Annual Report to

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and US EPA, ORD, Corvallis OR

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Determining the effectiveness of the Clean Air Act and Amendments on the recovery of surface waters in the northeastern US

IAG 06HQGR0143

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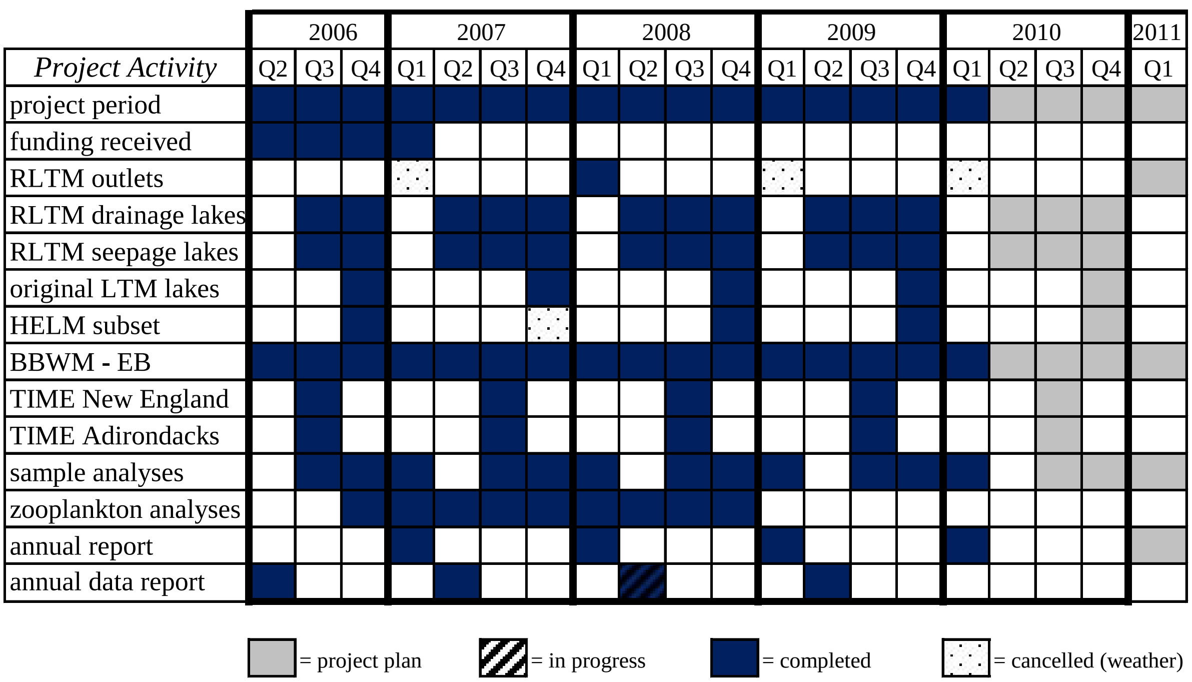
Overview of activities during 2009. A schematic summary of progress on the project plan is provided below and discussed on the following pages. We are concluding the fourth year of this five year project that supports the continuing needs of EPA for meeting the Congressional mandate for the agency to assess the effectiveness of the Clean Air Act Amendments of 1990. Field work and data assessment are on schedule, and the supplemental zooplankton component is well underway.

The lead organization for the project is now the University of New Hampshire. The 2009 transition from PSU to UNH was successful both fiscally and logistically. Field and laboratory assistance continues from the University of Maine.



Project background

Objectives. This proposed research is part of the EPA program to collect long-term data on the trends and patterns of response in surface waters sensitive to acidic deposition. The goals and methods are hierarchical from intensive site-specific to regional statistical populations. The objectives are to:

1) document the changes and patterns in aquatic chemistry for defined sub-populations and sites that are known to be susceptible to acidification or recovery;
2) evaluate linkages in changes in surface waters, if any, to changes in deposition that are related to regulatory goals;
3) characterize the effectiveness of the Clean Air Act Amendments in meeting goals of reducing acidification of surface waters and improving biologically-relevant chemistry in the northeastern US; and
4) provide information for assessment of the need for future reductions in atmospheric deposition based on the rate of recovery (or not) of the systems under study.

