

Atmospheric Mercury in Vermont and New England: Measurement of deposition, surface exchanges and assimilation in terrestrial ecosystems

Final Project Report – Additional Project Activities – 1/16/2009

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Coordination with national, regional, and state mercury research

The project personnel successfully coordinated with other mercury research efforts at local, regional and national levels. We used the new Hg information from the Underhill site and this project to estimate mercury deposition not only to the forested watersheds of Vermont, but also specifically to Lake Champlain in collaboration with researchers supported by the NOAA funded, Lake Champlain Research Consortium. More broadly, Drs. Miller and Keeler participated in the Northeast Mercury Research Group, a regional research group funded by the USDA Forest service (www.briloon.org/mercury). The three-way, west-east (WA-VT), precipitation mercury collector intercomparison was a collaboration designed to inform the national MDN program on potential improvements to collector design as well as to facilitate Underhill's transition from the UMAQL to the MDN system.

Observations and analyses of mercury concentrations and fluxes at Underhill are providing benefits to several mercury research groups working to model emissions-transport-deposition cycles at regional, national and global scales. The results of our trend analysis (no trend detected) for wet deposition and our source identification efforts for wet and dry deposition have informed state, regional, national, and international air-quality planning bodies about the identity of sources contributing mercury to the biologically sensitive New England region. Our pioneering assessment of mercury in a terrestrial food-web has highlighted the need for expanded consideration of the risks posed by atmospheric mercury deposition to terrestrial environments.

Dr. Miller participated in the technical working group designing protocols and operations standards for the proposed MTN (Mercury Trends Network), a new mercury dry deposition network being established by NADP. Dr. Miller provided detailed information on our operating procedures and data management process for use in developing the network SOP. Underhill served as demonstration site for the network and we hosted a field trip for NADP personnel to observe our operations in 2007. The Underhill site was one of the initial four sites funded by EPA-OAR-CAMD for start-up of the network in January of 2008.

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Scientific communication and public outreach

In addition to the research coordination activities described above project personnel made numerous presentations about project activities and results at regional and national meetings. Dr. Miller produced a public-outreach overview document describing mercury research activities at Underhill in conjunction with the VMC. Interviews were granted to print and radio media to convey project results to the public. Several peer-reviewed scientific publications were prepared, accepted and published that made use of project data. Additional manuscripts are currently being prepared for submission by the project team. The final results of the project (which are the subjects of these manuscripts) will be presented at national meetings and communicated to the air-quality management community.

Meetings, Public Presentations, and Publications (chronological)

- Dr. Keeler presented results from a coordinated, long-term event wet-deposition study (funding sources including NESCAUM and NOAA) at the October 2004 Vermont Monitoring Cooperative Research Symposium in Burlington, VT.
- Dr. Miller presented preliminary results from the air-concentration and flux measurement activities at the October 2004 Vermont Monitoring Cooperative Research Symposium in Burlington, VT.
- Dr. Miller presented regional mercury deposition estimates based, in part, on the Underhill observation record at the 2004 NADP/MDN meeting in Halifax, Nova Scotia.
- Dr. Miller attended the 2005 NADP meetings and presented preliminary results from the 2005 mercury speciation studies, and 2004 vapor phase exchange measurements. He also described the 3-way precipitation Hg collector comparison study, but no preliminary results were presented at that time.
- The precipitation event mercury record was analyzed and published as part of three different manuscripts:

Keeler, G.J., Gratz, L.E, and Al-Whali, K. (2005) Long-term Atmospheric Mercury Wet Deposition at Underhill, Vermont. *Ecotoxicology* 14, 71-83.

Miller, E.K., VanArsdale, A., Keeler, G.J., Chalmers, A., Poissant, L., Kamman, N., and Brulotte, R. (2005) Estimation and Mapping of Wet and Dry Mercury Deposition across Northeastern North America. *Ecotoxicology* 14, 53-70.

VanArsdale, A., Weiss, J., Keeler, G.J., Miller, E.K., Boulet, G., Brulotte, R., Poissant, L., and Puckett, K. (2005) Patterns of Mercury Deposition in Northeastern North America (1996-2002). *Ecotoxicology* 14, 37-52.

