Acid Deposition Research Needs in Massachusetts

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Marie-Françoise Walk

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MA Water Resources Research Center
Blaisdell House - 310 Hicks Way
University of Massachusetts
Amherst, MA 01003
www.umass.edu/tei/wrrc
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Executive Summary

The Massachusetts Water Resources Research Center, on a contract for the Massachusetts Department of Environmental Protection, convened a summit on research needs for acid deposition in Massachusetts on January 25, 2007 at UMass Amherst.

Fifteen experts participated and identified gaps in current knowledge and future research needs for acid deposition. While there has been much research in the past 30 years devoted to acid deposition and its effect on the environment, changes in pollution emissions and the recognition of climate change impacts expose gaps in past studies and point to new research needs.

This report is arranged into seven areas of environmental interest including emissions, soils and forests, surface water, estuaries, fish and wildlife, groundwater, and climate change, and includes recommendations for further study regarding acid deposition in Massachusetts.

The research areas that surface as most immediate and within reach of the MA DEP include:

Emissions
Monitor trends and provenance of NOx emissions, quantify local emissions, and add trace elements including mercury.

Soils/Forests
Monitor soils, especially nutrients and the effect of emissions on soil respiration and carbon; compare present forest canopy to 1980s data.

Surface Waters
Continue long-term monitoring at sensitive sites, add mercury and nitrate monitoring in roadside streams.

Fish and Wildlife
Continue long-term research, add biological monitoring (keep tabs on indicator fauna species), study determinant mercury levels in fish.

Estuaries
Determine percent N due to acid deposition in estuaries.
Introduction

On behalf of the MA Department of Environmental Protection (MADEP), the MA Water Resources Research Center convened a summit on research needs for acid deposition in Massachusetts. The summit was held at the University of Massachusetts Amherst on January 25, 2007.

The purpose of this summit was to assemble experts\(^1\) in the field of acid deposition research to discuss the current knowledge on this topic, and from this baseline, identify gaps in current knowledge and future research needs. The goal was to provide the Commonwealth of Massachusetts with recommendations for future funding. This report summarizes the results of the summit, covering what is known, unknown, and recommendations for further study regarding acid deposition in Massachusetts.

The discussion is arranged into seven topics of interest including emissions, soils and forests, surface water, estuaries, fish and wildlife, groundwater, and climate change.

1. Emissions

Current Knowledge
The 1990 Clean Air Act Amendments mandated a 50% reduction in SO\(_2\) emissions and a two million ton reduction in NO\(_x\) emissions compared to their 1980 levels. As of several years ago, power-generated emissions had indeed decreased significantly: by 35% for sulfur and 18% for nitrogen (Kahl et al, 2004). Emissions in Massachusetts are down 62% for NO\(_2\) and 64% for SO\(_2\) since 1985, and are down 70% for mercury since 1998 (EOEA 2006). Deposition rates of S and N have decreased as well although at lower rates than emission reductions. However, both in the region and Massachusetts in particular, nitrogen emissions from vehicle sources have not decreased and have even increased.

The following two graphs illustrate emission trends over the past 15 years.

\(^1\) The list of summit participants can be found on page 17 of this report
The National Atmospheric Deposition Program (NADP) is a nationwide network of precipitation monitoring sites and has documented geographical and temporal long-term trends in precipitation chemistry since 1978. There are three NADP sites in Massachusetts: MA08 at the Quabbin Reservoir in Belchertown, MA13 in Lexington, and MA01 in Wellfleet on Cape Cod (see map below).

Measurements from 1981 to 2005 of pH, SO$_2$, and NO$_x$ at these three sites show an increase in pH and a decrease in sulfur dioxide and nitrogen oxides (see next three pages).
Despite these encouraging trends, scientists and government agencies agree that further emission reductions are needed for the recovery of the environment and to prevent further acidification. Furthermore, it is now recognized that particulate matter PM$_{2.5}$ (of which acid aerosols are one component) is important regarding health effects, and should be addressed in emission controls.