We continue to explore changes in biological condition using zooplankton collected in 2004 under separate funding from 145 ELS-II lakes in the northeast, as part of our 20th anniversary re-analysis of the Eastern Lake Survey (see Rosfjord *et al*., 2007). This re-sampling included total and methyl mercury analyses for lake water, GIS analyses of lake context, and creation of an integrated GIS-chemistry database for the 1986 ELS-II lakes and 2004 re-sampling data.

Approach. The schedule of tasks ranges from weekly to annual, continuing data records that now range from 16 to 25 years. We evaluate chemistry on a weekly basis year-round at the small watershed-scale at BBWM, weekly during the spring melt period at LTM lake outlets when seasonal conditions warrant, quarterly in LTM, and annually during the historical index period for the HELM and TIME lakes. These project components provide a *statistical framework* for inferring regional chemical patterns using TIME and LTM (and ELS-II under separate funding). The *long-term records* of LTM, HELM and BBWM provide information on seasonal and annual variability, and thus provide a seasonal context for the annual surveys.

Expected Results. This information is fundamental for EPA to meet the Congressional mandate for reporting on the effectiveness of the Clean Air Act Amendments (CAAA). The combination of site-specific data within the regional context will provide for an effective assessment of the effects of declining declining pollutant emissions on SO_4 concentrations, base cation depletion, and changes in N-saturation or DOC contributions to acid-base status. The results are also central to the decisions on additional emission reductions that may be needed to produce recovery.

Name	Class	Town (Gazmap)	Area (ha.)	Depth (m)	Elev (m)
Regular RLTM sites:					
Abol Pond	thick till	T 2 R 9 (50)	36	10	181
Bean Pond	med. till, high DOC	Pleas Ridge (30)	12	9	381
Bracey Pond	GW seepage	T 34 MD (34)	8	9	117
Crystal Pond	perched/seepage	T 40 MD (34)	10	9	113
Duck Pond	perched/seepage	T 22 MD (24)	2	3	82
Jellison Pond	med. till, low DOC	Amherst (24)	18	17	123
Newbert Pond	thin till, high DOC	Appleton (14)	13	4	89
Mud Pond	thin till, low DOC	T 10 SD (24)	1	15	104
Partridge Pond	thin till, low DOC	Amherst 24)	9	7	175
Salmon Pond	med. till, low DOC	T 10 SD (25)	4	10	94
Second Pond	thin till, high DOC	Dedham (23)	27	13	126
Wiley Pond	med. till, high DOC	Patten (51)	11	6	235
LTM Supplemental Lakes:					
Anderson Pond	thin till, low DOC	T 10 SD (25)	5	6	66
Little Long Pond	thin till, low DOC	T 10 SD (25)	24	25	75
Tilden Pond	thin till, low DOC	T 10 SD (25)	15	9	72

LTM/TIME annual field schedule

LTM-Maine	Apr				May				Jun				Jul				Aug				Sep				Oct				Nov
1 Bean					X												X									w/ HELM			
2 Partridge				X											X													X	
3 Jellison Hill				X											X													X	
4 Crystal				X											X												X		
5 Duck				X											X												X		
6 Bracey				X											X												X		
7 Newbert			X												X														X
8 Wiley					X												X										X		
9 Abol					X												X										X		
10 Mud					X											X												X	
11 Salmon					X											X												X	
12 Tunk outlet					X											X												X	
13 Second					X											X												X	
14 Little Long																												X	
15 Tilden																													
16 Anderson																													
TIME-Maine																													
1 Round Pond				w/ TIME NH															X										
2 Mountain Pd				fly with HELM																						fly w/ HELM			
3 Bog Pd Hartland				w/ Bean in spring and summer														X											
4 East Branch Lake				[Penobscot Nation sample]														X											
TIME NH																			X	X	X								
TIME New Engl																				X	X								
TIME NY																				X	X	X	X	X					
HELM																										X	X		

Spring weekly drainage lake samples: weather and snowcover dependent

Project Status: Water Chemistry

Field sampling. All project field objectives in 2009 were accomplished as planned with the exception of the spring drainage lake samples. Maine experienced an extremely wet spring making spring sampling logistically difficult and potentially unsafe for field crews.

