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## Profiles of Mercury in the snow pack at Station Nord, Greenland shortly after polar sunrise

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[1] Mercury (Hg) species have been measured in the snow pack at Station Nord, Greenland both in the snow and in the air of snow from February 25 to March 15, 2002, during twilight and low solar irradiation periods. More than 99% of Hg is in the snow itself ( $\sim 94\text{--}97\%$  as  $\text{Hg}^{2+}$  and  $\sim 5\%$  as  $\text{MeHg}^+$ ) while less than 1% is in the interstitial air of snow as  $\text{Hg}^{\circ}$ . Gaseous Elemental Mercury ( $\text{Hg}^{\circ}$ ) concentrations, decreased exponentially with depth from  $\sim 1.5 \text{ ng/m}^3$  outside to  $\sim 0.1 \text{ ng/m}^3$  at 120 cm depth in the snow air.  $\text{Hg}^{\circ}$  incorporation flux to the snow pack has been evaluated to  $\sim 5.8\text{--}7.0 \text{ pg/m}^2/\text{h}$  which is weak, indicating that this process does not change significantly the Hg content in the snow. We believe that this decrease in the air of snow is the result of fast oxidation processes of  $\text{Hg}^{\circ}$  rather than adsorption of  $\text{Hg}^{\circ}$  onto snow surfaces. *INDEX TERMS:* 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 0330 Atmospheric Composition and Structure: Geochemical cycles. **Citation:** Ferrari, C. P., A. Dommergue, C. F. Boutron, P. Jitaru, and F. C. Adams (2004), Profiles of Mercury in the snow pack at Station Nord, Greenland shortly after polar sunrise, *Geophys. Res. Lett.*, *31*, L03401, doi:10.1029/2003GL018961.

### 1. Introduction

[2] The study of the global cycle of mercury in polar environments is of great importance as i) mercury appears to be highly accumulated in the Arctic environment and biota [Wagemann *et al.*, 1996], especially in fish and sea mammals which represent a high component of native populations diet; ii) the snow pack represents an important reservoir of this toxin, as it can cover up to 50% of land masses in the Northern Hemisphere in winter.

[3] Many studies have been devoted to Atmospheric Mercury Depletion Events (AMDE) occurring after polar sunrise in the atmosphere both in the Arctic [Schroeder *et al.*, 1998] and in Antarctica [Ebinghaus *et al.*, 2002] leading to a fast deposition of Hg onto snow surfaces [Lindberg *et al.*, 2002]. The snow pack appears during these events to be

an environmental sink of mercury, as it can accumulate Hg in the first snow layers after deposition. However, it can also act as a source to the atmosphere [Dommergue *et al.*, 2003a] and to aquatic reservoir at the snowmelt [Dommergue *et al.*, 2003b]. The snow pack seems to be an active reservoir for Hg since two recent studies have pointed out a decrease of  $\text{Hg}^{\circ}$  concentration in the air of the snow in Arctic sites [Kemp *et al.*, 2000; Dommergue *et al.*, 2003a, 2003c]. In this study, we investigated the distribution of Hg speciation ( $\text{Hg}^{\circ}$  in the air of snow,  $\text{Hg}^{2+}$  and  $\text{MeHg}^+$  in the snow) in the snow pack in Station Nord, Greenland over a three weeks period shortly after polar sunrise in order to assess  $\text{Hg}^{\circ}$  incorporation flux in the snow pack.

### 2. Experimental

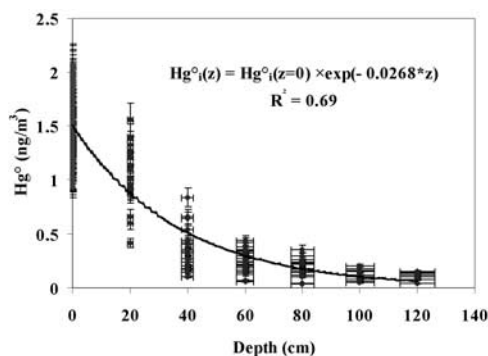
[4] Experiments were conducted in Station Nord, Greenland, a Danish military base located in the Northern-East part of Greenland ( $81^{\circ}36'09''\text{N}$ ,  $16^{\circ}40'12''\text{W}$ ) from February 25th to March 15th, 2002, on the shore of the Wandel Sea, which was totally frozen during the experimental period.