**Knowledge Gaps**

**Emission source apportionment needs better understanding, via trace element analysis.** The Casco Bay Estuary Partnership (<http://www.cascobay.usm.maine.edu/oldsite/toxics.html>) has studied precipitation and trace elements (nitrogen, mercury and fine particulate matter pollution) and recommends that other states including Massachusetts join them in monitoring mercury emissions. Measurement of ambient inorganic nitrogen concentrations is also needed (Ryan et al, 2003). EPA Region 1 is working with NADP’s National Trends Network (NTN) program with consultants and recommends combined aerosol and wet precipitation event information to obtain better information. In the Northeast, potential emission sources may come from the TVA to the Mississippi River and east. However, the net effect may not be as much as expected because some may be due to out-of-area influences.

There is also a need to augment existing data sets for trace elements and obtain episodic data, particularly for nitrogen emission sources, because unlike sulfur emissions which are clearly decreasing on a regional basis, nitrogen emissions are variable.

A good model to collect future emission data is GEOSS: Global Earth Observation System of Systems (<http://www.epa.gov/geoss/>). The idea is to collect modeling and observational information and as many data sets as possible, into a common reporting system (e.g. incorporate NASA, NOAA, EPA).

**Critical loads for sulfates and nitrogen need to be determined in three areas: estuaries, forests, and forest/soil/water interface.** Recovery targets should be reconciled with regulator tools. SIPS (state implementation plans), which are used to comply with the Clean Air Act, are involved. EPA has studied how much reduction is needed to restore lakes and streams. In its 2005 Acid Rain Program Report, EPA describes further caps on SO$_2$, NO$_x$, and mercury emissions (EPA, 2005). However, scientists feel that emission regulations should be more closely tied to critical load calculations.

**Research Needs**

- There is a clear need to document the trend in NO$_x$ emissions, as nitrate has not been responding as much as sulfate to controls. It will be important for future regulations to determine the origin of N emissions, using for example isotope tracers of N or developing new tracers to find sources of N. Wet and dry deposition data should be combined to obtain a clearer picture of emission trends. NO$_x$ emission source data should be plotted and compared with nitrate deposition rates.

- Trace elements, including mercury, need to be added to acid deposition monitoring programs. For example, fund additional monitoring at the Quabbin site – a few times at least, preferably doubling up the number of event-based data points. An annual amount of $50,000 was estimated to cover the cost of this additional monitoring.
- **Mercury release inventories** are needed, especially on mercury product breakage and mobile sources.
- **Documentation of deposition sources**, including in-region and out-of-region contributors.
- **Quantify local emissions, especially from automobiles.** Such low-level emissions are not adequately quantified in the current deposition monitoring programs. Since many highways parallel streams in the state, low-level emissions are likely to have a more direct impact on streams than emissions from long-range sources. This is also true for dry deposition and likely an even greater source of nitrogen than wet deposition. The fact that highways are designed to efficiently channel water off the roadways to nearby streams further enhances the likelihood that these emissions will move directly into surface waters with little contact (and retention) with terrestrial surfaces. Though research is needed to translate critical loads into necessary control programs, since NADP already has a funding program for critical load calculations, this is probably not an area that MADEP needs to fund.

2. **Soils and Forests**

**Current Knowledge**
An important problem caused by acid deposition is due to the depletion of nutrient base cations (Ca\(^{2+}\), Mg\(^{2+}\)) compounded by the mobilization of Al and the accumulation of S in soils. The continued acidification of soils and losses of Ca and Mg harms forest health, reducing the supply of available nutrients for tree growth (both because of fewer nutrients uptake from the soil and more nutrients leached from leaves). “The change in soil nutrient ratios (lower Ca/higher Al) can disrupt physiological processes that are important to maintaining forest health. These changes lower resistance to natural stresses, such as insect, disease and climatic extremes” (NAPAP, 2005). In Massachusetts, Miller (2003) shows that “48% of Massachusetts forests are sensitive to acid deposition: deposition of sulfur and nitrogen exceeds the level at which harmful ecological effects occur (critical load).” Soil chemistry changes cause concern for high and mid-elevation spruce-fir forests in New England, and “recent evidence of damage to sugar maples in the Northeast has also emerged” (Chestnut, 2005).
Increasing nitrate deposition may cause soil respiration problems, which may be an important issue. The Hubbard Brook research in New Hampshire shows significant nitrate leakage – increasing during the 1980s and maintaining similar levels during the 1990s.