Analytical. Analyses are complete for all samples collected through 2009, except for total aluminum. Aluminum samples are currently being analyzed by ICP and the USDA Forest Service Region 1 laboratory in Durham, NH. Analysis delays are the result of laboratory remodeling and instrument repair. We anticipate completion of the Al analyses by the end of April 2009.

Samples from East Bear Brook at BBWM, which are collected on a regular basis year around, continue to be analyzed in a contract laboratory at UMaine.

Data reporting. All data collected through 2008 have been delivered to EPA. The next delivery of data to EPA is expected in spring 2010, after evaluation of inter-laboratory comparisons and regular QA analyses by UNH and UMaine. Substantial unexpected (and unbudgeted) effort in 2009 went into moving our long-term database from mainframe SAS at UMaine to PC SAS, caused by the discontinuation of the mainframe at UMaine.

Presentation of findings. Several publications and presentations have resulted from this project since the final report for the previous LTM/TIME grant, listed at the end of this report. We recently presented an overview of Maine high elevation lakes at the 2009 North American Lake Management Society International Symposium (Baumann and Kahl, 2009). These results will be incorporated in Baumann's MS thesis to be completed in 2010. At least two more publications are expected to result from the thesis. The first will address organic acidity in high elevation lakes (see appendix).

New developments: Co-PI Webster is now affiliated with Michigan State University, and will continue her role in publishing on this grant.

***Recent publications** using related project information*

Baumann, A.J. and J.S. Kahl, 2007. Chemical trends in Maine High Elevation Lakes. LakeLine 27:30-34.

Campbell, J, J. Hornbeck, M. Mitchell, M. Adams, M. Castro, C. Driscoll, J.S. Kahl, and others, 2004. Input-output budgets for inorganic nitrogen for 24 watersheds in the northeastern United States. Water Air Soil Pollut., 151:373-396.

Dupont, J., T. Clair, C. Gagnon, D. Jeffries, J.S. Kahl, S. Nelson, and J Peckenham, 2005. Estimation of critical loads of acidity in the northeastern US and eastern Canada. Environ. Monit. Assess. 109:275-291.

Hunt, K., J.S. Kahl, J. Rubin, and D. Mageean, 2007. Assessing the science-based needs of stakeholders; a case study on acid rain research and policy. Journal of Contemporary Water Research and Education, 136: 68-79.

Kahl, J.S., J. Stoddard, R. Haeuber, S. Paulsen, R. Birnbaum, F. Deviney, D. DeWalle, C. Driscoll, A. Herlihy, J. Kellogg, P. Murdoch, K. Roy, W. Sharpe, S. Urquhart, R. Webb, and K. Webster, 2004. Response of surface water chemistry to changes in acidic deposition: implications for future amendments to Clean Air Act. Environmental Science and Technology, Feature Article 38:484A-490A.

Lawler, J., J. Rubin, B.J. Cosby, I. Fernandez, J.S. Kahl, S. Norton, 2005. Predicting recovery from acidic deposition: Applying a modified TAF (Tracking Analysis Framework) Model to Maine High Elevation Lakes, Water Air Soil Pollut. 164:383-389.

Nelson, S.J., W.H. Halteman, J.S. Kahl, N.C. Kamman, D.P. Krabbenhoft, 2010. Predicting mercury concentrations in northeast lakes using hydrogeomorphic features, landscape setting, and chemical co-variates. Environmental Science and Technology, in review.

Norton, S., I. Fernandez, J.S. Kahl, and R. Reinhardt, 2004. Acidification trends and the evolution of neutralization mechanisms through time at the Bear Brook Watershed, Maine, USA. Water, Air, Soil, Pollution Focus 4:289-310.

Rosfjord, C., K. Webster, J.S. Kahl, S.A. Norton, I. Fernandez, and A. Herlihy, 2007. Anthropogenically-driven changes in chloride complicate interpretation of base cation trends in lakes recovering from acidic deposition. Environ Sci Technol, 41:7688 -7693.

