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

Final Project Report – GEM Flux Measurements – 1/16/2009

PI: Melody Brown Burkins, University of Vermont (UVM)
Co-PIs: Eric K. Miller[1], Ecosystems Research Group, Ltd.; and Jamie Shanley, US Geological Survey
Collaborators: Sean Lawson, VTANR-VMC; Mim Pendelton, Carl Waite, UVM;
Rich Poirot, VTANR-APCD; Alan VanArsdale, USEPA; Mark Cohen, NOAA
Project Officer: Eric Hall, USEPA

Measurements of GEM exchanges over a forest canopy

At the outset of the project there was tremendous uncertainty about the magnitude and the mechanisms governing net-gaseous Hg assimilation by forest canopies (Miller 2002, Lindberg et al. 1998). The most widely used inferential model for GEM deposition at the time (Lindberg et al. 1992) did not represent the known bi-directional nature of the GEM flux. It was clear from limited direct observations that at times GEM deposits and at times it is emitted from the forest canopy (Lindberg et al. 1998). It was also impossible to reconcile the large GEM fluxes implied by the Lindberg et al. (1992) model with the much smaller fluxes indicated by measurements of leaf-assimilated Hg (see Miller 2002, 2005). It appeared likely that Hg deposition/emission is governed by a compensation point, an ambient concentration above which deposition occurs and below which emission occurs (Hansen et al. 1995). Direct measurements of atmosphere-forest exchanges of GEM were needed to resolve this discrepancy, identify a potential field compensation point and to help elucidate the physical and physiological processes regulating GEM deposition or emission.

Direct, micrometeorological measurements of atmosphere-canopy exchanges of GEM were made using the modified Bowen-ratio method (Lindberg et al. 2002, Lindberg and Meyers 2001, Lee et al. 2000). Briefly, this method involves measuring the concentration gradient of mercury-vapor above the surface while concurrently measuring the gradient of either temperature (Lee et al. 2000) or water-vapor (Lindberg et al. 2002) and the turbulent flux of sensible or latent heat over the same height interval. The turbulent transfer coefficient derived from, for example, the latent heat flux and the water-vapor gradient is then assumed to apply to mercury vapor (Lindberg and Meyers 2001). The turbulent fluxes of latent and sensible heat were measured using the Bowen-ratio method and confirmed with sensible heat-fluxes measured by the eddy correlation method. The mercury gradient was measured with a Tekran 2537A. The GEM flux measurements were conducted from the VMC forest canopy observation tower at the Proctor Maple Research Center (Figures 1 and 2). Simultaneous measurements of the turbulent fluxes of CO_2 and H_2O were made in conjunction with mercury flux measurements over the forest canopy. These measurements provided insight into the physiological and physical mechanisms governing mercury exchange.

Initial flux measurements were begun in late summer of 2004. In December of 2004 a severe windstorm following heavy rains blew over the 100-foot tower that supported the mercury concentration and flux measurements (Figure 3). Shortly after the tower collapsed, typical cold winter weather set in and the ground froze solid. Difficult winter conditions limited salvage operations. After winter conditions eased, equipment was salvaged and repaired. Concentration

[1] Corresponding author for the final project report. Email: ekmiller at ecoystems-research.com Voice: 802-649-5550

measurements were reestablished in April, 2005 based out of an instrument shelter at the NADP/MDN site located ~1km north of the tower site. We were delayed in restarting the flux measurement program at the tower site for several reasons. University of Vermont Facilities Management controlled the schedule of salvage, repair, and insurance claims for the tower damage. There were considerable logistical problems to overcome in designing and constructing new anchors and a new tower. The tower reinstallation was completed on October 4th, 2005.

