

## The fate of mercury species in a sub-arctic snowpack during snowmelt

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### Abstract

An extensive mercury study was conducted in April 2002 prior to and during the annual melting of a snowpack in a sub-arctic site along the Hudson Bay (Canada). Gas-phase measurements show that the snowmelt coincides with an elemental mercury ( $\text{Hg}^{\circ}$ ) pulse in the snowpack air far above ambient levels. Additional measurements of inorganic mercury ( $\text{Hg}^{2+}$ ) and methylmercury ( $\text{MeHg}^{+}$ ) in snow pits, in surface snow and in a meltwater sample clearly reveal that most of Hg is removed from the snow during the first days of snowmelt. We estimate that gas-phase exchanges contribute poorly to remove Hg from the snowpack; consequently during a snowmelt day more than 90% of Hg present in the snow surface is likely released with the meltwater. In arctic areas, where Hg accumulates at an accelerated rate in the snow surfaces [Lu *et al.*, 2001] during mercury depletion events (MDE), the

discharge of this toxic and bio-accumulating pollutant in water systems could be a threat to ecosystems and local indigenous populations.

## **Introduction**

Contamination of arctic ecosystems is one of the central questions dealing with Mercury Depletion Events (MDE). Annually, at the polar sunrise elemental mercury ( $\text{Hg}^0$ ) is depleted in the atmosphere of polar and sub-polar regions [Schroeder *et al.*, 1998].  $\text{Hg}^0$  is rapidly oxidized to reactive divalent species and is subsequently deposited onto snow surfaces in a bioavailable form. Environmental implications of MDE are broad since large amounts of Hg could enter the arctic ecosystem via snowmelt water. Fish and marine mammals and consequently indigenous populations living of marine and freshwater resources could be exposed to this bioaccumulating and toxic compound.

It has been suggested recently that Hg is chemically active in the snowpack and could be released into the atmosphere via a sunlight-induced mechanism [Lalonde *et al.*, 2002]. During snowmelt, it is not clear whether mercury is incorporated in meltwater or reemitted to the atmosphere. Therefore, we investigated the fate of mercury species in a sub-arctic snowpack at the onset of annual snowmelt.

## **Experimental**

The studies were performed at Kuujjuarapik/Whapmagoostui (55°17'N, 77°46'W) on the east shore of the Hudson Bay (Canada) in April 2002. The Gaseous Mercury in interstitial Air in Snow (GAMAS) coupled with a Gardis-3 Hg analyzer is a new device for sampling and analysis of  $\text{Hg}^0$  in the air of snowpacks [Dommergue *et al.*, 2003a]. It was applied to continuously monitor temperatures and  $\text{Hg}^0$  in the air of a 1.02-m thick snowpack at depths of 0.25, 0.37, 0.54, 0.80 and 1.02 m successively. Five liters (STP) of air and a 1 l min<sup>-1</sup> flow

rate were used allowing a detection limit of about  $0.04 \text{ ng m}^{-3}$ . Uncertainty ( $1\sigma$ ) among replicates taken from the same depths was around 15%. Atmospheric  $\text{Hg}^{\circ}$  was measured at 2 m height with a Tekran 2537A analyzer operating with a 15-min sampling interval and a flow rate of  $1.5 \text{ l min}^{-1}$ .

Duplicate samples of surface snow were collected daily at 8 AM and 8 PM in 1 l Teflon bottles. On the nights of April 7 and 14, samples were collected in two snow pits in 1 l LDPE bottles for depths of approximately 0.20, 0.40 and 0.60 m ( $\pm 5 \text{ cm}$ ). For both sampling methods, acid-cleaned materials were used and gloves and full clean room clothing were worn. Samples were packed inside sealed polyethylene bags and were kept frozen in the dark until melted before analysis. An attempt was made to collect meltwater using an acid-cleaned  $0.40\text{m} \times 0.40\text{m}$  Teflon pan, inserted horizontally at a depth of 10 cm. The outflow was carried in a Teflon tube by gravity to a 1 l LDPE bottle.

