercury emissions during the 1970s

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Mercury (Hg) is an extremely toxic pollutant, and its biogeochemical cycle has been perturbed by anthropogenic emissions during recent centuries. In the atmosphere, gaseous elemental mercury (GEM; Hg\textsuperscript{0}) is the predominant form of mercury (up to 95%). Here we report the evolution of atmospheric levels of GEM in mid- to high-northern latitudes inferred from the interstitial air of firn (perennial snowpack) at Summit, Greenland. GEM concentrations increased rapidly after World War II from \textsuperscript{1.5} ng m\textsuperscript{-3} reaching a maximum of \textsuperscript{3} ng m\textsuperscript{-3} around 1970 and decreased until stabilizing at \textsuperscript{1.7} ng m\textsuperscript{-3} around 1995. This reconstruction reproduces real-time measurements available from the Arctic since 1995 and exhibits the same general trend observed in Europe since 1990. Anthropogenic emissions caused a two-fold rise in boreal atmospheric GEM concentrations before the 1970s, which likely contributed to higher deposition of mercury in both industrialized and remote areas. Once deposited, this toxin becomes available for methylation and, subsequently, the contamination of ecosystems. Implementation of air pollution regulations, however, enabled a large-scale decline in atmospheric mercury levels during the 1980s. The results shown here suggest that potential increases in emissions in the coming decades could have a similar large-scale impact on atmospheric Hg levels.

Results and Discussion

The potential of polar ice sheets to serve as an archive for reconstruction of past atmospheric compositions has been well


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established (17, 18). From the top surface to a depth of approximately the same rate as the surrounding ice. At still greater depths, when the overburden is sufficient, all pores become closed, and air can no longer be extracted. This defined the transition from firm to ice referred to as close-off and is located at approximately 69 m depth at Summit Station (20).

To infer the atmospheric record of GEM from firn air concentrations presented in Fig. 1, we determined a site specific, diffusivity-depth relationship. No changes in the physical structure of the firm required consideration: surface temperature and accumulation are the main parameters determining firm structure, and their limited evolution at Summit during the last century (22, 23) did not induce significant changes in either density or porosity of the firm.

Fig. 1. Gaseous elemental mercury (GEM; Hg°) concentrations measured in firn air at Summit Station from 15 to 79.5 m depth. The atmospheric level reported is a mean concentration from three weeks of measurements during both summer 2005 and spring 2006 (21). Above 69 m depth in the firm is the diffusion zone, where gas can diffuse rapidly. From 69 to 80 m is the lock-in zone, where impermeable winter layers prevent vertical diffusion of air, but persistent open porosity allows air pumping. The bars represent one standard deviation confidence interval.

Due to physical effects and, in some instances, chemical effects, the composition of firm air at any depth does not correspond exactly to the atmospheric composition at some time in the past (20). Thus, evaluation of all physical and chemical processes that may affect GEM between the shallow snowpack and the deep firm is required to correctly infer an atmospheric history from firn air concentrations (reported in Fig. 1). We recently investigated this transfer function for GEM under present conditions and proposed a specific conceptual model of the firm at Summit (21). Three primary zones were categorized: the chemical zone, the diffusion zone, and the lock-in zone. In the near surface snow air, we observed during late spring and summer variability of GEM concentrations on a daily timescale induced by chemical processes, as well as a seasonal shift in these chemical mechanisms. Such processes defined the chemical zone, which extends from the surface to ~2.5 m depth. Daily variations of GEM concentrations are too rapid to influence the firm record, however, and the influence of seasonal variations does not extend below the first 15 m of the firm. The diffusion zone extends from the bottom of the chemical zone to the top of the lock-in zone and is comprised of an openly porous and permeable matrix in which air composition is solely determined by physical processes. Within the lock-in zone, the impermeable winter layer prevents vertical diffusion of air, but persistent open porosity in the summer layer allows extraction of samples. Hence, air in the lock-in zone cannot equilibrate with the overlying diffusive column. The transition between the diffusion zone and the lock-in zone is located at ~69 m depth during this study. Above 69 m depth, the diffusion zone allows a smooth record of atmospheric changes. Below 69 m depth, the air ages at approximately the same rate as the surrounding ice. At still greater depths, when the overburden is sufficient, all pores become closed, and air can no longer be extracted. This defined the transition from firm to ice referred to as close-off and is located at ~79.5 m depth at Summit Station (20).

