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Archives of total mercury reconstructed with ice and snow from Greenland and the Canadian High Arctic

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HIGHLIGHTS

- THg archive with a high resolution reconstructed with ice and snow samples from the polar regions.
- First comparable and reproducible THg archives from Greenland and the Canadian High Arctic.
- A broad THg and F_{THg} peak between late 1980s and late 1990s was common to all 3 locations studied.
- Volcanic contribution of Hg in glacier ice has not been found outstanding.
- F_{THg} found in studied areas is much lower than those found in lake sediments and peat bogs.

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ABSTRACT

This study reports total Hg concentration and atmospheric flux data from ice cores and snow/ice shallow pits from two Canadian Arctic and one Greenland glaciers, with the aim of reconstructing a high resolution record of THg deposition extending back into the pre-industrial period. An 88-m ice core (653 samples) from the NEEM glacier site in Northwest Greenland was retrieved in August 2010. The bottom sample was dated to 1748, resulting in a 262 year archive. Snow and ice samples (143 samples) were recovered from a 10.3-m pit dug on the Mt. Oxford Icefield, Nunavut, in May 2008, covering 30 years. Another 15.5-m short core drilled on the Agassiz Ice Cap, Nunavut, in April 2009 yielded 191 samples covering 74 years. Net rates of atmospheric THg deposition (F_{THg}) were calculated based on THg concentrations and snow accumulation rates. Results from NEEM site show that THg and F_{THg} range from sub-pg g^{-1} to 120.6 pg g^{-1} (mean = 1.5 pg g^{-1} , $n = 653$) and from 0.06 to 1.42 $\mu\text{g m}^{-2} \text{year}^{-1}$ (mean = 0.25 $\mu\text{g m}^{-2} \text{year}^{-1}$, $n = 218$) respectively, much lower than those found in other natural media such as sediments, peat bogs and wet precipitation. The discrepancy of F_{THg} found in glaciers from other natural media could mainly be due to the more severe photo-reduction and reemission of deposited oxidized Hg. This study also demonstrates that reproducible THg archives can be reconstructed with glacier ice and snow samples from Greenland and the Canadian High Arctic. The THg archive reconstructed with the short core from NEEM site is so far the longest with the highest resolution in Greenland and the Canadian High Arctic.

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1. Introduction

Mercury (Hg) is currently one of the most problematic elements in the northern environment as it has significant adverse impacts on human health and the environment (AMAP, 2011). When inorganic Hg is converted into organic methyl Hg, it becomes highly toxic and able to be biomagnified through food chains. Mercury in the Arctic environment is therefore potentially of risk to people and wildlife (AMAP, 2011). To understand how much Hg in the environment comes from anthropogenic sources, and to assess Hg flux, fate and

trends now and in the future, it is desirable to understand the temporal and spatial distributions of total Hg (THg) and annual THg accumulation rates (F_{THg}) in various natural media.

Many Hg archive reconstructions have been done with sediments (Marvin et al., 2004; Muir et al., 2009), peat bogs (Biester et al., 2002; Roos-Barraclough et al., 2002; Shotyk et al., 2003; Allan et al., 2013), wet precipitation (Sanei et al., 2010) and glacier ice (Boutron et al., 1998; Schuster et al., 2002; Zheng et al., 2014). F_{THg} estimated from those archives and measurements of wet depositions ranges from less than 1/10th of $\mu\text{g m}^{-2} \text{year}^{-1}$ (Zheng et al., 2014) up to 200 $\mu\text{g m}^{-2} \text{year}^{-1}$ (Allan et al., 2013). Those values with a large range reflect the reality of differences of sites where samples were taken and media that were used for the archive reconstruction. The large difference of F_{THg} results from sediments, bogs, wet deposition

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and ice/firn cores demonstrates that F_{THg} could be site and media specified. Muir et al. (2009) found that F_{THg} decreases with increasing latitude based on results retrieved from lake sediments.

In high latitude regions, especially those areas that are covered by year-round ice and snow, Hg on surfaces of glaciers and ice caps from atmosphere deposition does not mean the deposited Hg could stay with the snow permanently. Instead, most of the deposited Hg could be photo-reduced and re-emitted to the air with a very small fraction staying buried at depth, resulting in an annual sequestration of THg. This phenomenon has been observed and confirmed by experiments in the field and in laboratory (Lalonde et al., 2003; Kirk et al., 2006; Dommergue et al., 2007; Brooks et al., 2008, 2011). Therefore, the net deposition rate of Hg or F_{THg} on glaciers could be much less, due to the specific environment conditions in the high latitude regions, than those areas where there is none or less snow or ice coverage. In other words, F_{THg} in glacier areas could be quite different from those in southern areas or those areas where snow coverage is not year-round. Ice covered area in the polar regions accounts for a significant portion of earth surface, and therefore, a precise estimation of F_{THg} in the ice covered area is desirable and needed for a sound assessment of global Hg budget and global Hg cycle.