Total Hg ( $\text{Hg}_T$ ) in surface snow was measured at Environment Canada (Montréal, Canada) using preconcentration by amalgamation of  $\text{Hg}^{\circ}$  on a gold trap followed by its detection using atomic fluorescence. The sample was prepared by dissociating Hg complexes using  $\text{BrCl}$  and reducing  $\text{Hg(II)}$  to  $\text{Hg}^{\circ}$  using  $\text{SnCl}_2$ . A detection limit of  $54 \text{ pg l}^{-1}$  (3 times the standard deviation) was achieved.

Inorganic ( $\text{Hg}^{2+}$ ) and methylmercury ( $\text{MeHg}^+$ ) in pit samples and in the meltwater were measured at the Micro and Trace Analysis Center (Antwerpen, Belgium) by multicapillary gas chromatography (MCGC) with inductively coupled plasma time-of-flight mass spectrometry (ICP-TOFMS). A purge-and-trap injection (PTI) system was used for the *in situ* derivatization, extraction and injection. Propylmercury was used as internal standard. Detection limits of  $16 \text{ fg g}^{-1}$  (as Hg) and  $256 \text{ fg g}^{-1}$  were obtained for  $\text{MeHg}^+$  and  $\text{Hg}^{2+}$  respectively. The analytical precision (RSD) for ten successive injections of a standard mixture containing  $10 \text{ pg MeHg}^+$  and  $\text{Hg}^{2+}$  was 1.2% for  $\text{MeHg}^+$  and 4.1% for  $\text{Hg}^{2+}$ .

## Results

Figures 1a, 1b and 1c show net global radiation and temperatures inside the snowpack on April 11-13, 2002. Sunrise occurred at 6:30 AM (local time) and sunset at 8 PM with a maximum irradiation at about 3 PM. On April 11 and the three previous days (not shown here, see [Dommergue *et al.*, 2003b]) snowpack temperatures were below 0°C without any diurnal pattern except at the 0.25 m depth. On April 12, melting occurred in the entire snowpack. Between 1 and 3 PM, temperatures in the first 60 cm rapidly reached 0°C. The temperature measured at the bottom of the pack (1.02 m) reached 0°C around 6 PM. The temperature at 0.80 m remained unchanged, but started to increase the following day and reached 0°C on April 14 (not shown). Because we recorded temperature using a small probe, an isolated hard layer of snow (*e.g.* a refrozen meltlayer) could have temporally insulated the probe from the surrounding layers of melting snow. All temperatures (except for depth 0.80 m) were at 0°C on April 13.

Gas-phase Hg<sup>0</sup> measurements are shown in figures 1d and 1e. Prior to the melting (on April 11 and previous days), Hg<sup>0</sup> production was occurring in the first layers of the snowpack as evidenced by Hg<sup>0</sup> concentrations above ambient levels. These measurements exhibited a well-marked diurnal cycle following variations in solar radiations. Given that Hg<sup>0</sup> desorption is negligible and soil emissions of Hg were insignificant in our field conditions, photoreduction and/or reduction of Hg(II) complexes by unidentified photochemically produced compounds may explain the Hg<sup>0</sup> cycle [Dommergue *et al.*, 2003b]. For these days, Hg<sup>0</sup> was below 5 ng m<sup>-3</sup> in the first three layers and below 1 ng m<sup>-3</sup> in the two bottom layers. On the first day of melting (April 12), maximum Hg<sup>0</sup> concentrations were 4-6 times greater than on the previous days. Hg<sup>0</sup> drastically increased to 18-20 ng m<sup>-3</sup> in the first 40 cm and to ~8 ng m<sup>-3</sup> at 0.54 m. Such high values have never been observed in arctic and sub-arctic

atmospheres. Simultaneously, ambient  $\text{Hg}^\circ$  showed higher concentrations around  $4 \text{ ng m}^{-3}$ . At depths 0.80 and 1.02 m,  $\text{Hg}^\circ$  increased above atmospheric levels, however experiments were stopped due to technical problems. At all depths, the highest  $\text{Hg}^\circ$  concentrations were recorded subsequently the temperature jumps, suggesting that the melting is partly responsible for enhanced  $\text{Hg}^\circ$  production. On April 13,  $\text{Hg}^\circ$  concentrations in the first 60 cm of the snowpack were lower, but still twice as high as on non-melting days (7-11 April).