- Initial Bicknell's thrush blood mercury data were analyzed and published in:
Rimmer, C.C., McFarland, K.P., Evers, D.C., Miller, E.K., Aubry, Y., Busby, D., and Taylor, R.J. (2005) Mercury Levels in Bicknell's Thrush and Other Insectivorous Passerines in Montane Forests of Northeastern North America. *Ecotoxicology* 14, 223-240.

- Dr. Miller gave an informal presentation to the NESCAUM/EPA Region 1 Mercury working group at the NESCAUM offices in Boston on January 11, 2006. He presented preliminary results from the 2005 mercury speciation studies, and the 3-way precipitation Hg collector comparison study.
- Drs. Shanley, Miller, and Keeler were coauthors of a study published in *Environmental Science and Technology*.

Gao, N., N.G. Armatas, J.B. Shanley, N.C. Kamman, E.K. Miller, G.J. Keeler, T. Scherbatskoy, T.M. Holsen, T. Young, L. McIlroy, S. Drake, B. Olsen, and C. Cady. (2006) A mass balance assessment for mercury in Lake Champlain. *Environ. Sci. and Technol.* 40: 82-89.

This study used data from the Underhill site and other sites in the region to estimate atmospheric deposition to Lake Champlain as part of a mass-balance assessment for mercury in the lake.

- Dr. Miller gave a presentation at the New England Interstate Water Pollution Control Commission 2006 Mercury Science and Policy Conference in Newport, RI. The presentations from the conference can now be found online at: http://www.neiwppcc.org/hgconference/hg_archives.htm
- Dr. Miller attended a scoping meeting for the NADP proposed Mercury Trends Network (MTN) in 2006. The MTN is being proposed within NADP with support from USEPA Air Markets. The MTN is proposed to provide measurements of speciated ambient air mercury concentrations and estimates of mercury dry deposition. The Underhill site is considered to be an example of how the network sites could be configured and operated.
- Dr. Miller gave a presentation at the Lake Champlain Research Consortium annual meeting on September 29th, 2006. This presentation reviewed the atmospheric mercury research of interest to the LCRC members.
- Dr. Miller gave a presentation at the NADP annual meeting in October, 2006. This presentation reviewed the latest atmospheric mercury research at Underhill. Dr. Miller also presented a poster at the October NADP meetings describing the results of the 3-way mercury collector comparison. An identical poster was also presented at the VMC Annual Meeting, October 30th, 2006. Eric Miller and Sean Lawson gave a presentation on the Atmospheric Mercury Research project to members of the International Society of Environmental Journalists. The society held their annual meeting in Burlington, VT. Approximately 40 journalists participated in a field trip to UVM's Proctor Maple Research Center.
- The project team and the Vermont Monitoring Cooperative participated in hosting the spring 2007 NADP meeting in Burlington, VT. Dr. Miller made a presentation on operational considerations to a session of the meeting. Miller, Lawson, and Pendleton led a field trip to the Underhill Atmospheric Mercury Site as part of the meeting

Manuscripts in Preparation

1. Comparison of precipitation mercury samplers (Miller and others – target journal : *Environmental Monitoring and Assessment*)
2. Possible explanations for the lack of trend in wet deposition of mercury at Underhill, Vermont during a period of significant estimated mercury emissions reductions. (Miller, Poirot, Shanley, Cohen, Gratz, Pendelton, Lawson, Burkings, and Keeler – target journal: *Environmental Science and Technology*)
3. Climatology and Potential Sources of GEM, RGM, and Particulate Mercury at Underhill, VT (Miller, Poirot, Cohen, VanArsdale, others – target journal: *Environmental Science and Technology*)
4. Climatology and Potential Sources of Methyl-Mercury in Precipitation at Underhill, VT (Miller, Poirot, Shanley, others – target journal: to be determined)
5. Wet- and Dry-Deposition of Speciated Mercury at Underhill, VT (Miller, Poirot, Shanley, others – target journal: to be determined) *speciated wet = methyl and total, speciated dry = GEM, RGM, HGP.
6. Mercury Bioaccumulation in the Terrestrial Food Web of Montane Forests (Rimmer, McFarland, Miller, Faccio, and Taylor - target journal *Ecotoxicology*).