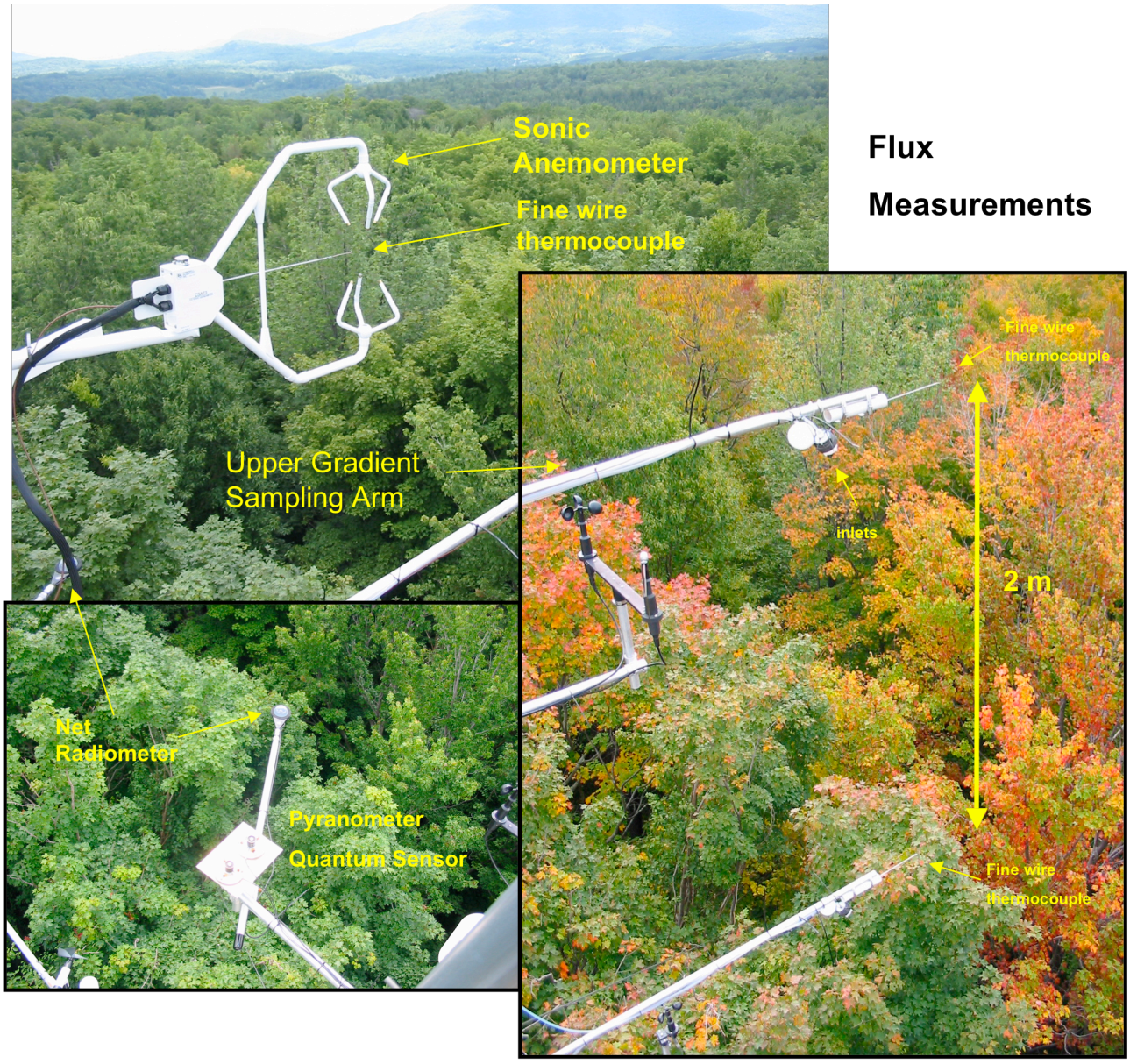

Figure 1. Configuration of inlets and sensors for the GEM flux measurement system deployed on top of the forest canopy observation tower.