Studies of THg and estimation of F_{THg} using ice and snow from ice caps and glaciers have been carried out by a number of researchers. Faïn et al. (2008, 2009) carried out measurements of gaseous Hg trapped in central Greenland firn and estimated that anthropogenic emissions of Hg caused a 2-fold rise in gaseous elemental mercury (GEM) concentrations before the 1970s. With two ice cores from the Upper Fremont Glacier (UFG) (USA), Schuster et al. (2002) reconstructed a non-continuous archive of 270-year atmospheric Hg deposition with 97 ice samples, and concluded a 20-fold increase of THg deposition in the mid 1980s compared to the level before the Industrial Revolution (IR). Ferrari et al. (2008) carried out a series of total Hg measurements in surface snow samples. Kirk et al. (2006) found rapid reduction and reemission of Hg to and from snow during atmospheric mercury depletion events (AMDEs). Results of THg from those studies with snow and ice samples range from sub-pg g^{-1} to over 650 pg g^{-1} (Kirk et al., 2006), while estimation of F_{THg} , from $\leq 1 \mu\text{g m}^{-2} \text{year}^{-1}$ to over 20 $\mu\text{g m}^{-2} \text{year}^{-1}$ (Schuster et al., 2002; Zhang et al., 2012). As the 270-year THg archive with two UFG ice cores (Schuster et al., 2002) was only reconstructed with 97 samples and the archive's early 100 years (1720–1820) was only represented by about 15 samples (judged from Fig. 3, Schuster et al., 2002), the archive is virtually in a low resolution. Therefore, there are virtually no long-term archives with a high resolution of THg and/or F_{THg} reconstructed with ice and snow in the world so far. This study is therefore aiming to reconstruct long-term archives of THg and F_{THg} with ice and snow samples from multi-sites of glaciers and/or ice caps in Greenland and in the Canadian High Arctic, and to investigate if THg archives are reproducible.

2. Experiment

Field sampling, sample protection, shipping, processing and analysis as well as contamination control and quality control in this study followed the methodology and protocol specifically developed for quantification of THg in ice and snow samples at Natural Resources Canada (NRCAN). Details can be found in the publication by Zheng et al. (2014). Therefore, only a brief introduction and summary of lab conditions, reagents, instrumentation and analysis are described below.

2.1. Mercury-controlled cold clean room and room-temperature clean laboratory

A cold clean room (Class-1000 with a Class-100 working station) and room temperature clean rooms (Class-1000 with all Class 100 benches and a Class-10 polypropylene fully exhausted vertical laminar flow hood, $\sim 2 \text{ m}^2$ working surface) at NRCAN have been periodically

certified by commercial companies. The Hg-controlled cold clean room was also constantly monitored with a portable particle counter, Handheld 3016 (Lighthouse Worldwide Solutions). Both the cold clean room and the room temperature clean rooms were in good conditions during this study.

All sub-sampling and/or ice/firn sample processing for THg quantification were carried out in the Hg-controlled cold clean room when necessary. During sample analysis, all THg samples were thawed and processed in the Class-10 polypropylene fully exhausted vertical laminar flow hood. The stationary THg analyzer used in this study was located on a Class-100 counter within the room temperature clean rooms.

2.2. Utensil cleaning, sample containers, reagents, and sample preparation for THg analysis

Laboratory water used in this study was produced using a reverse osmosis-de-ionized (RO-DI) Milli-Q system (18.2 M Ω Milli-Q Plus water system with Milli RO12 Plus feed, MA, USA). Only glass bottles (400-ml, Fisherbrand Clear French Squares with PTFE-Faced PE-Lined Closures) were used for this study. A simplified washing procedure was applied to the glass bottles by rinsing them with RO-DI water at least three times and letting them dry inside a Class-100 bench. Background of THg in 12 randomly selected new glass bottles, cleaned using this simplified procedure, were estimated to be $0.08 \pm 0.02 \text{ pg g}^{-1}$ ($n = 12$) based on their signal intensities, all lower than the limit of detection (LoD) of the Hg analyzer used (Zheng et al., 2014).

Utensils used in this study, including polyethylene scoops and titanium chisels used for field sampling and/or sample processing in laboratories, were all treated and cleaned following a strict ultra-clean protocol to avoid any possible contamination.

All reagents used in this study were so handled to best possibly remove potential Hg from the reagents following standard procedures. Sample thawing and preparation for analysis followed the strict NRCAN THg analysis protocol.

Measurements of laboratory blanks during each batch of sample analysis ($n = 33$) were all less than LoDs (0.2 pg g^{-1} before 2010 and less than 0.1 pg g^{-1} after January 2010) of Tekran 2600 Hg analyzer (Zheng et al., 2014).

2.3. Instrumentation, analytical method, quality control and sample analysis

Instrument used in this study was Tekran 2600 (Toronto, Canada) bench top THg analyzer. A modified U.S. EPA method 1631e for THg quantification in water was used in this study. Sodium borohydride (NaBH_4 0.15%w/v in 0.03% w/v NaOH) was used as the reducing agent instead of stannous chloride. The reason for this modification is NaBH_4 reduction is more efficient with a smaller volume of reagent required for ice and snow samples analyzed by the Tekran 2600 system. Tekran 2600 using this modified method has demonstrated to produce precise and accurate results as well as a consistent performance when analyzing blanks and NIST SRM 1641d (The National Institute of Standards and Technology Standard Reference Material 1641d). SRM 1641d was analyzed approximately every 10 samples throughout this study. All analyses of SRM 1641d were within the certificate value (certified $7.78 \pm 0.14 \text{ pg g}^{-1}$ with 200,000 times dilution, analyzed $7.81 \pm 0.07 \text{ pg g}^{-1}$, $n = 35$) (Zheng et al., 2014). The precise quantification of SRM 1641d reference material confirmed the quality and accuracy of sample analysis in this study.

To verify the stability and reproducibility of the analyzer Tekran 2600 and the modified EPA method 1631e, 59 randomly selected samples from the full set of samples were reanalyzed 16–24 h after their first analyses. The results confirm an excellent precision with the Tekran 2600 analyzer and the modified EPA method 1631e ($0.90 \pm 0.74 \text{ pg g}^{-1}$ for the first run analysis versus $0.87 \pm 0.73 \text{ pg g}^{-1}$ for the repeat

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