Time series of  $\text{Hg}_T$  concentration in surface snow are shown on figure 2a. It is notable that the largest  $\text{Hg}_T$  difference between morning and evening samples was observed on April 12 when concentration dropped from  $17.2 \text{ ng l}^{-1}$  down to  $2.3 \text{ ng l}^{-1}$ . On this day, concentrations of  $\text{Hg}^{2+}$  and  $\text{MeHg}^+$  were  $11.9 \text{ ng l}^{-1}$  and  $0.09 \text{ ng l}^{-1}$  respectively in a meltwater sample collected inside the snowpack (depth 10 cm). One can notice the significant increase in the Hg concentration in surface snow from April 9 to April 11. A mercury depletion event (see time series of ambient  $\text{Hg}^\circ$  in Fig. 1d) occurred from April 10 (noon) to the morning of April 11. MDE are known to convert rapidly  $\text{Hg}^\circ$  into divalent Hg, which is subsequently deposited onto snow surfaces. The Hg content of the snow should then increase. Before the MDE, it is not clear where does this higher Hg content come from. The presence of dry deposited particulate matter of natural or anthropogenic origin may explain that. Snow pits constructed on April 6 and 14 (Fig. 2b) showed similar  $\text{Hg}_T$  content at a depth of 0.6 m whereas  $\text{Hg}_T$  concentration varied significantly between the two pits at the 0.2 and 0.4 m depths. At these depths, the  $\text{Hg}^{2+}$  concentration decreased from  $13.1\text{-}15.4 \text{ ng l}^{-1}$  prior to any melting events to  $1.5\text{-}3.6 \text{ ng l}^{-1}$ . For these pits,  $\text{MeHg}^+$  concentrations were in the range of  $0.04\text{-}0.12 \text{ ng l}^{-1}$ .

Using these data, we estimate that prior to the melting (on April 6), the top 40 cm of the snowpack (snow density  $0.3\text{-}0.4 \text{ g cm}^{-3}$ ) contained roughly  $1.7\text{-}2.3 \text{ } \mu\text{g m}^{-2}$  of  $\text{Hg}_T$  ( $10\text{-}50 \text{ ng m}^{-2}$  of  $\text{MeHg}^+$ ). In the most extreme case (*i.e.* using maximum  $\text{Hg}^\circ$  concentration of about  $20 \text{ ng m}^{-3}$  recorded on April 12),  $4\text{-}6 \text{ ng m}^{-2}$  of  $\text{Hg}^\circ$  were present in the same layer, that is less

than 1% of the whole Hg content. On April 14 and after the first day of melting, 0.5-0.9  $\mu\text{g m}^{-2}$  of  $\text{Hg}_T$  were still present in the snowpack. Thus, according to the two pits experiments, 50-70% of  $\text{Hg}_T$  was removed from the snowpack. This estimate is a lower value since it does not really take into account the increase of  $\text{Hg}_T$  concentration in the snow surface mentioned above. Estimates using surface snow data from the first day of melting (April 12) show that 70-90% (0.5-1.1  $\mu\text{g m}^{-2}$ ) of  $\text{Hg}_T$  was lost in the top 10 cm of the snowpack on that day.

## Discussion

$\text{Hg}_T$  measurements in snow (Fig.2) show evidence that the first day of snowmelt significantly affected the Hg balance in the snowpack. As the snowpack warms, there is a growth in the size of the snow grains, resulting in a decrease of the specific surface area available for sorption processes. The thickness of the liquid-like region at the ice air interface, where most of the chemical reactions are believed to occur, increases. Once the snow is warmed to 0°C, liquid water is increasingly present around residual ice grains inducing some changes in the physical properties of the snowpack. During snow precipitation and formation, trace compounds are incorporated inside snow particles. Because ionic solutes are segregated toward the exterior of the snow grain during snow formation and metamorphism [Harrington and Bales, 1998] a large fraction of ionic species is released in the earliest fraction of meltwater (the so-called “ionic pulse”). We believe that Hg species are released from the snowpack via these processes at the onset of snowmelt. Due to their different properties,  $\text{Hg}^0$  should be released rapidly to the air and  $\text{Hg}^{2+}$  to the liquid water of the snowpack. Data on the solubility and solid state diffusion coefficient of Hg in ice are missing; thus we are not able to assess the magnitude of this supposed release. During snowmelt, the reservoir of  $\text{Hg}^{2+}$  present in the liquid water is certainly larger than on previous days. Consequently,  $\text{Hg}^0$  released by photoreduction and/or reduction by photochemically produced compounds (such as  $\text{HO}_2$