During the last 200 years (27), and there has been no evidence for a large-scale destruction of GEM in the interstitial air of snowpacks depend on environmental conditions (irradiation, temperature, and presence of water layer around snow grains) and the chemical composition of snow (e.g., presence of halogens or divalent mercury species as a consequence of surface deposition) (25).

Greenland surface air temperature trends, including at the Summit site, have not shown persistent warming since 1930 in contrast to global average surface temperature (23). Liquid water in snowpacks could enhance GEM emission (e.g., during snowmelt) (26), but occurrences of surface snow melting at Summit are extremely improbable. Similarly, important changes in solar irradiation at Summit during the last century are unlikely. Direct radiative forcing induced by natural variability of solar activity has been extremely low during the last 200 years (27), and there has been no evidence for a large-scale destruction of stratospheric ozone in the Northern hemisphere (28). Arctic ozone levels exhibit high natural seasonal and interannual variability, however, driven primarily by atmospheric dynamics; and between 1979 and 2000, the trend in mean annual total column ozone over the Arctic was about ~3% per decade (7% accumulated loss) (29). Photo-reduction of divalent mercury species in snow is enhanced by UV irradiation (30, 31). Although the amount of UV radiation reaching the snow surface at Summit is notably influenced by stratospheric ozone levels, the UV measurement time series available in the Arctic is not yet long enough to allow trends to be detected. A recent laboratory study showed that a 10% increase in UV radiation would have
A negligible effect on GEM production in the interstitial air of alpine snow (31). How snow composition impacts mercury chemistry is still under challenge (32). Higher levels of sea salts favor reduction processes and thus GEM destruction in the snow interstitial air (30), but no long-term trends have been reported for sea salt impurities deposited on the Greenland ice sheet during the last century (33). Recent evolution of surface snow composition at Summit has been dominated by human-driven pollutants, but Hg\textsuperscript{2+} concentrations measured in a firn core dated from 1949 to 1989 exhibited low values and limited variation (range: 0.05–2.0 pg g\textsuperscript{-1}; mean: 0.43 pg g\textsuperscript{-1}) (13).

GEM does not adsorb on ice surfaces, as demonstrated by both field and laboratory measurements (34, 35), and we showed that present chemical processes in the shallow firn do not perturb the long-term record of past atmospheric GEM (21). We cannot eliminate the possibility that slow evolution of environmental conditions (e.g., surface temperature and irradiation) and snow composition (e.g., sea salts levels) induced slight evolution in chemical processes involving GEM in the shallow firn during recent decades. However, such processes are unlikely during winter and occur during a limited period of the year (typically from late spring to early fall, 21), and we argue that their past evolution will have a negligible impact on the deep firn atmospheric record. Thus, we conclude the firn air is a reliable archive to investigate historical changes of GEM atmospheric concentrations during the last ~70 years.

Atmospheric trends inferred from polar archives inform about boundary layer levels, and the lack of information about free tropospheric background could be a concern for many gaseous species. Conversely, specific oxidation processes can lead to depletion of GEM in the upper troposphere and lower stratosphere (36), and GEM concentrations in the boundary layer could be relevant information when measured far from large pollution sources (e.g., at Summit). Active atmospheric chemistry has been reported for polar coastal areas: the so-called Atmospheric Mercury Depletion Events (AMDEs), occurring simultaneously with the post-solar sunrise destruction of ozone, can lead to complete depletions of atmospheric GEM during springtime (32). No atmospheric ozone depletion events were observed at Summit (37); and, consequently, AMDEs are unlikely to occur in central Greenland. No strong variations in atmospheric GEM concentrations are thus expected throughout the year at Summit. Furthermore, firn air in the lock-in zone has the useful property that it averages surface gas concentrations for a decade or so, due to diffusion in the firm.

GEM atmospheric monitoring available from the Arctic at Alert (82°N, 62°E, 205 m, Canada) (38) and Ny-Ålesund (78°N, 11°E, 474 m, Spitzbergen) (8) as well as the GEM history reconstructed from the firn air at Summit exhibit stable GEM concentrations of about 1.7 ng m\textsuperscript{-3} during the last 10 years (Fig. 3). In Europe, atmospheric GEM monitoring started at the summit of the Wank mountain (47°N, 11°E, 1780 m, Germany), at Lista (58°N, 6°E, 10 m, Norway), and at Mace Head (53°N, 9°W, 10 m, Ireland) in 1990 (39), 1993 (40), and 1996 (41), respectively. The Wank station is located in a central and continental area and is more strongly influenced by regional sources than the Lista and Mace Head stations. From 1990 to 1996, a decrease in atmospheric GEM concentrations was observed at both the Wank and Lista stations, with higher levels at Wank (Fig. 3). This trend has been related to the 45% decrease in European anthropogenic emissions reported between 1990 and 1995 (42). Atmospheric GEM history reconstructed from
the Summit record reproduces this decrease, but to a lesser extent. It is likely that the influence of European anthropogenic sources is diluted at northern latitudes, but the signal is still evident. Since 1996, Mace Head and Lista records exhibit stable GEM atmospheric levels around 1.7 ng m⁻³, which are well reconstructed using firn air from Summit.

