II. Summary of Findings from EMEP-Supported Projects Related to the Deposition of Sulfur, Nitrogen, and Mercury

Preface

This summary, which focuses on the deposition of sulfur, nitrogen, and mercury, and ecosystem response, was prepared by NYSERDA staff to assist with program evaluation efforts and facilitate discussions at the upcoming evaluation and planning meeting with EMEP Program Advisors and Science Advisors. Conclusions drawn from this effort will assist in determining research needs related to the EMEP program and help focus research solicitations.

Introduction

The EMEP multi-year plan (2002) identified acid deposition and mercury pollution as important energy-related environmental management challenges facing New York State. The general research objectives were to:

1. develop and evaluate the effectiveness of pollution control strategies for acid deposition and mercury;
2. quantify local sources versus regional transport of acid deposition precursors and mercury to develop more equitable pollution control strategies;
3. identify alternative environmental protection and mitigation strategies to reduce the impacts of acidification and exposure to mercury in New York; and
4. develop emerging multi-media/multi-pollutant environmental protection strategies.

Through a series of competitive solicitations targeted at critical research gaps identified in the EMEP research plan, approximately 20 research projects focusing on acid deposition and mercury were selected and funded by the EMEP Program. The total cost of this research was $20 million. The contribution of EMEP funds to these research projects was $9.5 million. The broad implications of each of these research projects are identified on the Project Update sheets [see Appendix D]. Below is a summary of knowledge created from the completed and ongoing projects.

1. Acid Deposition

Summary

With implementation of the Clean Air Act Amendments of 1990 (CAAA), it was expected that increases in precipitation pH would result in increases in pH and acid neutralizing capacity (ANC) in sensitive New York State surface waters, and decreases in sulfate (SO_4^{2-}), nitrate (NO^{3-}), and aluminum (Al) concentrations. This was expected to allow for the recovery of species diversity in biota affected by surface-water acidification during the 20th century. Adequate data on surface-water chemistry are now available to evaluate whether these intentions are being realized. The results of studies indicate that surface waters are slowly improving due to reduced levels of acid precipitation, but that additional improvements may require reductions in SO_2 and NO_X emissions greater than those mandated under the 1990 CAAA.

1.1 Adirondacks

Findings from the Adirondack Lakes Survey Corporation (ALSC) Long-Term Monitoring (ALTM) project indicate that when declines in emissions result in declines in acidic deposition, these improvements can translate into changes in water chemistry. Time-series analysis of water chemistry conducted from 1982-2000 show that improvements have occurred in a number of lakes, albeit slightly, as a result of changes in atmospheric deposition. More recently in 2005, another time-series analysis was conducted with four years of additional data. These results show that while a greater number of lakes are exhibiting positive trends, the slopes or the rates of those trends have slowed. The patterns and mechanisms for these changes are being further analyzed across lake types.

Although some recovery is evident, the Adirondack region continues to be at high risk from acidic deposition. Atmospheric deposition was the dominant source of acidity (>90%), while mineral weathering was the major source of improved acid neutralizing capacity for all study sites. Simulations indicated that these ecosystems would show little recovery after 2010 under the 1990 Clean Air Act Amendments. The effects of changes in atmospheric pollutant concentrations need to be placed in the context of other environmental influences, such as changes in climate and forest biota, in order for us to understand the major factors influencing the environmental health of the Adirondacks. These results, and continued monitoring, will not only provide critical information for assessments of forest and aquatic resources in the region, but also prove useful in predicting and evaluating the impact of policy options and future legislation on Adirondack air and water quality.

The following observations and conclusions were primarily drawn from EMEP research projects:

Deposition

Precipitation pH has increased an average of about 0.02 units in the Adirondacks and Catskills from 1984 to 2001; three-fourths of this increase has resulted from decreases in SO_4^{2-} concentrations and about one-fourth from decreases in NO^{3-} concentrations. These changes are paralleled by, and are assumed to result from, similar decreases in sulfur and nitrogen oxide emissions largely from power plants in the predominant source region of acidic precipitation for the state.

Sulfate

Sulfate concentrations in lake discharge markedly decreased in the 1990s, reflecting declining patterns in both wet and dry sulfur deposition. Despite decreases in atmospheric deposition and hydrologic losses, sulfate was still the dominant negatively charged ion in drainage water and was therefore a cause of declines in concentrations of positively charged ions. Declining surface water concentrations of base positively charged ions will have a great impact on future trends in surface water recovery from acidification.