[5] The sun rose on February 26th to reach a solar zenith angle of about  $0.5^{\circ}$ . During the study period (February 25 to March 15), all the mercury measurements in the air of the snow pack were conducted during daylight hours (February 26 to March 15) or during twilight (February 25) between 9:00 AM to 5:00 PM local hours. Solar irradiation was weak and increased from a few  $\text{W/m}^2$  on February 25 to reach a maximum of  $\sim 80 \text{ W/m}^2$  on March 15.

[6] Measurements of mercury in the snow pack interstitial air were performed using GAMAS, Gaseous Mercury in Interstitial Air in Snow, coupled with a Hg vapour analyser which is a new tool dedicated to sampling and analysis of mercury and temperature measurements in the air of snow packs [Dommergue *et al.*, 2003c]. Seven probes were installed every 20 cm of depth from the surface to the bottom of the snow packs (i.e.,  $\sim 120 \text{ cm}$ ) at  $\sim 10 \text{ m}$  from the field laboratory where a Gardis 3 (Ekoservis) mercury vapour analyser and snow temperature controller were housed. The probes were connected successively to the mercury analyser with a 10 m unheated Teflon line. The sampling flow rate was 1 L/min and the volume of air sampled was between 5 and 15 litres. Reliability and performances of the analyser have already been verified through intercomparison exercises [Munthe *et al.*, 2001]. During the experiment, the Gardis 3 analyser previously calibrated had an absolute detection limit of  $0.2 \text{ pg}$  of Hg approximately. Hence, a detection limit

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**Figure 1.**  $\text{Hg}^\circ$  concentrations ( $\text{ng}/\text{m}^3$ ) in the interstitial air of snow pack for different depths.  $\text{Hg}^\circ$  concentrations measured at the surface correspond to one hour average sampling time.

around  $0.04 \text{ ng}/\text{m}^3$  is expected when measuring  $\text{Hg}^\circ$  in the air of snow. The R.S.D. between replicates was about 15%. Seventeen  $\text{Hg}^\circ$  concentration profiles in the air of snow were constructed during the experiment period, under twilight and daylight hours. For each profile,  $\text{Hg}^\circ$  concentration measurements were made above the snow and at the successive depths of 20, 40, 60, 80, 100, and 120 cm. Triplicate measurements were taken at each depth.

[7] Snow samples were collected in two snow pits dug on February 25 and March 5, 2002 near the field laboratory. One litre LDPE bottles were inserted horizontally in a clean face of a pit to collect samples at depths 0–0.10 m, 0.20–0.30 m, 0.40–0.50 m, 0.60–0.70 m, and 0.80–0.90 m. Acid clean materials were used and gloves and full clean room clothing were worn. Samples were kept frozen, in the dark, and were packed inside sealed polyethylene bags until they were melted before analysis.

[8] Inorganic mercury ( $\text{Hg}^{2+}$ ) and methylmercury ( $\text{MeHg}^+$ ) in snow pit samples were measured in the Micro and Trace Analysis Center (Antwerpen, Belgium) by multi-capillary gas chromatography (MCGC) with inductively coupled plasma time-of-flight mass spectrometry (ICP-TOFMS) described in details in *Jitaru et al.* [2003].

### 3. Results

[9] The temperature in the snow pack increased with depth from 238.7–246.7 K at 20 cm to 258.7–259.6 K at 120 cm with a variability decreasing with depth (i.e., 2.7 K at 60 cm, 1.4 K at 80 cm, 1.3 K at 100 cm and 0.7 K at 120 cm).

[10] Results of  $\text{Hg}^\circ$  measurements in interstitial air of the snow pack for the whole experimental period are given in Figure 1.  $\text{Hg}^\circ$  concentrations ranged from  $\sim 0.93$  to  $2.10 \text{ ng}/\text{m}^3$  above the snow surface, and  $\sim 0.41$  to  $1.56 \text{ ng}/\text{m}^3$  at 20 cm depth, and continuously decreased with snow pack depth to  $\sim 0.04$  to  $0.15 \text{ ng}/\text{m}^3$  at 120 cm. There is a significant range of  $\text{Hg}^\circ$  concentrations in the 0–40 cm snow pack depth interval as this interval is rapidly mixed with atmospheric air. Equation (1) was developed from the data presented in Figure 1 and describes relation between  $\text{Hg}^\circ$  in the interstitial air and depth:

$$\text{Hg}^\circ(z) = \text{Hg}^\circ(z=0) \times \exp(-0.0268 \times z) \quad (r^2 = 0.69, n = 146), \quad (1)$$

