Mercury storage in sub-arctic lake sediments in Stordalen Mire, Abisko Sweden

M.F. Fahnestock[1], L. Erickson[2], M. Wik[3], J.P. DeStasio[1], M.J. Halloran[2], J.B. Setera[4], C.K. McCalley[5], J.G. Bryce[1], R.K. Varner[1,6], J. E. Johnson[1], P. M. Crill[3]

[1]Department of Earth Sciences, University of New Hampshire, Durham, NH; [2]Northern Ecosystem Research for Undergraduates, University of New Hampshire, Durham, NH;
[3]Department of Geological Sciences, Stockholm University, Stockholm, Sweden; [4]Department of Earth and Planetary Science, Rutgers School of Arts and Sciences, New Brunswick, NJ;
[5]Thomas H. Gosnell School of Life Sciences, Rochester Institute of Technology, Rochester NY; [6]Institute for the Study of Earth, Oceans and Space, University of New Hampshire, Durham, NH, USA

MOTIVATION

- Warming temperatures in the Abisko region over the past decades are correlated with increased active layer depth and shifts in timing of lake freeze and ice break-up.[1]
- The loss of permafrost in Stordalen Mire drives palsa degradation, expansion of thaw ponds and shorter ice-covered seasons.[2,3] Previous studies have reported significant loss of carbon to the atmosphere and hydrosphere in the form of carbon dioxide (CO_2) and methane (CH_4).
- Due to its strong affinity for carbon, previously sequestered mercury (Hg) in thawing permafrost can be released to the atmosphere and hydrosphere.[4,5]
- We evaluate the amount of Hg stored in lake sediments in three sub-arctic lakes, to further our understanding of the fate of Hg in an ecosystem evolving due to changes in the carbon storage.

STUDY SITE & APPROACH

- Stordalen Mire (68°21'N, 19°02'E) is located 11 km east of Abisko, Sweden. The mire is 350 m above sea level.
- Mean annual temperature of 0.07°C and 308 mm of precipitation at Abisko (20-year mean; 1986–2006).
- Lakes are underlain by sporadic permafrost visible as ombrotrophic palsas rising above semi-wet and wet minerotrophic fens, streams, and shallow lakes.[6,7]

- Three lakes chosen for their proximity to a thawing permafrost peatland and sampled in order to study intra- and inter-lake variability in total sediment Hg (THg).
  - **Villasjön**: a shallow lake chosen for its previously established methane ebullition gradient.[8]
  - **Mellarsta Harrsjön** (aka Mellan Harrsjön): Water enters Mellan Harrsjön from the catchment's mainstream and from Yttre Harrsjön.
  - **Inre Harrsjön**, connects to Mellarsta Harrsjön via Yttre Harrsjön.
  - Lakes contain limited erosional margins and mostly surrounded by wet fens dominated by *Eriophorum angustifolium*, *Carex rostrata* and *Sphagnum spp.* and semiwet subalpine birch forests.[6]

METHODS

- Sediment cores were taken using a modified AMS hammer corer; sub-sampled for total Hg (THg) every 2cm and for total carbon, nitrogen and sulfur every 5 cm.
- Twelve cores were taken in Villasjön, where known high and low ebullition sites correspond to high and low CH_4 concentrations. Two transects were chosen: M' and P'
- Both a shallow and a deep water cores were taken at Mellarsta and Inre Harrsjön.
- Samples for Hg analysis were freeze dried and CNS samples oven dried.
- Analysis for THg was carried out via thermal decomposition, amalgamation/atomic absorption spectrophotometer (TDA/AAS) with the Milestone DMA-80 Direct Mercury Analyzer (Milestone, Microwave Laboratory Systems, Shelton, CT), at UNH following US EPA Method 7473.
  - DORM-4, NIST1547 quality controls standards included every 10 samples for assessing accuracy and precision.
- Total carbon, nitrogen and sulfur contents measured via Perkin-Elmer CHNS 2400 analyzer at UNH.