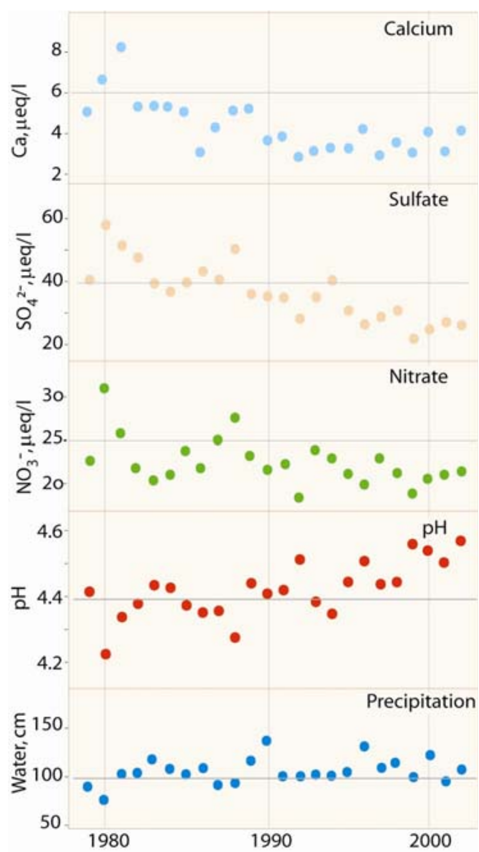

Figure 1. Wet deposition data from the National Trends Network station at Huntington Forest; the points give the annual volume-weighted averages of weekly observations. Sulfate has declined steadily and pH risen steadily. Calcium decreased in the 1980s and since then has been mostly constant; nitrate has perhaps declined, but the variability in the record makes it difficult to be sure.

Nitrate

In contrast to the case of sulfates, no long-term trend in nitrate concentrations in lake discharge was found in the 1990s. Patterns of nitrate cannot be attributed to changes in nitrate deposition or explained by nitrogen saturation models. However, nitrate concentrations were found to be correlated to interannual changes in air temperature (1983–2001). The strong relationship between air temperature and watershed nitrate loss (January–March) suggests that snowmelt responses to winter temperature fluctuations play a critical role in nitrate-loss variability in different years. The contrasting seasonal patterns that were observed might be explained by the high biological demand for nitrate and the microbial production of dissolved organic nitrogen (DON) during warm summer months. The close relationship observed between temperature and nitrate/DON loss suggests that the biotic and hydrological processes affecting nitrogen loss may be especially sensitive to changes in climate, as well as to changes in atmospheric nitrogen deposition. This response of watershed nitrogen to variations in climate and atmospheric deposition may determine the long-term patterns of nitrogen loss from the watershed.

The inclusion of both dry deposition and organic nitrogen substantially elevates estimates of nitrogen retention in Adirondack waters. This result, along with very low summer NO^{3-} concentrations in drainage water, suggests that there is a strong biological retention of both organic and inorganic nitrogen in Adirondack watersheds. In addition, the lowest NO^{3-} concentrations have been found in subcatchments with the highest proportion of wetlands, suggesting the importance of wetlands as sites of denitrification.

ANC

ANC improvement was detected in 29 out of 48 ALTM lakes. The rates of change are small and slow, suggesting it will take decades at current deposition rates to reach 50 meq/L, a level suitable for aquatic biota. Overall lake ANC levels remain a concern, with 34 lakes showing average ANC values of less than 50 meq/L, including 10 lakes with ANC levels below zero.

pH

Improvements (increased pH) were observed in 18 of the ALTM lakes, with two lakes declining. Overall averages were still critical to aquatic biota, with 23 lakes showing an annual mean pH of less than 5.5.

Aluminum

Episodic acidification is extremely damaging to Adirondack lake and stream ecosystems. High acidity during snowmelt and high-flow events causes aluminum levels to rise, which has been well documented to cause widespread fish mortality. An ongoing EMEP-funded study has found that over 50% of western Adirondack streams have aluminum levels during high-flow events that are sufficiently elevated to cause fish mortality. Of the 52 ALTM lakes, decreases in toxic forms of aluminum were detected in 28 lakes. While a shift was observed from toxic inorganic forms toward less toxic organic forms, toxic levels remain above those known to be toxic to juvenile forms of native Adirondack fish.

Significant Trends in 48 ALTM Lakes Values are Average Rates of Change							
	SO_4^{2-} ↓	NO_3^- ↓	C_B ↓	ANC ↑	pH ↑	Al_{im} ↓	DOC ↑
1992-2000	**44** **-2.57**	**15** **-1.03**	**26** **-3.33**	**29** **1.60**	**18** **0.04**	**28** **-0.31**	**7** **15.7**
1992-2004	**47** **-2.11**	**22** **-0.50**	**24** **-1.62**	**37** **1.13**	**29** **0.02**	**40** **-0.16**	**12** **9.6**

Table 1. Trends in selected analytes measured by the Adirondack Lakes Survey Corporation

Forest Species Composition

Species composition is a critical factor in forest nutrient dynamics, especially with respect to the processing of N. Thus, factors that change the forest community (e.g., pests, climate change) could have a dramatic impact on how New York forests respond to atmospheric pollutant deposition. The influence of species composition has been shown clearly in a study of subcatchments within the Arbutus Watershed (near Newcomb in the Adirondacks). A subcatchment with a large component of sugar maple and white ash (species that show high rates of nitrogen mineralization and nitrification) exhibited greater losses of nitrate than other subcatchments. While American beech and sugar maple were the predominant species in terms of biomass, red maple was the only species to increase in biomass. However, total aboveground biomass decreased during the period, while annual litter production remained constant.