From 1980 to 1990, atmospheric GEM concentrations in the Northern Hemisphere were only investigated at one station, Rorvik, near Gothenburg (53°N, 11°E, 10 m, Sweden), which is likely to have been strongly affected by regional Hg sources (i.e., Eastern Europe before the breakup of the USSR). The Rorvik record exhibits maximum GEM concentrations in the late 1980s but is likely only representative of a local pattern as pointed out by a panel of international experts convened in Madison, Wisconsin as part of the 8th International Conference on Mercury as a Global Pollutant (August 2006) (1). Thus, the Rorvik record is not directly comparable to our Greenland firn air reconstruction. Before 1980, the only atmospheric GEM data were obtained during short ship cruises (i.e., a few weeks) across the Atlantic Ocean. These measurements exhibit strong latitudinal gradients with peak values around 40°N and mean values of ~2 ng m⁻³ in the northern hemisphere (7, 43–45). While atmospheric GEM concentrations reconstructed from Summit firn air are higher than mean hemispheric values, they reproduce GEM levels observed shipboard between 40° and 50°N (see Fig. S1).

Reconstructions of the worldwide trend of GEM concentrations have been attempted using all direct measurements of atmospheric GEM available since 1977. Slentz et al. (8) suggested that GEM concentrations in the northern atmosphere had been increasing since 1977 to reach a maximum in the late 1980s. More recently, Lindberg et al. (1) concluded that a null hypothesis (i.e., little change in atmospheric GEM since 1977) could be as likely as the history proposed by Slentz et al. (8). Both reconstructions strongly relied on mean hemispheric GEM levels inferred from oceanographic expedition measurements. These ship-based records, however, are strongly influenced by short timescale variations of both natural and anthropogenic GEM sources and do not allow quantitative modeling of latitudinal distribution of GEM concentrations, even though they appear to exhibit a north-south structure.

Atmospheric GEM concentrations inferred from Greenland firn air reproduce the scarce GEM measurements reported from 1977 to 1980 across the Atlantic Ocean at mid-northern latitudes (7) (see Fig. S1 and SI Text for more details), confirming a spatially heterogeneous evolution of the Northern Hemisphere atmospheric mercury burden during recent decades. We conclude, therefore, that evolution of GEM concentrations reconstructed above Greenland cannot be generalized to the whole Northern Hemisphere, but is instead likely to be representative of middle and high northern latitudes. Most anthropogenic sources have been located at temperate northern latitudes, and global atmospheric circulation transfers to Greenland air masses from North America, Europe, and, to a lesser extent, Asia (46). Thus, firn air collected at Summit becomes a powerful archive to infer the impact of human emissions on the atmospheric reservoir during recent decades.

Globally, approximately half a million tons of metallic Hg have been produced from cinnabar and other ores for industrial applications (mainly nonferrous metal smelting, manufacturing, and chlor-alkali plants) since 1925 (47). Only a fraction of the mercury extracted from geological stocks has been emitted directly to the atmosphere by industrial processes (~30% in recent decades (48)). In both the United States and Europe, however, most sediment and peat record reconstructions show higher deposition of atmospheric diurnal mercury during the 1970s (10, 11, 14), simultaneous with the peak in worldwide production of mercury of ~10⁷ kg year⁻¹ (Fig. 2B) (49). Worldwide mercury production is, therefore, an indirect indicator of anthropogenic Hg emissions to the atmosphere. Such an indicator is extremely useful when direct estimates of anthropogenic mercury emissions by source inventories are not available (i.e., before 1980).