Rosfjord, C., J.S. Kahl, K. Webster, S. Nelson, I. Fernandez, L. Rustad, and R. Stemberger, 2006. Acidic deposition-relevant changes in lake chemistry in the EPA Eastern Lake Survey, 1984-2004. Final report to USDA NSRC, Durham, NH. 69 p.

Recent presentations using project information

Baumann, A.J., and J.S. Kahl, 2009. Assessing the effectiveness of federal acid rain policy using remote and high elevation lakes in northern New England. North American Lake Management Society International Symposium, Hartford, CT, October 29, 2009.

Kahl, J.S., 2009. Changes in base cations related to long-term changes in Cl distribution in northeastern lakes. Gordon Research Conference, Forested Catchments, July 12-17, 2009, Proctor Academy, NH.

Kahl, J.S., 2008 (invited). Twenty year changes in spatial patterns of Cl distribution in the northeastern US. NH Water Conference, April, 2008.

Kahl, J.S., 2007 (invited). Using societal-based incentives to address new threats to New England Lakes. Day-long short course in New England Lake Science Academy, Camp Kieve, Maine. July, 2007.

Kahl, S., K. Webster, D. Sassan, C. Rosfjord, S. Nelson, M. Greenawalt-Yelle, 2007. Increasing Cl in northeastern surface waters: an indicator of increasing development pressure. Maine Water Conference, Augusta, ME, March 21, 2007.

Kahl, J.S. 2006 (invited). Acid rain in New England: using high elevation lakes as sentinels of change. Maine Mountain Conference, October 21, 2006. Rangeley, Maine

Kahl, J.S., *et al.*, 2006 (invited). The design of a national mercury monitoring network: Learning from the EPA acid rain experience. The Eighth International Mercury Conference, Madison WI, August 8, 2006.

Kahl, J.S. *et al*., 2006. Obfuscation of trends in base cations by regional salt contamination. Hubbard Brook Committee of Scientists annual meeting, July 12, 2006.

Kahl, J.S., 2006 (invited). 'Natural and human-derived sources of acidity in Maine Atlantic Salmon Rivers'. Atlantic Salmon Commission workshop on acidity, Bangor ME. April 10, 2006.

Kahl, J.S., 2005 (invited). The intersection of environmental science and environmental policy. NH Charitable Foundation Lakes Region annual meeting, Meredith, NH, September, 2005.

Kahl, J.S., 2005 (invited). Tracking response and recovery in surface waters in the northeastern US. Annual meeting of the Ecological Society of America, Montreal, August, 2005.

Kahl, J.S., and Catherine Rosfjord, 2005 (invited). Acid rain and the Clean Air Act in the northeastern US. Annual meeting of the NH-ME Androscoggin River Watershed Council, Bethel, June, 2005

Kahl, J.S., 2005 (invited). Developing a lake research agenda for NH. NSF workshop on lake research infrastructure in the northeast, Colby Sawyer College, April 2005.

Kahl, J.S., S. Nelson, and A. Grygo, 2004. Surface water chemistry data for the northeastern US for interpreting climate and acid rain trends. Northeast Ecosystems Research Consortium meeting, Durham, NH, October, 2004.

Kahl, J.S., K. Webster, M. Diehl, and C. Rosfjord, 2004. Successes of the Clean Air Act Amendments of 1990. Maine Water Conference invited plenary talk, Augusta, ME, 2004.

Kahl, J.S. and K. Johnson, 2004. Acid-Base Chemistry and Historical Trends in Downeast Salmon Rivers. Maine Water Conference, Augusta ME, April 2004.

Kahl, J.S., 2004 (invited). The Clean Air Act Amendments of 1990; testing a program designed to evaluate environmental policy. Lecture, Colby College. April, 2004

S.J. Nelson, J.S. Kahl, N.C. Kamman, D.P. Krabbenhoft, W.H. Halteman, 2009. (Poster) Predicting mercury concentrations in northeast lakes using hydrogeomorphic features, landscape setting and chemical co-variates. Gordon Research Conference, Forested Catchments, July 12-17, 2009, Proctor Academy, NH.