Speckled Alder

Speckled alder fixes substantial quantities of nitrogen along the lake inlets. Wetlands with abundant alder had 6 times higher nitrate accumulation than reference wetlands without alder. Surface waters in alder-dominated wetlands contained approximately 3 times more nitrate than non-alder counterparts. In specific alder-dominated wetlands, nitrogen addition from alder exceeds atmospheric deposition by as much as 3 times. These findings highlight the importance of several factors that affect aquatic N concentrations, such as the biogeochemical attributes of the watershed and landscape, including the presence of alder-dominated wetlands. The effectiveness of measures to reduce nitrogen emissions and deposition will depend on these factors.

Fish Species Richness

Comparisons with earlier studies show that declines in species richness have stabilized and that the trend may be reversing. Future surveys could ascertain whether there is a pattern of biological recovery following decreases in acidic deposition and improvements in water chemistry.

Soils

More than 75% of the 1,320 Adirondack lakes in the target population studied receive surface water drainage from watershed soils that have low exchangeable calcium concentration (less than 0.52 $cmol_c\ kg^{-1}$), base saturation (less than 10.3%; a measure of the soil's ability to neutralize acids), and pH (less than 4.5). Comparison of data developed in an EMEP study and data collected by the U.S. Environmental Protection Agency in the mid-1980s suggested that while lakewater chemistry was improving subsequent to large decreases in acid rain, Adirondack soil acid-base chemistry may have been continuing to deteriorate in most of the acid-sensitive watersheds. It appears that this deterioration in soil condition may have occurred even while lake chemistry was getting better. Such an effect would be expected to restrict the extent to which lakes will be able to recover in the future from acidification and might contribute to future adverse impacts on forest vegetation. Watershed soil is clearly the key to determining the extent to which Adirondack lakes will recover.

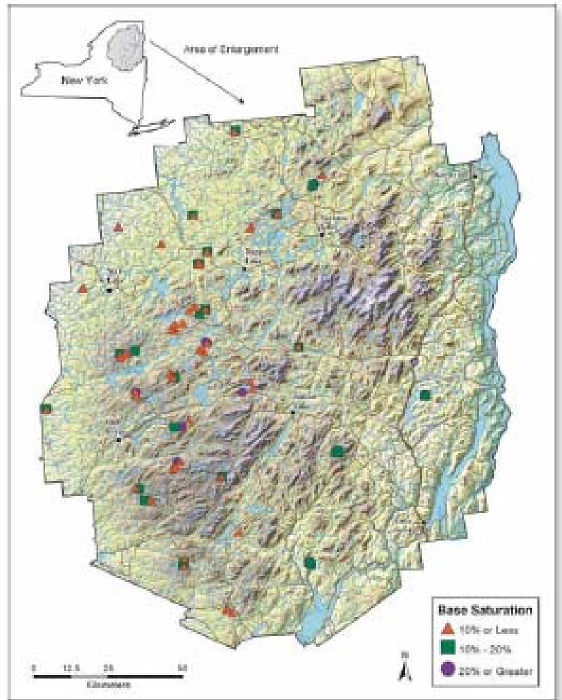

Figure 2. Soil base saturation for soil samples collected within the statistically-selected watersheds in the Adirondacks.

<u>*Supporting EMEP Projects:*</u>
Long-Term Monitoring Program for Evaluating Changes in Water Quality in Adirondack Lakes, Karen Roy, et al., No. 4915

Evaluation of the Recovery from Acidification of Surface Waters in the Adirondacks, Myron Mitchell, et al., No. 4917

Effects of Atmospheric Deposition of S, N, Hg on Adirondack Ecosystems, Dudley Raynal, et al., No. 6806

Assessment of Extent to Which Intensively-studied Lakes are Representative of the Adirondack Mountain Region, Tim Sullivan, et al., No. 7605

Assessment of Extent to Which Intensively-studied Lakes are Representative of the Adirondack Mountain Region

1.2 Catskill/Lower Hudson Valley Region

Atmospheric Deposition

This area has shown a decline in total (wet and dry) atmospheric deposition of both nitrogen and sulfur since 1988. Total sulfur deposition has declined substantially (1988–2000), as a result of reductions of regional SO_2 emissions. However, sulfur emissions from power plants in the Hudson Valley area have decreased much less than emissions from power plants in the Northeast as a whole. Statistical correlations indicate that deposition trends at the site were more influenced by regional than by local emissions. Atmospheric deposition of nitrogen has also generally declined in this area since 1988.