Figure 1a

	Inre Harrsjön	Mellarsta Harrsjön	Villasjön
Area (ha)	2.2	1.1	17
Mean Water Depth[1] (m)	2	1.8	0.7
Max Water Depth[1] (m)	5	7	1.3
Radiocarbon dating (pre B.P.)[2]	2650		3400
Bedrock[3]	Schist	Schist	Granite
Inflow	Spring fed from the north & south	Stream fed	Spring and stream fed, water upwelling observed
Outflow	Share water via Yttre Harrsjön		Drains into Inre during high flow through fen
Ice free	mid-May to mid-October		
Vegetation Type[4]	*Potamogeton alpinus ottsulifolius* and *Myriophyllum alternifloru*	*Sparganium hyperborneum* and *Hippuris vulgaris* in shallower regions down to 1.5 m	Submerged Peat

*The water depths in Villasjön were measured by Jackowicz-Korczynski et al., 2010, and the depths in Inre Harrsjön and Mellarsta Harrsjön were measured by us.
[1]Karin Bergström by P. Kuuper, personal communication, 2010

Table 1 Lake information

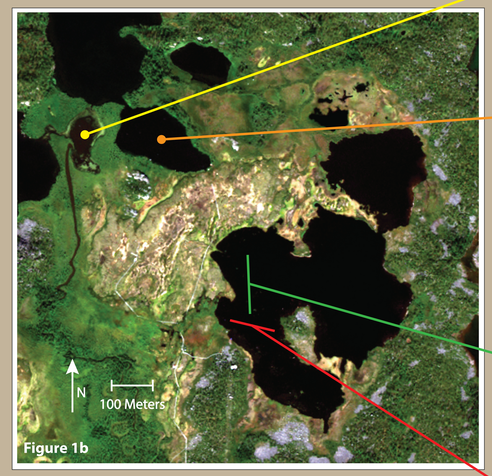
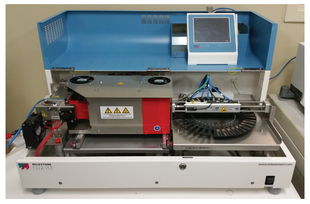


Figure 1b







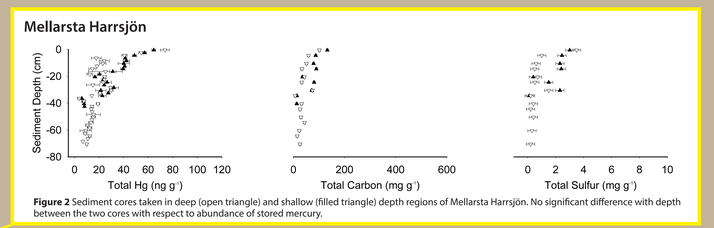


Figure 2 Sediment cores taken in deep (open triangle) and shallow (filled triangle) depth regions of Mellarsta Harrsjön. No significant difference with depth between the two cores with respect to abundance of stored mercury.

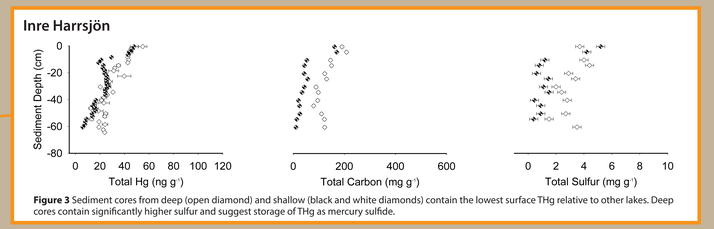


Figure 3 Sediment cores from deep (open diamond) and shallow (black and white diamonds) contain the lowest surface THg relative to other lakes. Deep cores contain significantly higher sulfur and suggest storage of THg as mercury sulfide.

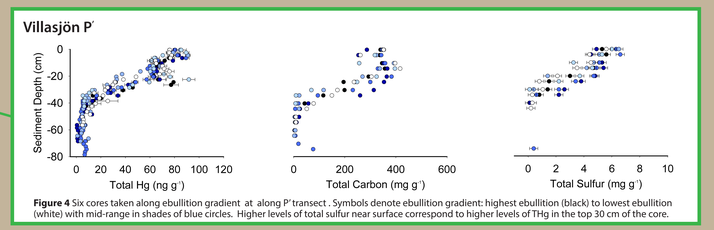


Figure 4 Six cores taken along ebullition gradient at along P' transect . Symbols denote ebullition gradient: highest ebullition (black) to lowest ebullition (white) with mid-range in shades of blue circles. Higher levels of total sulfur near surface correspond to higher levels of THg in the top 30 cm of the core.