Atmospheric GEM concentrations, reconstructed from Greenland firn air and the worldwide production of mercury, peak at roughly the same time during the 1970s (Fig. 2). Later, from 1980 to 2000, the atmospheric trend of GEM concentrations and global estimates of anthropogenic emissions of mercury to the atmosphere (mainly emissions from coal combustion) exhibit a similar trend: a large decrease during the 1980s and then stabilization between 1990 and 2000 (3, 50, 51). Hence, atmospheric GEM concentrations inferred from Greenland firn air and global anthropogenic Hg emissions have exhibited consistently similar trends during the most recent decades (Fig. 2), suggesting that the atmospheric reservoir of mercury at mid- and high-northern latitudes has been driven mainly by anthropogenic emissions during the last decades. Consequently, rapid industrial development in the Northern Hemisphere may have caused the significant two-fold rise in boreal GEM atmospheric levels reported after World War II. The decline observed from the 1970s to the early 1990s is likely a response to emission controls implemented through the United States Clean Air Act of 1970 and the 1977 amendments. During the same period, air pollution regulations in Europe gradually eliminated uncontrolled Hg emission sources that were common in urban areas.

The atmospheric lifetime of mercury is long enough to allow transport of GEM from emission areas, mostly mid-northern latitudes, to remote locations. In the Arctic, the record of atmospheric deposition of Hg²⁺ species in lake sediments exhibits a similar global trend as atmospheric GEM concentrations inferred from Greenland firn: lake sediments show an increase in mercury deposition which parallels increasing industrialization (see ref. 15 for a critical review). However, deposition patterns differ among Arctic lakes, particularly within the last few decades where either increases or decreases in Hg accumulation rates have been reported in Alaska (52) as well as in Greenland (11). Cycling within individual lake and transport from surrounding catchment soils may amplify or diminish the atmospheric deposition signal. Discrepancy between sediment archives also could be related to recent climate warming that may have affected the limnology of High Arctic lakes (16). However, both sediment records of atmospheric deposition of Hg²⁺ species at high northern latitudes and atmospheric GEM concentrations inferred from Greenland firn air support the conclusion that transfer of anthropogenic inorganic mercury through the atmosphere to terrestrial and marine reservoirs occurs on a large scale. The connection between deposition of inorganic mercury, enhanced during the 1970s, and contamination of ecosystems, due to methylation processes is more complex. Mercury levels in lakes respond rapidly (within years) to changes in mercury deposition directly to their surfaces, but much more slowly (in decades) to changing inputs to their watersheds (6). Although decreases in fish mercury have been reported recently for lakes exposed to reduced mercury deposition (53), other aquatic ecosystems may continue to bio-accumulate Hg due to higher anthropogenic emissions in the 1970s.

Conclusion

Mercury emissions have decreased recently in Europe and North America, but these declines have been offset by increases in Asia, one of the fastest growing regions in the world (3). Asia has become the largest contributor of anthropogenic atmospheric Hg, responsible for more than half of global emissions, and a significant increase in emissions from this region is expected in the next few decades due to rapid economic and industrial development (54). Notably, anticipated growth in coal combustion, steel production, gold mining, and disposal of Hg-
Materials and Methods

Firm Air Sampling. From May 25 to May 31, 2006, we investigated GEM in the firm air at a remote location 10 km away from Summit Station, central Greenland (72.6°N, 38.5°W, 3200 m elevation). We sampled firm air using established methods (18, 19, 56) from one borehole at depths of 15, 25, 30, 40, 50, 58, 63, 66, 70, 72, 74, 76, 78, and 79.5 m. A 4-m long bladder was lowered into the borehole after drilling to the sampling depth and pressurized with air from the bottom of the hole, effectively sealing the hole. Two Dekabon (polyethylene/aluminum composite) lines were used to pump firm air from a space left immediately below the bladder. These lines drew air from two separate openings above and below a horizontal stainless steel baffle that was nearly as wide as the borehole. Air was pumped from the upper opening at 20 L min⁻¹ and directed to waste after measuring CO₂ concentration (in situ measurements using an NDIR analyzer). When CO₂ levels stabilized, indicating effective removal of contamination by younger and/or ambient air, sampling from the lower opening was initiated. Firm air was first collected in flasks for additional measurements (e.g., CH₄ and CFCs) before GEM sampling actually started. Pumping from the upper opening continued during sampling (at 20 L min⁻¹) to remove any air leaking from within or around the bladder. This technique also served to keep sampled air out of contact with the bladder itself.