Sulfur
Sulfate concentrations in throughfall (water that falls to the ground following interaction with the forest canopy) and soil solution indicate a lower flux of sulfur through the ecosystem. Soil solution sulfur concentrations are much higher than throughfall concentrations and indicate a substantial internal source of sulfur in these forests, most likely from the weathering of the shale and slate bedrock in the area. Sulfate and calcium are highly correlated in throughfall and soil solution. The decline in atmospheric deposition of sulfur has decreased the leaching of calcium from the canopy and soil, despite the fact that mineral weathering adds a substantial amount of sulfur to this ecosystem.

Nitrogen
No decrease in inorganic nitrogen concentration in soil solution has been observed in the Catskill region. Under current nitrogen deposition conditions, there is minimal loss of nitrogen in Catskill drainage waters, suggesting almost complete retention of nitrogen. Nitrogen is strongly retained both in the forest canopy and in the soil, through complex plant, microbial, and abiotic processes. Overall, the nitrogen retention capacity for many forests in the Hudson Valley would be expected to be quite high because of the area's history of widespread burning and harvesting in the 19th century and the dominance of oaks, which tend to have low rates of nitrogen cycling and loss.

Forest Dynamics and Exotic Pests
These forests are affected by multiple biotic stresses, primarily owing to the introduction of exotic pests. The two most problematic, the gypsy moth and the hemlock woolly adelgid, have substantial effects on nitrogen cycling and retention. Gypsy moth defoliation can radically alter the forest nitrogen cycle. While this perturbation usually causes little nitrogen loss from the forest in drainage waters, under certain conditions, such as nitrogen saturation, nitrogen losses can be significant.

N Cycling Experiment
In the Hudson Valley region (specifically, the Institute of Ecosystem Studies forest in Millbrook), fertilized plots show signs of significant increases in foliar and litterfall nitrogen and in NO^{3-} leaching. The surprisingly high NO^{3-} leaching indicates that nitrogen saturation may occur at this site much earlier than expected based on other studies, suggesting that the ecosystem is susceptible to nitrogen saturation. This accelerated trend toward nitrogen saturation may result from characteristics of the site, which is an oak forest on thin soils in a ridgetop location. However, the site is representative of a major ecosystem type in southern New York State and is typical of sites attacked by the gypsy moth. This susceptibility to both nitrogen saturation and gypsy moth defoliation indicates that the interaction of these stresses should be further studied.

Catskill Streams
Data collected by the NYS Department of Environmental Conservation and U.S. Geological Survey in 1987 indicated that stream pH and ANC values are strongly related to the number of mayfly species in the Neversink River watershed. These data suggest that mayfly species diversity may be a good indicator of recovery following pH and ANC increases resulting from reduced levels of acidic deposition. Preliminary analysis of the 2003 data and comparisons with the 1987 data indicate a similar general pattern of downstream change in mayfly and fish species as pH increases in the Neversink River. There are, however, some differences between the two study years. In the process of comparing data from the two years and attributing any changes to reduced acidic deposition, the fact that 2003 was a very wet year has proven to be a challenge. Summer streamflow in 2003 in the Neversink River was the highest of any summer in the past 30 years. Current efforts are aimed at determining whether the primary cause of the subtle differences in stream chemistry and biota is, in fact, changes in acidic deposition, or simply the result of the very wet summer and the physical and chemical changes in stream habitat that accompany frequent high flows.

While the widespread recovery of streams in the Catskills is as yet unconfirmed, recent data do suggest that recovery may be underway in waters with ANC values in the range of 30–70 μeq/L. These potential early improvements in stream chemistry may eventually result in the recovery of acid-intolerant biota in affected streams. This project's comparison of 1987 and 2003 data from the Neversink River will contribute to our understanding of how reductions in the acidity of deposition affect water chemistry and biota. Additionally, the data accumulated by the project will provide a basis for comparison against which future changes in the acidity of precipitation and in the acid-base status of surface waters in the Neversink basin may be evaluated.