radicals) [Dommergue *et al.*, 2003b] should be greater. Photochemical processes may occur deeper within the snowpack as a result of enhanced penetration of light. Additionally, reduction should take place in the aqueous phase with subsequently higher kinetic rates. Gas-phase measurements in the air of the snowpack and the Hg<sup>0</sup> pulse observed the first day of melting support these hypotheses. By estimating the slope of the rising part of the Hg<sup>0</sup> time series inside the snow-pack (Fig. 1d), we can approximate Hg<sup>0</sup> production rates prior to the melting as  $1\text{-}2\cdot 10^2$ ,  $3\cdot 10^2$  and  $2\cdot 10^2$  molec cm<sup>-3</sup> s<sup>-1</sup> at depths of 0.25, 0.37 and 0.54 m respectively. For this calculation, we assume that there is no diffusion from one layer to another. At the onset of snowmelt (April 12), these rates suddenly increased from  $2\cdot 10^2$ ,  $3\cdot 10^2$  and  $1\cdot 10^2$  molec cm<sup>-3</sup> s<sup>-1</sup> (estimation derived from the first hours of sunlight to ~10:30 AM) to  $2\cdot 10^3$ ,  $3\cdot 10^3$  and  $1\cdot 10^3$  molec cm<sup>-3</sup> s<sup>-1</sup> at depths of 0.25, 0.37 and 0.54 m respectively. Because the Hg<sup>0</sup> production rates were at least 10 times greater than prior to snowmelt and Hg<sup>0</sup> concentrations in the air of the snowpack were high during snowmelt, we believe that the snowmelt is at the origin of an enhanced Hg<sup>0</sup> production. Hg<sup>0</sup> is poorly adsorbed onto snow surfaces [Ferrari *et al.*, in review] and should be poorly dissolved in ice due to its weak solubility in water. Nevertheless, it is not clear whether Hg<sup>0</sup> is directly released by physical processes or is the by-product of photoreduction and/or reduction of Hg(II) complexes released in aqueous phase at the onset of snowmelt.

Hg<sup>0</sup> production and Hg<sup>0</sup> pulse in the air of the snowpack at the onset of snowmelt raise two questions: does this pulse influence the atmospheric budget of Hg, and does it explain the observed removal of Hg<sub>T</sub> from the snow? As an upper boundary, the concentration gradient of Hg<sup>0</sup> in the first 40 cm of the snowpack (where the production is the strongest) is 43-56 ng m<sup>-3</sup> m<sup>-1</sup>. Using the molecular diffusivity of Hg<sup>0</sup> in air [Massman, 1999], a snow density around 0.3-0.4 g cm<sup>-3</sup> and equations of gas diffusivity in firn [Fabre *et al.*, 2000], we derive the Hg<sup>0</sup> molecular diffusivity in the snow of about  $1.0\text{-}1.2\cdot 10^{-5}$  m<sup>2</sup> s<sup>-1</sup> and maximum Hg<sup>0</sup> fluxes of 1.5-