Valve Box, CO2/H2O and Hg Analyzers

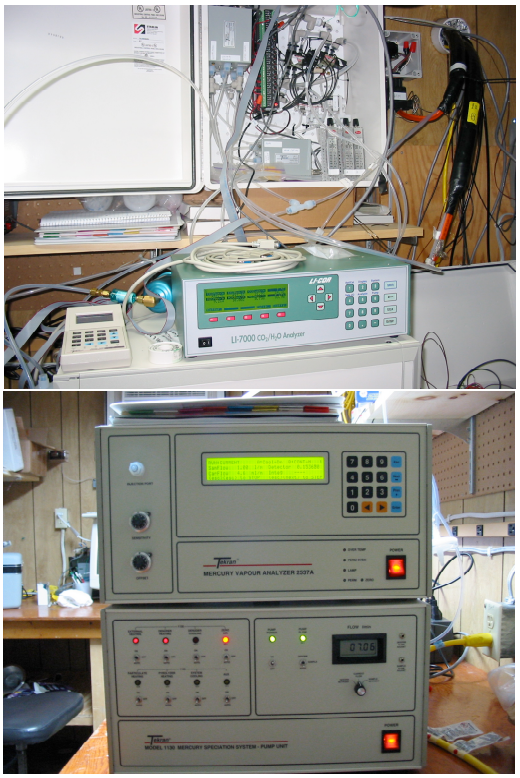

Figure 2. Analytical equipment housed in the trailer at the base of the forest canopy observation tower.

Figure 3. The atmospheric measurement tower after the severe wind storm (left). New forest canopy tower under construction (right).

Bi-directional fluxes (both emission and deposition) were observed (Figure 4). Net emission and deposition fluxes ranged up to 1000 ng/m^2/h. Deposition was typically observed during the day with high solar fluxes, while emission typically occurred at night or during cloudy periods. Companion measurements of CO_2 and water-vapor exchanges suggested peak deposition values occurred in conjunction with strong photosynthesis (Figure 5).

GEM exchange measurements and their analysis were limited by the rescission of funding for years 4 and 5 of the project and due to the unfortunate collapse of the forest canopy tower in the winter of 2004 which caused a significant delay in the measurement program. We hope to complete additional analyses of GEM flux measurements in the near future under separate funding.

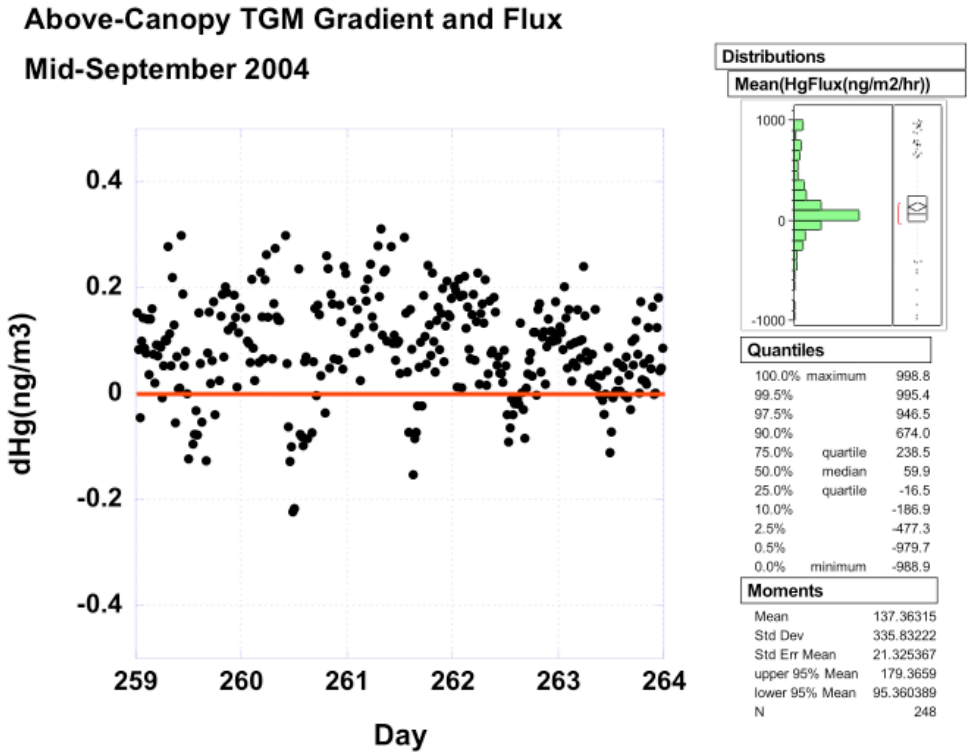

Figure 4. (Left) Example of diurnal and multi-day pattern of above-canopy mercury gradient. Positive values indicate emission of GEM and negative values indicated deposition to the forest canopy. (Right) Frequency distribution of fluxes.