Summary of Project Results and Benefits

This project achieved the extension of the longest continuous record of event-based mercury deposition in the world. The collector comparison study facilitated a successful transition from the UMAQL network to the national MDN network. Results from the comparison study provided critical guidance for collector improvements and interpretation of the data collected by various networks. As has been clearly demonstrated through analysis of the impact of reduced sulfur emissions, long-term records are invaluable for trend detection and confirmation of the efficacy of emissions controls. The long-term record of atmospheric mercury levels at Underhill, VT represents one of the few opportunities for this kind of assessment with respect to mercury emissions reductions. Initial results from the period 1993-2007 show no measurable impact of the substantial reductions in municipal-waste and medical-waste incinerator emissions that occurred regionally and nationally during this time period. Instead, the long-term record and studies of potential source contributions using air-mass back-trajectory methods and the observations at Underhill demonstrated that northern New England receives the majority of its mercury deposition from long-range industrial and EGU sources that have not reduced emissions over the observation period.

The project's measurements of bi-directional GEM exchanges over New England forests helped to close the gap in understanding of the magnitude and driving factors for bi-directional GEM fluxes in forests. As new anthropogenic mercury emissions continue to be curtailed, the importance of diffuse sources of re-emission of prior mercury pollution will become more important to the atmospheric mercury burden. Understanding the magnitude and processes governing the canopy re-emission flux is essential to accurate modeling of the persistence of mercury pollution in the environment.

Previous studies at Underhill, VT demonstrated that significant amounts of Hg accumulate in the foliage of forest trees. Investigators speculated that this represented assimilation of atmospheric mercury. The GEM flux measurements conducted in this project confirmed that this process is occurring and represents a large annual transfer of Hg from the atmosphere to the landscape, roughly equal in magnitude to wet deposition.

Given the substantial accumulation of mercury in terrestrial ecosystems due to wet and dry atmospheric deposition, it was imperative to assess the extent to which mercury enters the terrestrial food web. Our measurements of mercury levels in foliage-eating insects and the birds that feed on those insects provided new insights into the magnitude of mercury bioaccumulation and biomagnification in terrestrial ecosystems. Our study has provided the most comprehensive and compelling evidence that biota at the mid- and top-levels of the terrestrial food web are at significant risk from atmospheric deposition of mercury.

Taken together, the results of mercury studies accomplished by the project greatly enhanced the scientific community's ability to assess current and future mercury deposition, re-emission, net ecosystem retention and risk to terrestrial biota across New England.

In-kind and Leveraged Funding and Companion Projects

The scope of project activities described above is broad. The funding provided by EPA-ORD for this project was substantially augmented with in-kind contributions from investigators and collaborators as well as by funds directed toward this research effort as a result of the leverage created by the cooperative agreement. Below we highlight major additional sources of support that will permitted the realization of the full scope of the research described above. The leveraged funds and in-kind support amounted to over \$250,000. The leveraged funding and many companion projects would not have been feasible with out the foundation of support for atmospheric mercury research provided by the EPA-ORD cooperative agreement.

- NOAA Sea Grant – LCRC – (\$40,000) support for analysis of archived ambient Hg samples
- VTANR-DEC-AQD-Air Toxics Program – (\$20,000) support for Tekran RGM system
- VTANR-DEC-WQD – laboratory services and analytical support
- VMC –VTANR-FP&R – targeted research funds (\$60,000) supporting E. Miller’s time
- VMC – UVM– in-kind support: facilities and sampling operations at PMRC
- ERG – in-kind support: micrometeorological flux measurement system, reduced indirect rate
- VINS/VCE – in-kind support: use of facilities and sampling equipment
- NOAA – (\$20,000) intercomparison study
- NOAA Sea Grant – LCRC – Funding for MDN event wet-deposition at Underhill, VT
- NOAA Sea Grant – LCRC – Lake Champlain Mercury Mass-Balance Study
- VTANR-DEC-AQD – Ticonderoga Mill Test Burn Study
- VTANR-DEC-AQD / EPA Region 1 – Mobile Mercury Facility Grant