2.5 ng m<sup>-2</sup> h<sup>-1</sup> for the first day of snowmelt. True values are certainly higher because we neglect ventilation by wind with a turbulent diffusivity in the range 10<sup>-3</sup> to 1 m<sup>2</sup> s<sup>-1</sup> [Hutterli *et al.*, 1999]. Snow-to-atmosphere flux measurements performed using a dynamic flux chamber [Poissant and Casimir, 1998] 200 m from our measurement site show a maximum value of approximately 6.5 ng m<sup>-2</sup> h<sup>-1</sup> on the same day [Poissant, unpublished data]. Though the two values are comparable, we caution that because of the different sites location many parameters such as wind speed, irradiation, chemical and physical characteristics of the snowpack could have been different. We will thus use both values in the following discussion. By integrating fluxes over the snowmelt day, we estimate an Hg<sup>o</sup> flux of 10-35 ng m<sup>-2</sup> day<sup>-1</sup> on April 12. If affected by a similar snowmelt day, a surface as large as the Hudson Bay (~5\*10<sup>11</sup> m<sup>2</sup>) should emit a pulse of 5-17 kg of Hg<sup>o</sup> into the atmosphere. In some arctic locations, continuous snowmelt and higher Hg<sup>o</sup> fluxes seem to directly affect ambient Hg<sup>o</sup> levels [Lindberg *et al.*, 2001]. Though we registered higher ambient Hg<sup>o</sup> values on April 12, the relationship between snowmelt and the increase of ambient Hg<sup>o</sup> is here only speculative. Even if observed Hg<sup>o</sup> levels in snowpack air are several times higher than ambient ones, resulting fluxes are weak. Thus, the Hg<sup>o</sup> pulse in the snowpack seems to weakly impact the atmosphere above Kuujjuarapik. However, if frequent snowmelt days occur until the disappearance of the snowpack, this impact should be re-evaluated.

Between the two snow pits experiments (8 days), we estimate, by integrating the calculated and measured fluxes over that period, that at maximum 40-100 ng m<sup>-2</sup> of Hg<sup>o</sup> were emitted from the snowpack. Such emission fluxes explain only 3-13% of Hg removal as inferred by snow pits data. These values are really an upper limit because we do not consider the fact that the snowpack can be sometimes a sink for Hg<sup>o</sup> during nighttimes. During that time, ambient Hg<sup>o</sup> can be higher than the snowpack air concentration resulting in a downward flux of Hg<sup>o</sup> into the snowpack air. Similarly, Hg<sub>T</sub> removal in surface snow on April 12 is not explained by

the  $\text{Hg}^\circ$  pulse which comprised a maximum release of  $35 \text{ ng m}^{-2}$  that is less than 7% of Hg loss from the snow. Therefore,  $\text{Hg}^\circ$  evasion from the snowpack is not an important pathway explaining Hg depletion in snow. Instead, Hg likely leaves the snowpack with the meltwater. A 150 ml volume of meltwater collected inside the snowpack at 10 cm had  $\text{Hg}^{2+}$  concentration of  $11.9 \text{ ng l}^{-1}$  corresponding to a loss of  $\sim 11 \text{ ng m}^{-2}$  of  $\text{Hg}_T$  at a 10 cm depth. Because our water sampling system was certainly not properly designed or placed in the snowpack to collect water that can move in preferential channels, we missed a large volume of meltwater. Thus, the Hg concentration is not representative of Hg loss from the snowpack.

### **Summary and conclusion**

Our studies of Hg species in the snowpack along the Hudson Bay at the onset of snowmelt in April 2002 have revealed that the first day of snowmelt coincides with an  $\text{Hg}^\circ$  pulse in the air of the snowpack and a removal of  $\text{Hg}_T$  from surface snow and in the first 40 cm of the snowpack. Several lines of evidence support the hypothesis that  $\text{Hg}^\circ$  evasion from the snowpack is not a predominant pathway in Hg removal from snow. Consequently most of the Hg present in snow is being transported in meltwater and runoff waters. If exposed to a similar snowmelt day, the top 10 cm of a surface as large as the Hudson bay could discharge 250-550 kg of Hg into meltwater. As inferred from pits data, the discharge could be in the range of 400-700 kg in 8 days. Moreover, a few hundreds of kg of Hg could be further discharged in water systems when the snowpack totally melts. Hg discharge could be even greater in arctic regions severely impacted by mercury depletion events showing higher Hg concentration in surface snow [Lu *et al.*, 2001]. From our observations, we suggest that the beginning of annual snowmelt initiates a massive influx of a bioaccumulating and toxic pollutant to water systems, leading to a potentially alarming contamination of ecosystems and populations.