Nelson, S.J., I. Fernandez, S. Norton, B. Wiersma, L. Rustad , J.S. Kahl, 2008. The Bear Brook Watershed in Maine: Long-term research supporting climate change inquiry. Hydroclimatic effects on ecosystem response: participant workshop, Syracuse, NY, September 19, 2008.

Nelson, S.J., N. Kamman, D. Krabbenhoft, J.S. Kahl, K. Webster, 2008. Evaluating spatial patterns in mercury and methyl mercury in northeastern lakes: Landscape setting, chemical climate, and human influences. Northeastern Ecosystem Research Cooperative Conference, Durham, NH, November 12-13, 2008.

Nelson, S.J. 2008. Evaluating spatial patterns in mercury and methyl mercury in northeastern lakes: landscape setting, chemical climate, and human influences. Maine Water Conference, Augusta, ME, March 19, 2008.

Appendix I. Draft abstract from MS candidate Baumann thesis (partially supported by this IAG).

Increasing organic acidity as part of recovery from acidic deposition in Maine high elevation lakes.

Abstract

Recovery from acidic deposition in the northeastern U.S. has led to improvements in surface water quality in some of the most affected waters. Nowhere is this more apparent than in the high elevation lakes of Maine (HELM). An important consequence of decreased acid rain has been an increase in dissolved organic carbon (DOC) in recovering surface waters across the northern hemisphere. This result has led to a transition from inorganic (acid rain) to natural (DOC) sources. The rapid response of the acid sensitive HELM lakes to changes in deposition makes them well suited for assessing increased DOC as part of recovery from acid rain. The 'Sulfate Fraction' (SF, the relative contribution of SO_4 to the total amount of anions) has decreased ~ 40% in these lakes since the mid 1980's, yet no significant change in pH has occurred (Figure 1). Over the same time period, DOC has been increasing at ~0.1mg/L/yr (~+20% since the mid-1980s), making up an increasing portion of the ionic strength of the HELM waters (Figure 2) that are simultaneously becoming more dilute (Figure 3). This shift in acidity is responsible for the relatively unchanged pH status of acid sensitive waters despite significant decreases in inputs of acid rain to the systems. Our conclusion is that while recovery in ANC is still incomplete, recovery in pH may be nearing completion due to replacement of mineral acidity with organic acidity (Figure 2). This conclusion is consistent with ample paleolimnological evidence that shows that lakes classified as 'acidified' in the 1980s (pH < 5) typically had pre-historical pH values that were between pH 4.8 and 5.8. pH 'recovery' of these systems will (has?) been represented by modest increases in pH because of the acidic nature of these systems long before human impact from acidic deposition. The policy-relevant importance of these results is recognition that natural organic acidity must be included in critical load calculations, which will reduce the amount of S reduction needed in emissions/deposition to meet pH targets.

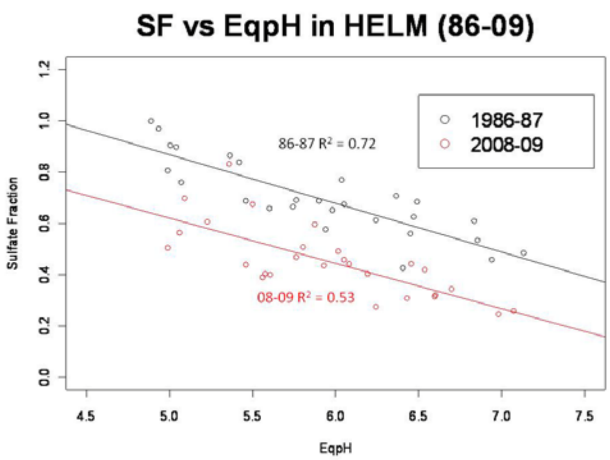


Figure 1. Reduction in the contribution of sulfate to total anion concentration, but without an increase in pH

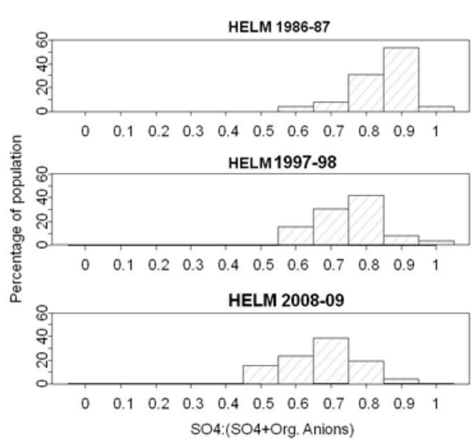


Figure 2. Temporal evolution towards increasing importance of organic acidity in HELM lakes.