where  $\text{Hg}^\circ$  ( $\text{ng}/\text{m}^3$ ) is a function of depth ( $z$ ) in cm. When  $z = 0$ ,  $\text{Hg}^\circ$  is the  $\text{Hg}^\circ$  concentration above snow. *Dommergue et al.* [2003a] have shown in a sub-arctic site, Kuujuarapik, Québec a similar profile for  $\text{Hg}^\circ$ , which fitted approximately equation (1). Results for both  $\text{MeHg}^+$  and  $\text{Hg}^{2+}$  in snow are presented in Table 1 for two snow pits dug on February 25th and March 5th. For snow sampled during twilight on February 25th,  $\text{Hg}^{2+}$  concentrations were increasing with depth from extremely low levels (below the detection limit of the method) for snow between the surface and 25 cm depth, to  $(4.23 \pm 0.33) \text{ ng}/\text{L}$  for bottom snow.  $\text{MeHg}^+$  concentrations were 4–50 times lower and fairly constant with depth. For snow sampled on March 5th,  $\text{Hg}^{2+}$  concentrations with depth followed the same pattern as observed on February 25th, but with lower concentrations in the deepest layers. For  $\text{MeHg}^+$ , the concentrations were 5 times lower than observed for February 25th except for the upper layer of the snow pack, where  $\text{MeHg}^+$  concentrations were fairly similar.

### 4. Discussion

[11] With the results of Hg speciation in the snow derived from the two snow pits (Table 1), and the integration of equation (1) for interstitial  $\text{Hg}^\circ$ , we can estimate that  $1 \text{ m}^3$  of snow (with an average density of 0.25) contained  $\sim 95 \text{ pg}$  of interstitial  $\text{Hg}^\circ$ ,  $\sim (8\text{--}50) \text{ ng}$  of  $\text{MeHg}^+$  and  $\sim (150\text{--}1550) \text{ ng}$  of  $\text{Hg}^{2+}$ . Hg in the snow pack consisted mainly of  $\text{Hg}^{2+}$  in the snow ( $\sim 94\text{--}97\%$ ) with a contribution of  $\text{MeHg}^+$  of  $\sim 2\text{--}5\%$  while interstitial  $\text{Hg}^\circ$  represented less than  $\sim 0.01\%$ . Incorporation flux of Hg in the snow pack, neglecting the ventilation by wind, can be calculated using the following equation:

$$F = -K_d \times \left( \frac{\partial C}{\partial z} \right)$$

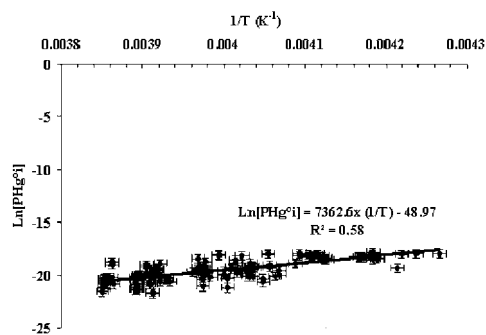
where  $\left( \frac{\partial C}{\partial z} \right)$  is the concentration gradient for  $\text{Hg}^\circ$  between the air above snow and the interstitial air at a depth  $z$  (in our case given by equation (1)), and  $K_d$  the  $\text{Hg}^\circ$  molecular diffusivity in snow ( $\sim 1.0\text{--}1.2 \times 10^{-5} \text{ m}^2 \cdot \text{s}^{-1}$ ) given by *Dommergue et al.* [2003b]. The calculated incorporation flux of  $\text{Hg}^\circ$  in the snow pack was  $\sim 5.8\text{--}7.0 \text{ pg}/\text{m}^2/\text{h}$  which is extremely low ( $\sim 1000$  fold) related to the emission fluxes measured or calculated for sub-arctic and arctic sites (i.e.,  $\sim 1.5\text{--}2.5 \text{ ng}/\text{m}^2/\text{h}$  by *Dommergue et al.* [2003b],  $\sim 1\text{--}8 \text{ ng}/\text{m}^2/\text{h}$  by *Schroeder et al.* [2003]). This very weak value of flux implies that incorporation of atmospheric  $\text{Hg}^\circ$  to the snow pack was not an important sink of Hg in Station Nord.