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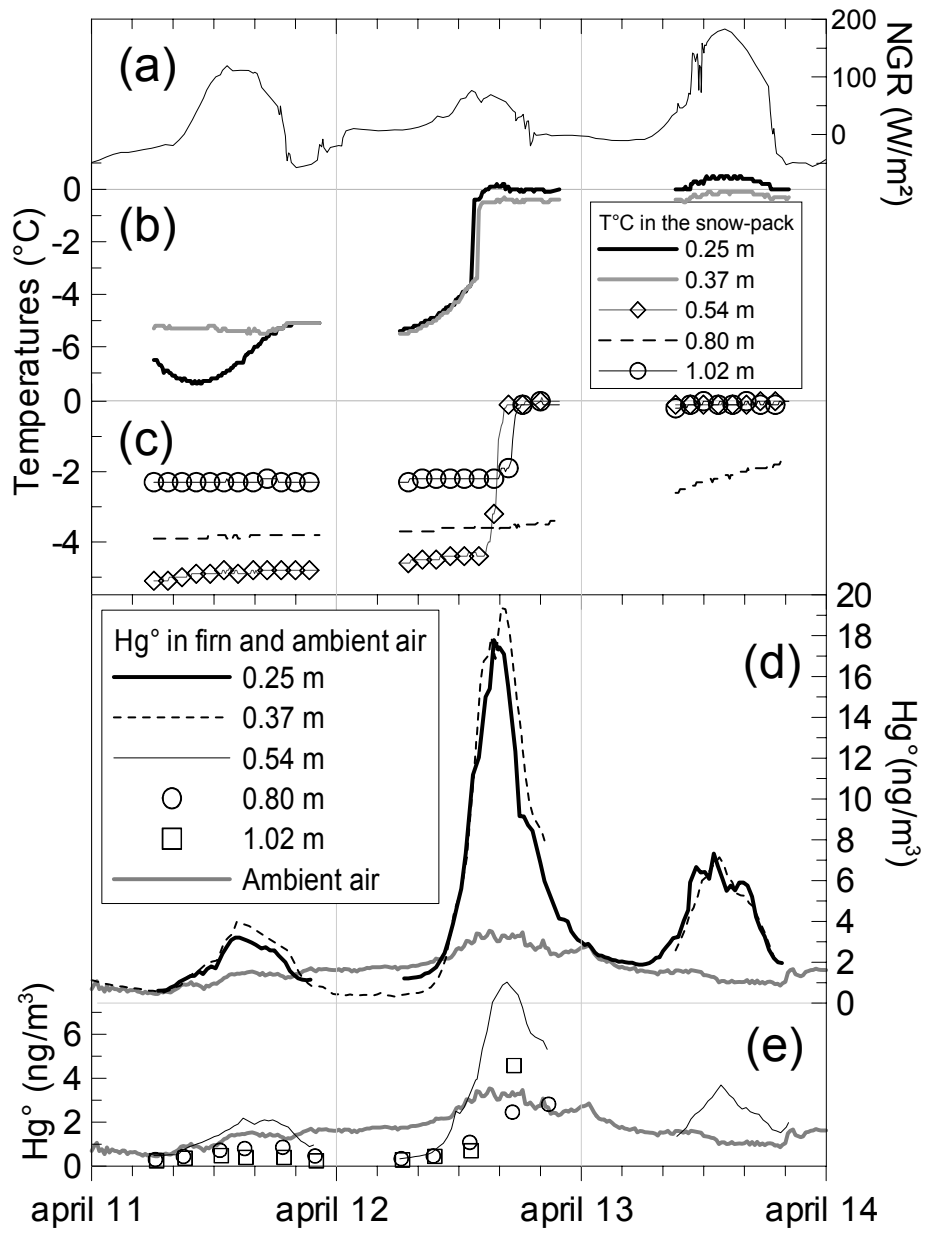
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**Figure 1:** Time series of (a) Net Global Radiation NGR ( $\text{W m}^{-2}$ ) and snowpack temperature ( $^{\circ}\text{C}$ ) at the depths of (b) 0.25 and 0.37 m and of (c) 0.54, 0.80 and 1.02 m. Time series of  $\text{Hg}^{\circ}$  concentration ( $\text{ng m}^{-3}$ ) in the snowpack air at the depths of (d) 0.25 and 0.37 m and of (e) 0.54, 0.80 and 1.02 m on April 11-13, 2003. Ambient  $\text{Hg}^{\circ}$  is shown on graphs (d) and (e).