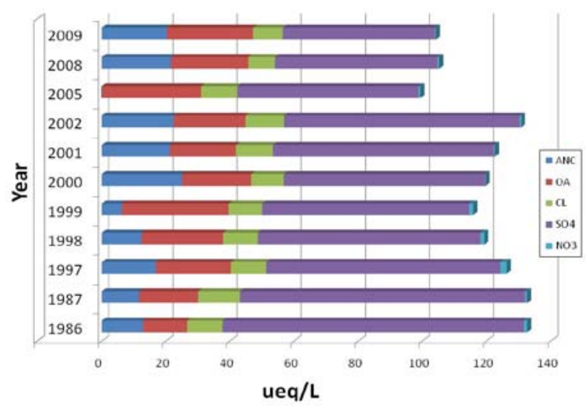


Figure 3. Changes in ionic composition of HELM lakes, showing reduction in ionic strength.

Appendix II. Draft abstract from MS candidate Doogan thesis
(supported under separate funding from this IAG)

Temporal Trends and Landscape Controls on Al in the Hubbard Brook Experimental Forest and White Mountains of NH

Christian Doogan, MS candidate, Plymouth State University, 2010

Draft Abstract

Stream chemistry is influenced by many factors such as vegetation cover, precipitation chemistry, soils, geology, and mineral weathering. The differences in the distribution of flow paths and water residence times throughout the landscape also affect stream chemistry. Acidification of stream water posses a possible threat to many species and the extent of this impact in the White Mountains of New Hampshire remains unclear despite abundant historical information collected over the past 30 years. Motivated by the challenge of understanding the relationship of the landscape to stream chemistry in this region, this project sought to a) describe the stream chemistry variability in the region and its relationship to the landscape, and b) determine trends in episodic and baseflow Al concentrations. The objective was to investigate stream chemistry under contrasting high to low flow regimes of watersheds varying in basin size and spatially across the region to investigate a wide range of landscape characteristics. This analysis was done by comparing stream chemistry samples collected in a synoptic strategy under spring high and summer low flows to a) landscape variables derived from terrain analysis of surface topography, and b) to historical data from previous surveys.

Stream chemistry from a second data set collected under non-event conditions at Hubbard Brook was compared to similar landscape variables derived from terrain analysis. The Hubbard Brook dataset provided an opportunity to test the landscape variables on a large number of nested sample locations collected under higher spatially resolution including basin sizes far smaller than the region wide analysis. Landscape variables were compared with stream chemistry of both datasets by Spearman rank correlations and linear stepwise regression analysis.

Results from this study show that although many relationships could be found between the landscape variables analyzed and stream chemistry, many were only weakly correlated. The landscape variables could only partially predict stream chemistry under either high or low flow for the 2008 White Mountain dataset or under the non-event fall 2001 Hubbard Brook Valley-Wide dataset. At best, 60% of stream chemistry variables could be explained for either dataset by the landscape variables. However, it was found that many of the landscape variables

correlated with stream chemistry under spring high flows were also correlated to summer low flow so that no distinct difference in the landscape variables important to flow regime was observed. Percent till calcium concentration was found to be an important landscape variable for the White Mountain region. For Hubbard Brook it was elevation and forest cover that were the most important landscape variables. Forest cover was correlated to total aluminum (Al) and inorganic aluminum (Al_i) for both the White Mountain region and at Hubbard Brook. The results show that easily derived topographic data and simple terrain analysis cannot characterize landscape control on stream chemistry variability and the landscape variables analyzed do not act as accurate surrogates for the hydrologic processes that influence stream chemistry.