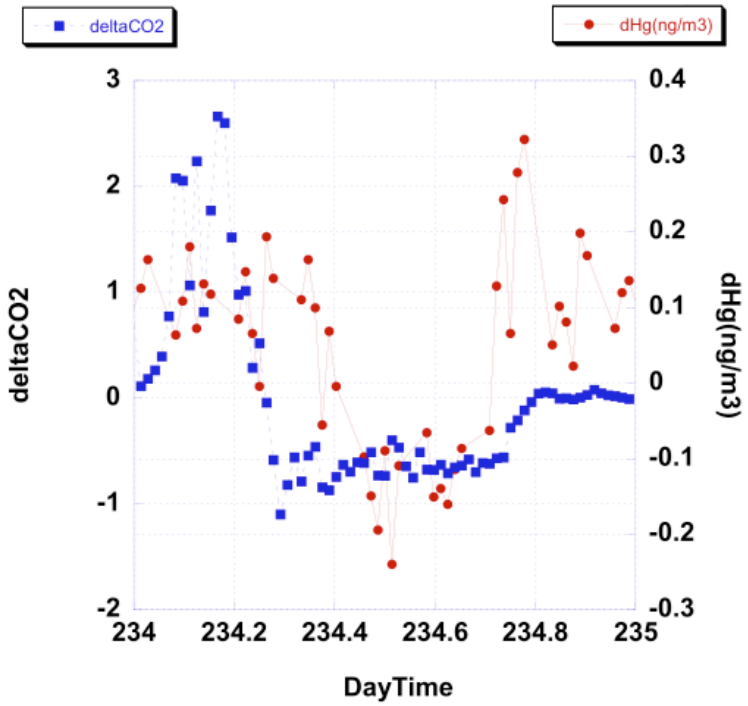

Figure 5. Example diurnal cycle of GEM and CO_2 gradients measured above the forest canopy. GEM deposition is indicated by the negative GEM gradients (red). GEM deposition is strongest at mid-day during intense photosynthesis (indicated by the strong negative CO_2 gradient - blue).

References

Hanson, P.J., S.E. Lindberg, T.A. Tabberer, J.G. Owens, K.-H. Kim. 1995. Foliar exchange of mercury vapor: evidence for a compensation point. *Water, Air , and Soil Pollution* **80**:373-382.

Lee, X., G. Benoit, X. Hu. 2000. Total gaseous mercury concentration and flux over a coastal saltmarsh vegetation in Connecticut, USA. *Atmos. Environ.* **34**: 4205-4213.

Lindberg, S.E., T.P. Meyers, G.E. Taylor, R.R. Turner, and W.H. Schroeder (1992) Atmosphere-Surface Exchange of Mercury in a Forest: Results of Modeling and Gradient Approaches. *Journal of Geophysical Research* **97**:2519-2528.

Lindberg, S.E. and T.P. Meyers. 2001. Development of an automated micrometeorological method for measuring the emission of mercury vapor from wetland vegetation. *Wetlands Ecology and Management* **9**:333-347.

Lindberg, S.E., W. Dong, and T. Meyers. 2002. Transpiration of gaseous mercury through vegetation in a subtropical wetland in Florida. *Atmos. Envir.* 36:5200-5219.

Lindberg, S.E., P.J. Hanson, T.P. Meyers, and K-Y Kim. 1998. Micrometeorological studies of air/surface exchange of mercury over forest vegetation and a reassessment of continental biogenic mercury emissions. *Atmos. Envir.* 32:895-908.

Lindberg, S. E., and W.J. Stratton. 1998. Atmospheric mercury speciation: Concentrations and behavior of reactive gaseous mercury in ambient air. *Envir. Sci. & Technol.* 32:49-57.

Miller, E.K. 2002. Estimation and Mapping of Wet and Dry Mercury Deposition Across the VT-NH Region. Project report submitted to Neil Kamman, VTDEC, Water Quality Division, 103 South Main St., Building 10 North, Waterbury, VT 05671-0408. Ecosystems Research Group, Ltd., Norwich, VT.

Miller, E. K., A. VanArsdale, J. G. Keeler, A. Chalmers, L. Poissant, N. C. Kamman, and R. Brulotte. 2005. Estimation and mapping of wet and dry mercury deposition across northeastern North America. *Ecotoxicology* 14:53-70.