**Figure 2:** Concentrations of (a) total Hg ( $\text{ng l}^{-1}$ ) in the surface snow (6-14 April, 2003) and in a meltwater sample (April 12) and of (b)  $\text{Hg}_T$  and methylmercury ( $\text{ng l}^{-1}$ ) in snow pits constructed on April 6 and 14. The estimated thickness of the sampled snow layer is shown by the bar width. A sample from the first pit (depth 40 cm) was below the detection limit for  $\text{MeHg}^+$ .

FIGURE 1



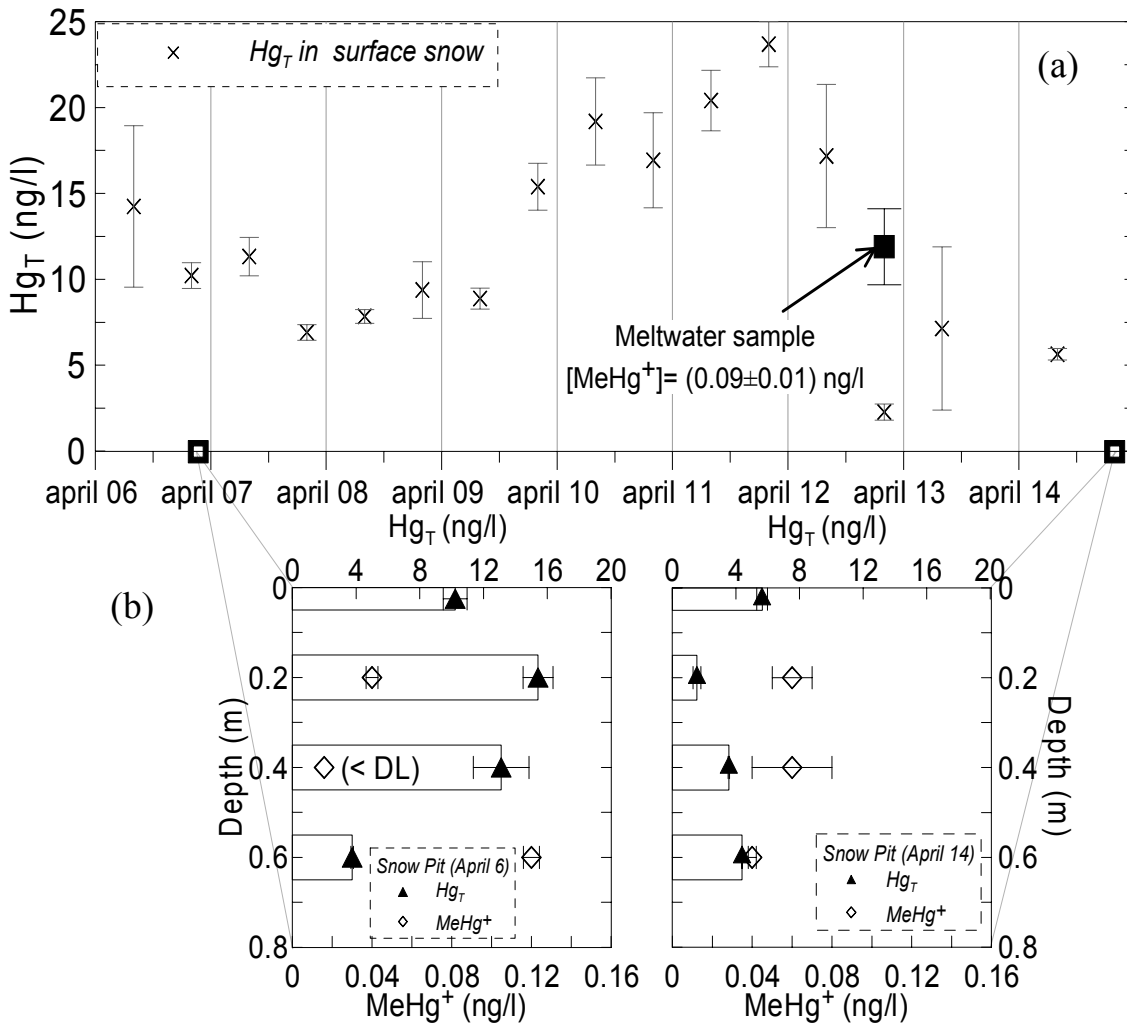


FIGURE 2