Supporting EMEP Projects:
Integrated Assessment of the Recovery of Surface Waters from Reduced Levels of Acid Deposition in the Catskills and Adirondacks, Burns, et al., No. 6486/6490

Monitoring Deposition and Effects of Air Pollution in the Hudson Valley, Lovett, et al. 6819

Potential Recovery of Water Chemistry and Stream Biota from Reduced Acid Deposition at a Sensitive Watershed in the Catskills, Burns, et al., No. 7606

1.3 Statewide Issues Related to Acid Deposition

Nitrogen

Reactive nitrogen originates from numerous sources and has complex relationships with other pollutants. It therefore requires integrated management strategies and policies addressing multiple rather than individual sources. The assessment of nitrogen pollution in the Northeast region shows that the current CAAA has not yet had a substantial effect on airborne nitrogen emissions. It is important to note that there is currently neither an air quality standard for ammonia nor a water quality standard that limits the total loading of reactive nitrogen to surface waters. Together with efforts to reduce SO_2, CO_2, and other pollutants, nitrogen in the Northeast can be further decreased through a number of strategies: reducing power-plant nitrogen emissions, improving wastewater treatment to remove nitrogen from effluent, reducing the use and increasing the efficiency of nitrogen fertilizers, and creating and restoring natural nitrogen sinks in wetlands and floodplains. A number of innovative nutrient management projects currently being implemented on farms throughout the region provide hopeful examples.

Modeling results offer insight into the relationship between different emissions reduction scenarios and ecosystem recovery. Appraisal of the rate of recovery of different Northeast ecosystems under a variety of policy scenarios indicates that:

- controls on vehicle and electric utility NOx emissions produce the largest reductions in airborne nitrogen pollution,
- nitrogen removal from wastewater at a basin-wide scale is the single most effective means of reducing nitrogen loading to estuaries in the Northeast.

The Role of Sugar Maple in the Nitrogen Cycle
In sugar maple stands, rates of nitrification and nitrate leaching are higher than in those of other common hardwood and softwood species. A number of factors may account for this phenomenon, including the rate at which leaf litter decomposes in the soil and soil chemistry in the stands. Although sugar maple is the most dominant species in northern hardwood forests of New York, conditions such as beech bark disease, climate change, soil calcium depletion, and pest infestations may affect the future abundance of sugar maple and consequently nitrogen cycling and ANC trends in these regions. Thus, future trends in

nitrate concentrations in surface waters may depend on changes in tree species composition, which is determined by multiple environmental factors.

Soils—Statewide

EMEP research studying soils statewide is still in its early stage. It is known that an accurate assessment of the threat of Ca depletion by acid rain and forest harvesting depends on understanding the contribution of apatite weathering to Ca cycling in forest soils. Apatite is important in granitoid parent materials (igneous rock) but not in sedimentary rocks. The soil parent materials derived from clastic sedimentary rocks averaged 80 ppm Ca from apatite, compared to 720 ppm in the parent materials derived from igneous rocks. Parent material is likely more important than glaciation in determining apatite availability in soils. Sugar maple decline has been observed to occur on unglaciated soils of the Allegheny Plateau; glaciers tend to rejuvenate reservoirs of fresh minerals available for weathering. Roots and fungi, including mycorrhizae, play a role in mineral weathering, and it has been suggested that apatite weathering is at least in part under biotic control.

Information concerning the distribution of sources of Ca and the ability of tree species to access Ca is essential for predicting the sensitivity of forests across New York State to Ca depletion. The data accumulated through EMEP-funded research will provide a more accurate assessment of the likelihood that New York State forests will be adversely affected by the depletion of calcium and other exchangeable cations as a result of acidic deposition. This information is vital for the development of sound policies for managing and protecting New York's forest resources. Additionally, carbon storage in forest soils is of increasing interest because of the need to account for C storage in soils when using forests to offset emissions of CO_2.

Supporting EMEP Projects:

Status and Effects of Nitrogen Pollution in North Eastern United States , Lambert, et al., No. 6487

Evaluation of the Recovery from Acidification of Surface Waters in the Adirondacks, Myron Mitchell, et al., No. 4917

Assessing the Sensitivity of New York Forests to Cation Depletion, Yanai, et al., No. 8649

2. Mercury

2.1 Ambient Measurement and Deposition

Historical Patterns of Mercury Deposition in the Adirondacks

Concentrations of total Hg were measured in sediment cores collected from eight Adirondack lakes. Historical profiles were determined for each lake showing changes in sediment Hg deposition over the past 200 years. Although there were lake-to-lake variations, on average sites showed a 5.8-fold increase in sediment Hg deposition from background values (before 1900) to peak values. Mercury deposition peaked from 1973 to 1995 and decreased in recent years. Conducting this analysis for pre-anthropogenic conditions, the deposition of Hg to the surface of a perched seepage lake (a lake that receives all of its inputs directly from precipitation, with no watershed inputs) was estimated to be 3.4 ± 1.1 $\mu g/m^2$-yr. For modern conditions, the deposition of Hg to the surface of a perched seepage lake was determined to be 8.6 ± 2.4 $\mu g/m^2$-yr; a value similar to current estimates of wet Hg deposition for the Adirondacks. Watershed Hg retention by year was estimated using sediment deposition data across the study lakes. It was observed that the retention of Hg in Adirondack lake-watersheds has decreased over the past 200 years. The mechanism responsible for this decline is unclear.