**Knowledge Gaps**

Better understanding of the **impact of nitrogen decrease on biological effects** is needed. “Nitrogen is a limiting nutrient to most terrestrial ecosystems, but may be a pollutant in aquatic ecosystems and drinking water. Therefore there are many fundamental and applied questions regarding the impact of long-term additions of nitrogen to our landscape” (Harvard Forest, 2007 web site). Nitrogen may play a dual role in critical loading: as a nutrient and as an acidifier. If N emissions are reduced, this might affect biological responses and have an impact on ecosystem processes. The nutrient part of the equation needs to be better understood. More study is needed to understand nitrogen effect on soil respiration and whether increased N levels due to acid deposition lead to an increase in soil respiration. In the Midwest and at Harvard Forest in Massachusetts (Micks et al, 2004), research has shown that nitrogen increases affect soil respiration. Will decreases in N emissions lead to forest stress?

Another issue is **whether acid deposition is leading to forest change and is responsible for tree decline in Massachusetts**. Some satellite research has found that acid deposition is related to red spruce damage on Mount Greylock. Tree decline has been shown to be concurrent with increases in acid-tolerant ground vegetation, but scientists are not sure how to interpret this phenomenon and the connection needs more study. Soil acidity is also causing changes in forest composition and age.
The role of forest changes is unclear. There seems to be soil acidification once a forest is disturbed or this may be due to the forest maturation cycle. Conventional wisdom is that tree uptake of nitrates and nutrients gradually decline with forest maturation, and that more nitrogen migrates to water. Land use can also have an impact (e.g., agriculture, fires) so future research needs to control for land use variation.

More soil data related to acid deposition is needed, as well as long-term studies, as the time scale known to replace cations in depleted soils is on the order of centuries.

**Research Needs**
- **More soil monitoring is needed, both in solid and solution phases.** Usually, too many samples are needed to practically monitor, so sampling protocols are needed. Coordinate with the Northeastern Soil Monitoring Cooperative <http://www.czen.org/node/364>.
- More research should be conducted on soil nutrients, such as nitrogen, and the effect of changes in emissions on soil respiration and carbon.
- Soil sampling should be added to volunteer monitoring programs and the soil samples archived for later analysis when resources are available.
- Forest health needs to be assessed; One suggestion would be to work on the plots surveyed by the Forest Inventory Analysis Project <http://www.fs.fed.us/ne/fia/states/ma/index.html>. Tree cores can provide some record of soil history (see Mt. Holyoke Range tree cores). Another suggested project would be to update the 1980s infrared overflights to compare the current forest canopy with the 1980s images.

**3. Surface Water**

**Current Knowledge**
The decrease in base cations in soils prevents the neutralization of acid inputs in soil water, leading to acidification of recipient surface waters. “Acid episodes will continue until the availability of S and N is further reduced and the base status of the mineral soil is improved” (Lawrence, 2002).

Though there are fewer acidic lakes in the Adirondacks now, the change in New England lakes since the early 1990s is insignificant (5.5% of lakes are acidic now compared to 5.6% in 1990, NAPAP, 2005). This is despite a moderate but significant increase in ANC and pH in most regions, and modest decreases in Al concentrations. In Northeast surface waters, base cations such as Ca\(^{2+}\) and Mg\(^{2+}\) declined at a rate between -1.5 and -2.5 μeq/L/yr; and sulfate decreased -2.5 μeq/L/yr and nitrate decreased by 0.5μeq/L/yr (Kahl et al, 2004).