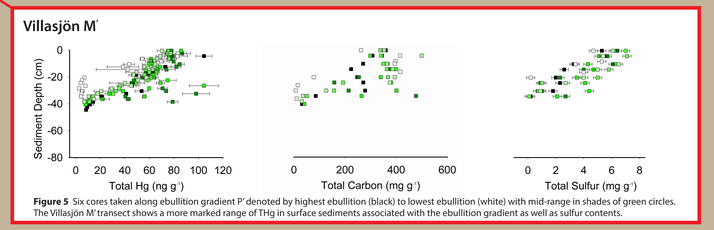


Figure 5 Six cores taken along ebullition gradient P' denoted by highest ebullition (black) to lowest ebullition (white) with mid-range in shades of green circles. The Villasjön M' transect shows a more marked range of THg in surface sediments associated with the ebullition gradient as well as sulfur contents.

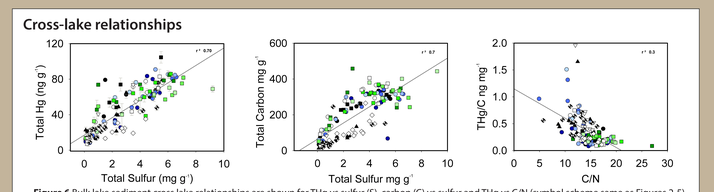


Figure 6 Bulk lake sediment cross lake relationships are shown for THg vs sulfur (S), carbon (C) vs sulfur and THg vs C/N (symbol scheme same as Figures 2-5). Reduced sulfur has been shown to be increasing in high arctic lakes in response to labile carbon availability. Ratios of THg/C vs C/N are used to evaluate the influence of terrestrial vs algal inputs of carbon which can help identify the source of THg to lake sediments.

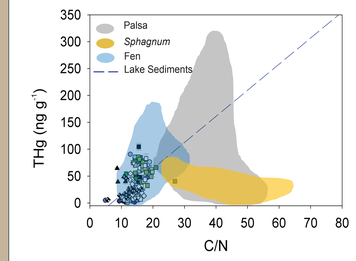


Figure 7 (left) Lake sediment THg from all three lakes is shown versus C/N ratio along with terrestrial THg data from cores taken in the palsa, *Sphagnum* and fen sites in Stordalen Mire. Both the lake sediments and fen sites show C/N ratios shifted to aquatic signatures.

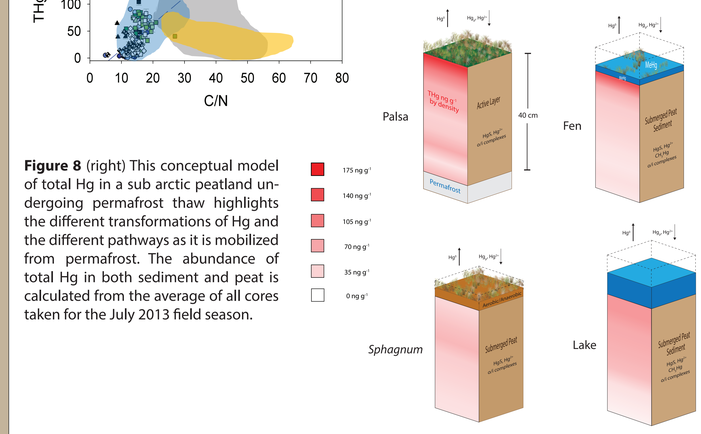


Figure 8 (right) This conceptual model of total Hg in a sub arctic peatland undergoing permafrost thaw highlights the different transformations of Hg and the different pathways as it is mobilized from permafrost. The abundance of total Hg in both sediment and peat is calculated from the average of all cores taken for the July 2013 field season.