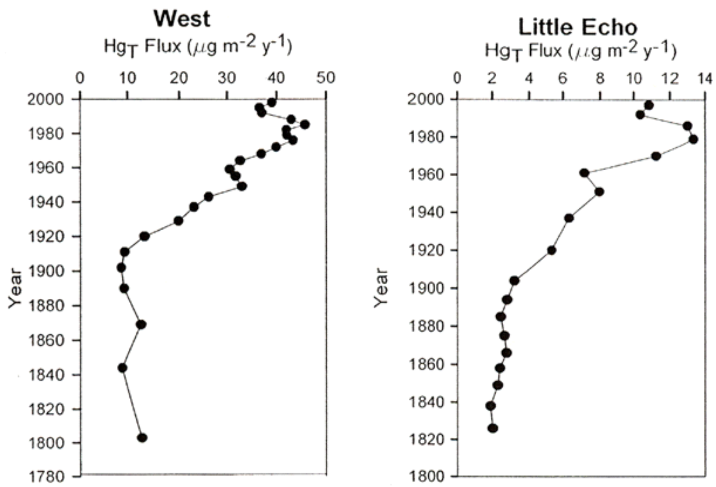

Figure 3. Sediment Hg deposition to West Pond and Little Echo Pond in the Adirondacks over time. Increases in sediment Hg deposition in the early 1900s have been followed by decreases in the latter portion of the century. Note Little Echo Pond is a perched seepage lake and is probably representative of direct changes in wet Hg deposition.

Mercury Deposition Network (MDN) Sites

The last complete year for which wet mercury data are available is 2004. The annual volume-weighted concentration of total mercury for Huntington Forest for 2004 is 6.7 ng/L, with values ranging from 0 to 27.7 ng/L. The annual wet deposition of mercury is 6.9 μg/m^2-yr, with weekly deposition ranging from 0 to 650 ng/m^2-wk. These values are similar to values reported in the network for other sites in eastern Northern America. The time-series of annual volume-weighted concentrations of mercury in wet deposition at Huntington Forest for the period the site has been in existence in shown in the figure below. These values are also compared to values at Biscuit Brook in the Catskills for 2004. There have been no significant trends in mercury deposition at the Huntington Forest since measurements were initiated at the site.

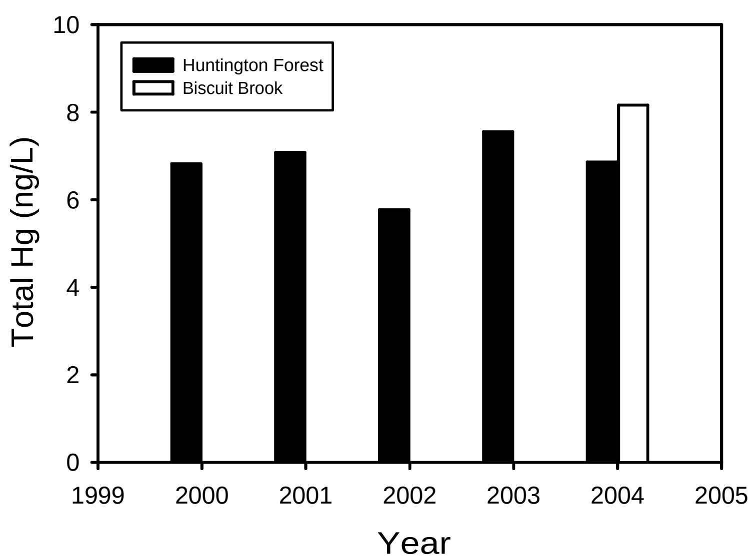

Figure 4. Time-series of annual volume-weighted concentrations of total mercury in wet deposition at Huntington Forest. The concentrations of total mercury at Huntington Forest are compared to values for Biscuit Brook for 2004.

Although the MDN site at the Huntington Forest has only been in existence for a short period, the data collected have been extremely valuable in the interpretation of mercury deposition for the region. Moreover, if there are additional reductions in atmospheric emissions of mercury, such as those proposed for electric utilities, data from this site could provide a reference from which it will be possible to detect changes in wet deposition if they occur.

Using information on wet deposition at Huntington Forest, and throughfall and litter deposition on mercury obtained from Sunday Lake and Huntington Forest, Dr. Charles Driscoll (Syracuse University) is developing maps of total mercury deposition for the entire Adirondack region.

The work performed at the MDN stations also benefits other studies of mercury in the region, such as the NYS Department of Environmental Conservation's extensive survey of mercury concentrations in fish in NYS waters. As part of the MDN, these sites will contribute to a national database of weekly concentrations of total mercury in precipitation and seasonal and annual fluxes of total mercury in wet deposition. Dry deposition of mercury is not currently measured at the New York State MDN sites.