In Massachusetts, the Acid Rain Monitoring (ARM) project shows no trends up or down in surface water pH. ANC represents the only obvious shift in trend slopes, having increased in 2000-2005 compared to 1980-1993. The significance of change in all parameters is based on paired t-tests of data from the 22 sites between the 1983 - 1993 period and the 2001 - 2005 period adjusted for season and hydrology as shown in the graph below.
Sulfate shows a strong and significant decline. Aluminum appears to be increasing but not significantly. Calcium, magnesium, manganese and potassium exhibit little change, though perhaps a slight downward trend. But sodium has a strong trend for increase, about the same slope as chloride. One might conclude that the base cations that are important for acid deposition recovery are not increasing whereas those resulting, very likely, from road salting practices but not relevant to acid deposition recovery are sharply increasing. Color also shows a significant increase. When acid deposition titrates a natural water body, usually we think about loss of ANC. But in New England, there is another buffer that is rarely considered - organic acids. These naturally make waters somewhat acidic and tea brown in color but they act as buffers against further lowering of the pH by mineral acids. So naturally colored waters have been titrating acid deposition and becoming less colored. ARM measured this as standard color units. The trend is for color to have significantly increased since ARM began, offering a hopeful sign that the other buffer system is also responding (Godfrey, 2005-2007). Godfrey concludes that our lakes may be nearing a return to conditions of the mid 1980s, when the situation of Massachusetts surface waters was serious with regards to population losses due to acid deposition and worsened until the late 1980s.

A similar trend in CI increases has been found in urbanized lakes in southern New England (Rosford et al, 2007). In these lakes, a corresponding increase in Ca was observed and was assumed to be due to an exchange with Na. This study found that base cation concentrations increased in lakes with high CI, but decreased in lakes with low CI. This pattern creates interference with the detection of acid deposition-caused trends in lake chemistry.

Most research looks at baseflow trends in surface waters. Episodic acidification (during snowmelt or large storms) is also a concern. According to Lawrence (2002), “Atmospheric deposition of S has decreased about 40% in the 10 years since completion of ERP data collection, yet acidic episodes in streams of upland regions in the northeastern United States persist and are likely to be much more widespread than chronic acidification.”

The EPA’s New England Wadeable Streams (<http://www.epa.gov/Region1/lab/news.html>) program assessed the ecological conditions of headwater streams across the New England region in 2001-2003 (three
sites in Massachusetts), processing pharmaceuticals, baseline chemistry, and biology for fish caught at hundreds of locations. As wadeable streams will be the first to respond to watershed changes, this provides a good database to build on.

**Knowledge Gaps**

DOC increases have been shown in Europe, N. America. A doubling of DOC was found in a Hudson River study. One issue is **whether observed DOC increases are masking an improvement in surface water pH.**

One factor that has largely been left out of current research is hydrology. The **effect of flow on water chemistry** is not clear. This is of particular interest for sulfur. Under low flow, sulfur is associated with the atmosphere. Under wetter conditions, it is more soil related, but it has been hard to detect flow related effects, as there isn’t much flow data to go with the chemistry data.

Another issue is **whether the Na and Cl increases documented in the ARM project mask more important decreases in base cations.** Other research confirms an increase in chloride, but not in sodium, as it can exchange for other base cations and aluminum.

There is currently little EPA-led stream research in New England. **Additional biological and chemical monitoring of streams** is needed in Massachusetts. Base cations in surface waters continue to be an issue. They are showing a slight decrease but more information is needed to understand why and whether they might be retained in soils.

**Lakes monitoring** shows widely variable change in base cations and a better understanding of the cause is needed. Land use change may be one reason. Site selection is very important in monitoring for acid deposition.

Monthly stream monitoring in Pennsylvania shows some recovery: nitrates, sulfate, and some cation reductions were observed in baseflow in streams, as well as in increase in base cations. But there is also an increase in silica, DIC, sodium, and a general downward trend in DOC. A better understanding of these changes is needed, particularly as these are relatively undisturbed small forested watersheds largely unaffected by forest cutting. Possible variables are rise in soil respiration, breakdown in organic matter, or changes in bio-geochemistry.

There are many roadside streams in Massachusetts and one question is whether there are localized impacts such as from automobile exhaust. **Comparisons** are needed with other **regional studies.**

**Research Needs**

- **Coordinate biological and chemical programs.**
- **Investigate the source of sodium and chloride increases in surface waters** further and whether sodium increase is correlated with a decrease in base cations. Data and sites should be examined carefully, particularly regarding road salt contamination, as high chloride can obscure the data set, and can also originate from other sources such as septic systems and water softeners.
- **Continue long-term research at acid sensitive sites** to determine cause and effect of the relationship between base cations and sulfate, and the
effects of cation loss on soil and surface water recovery, and relate to mercury.