GEM Analysis. A gas phase mercury analyzer 2537A (Tekran Inc.) was used for determination of GEM concentrations in firm air. The prefiltered air stream (0.2-μm Teflon particle filter) was collected on two gold cartridges. GEM was thermally desorbed and detected by cold vapor atomic fluorescence spectrometry at 253.7 nm. Use of dual gold cartridges allowed alternate sampling and desorption, resulting in continuous measurement of GEM on a predefined time base. Set-up, accuracy, and precision of this instrument have been evaluated previously during field comparisons at an urban/industrial site (57) and a remote marine background location (58). The Tekran analyzer was operated with a five-minute sampling frequency, and the air was sampled at a flowrate of 1 L min⁻¹. The analyzer was calibrated every 25 h with an internal automatic permeation source. The detection limit for GEM in this operation mode is about 0.15 ng m⁻³. Since the internal pump of the 2537A Tekran analyzer was not strong enough to pump firm air, particularly from deeper layers, a PTFE pump (M2-2C, Vacuubrand, Inc.) was connected to the firm sampling line, upstream to the Tekran analyzer. This pump delivered firm air to the mass spectrometer at a flowrate of 1 L min⁻¹. We measured blanks of the PTFE pump before and after sampling at all depths to quantify any contamination introduced by this additional pump.

At each depth, we sampled firm air for 40 min, resulting in approximately eight GEM measurements. We checked blanks of the Dekaban sampling line both at the beginning and end of the fieldwork; they were 0.08 ± 0.13 ng m⁻³ in 8, before sampling, on May 25, 2006) and 0.01 ± 0.06 ng m⁻³ (n = 18, after sampling, on June 1, 2006).

Diffusion Modeling. We used a one-dimensional gas diffusion model in Eulerian coordinates developed by Rommelaere et al. (24) to infer the atmospheric record of GEM from firm air measurements (Fig. 1). Processes taken into account included (i) air mixing by pressure and temperature gradients down to a few meters below the surface (i.e., the so-called convection zone); (ii) molecular diffusion in the open pore space and gravitational fractionation (entrainment toward the deeper firn depends on concentration gradients, diffusivities, and molar mass); and (iii) a downward air flux in the open porosity zone due to bubble closure removing air from the open pores. Note that this removed air has to be replaced by air coming from the upper parts of the firn, thus creating a downward flux. We set temperature and accumulation rate to their present-day values [respectively, 241 K (20) and 224 kg m⁻² year⁻¹ (20)] and assumed these rates were constant throughout the model run. We used firm structure parameters (density and closed porosity) from the EUROCORE drilling (20) (see Fig. S2).

GEM diffusivity in firm air depends on both GEM diffusivity in air and firm tortuosity. Tortuosity of a porous medium represents the complexity of the pathway and is commonly calculated as the ratio of the mean path length to the minimum possible (straight line) path length. We used an inverse method (60-62) to determine tortuosity of depths in the firn atmospheric GEM trend and from CO₂ concentrations measured in the firn at Summit. In other words, we obtained a site specific, tortuosity-depth relationship by adjusting diffusivity until the model reproduced the observed CO₂ firm-air profile when driven by the independently derived atmospheric CO₂ history (see Fig. S3). We sampled air for CO₂ analysis at the same depths as GEM.

We validated parameterization of the model diffusivity using CH₄ and three halocarbon species (CFC11, CFC113, and CCCL₄) for which atmospheric histories have been estimated from emission scenarios and real-time measurements (60-62). Concentration-depth profiles were determined for these four species using the adjusted diffusivity-depth profile in the diffusion model, and good agreement with concentrations-depth profiles actually measured in the firm sample was obtained (see Figs. S4 and S5).

Monte Carlo Modeling. Little information is available about past atmospheric levels of GEM, but real-time measurements reported since 1977 (8) and industrial production figures (49) suggest that GEM concentrations could have peaked within the last 50 years. Thus, we chose a mathematical parameterization of atmospheric GEM history from 1940 to 2006 (firm air model input)—which allowed for the possibility of a constant level, monotonic increase or decrease, or a GEM concentration oscillation during this timeframe—depending on the choice of parameter values. We tested a wide range of parameter values with the forward firm air model previously described (Monte Carlo approach), representing widely varying scenarios for atmospheric evolution of GEM concentrations. We then modeled a profile of GEM concentrations in the firn from each atmospheric history tested. Agreement between a modeled firm profile and the experimental firm profile (Fig. 1) was estimated using the χ² parameter (see SI Text). According to Tarantola theory (63), we were able to associate a probability density with each atmospheric scenario tested. We finally assessed probability distributions for GEM concentrations with a 1-year time resolution, and we calculated means and standard deviations from all distributions (i.e., every year from 1940 to 2006) (see SI Text). The envelope of atmospheric GEM concentrations presented in this study corresponds to the mean concentrations plus or minus one standard deviation.

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