**Table 1.** Speciation of Hg in Two Snow Pits Dug in February 25 and March 5

Depth (cm)	February 25, 2002		March 5, 2002	
	$\text{Hg}^{2+}$ (ng/L)	$\text{MeHg}^+$ (ng/L)	$\text{Hg}^{2+}$ (ng/L)	$\text{MeHg}^+$ (ng/L)
5	<L.D. <sup>a</sup>	$0.11 \pm 0.02$	n.d. <sup>b</sup>	$0.10 \pm 0.03$
25	$0.38 \pm 0.24$	$0.09 \pm 0.01$	$0.62 \pm 0.09$	$0.02 \pm 0.01$
45	$1.67 \pm 0.09$	$0.11 \pm 0.01$	$0.48 \pm 0.08$	$0.02 \pm 0.01$
65	$4.96 \pm 0.43$	$0.13 \pm 0.01$	$1.98 \pm 0.15$	$0.02 \pm 0.01$
85	$4.23 \pm 0.33$	$0.07 \pm 0.24$	$2.53 \pm 0.03$	n.d. <sup>b</sup>

<sup>a</sup>Below detection limit.

<sup>b</sup>Not determined.



**Figure 2.** Relation between  $\text{Ln} [P(\text{Hg}^\circ)]$  in interstitial air of snow and  $1/T$  ( $\text{K}^{-1}$ ) for the characterisation of  $\text{Hg}^\circ$  adsorption onto snow.

Consequently, with respect to Hg partition between snow and air and the resulting incorporation flux, we believe that variations of  $\text{Hg}^\circ$  concentration in the air of snow can not significantly affect the inorganic content of the snow over a short period. Furthermore, no direct input of  $\text{Hg}(\text{II})$  species in the reactive form or associated to particles to the snow pack has been pointed out since no AMDE's were observed during the experimental period. One can notice that the concentrations of both  $\text{Hg}^{2+}$  and  $\text{MeHg}^+$  in the snow (Table 1) decreased in 10 days after polar sunrise. This difference can be the result of spatial variability in concentrations, and/or difference in Hg content in snow layers deposited since the beginning of winter. For example, for 4 surface snow samples collected at the same time in the vicinity of the field laboratory,  $\text{Hg}^{2+}$  and  $\text{MeHg}^+$  concentrations were about (0.77–2.92) ng/L and (0.05–0.11) ng/L respectively. If the observed decrease in concentrations in the snow was not related to the spatial and historical variability, possible reductions processes of  $\text{Hg}^{2+}$  and  $\text{MeHg}^+$  have to be investigated.

[12] This decrease of  $\text{Hg}^\circ$  concentration in the interstitial air of the snow pack with depth imply either an adsorption of  $\text{Hg}^\circ$  onto the snow grains or homogeneous and/or heterogeneous transformations of  $\text{Hg}^\circ$  in the snow pack. The depletion of  $\text{Hg}^\circ$  inside the snow pack is probably driven by a fast process leading to a life time of  $\text{Hg}^\circ$  in the pack less than  $\sim 7$  hours.

[13] Indeed, using the rate of air diffusion in a snow-pack [Tang and McConnel, 1996] of  $\sim 18$  cm/h, we estimate the vertical diffusion of  $\text{Hg}^\circ$  from the snow surface to the bottom of the snow pack at Station Nord could last  $\sim 7$  hours. This value does not take into account the ventilation of the snow pack that increases the rate of transfer of chemical species in the snow pack. Consequently, the rate of transfer is probably less than  $\sim 7$  hours. Looking at Figure 1, we can estimate that the lifetime of  $\text{Hg}^\circ$  is probably lower ( $\sim 2$ – $3$  hours) as  $2/3$  of  $\text{Hg}^\circ$  concentration is already depleted at 40–60 cm depth.

[14] A possible explanation could be first the adsorption of  $\text{Hg}^\circ$  on snow grains in the snow pack. The adsorption depends on the concentration and partial pressure of  $\text{Hg}^\circ$  in the air of snow, the snow density ( $d$ ), the Specific Surface Area (S.S.A.) of the snow, the height of the snow column ( $h$ ) and the temperature ( $T$ ) of the snow. We can estimate roughly that the quantity of  $\text{Hg}^\circ$  adsorbed on a surface

(i.e., snow in our case),  $Q(\text{Hg}^\circ)_{\text{ads}}$ , is given by the following equation:

$$Q(\text{Hg}^\circ)_{\text{ads}} = P(\text{Hg}^\circ)_{\text{int}} \times S.S.A. \times d \times h \times \exp(-E/R \times T), \quad (2)$$