- Because recovery of water bodies is long-term, and it is difficult to find statistically robust connections, it makes sense to **continue to build data at existing sampling sites whenever possible.**
- **Monitor streams in relatively undisturbed forested watersheds.** Select sites not affected by land use changes and select sensitive watersheds or reservoirs at 20 locations, coordinated with ARM. Incorporate **both a broad brush approach** (e.g., ARM with many lakes) and **more intensive and long term monitoring** at a few sites (e.g., Cobble Mountain Reservoir, Quabbin, Upper Naukeag in Ashburnham) and monitor to determine if recovery is occurring or not, especially where there are USGS gauges. Work collaboratively with long-term broad-scale projects such as the Harvard Forest project.
  - Combine projects to **include both acid rain and mercury research.**
  - **Monitor mercury and nitrates in roadside streams.**
  - **Study ecological impacts of mercury.**

### 4. Estuaries

**Current Knowledge**

Another documented problem associated with acid deposition in the Northeast is the eutrophication of coastal estuaries, due in part to excess nitrogen contributed by air emissions of NOx (Chestnut et al, 2005).

The National Estuary Program is conducting much research in this area. In Massachusetts the central point of this effort is the Narragansett Bay Laboratory. Woods Hole Oceanographic Institution is also working in Waquoit Bay and Great Bay. The nitrogen issue is important in these efforts, but acid deposition is probably insignificant compared to N and sewage in terms of research focus.

**Knowledge Gaps**

The main **uncertainty is whether acid deposition is a significant source of excess nitrogen in estuaries and bays.**

**Research Needs**

- **Determine the percent of N due to acid deposition in estuaries** (use nitrate isotope N^{15}O^{18}).
- **Monitor mercury in estuaries and local saltwater fisheries**, including shellfish.
- **Create a mussel watch program.**

### 5. Fish and Wildlife

**Current Knowledge**

The status of aquatic organisms’ health affected by acid deposition does not seem to be well documented, though “Five years of testing conducted by MassDEP on average mercury levels in fish across the Commonwealth indicate a 15 to 32% drop in mercury concentrations” (EOEA, 2006). But an “inventory and analysis of sediment mercury (Hg) concentrations from 579 sites across northeastern North America data indicate that at least 44% of waters across the region have sediment
Total Mercury concentrations in excess of Canadian and United States minimum sediment contaminant guidelines for the protection of aquatic biota” (Kamman et al, 2005).

One study by Bowman et al (2006) in south-central Ontario concludes that although there has been short-term recovery of BMI communities, “overall improvements have been hampered by acid or metal toxicity associated with drought-induced re-acidification of the streams.”

Mercury levels have been monitored in Massachusetts since 1994. MA DEP has examined lake sediment chemistry and mercury in fish tissues. An area of concern was found in the Merrimack River valley near incinerators. Around 2000, aggressive state reduction programs began, resulting in up to 90% emission reductions in the areas of concern with less impact in other parts of the state. Long-term monitoring of mercury in fish tissue is ongoing with sampling at 10-20 lakes per year. Reductions in levels in yellow perch and largemouth bass have been identified. Low pH lakes were pulled out of the study because they were considered outliers. In future sampling they might be added back into the study for comparison with ARM data. MADEP is also looking at land use changes working with Harvard Forest.

A survey on zooplankton health in more than 100 lakes has recently been completed by Plymouth State University but the results are still pending.

Other relevant data discussed included data on mercury in otters (Dave Everts) and research dating back 25 years on the mercury/acid deposition connection.

Knowledge Gaps

Critical loads: Biological indicators are needed to confirm ecosystem recovery. Studies using a similar approach to MADEP would be helpful, particularly in coordinating with federal and other national data sets. MADEP needs to come up with a data set that is compatible with wide national databases to fit with EPA.

There is a need to develop source-receptor relationships to look for specific resources at risk and to target the most likely source(s) of the emissions impacting these sensitive resources. This approach can be very effective for local-scale emissions, but also effective in identifying sources outside the state that are impacting Massachusetts resources.