Ambient Mercury Measurement

New rules will soon be going into effect for Hg emissions, creating a need for both monitoring and research measurement to characterize and understand sources, sinks, and transformation processes of Hg. An added challenge is the various forms of Hg in the atmosphere: Hg, Hg (2+) or Reactive Gaseous Mercury (RGM), particulate mercury, and Hg in wet deposition. A comparison of manual measurements of gaseous mercury is being performed with an automated Tekran 2537A in the laboratory and at a field site in Postdam, NY. This project will likely be the basis of a Hg monitoring network in New York State. Based on the lessons learned in this project, the New York State Department of Environmental Conservation (NYSDEC) has submitted a proposal to EPA for expansion of Hg monitoring in New York State.

2.2 Mercury Transport

Computer modeling conducted in 2001 showed U.S. emissions (non-NYS) to be the largest source of total Hg deposition in New York State. Overall, results from local deposition and long-range transport scenarios vary only slightly from those of the baseline scenario. U.S. emissions were the largest source of total Hg deposition at all three New York State sites in the model. Dry deposition was dominated at all sites by NYS and U.S. emissions, while wet deposition showed a much greater contribution from sources outside North America.

Hg transport within and around NYS, and consequently the effects of individual point-source emissions on Hg deposition in NYS, are strongly influenced by small-scale meteorological features. As a result, fairly high resolution modeling of the air flows in NYS would be needed in order to accurately assess the impacts of point sources of mercury on deposition in NYS.

Major sources of uncertainty include emissions (including Hg species), the dry deposition of Hg(II), and, for lake mercury cycling models, the sediment burial rate. Knowledge of atmospheric Hg chemistry is still incomplete; laboratory experiments are needed to identify and characterize the most important reactions. Finally, there is a great need for Hg measurements with which to evaluate the models, including ambient atmospheric concentrations of Hg(0), Hg(II), and Hg(p), and wet and dry deposition rates of mercury.

2.3 Cycling of Mercury in Watersheds

The Sunday Lake watershed is located in the western part of the Adirondack Park in New York State, just west of the Stillwater Reservoir. This lake-watershed system was selected for mass balance analysis. Sunday Lake is a small lake surrounded by a much larger watershed; therefore, water moves through the lake rapidly, resulting in a detention time on the order of two weeks. Wetlands cover approximately 20 percent of the Sunday Lake watershed, with most located adjacent to lakes and streams (riparian wetlands). Wetlands border 65 percent of all the stream channels and much of the lake shorelines.

The dynamics of Hg were studied within upland coniferous and deciduous forest plots at the Sunday Lake watershed. The flux of Hg to the forest ecosystem was dominated by dry deposition, which was estimated as throughfall plus litterfall. These inputs accounted for 70% of total deposition. (Note: we have very little state/national data of what appears to be a dominant mercury flux—dry deposition.)

Concentrations of total Hg attached to soil varied in depth but were similar among plots with highest concentrations in the surface organic layer, lowest in the leached layer immediately below the organic layer; concentrations rose again in the upper mineral soil, and lower values were found in the deeper mineral soil. Total mercury concentrations in soil porewaters were highest in waters draining the forest floor, decreasing through the mineral horizons. Concentrations of MeHg attached to soil were relatively uniform throughout both forest soil profiles, whereas porewater MeHg concentrations were highly variable.

Hydrology

During the period of study, precipitation averaged 126.5 cm, compared to an average of 77.4 cm of stream flow. Total stream runoff was thus 61% of precipitation, with the remaining 39% assumed to be lost to evapotranspiration (ET). High flows tended to occur in the fall and spring, while the lowest flows generally occurred in late summer.

Most of the soils in the Sunday Lake watershed have relatively high infiltration capacities, which means that the vast majority of water that enters streams and lakes first moves through watershed soils. Approximately 20% of the total groundwater input enters the streams through the riparian wetland areas.

Groundwater levels fluctuate through the interface between the organic horizon and the mineral horizons of the wetland soils. The riparian wetlands discharge water to the streams from surrounding soils throughout most of the year. During rain storms and snowmelt events, the groundwater flow direction in the riparian wetland can be reversed, with water from the stream mixing with the groundwater. This may have the effect of moving more active forms of mercury and organic matter into wetlands with very active microbial communities that methylate mercury.