CONCLUSIONS

- Inter-and intra-lake variations in sediment THg were found to be most pronounced in the top 20 cm of the cores.
- Villasjön contained significantly more THg in the top 30 cm relative to Mellarsta and Inre Harrsjön. This is attributable to Villasjön being composed of predominantly submerged peat.
- In the Villasjön M' transect, the depth of maximum sediment Hg content varies across the ebullition gradient such that the cores from areas with high ebullition rates have elevated THg as deep as ~30 cm whereas maximum total sediment Hg in the low ebullition cores was located in the top 10 cm.
- Higher THg/C ratios in some sediments, mostly Villasjön were associated with lower C/N ratios, suggesting that the more aquatic carbon source is linked to higher in-situ Hg.
- THg depth profiles and surface levels of lake sediments from Stordalen Mire are similar to Toolik Lake LTER as well as other lakes in the Abisko area.

FUTURE WORK

- Pore water THg and methyl Hg would help determine if these lakes are mobilizing Hg that is reaching larger lakes where aquatic life may be bioaccumulating methyl Hg.
- Total gaseous Hg measurements from lake surface to evaluate flux of Hg to the atmosphere.
- Scaling up of THg inventories in lake sediments to improve estimates Hg stored in arctic lakes and projecting magnitude of Hg availability with expansion of permafrost thaw.

REFERENCES

1. Callaghan et al., (2010) A new climate era in the sub-Arctic: Accelerating climate changes and multiple impacts, *Geophys. Res. Lett.*, 37, L14705.
2. Christensen et al., (2004) Thawing subarctic permafrost: Effects on vegetation and methane emissions, *Geophys. Res. Lett.*, 31, L04501.
3. Johansson et al., (2006) Decadal vegetation changes in a northern peatland, greenhouse gas fluxes and net radiative forcing, *Global Change Biol.*, 12, 2352–2369.
4. Klaminder et al., (2008) An explorative study of mercury export from a thawing palsa mire. *Journal of Geophysical Research*. Volume 113.
5. Rydberg et al., (2010) Climate driven release of carbon and mercury from permafrost mires increases mercury loading to sub-arctic lakes. *Science of the Total Environment* 408, 4778-4783.
6. Malmer et al., (2005) Vegetation, climatic changes and net carbon sequestration in a North-Scandinavian subarctic mire over 30 years, *Global Change Biol.*, 11, 1895–1910.
7. Kokfelt et al., (2009) Ecosystem responses to increased precipitation and permafrost decay in subarctic Sweden inferred from peat and lake sediments, *Global Change Biol.*, 15, 1652–1663.
8. Wik et al., (2011) Bubbles trapped in arctic lake ice: Potential implications for methane emissions, *J. Geophys. Res.*, 116, G03044
9. Lindström et al., (1985) Geology of a part of the Tornetrask section of the Caledonian front, northern Sweden, in The Caledonide Orogen—Scandinavia and Related Areas, edited by D. G. Gee and B. A. Stuart, John Wiley, New York, pp. 507–513.
10. Jackowicz-Korczynski et al., (2010) Annual cycle of methane emission from a subarctic peatland, *J. Geophys. Res.*, 115, G02009.
11. Drevnick et al., (2010) Increased accumulation of sulfur in lake sediments of the high arctic, *Environ. Sci. Technol.*, 44 (22) 8415-8421.
12. Fitzgerald et al., (2005) Modern and historic atmospheric mercury fluxes in northern alaska: Global sources and arctic depletion, *Environ. Sci. Technol.*, 39, 557-568.
13. Lindeberg et al., (2007) Mercury pollution trends in subarctic lakes in the northern swedish mountains, *Ambio*, 36 (5) 401-405.

ACKNOWLEDGEMENTS Abisko Scientific Research Station (ANS) staff, Dr. Reiner Giesler for access to facilities for sample prep and assistance throughout our month long stay, Dr. Mike Palace for his assistance with w.v2 site image, Mr. Uriah Fahnestock for graphic design contributions and funding provided by NSF REU site EAR#1063037