Research Needs

- **Critical loads: starting with selection of appropriate indicators.** It is suggested that critical loads be linked with response indicators, such as a particular Massachusetts fauna species.
- **Compare low pH lakes in MADEP mercury study to the ARM results** to confirm whether the lakes that were removed from the study are truly outliers.
- **Continue long-term research programs and add biological monitoring to begin documenting potential biotic recovery.**
- **Study determinants of mercury levels in fish.**
6. Groundwater

While there was some study of groundwater/surface water interactions in relation to acid deposition 20 years ago (Richard Yuretich/UMass Amherst) there were no findings at that time that it was an important issue and subsequently there is not much data in this area. There is not a lot of interest in that arena, however it was suggested that older sites in the Yuretich study could be revisited to see if there has been any change in amount of acid deposition in watersheds over time.

7. Climate Change

Current Knowledge
In the past 20 years, a new issue has arisen in the form of climate change. EPA (2003) writes that “Other responses in surface waters may be partially attributable to factors other than atmospheric deposition, such as climate change and forest maturation. Continued long-term research and monitoring will be necessary to understand the causes, effects, and trends in these processes.” It is not clear yet how climate change will affect acid deposition and its effects. Elevated levels of CO₂ may lead to increased plant growth and uptake of N. However, higher temperatures may lead to increased nitrogen mineralization that in turn will cause enhanced N leaching. According to Sanderson (2005), “Climate change acts to worsen the problem of acidification by increasing the production of nitric acid from nitric oxide, and the proportion of ammonia converted into ammonium sulfate.” NAPAP (2005) adds that “Nitrogen export from recovering streams in the Catskill Mountains was reported to be greater during warm years than cool years, suggesting that a warmer climate could enhance acidification of surface waters if nitrogen deposition rates remained the same.”

Event-based precipitation samples from Central PA correlate chemistry with temperature and rainfall amounts. The study found temperature increases would produce concentration increase because of greater conversion rate of SO₂ in the atmosphere.

Knowledge Gaps
Precipitation is expected to increase in New England as a result of warmer climate and the effects on acid deposition are not well understood (e.g., will this lead to increased acid deposition or will there be more diluted deposition if there is a constant amount of pollutant?) These changes might mean that local emission sources are more important and long-range emissions less so.

An increase in DOC could be caused by climate change. **DOC is increasing with episodes, but the trend in surface waters is not known.**

Research needs:
- Climate or climate-related processes may counteract recovery by producing declines in base cations to offset a decline in sulfate, or by inducing an increase in natural organic acidity. These interactions of factors underscore the need to continue monitoring a subset of sensitive systems so as to understand the full suite of drivers and responses in ecosystems.
Conclusion

There has been much research devoted to acid deposition and its effect on the environment, but with the evolution of pollution emissions and the advent of climate change, old research gaps and new research needs have become evident.

Research areas that seem most immediate and within reach of the MA DEP include:

- Emissions: monitor trends and provenance of NOx emissions, quantify local emissions, and add trace elements including mercury.
- Soils/forests: Monitor soils, especially nutrients and the effect of emissions on soil respiration and carbon; compare present forest canopy to 1980s data.
- Surface waters: Continue long-term monitoring at sensitive sites, add mercury and nitrate monitoring in roadside streams.
- Fish and wildlife: Continue long-term research, add biological monitoring (keep tabs on indicator fauna species), study determinant mercury levels in fish.
- Estuaries: Determine percent N due to acid deposition in estuaries.

Summit Participants

David DeWalle, Penn State University Forest Hydrology
Sarah Dorner, MA Water Resources Research Center
Paul Godfrey, MA Water Resources Research Center
William Hagar, UMass Boston Sciences
Michael Hutcheson, MA DEP Office of Research and Standards
Steve Kahl, Plymouth State University Center for the Environment
Glenn Keith, MA DEP Bureau of Waste Prevention
Peter Kerr, UMass Amherst Environmental Analysis Laboratory
Gary Kleiman, NESCAUM/NESCCAF
Timothy Randhir, UMass Amherst Natural Resources Conservation
Jerry Schoen, MA Water Resources Research Center
William Sharpe, Penn State University Forest Hydrology
Alan Van Arsdale, US Environmental Protection Agency, Region 1
Marie-Françoise Walk, MA Water Resources Research Center
Richard Yuretich, UMass Amherst Geosciences
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