where  $P(\text{Hg}^\circ)_{\text{int}}$  is the partial pressure of  $\text{Hg}^\circ$  in the air of snow,  $E$  is the energy of adsorption and  $R = 8.32 \text{ J.K}^{-1}.\text{mol}^{-1}$  (ideal gas law constant). In Figure 2, a linear relation is obtained between  $\text{Ln}[P(\text{Hg}^\circ)_{\text{int}}]$  and  $1/T$  with a slope of  $\sim 7362.6$  and a value of  $-48.97$  for the intersection of the regression line and the Y axis. The slope allows us to reach the adsorption energy of  $\text{Hg}^\circ$  on snow calculated to be  $\sim 61 \text{ kJ.mol}^{-1}$ . This energy is closed to the energy of  $\text{Hg}^\circ$  volatilisation calculated with thermodynamical data and also the energy of  $\text{Hg}^\circ$  volatilisation from soils obtained with laboratory flux chamber [Bahlmann *et al.*, 2002, personal communication]. Taking into account the snow pack thickness of 120 cm, an average density of snow around  $0.25 \text{ g/cm}^3$ , an average S.S.A. for polar snow of  $500 \text{ cm}^2/\text{g}$  [Cabanes *et al.*, 2002], we obtain  $Q(\text{Hg}^\circ)_{\text{ads}} \sim 9.5 \times 10^{-23} \text{ Pa}$  in the Station Nord snow pack. The partial pressure of  $\text{Hg}^\circ$  in the air of a layer at 40 cm depth and 253 K is estimated to be  $\sim (1 \times 10^{-13} - 1 \times 10^{-14}) \text{ Pa}$ . The ratio between  $\text{Hg}^\circ$  adsorbed on snow and gaseous  $\text{Hg}^\circ$  in interstitial air in equilibrium clearly shows that  $\text{Hg}^\circ$  is weakly adsorbed on snow. This calculation is supported by the laboratory tests made by Bartels-Rausch *et al.* [2002] on  $\text{Hg}^\circ$  adsorption on crystalline ice indicating that reversible adsorption is the main process governing migration of  $\text{Hg}^\circ$  along the ice surface. Such a weak interaction of  $\text{Hg}^\circ$  with ice could be explained as this metallic compound is not capable to form hydrogen bonds.

[15] Then, a fast chemical reaction leading to the oxidation of  $\text{Hg}^\circ$  into  $\text{Hg}(\text{II})$  could then be at the origin of such  $\text{Hg}^\circ$  decrease in the snow pack. Atmospheric chemistry of  $\text{Hg}^\circ$  with current oxidants like ozone, OH radicals, cannot explain this fast process [Bergan and Rodhe, 2001].

[16] Interestingly, Albert *et al.* [2002] observed in Alert (Canada) for quite similar snow pack and light conditions (twilight and low light), a rapid depletion of ozone in the air of the snow. They suggested two mechanisms to explain this decrease based on a fast chemical reactivity of ozone in snow. The first mechanism was related to halogen species chemistry. Spicer *et al.* [2002] estimated that 1 pptv of halogen species could be responsible of the depletion of 1 ppbv of ozone. In polar atmospheres, simultaneous fast depletions of ozone and  $\text{Hg}^\circ$  have also been reported in spring [Schroeder *et al.*, 1998]. Chemical reactions involving halogen species (especially  $\text{Br}^\bullet$  and  $\text{BrO}^\bullet$ ) have been proposed to explain these concurrent depletions [Lindberg *et al.*, 2002]. The second mechanism that could deplete ozone in the air of snow could be the transformation of ozone into OH radicals onto black carbon surfaces contained into the snow deposited after long range transport of anthropogenic black carbon [Albert *et al.*, 2002]. As the kinetic constant between OH and  $\text{Hg}^\circ$  is low [Bergan and Rodhe, 2001], reaction between  $\text{Hg}^\circ$  and OH cannot explain the observed fast depletion of  $\text{Hg}^\circ$  in the snow. The reaction between  $\text{Hg}^\circ$  and  $\text{Br}^\bullet$  and  $\text{BrO}^\bullet$  is known to be fast in gaseous phase [Arya *et al.*, 2002]. Moreover, snow could

increase the rate of these reactions owing to the catalytic properties of snow surfaces [Ocampo *et al.*, 1987].

## 5. Summary and Conclusion

[17] Measurements of  $\text{Hg}^{\circ}$  concentrations in the snow show a decrease as a function of depth, indicating that the snow pack could then be a sink for  $\text{Hg}^{\circ}$  shortly after polar sunrise. The calculated incorporation flux of  $\text{Hg}^{\circ}$  in the snow pack, associated to this sink, is  $\sim 5.8\text{--}7.0\text{ pg/m}^2/\text{h}$ , which is weak minimizing the potential impact of this flux on  $\text{Hg(II)}$  snow content. Similarly with ozone depletion in the air of snow as well as during atmospheric ozone and Hg depletion events, we suggest that a specific chemistry involving halogen species in polar regions could be at the origin of a fast oxidation of  $\text{Hg}^{\circ}$  in the snow pack.

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