Lake Watershed Mass Balance

Wet Hg deposition was 10.3 $\mu g/m^2$-yr, with 0.6 percent of Hg occurring as MeHg. Forest vegetation was important in mediating the inputs of Hg to the forest floor. For the watershed as a whole, we estimate total deposition of Hg to the forest floor to be 33.9 $\mu g/m^2$-yr. This flux of Hg is considerably greater than estimates of Hg lost from the forest floor by drainage water (13.9 $\mu g/m^2$-yr). This discrepancy in Hg mass balance suggests that Hg is accumulating in the forest floor and/or Hg is lost by volatilization. Fluxes of HgT decreased with depth in soil water, coinciding with decreases in DOC. This pattern suggests that Hg is immobilized in the mineral soil by the deposition of organic matter. Concentrations of Hg in surface waters ranged from 1.9 to 4.6 ng/L. Atmospheric Hg deposition was retained in the watershed, and Sunday Lake was a sink for inputs of total Hg.

Considerable enrichment in MeHg was evident in throughfall and litterfall when compared to the flux in wet deposition. As observed for total Hg, drainage fluxes of MeHg were lower than drainage losses from the forest floor. This pattern suggests that the forest floor is an active zone of demethylation. Fluxes of MeHg were elevated in waters draining riparian wetlands. This flux of MeHg can largely explain the MeHg input to Sunday Lake. MeHg concentrations in surface waters of the Sunday Lake watershed ranged from 0.2 to 2.53 ng/L. The mass balance indicates that the watershed, particularly riparian wetlands, and the lake were net sources of MeHg to downstream surface waters.

2.4 Biota

Determination of levels of abiotic and biotic mercury exposure will contribute to the assessment of the risks posed by mercury pollution to human and ecological health in NYS and the Northeast. Of the loons sampled in the Adirondacks, 18% are estimated to be at risk for harmful effects from mercury contamination. Additional results from sampling in 2005–2006 will augment the database that is being developed. Behavioral impacts are now being summarized. Results will be coordinated with related projects, with the aim of improving assessment of the impacts of mercury exposure on loons. Findings should prove useful in broader evaluations of the impact of mercury toxicity and the extent of mercury distribution in northeastern aquatic ecosystems in relation to deposition. The evaluation of mercury levels in prey fish will also aid in identifying Adirondack lakes that should be evaluated for fish consumption advisories to prevent human exposure to mercury.

Fish and water samples have been collected from 131 lakes across New York State. More than 1700 fish samples and over 100 water samples have been analyzed. The NYS Department of Health has recently added numerous waterbodies, including all waters of the Adirondacks and Catskills, to their fish consumption advisories based on this data.

Supporting EMEP Projects:
Mercury in Adirondack/Catskills Wetlands, Lakes and Terrestrial Systems, Munson, et al., No. 4916

Contributions of Global and Regional Sources to Mercury Deposition in New York State, Seigneur, et al., No. 6485

Atmospheric Transport and Fate of Mercury in New York State, Walcek, et al., No. 6488

Mercury Deposition Monitoring in the Catskills, McHale, No. 6818

Mercury Deposition Monitoring in the Adirondacks, Driscoll, No. 8152

Long-term Monitoring and Assessment of Mercury Based on Integrated Sampling using the Common Loon, Prey Fish, Water, and Sediment, Schoch, et al., No. 7608

Strategic Monitoring of Mercury in New York State Fish, Simonin, et al., No. 7612/7716

Impact of In- and Out-of-State Power Plants on Semivolatile Pollutants in New York State, Hopke et al., No. 6083

3. Knowledge Gaps/Research Needs Identified Through the EMEP Projects

A quantitative understanding of the factors other than acid precipitation that may affect surface-water pH and ANC is necessary to separate the effects of clean air laws and atmospheric deposition from natural variability and disturbance.

While the acidity of deposition has declined in New York over the past 20 years, insufficient data exist for determining whether the acidity of streams has decreased in response to these developments. More adequate baseline data will also be needed for identifying and evaluating future trends in the acidity of soils and streams. In the absence of sufficient information, policy efforts at restoring and protecting these natural resources will have to be made with a considerable degree of uncertainty.

Uncertainties remain in our understanding of the atmospheric fate and transport of mercury. Major sources of uncertainty include emissions (including Hg species), the dry deposition of Hg(II), and, for lake mercury cycling models, the sediment burial rate. Knowledge of atmospheric Hg chemistry is still incomplete; laboratory experiments are needed to identify and characterize the most important reactions. There is also a great need for Hg measurements with which to evaluate the models, including ambient atmospheric concentrations of Hg(0), Hg(II), and Hg(p), and wet and dry deposition rates of mercury. Other needs also related to Hg include the fate of soil Hg, soil re-emissions, and the response of fish to changes in Hg deposition and acid deposition. Also of note is that the available mercury-in-fish database currently includes information on fish for only ~5% of NYS lakes.

Peer-Reviewed Publications Supporting this Summary

2000

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Hurd, T.M., D.J. Raynal and C.R. Schwintzer, 2001. Symbiotic N-fixation of Alnus incana ssp. rugosa in shrub wetlands of the Adirondack Mountains. *Oecologia* 126: 94-103.

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