

New York State Energy Research and Development Authority

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# Critical Loads of Sulfur and Nitrogen for Protection of Acid-Sensitive Aquatic and Terrestrial Resources in the Adirondack Mountains

Final Report

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# **Critical Loads of Sulfur and Nitrogen for Protection of Acid Sensitive Aquatic and Terrestrial Resources in the Adirondack Mountains**

## ***Final Report***

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# Notice

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## Keywords

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critical load, sulfur, nitrogen, acidification, acidic deposition

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# Acronyms and Abbreviations

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$\mu\text{eq}$	microequivalent
AEAP	Aquatic Effects Assessment Program
AIC	Aikake's Information Criteria
$\text{Al}_i$	dissolved inorganic aluminum
AirMon	Atmospheric Integrated Research Monitoring Network
Al	aluminum
ALS	Adirondack Lakes Survey
ALSC	Adirondack Lakes Survey Corporation
ALTM	Adirondack Long-term Monitoring Program
ANC	Acid neutralizing capacity
BS	Base saturation
BC	Base cation
$\text{Ca}^{2+}$	calcium
CAIR	Clean Air Interstate Rule
CALK	Calculated ANC
CASTNet	Clean Air Status and Trends Network
C	Carbon
CL	critical load
CMAQ	Community to Multiscale Air Quality model
DDRP	Direct Delayed Response Project
DIC	Dissolved inorganic carbon
DOC	Dissolved organic carbon
ELS	Eastern Lake Survey
EMAP	Environmental Monitoring and Assessment Program
EPA	Environmental Protection Agency
GIS	Geographic information system
GPS	global positioning system
ha	hectare
LTM	Long-term Monitoring program
m	meter
$\text{m}^2$	square meter
MAGIC	Model of Acidification of Groundwater in Catchments
meq	milliequivalents
$\mu\text{eq/L}$	microequivalents per liter
N	nitrogen
NAPAP	National Acid Precipitation Assessment Program
NADP	National Atmospheric Deposition Program
NADP/NTN	National Atmospheric Deposition Program/National Trends Network
NHD	National Hydrography Dataset
$\text{NO}_3^-$	nitrate
NYSDEC	New York State Department of Environmental Conservation
PCA	Principal components analysis
PM	Particulate matter
PnET-BGC	Photosynthesis and evapotranspiration biogeochemistry model
QA/QC	Quality assurance/quality control
RMSE	Root mean square error
S	sulfur
SAA	Sum of acid anions
SBC	Sum of base cations
SE	Standard Error
SIP	State Implementation Plan
$\text{SO}_4^{2-}$	sulfate
TL	Target Load

# Summary

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## S.1 Background

Most efforts to quantify aquatic and terrestrial ecosystem damage from sulfur (S) and nitrogen (N) air pollution in the Adirondack Mountains and to examine more recent ecosystem recovery in response to emissions control, have focused on lake water chemistry. However, relatively large decreases in regional upwind S emissions and generally comparable decreases in S deposition in the Adirondack Mountains over the past two to three decades have resulted in limited recovery of lake water acid-base chemistry. The limited nature of the observed surface water chemical recovery response to date has largely been attributed to depletion of exchangeable base cations on the soil.

Acidic deposition levels must be quantified in order to manage and affect full chemical, and hopefully also biological, recovery of both surface waters and soils. The critical load (CL) approach can provide such quantification. The CL is the level of sustained atmospheric deposition of S, N, or acidity below which harmful effects to sensitive ecosystems do not occur according to current scientific understanding. It is usually calculated under steady state conditions which may not occur for many decades or longer into the future. A dynamic CL can also be calculated that is specific to a particular point in time. This dynamic CL is often called a target load (TL). This report presents results of dynamic CL or TL simulations specific to the years 2050 and 2100. This research was undertaken to determine the CL values that will promote resource recovery in previously acidified aquatic and terrestrial ecosystems in New York State. The focus is on the Adirondack Ecoregion, which is an area recognized in New York State by the U.S. Environmental Protection Agency, and in particular, the intensively-monitored watersheds, many of which are in the southwestern portion of the Adirondack Ecoregion.

This research builds upon the work of Sullivan et al. (2006a), in which the same research team modeled the acid-base response of 70 Adirondack lakes in response to historic acidic deposition and several scenarios of future emissions controls using the Model of Acidification of Groundwater in Catchments (MAGIC) and the Photosynthesis and Evapo Transpiration Bio Geo Chemistry (PnET-BGC) model. Study lakes included statistically selected lakes from the U.S. Environmental Protection Agency's (EPA) Environmental Monitoring and Assessment Program (EMAP) and a group of intensively studied long-term monitoring lakes.

## S.2 Approach

Simulations for this project using the MAGIC model were based on two acidic deposition drivers (S and N), three sensitive receptors (lake water, soil, and soil solution), one or more chemical indicators for each, two to three critical threshold levels for each indicator, and three to four endpoint years or periods. Selection of these various CL parameters had important influence on the resulting CL calculations. Thus, we present a large matrix of CL results. The calculated CL values must be interpreted in the context of these choices.

For modeling the CL to protect or restore Adirondack lakes, we used the indicator acid neutralizing capacity (ANC), with critical threshold criteria values equal to 0, 20, and 50  $\mu\text{eq/L}$  (U.S. EPA 2008b). Additional analyses were conducted to estimate the deposition of N that would maintain lake nitrate ( $\text{NO}_3^-$ ) concentration below 10 and 20 microequivalents per liter ( $\mu\text{eq/L}$ ), values that may suggest N enrichment. Analyses to investigate the effects of nutrient (N) input on biodiversity were not conducted.

For protection of terrestrial resources, we modeled the CL to attain soil % base saturation (BS) values of 5, 10, and 15%. We also modeled the CL to attain a molar ratio of calcium (Ca) to aluminum (Al) or nutrient base cations (Bc) to Al in soil solution equal to 1.0 or 10.0. For these analyses, the Bc include  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{K}^+$  and excludes  $\text{Na}^+$ , which is not an important plant nutrient.

## S.3 Critical Load Patterns

Lower aquatic S CL values (indicating greater acid sensitivity) were generally found for Adirondack lakes that currently have low ANC. Furthermore, CL values tended to be lower if the objective was to protect lake water to a higher level (i.e., ANC = 50  $\mu\text{eq/L}$ ) as compared with a lower level of protection, such as protecting to ANC = 0  $\mu\text{eq/L}$ .

In general, the threshold value of 10 for the Ca:Al or Bc-to-Al ratio chemical indicators for soil solution did not prove to be useful for discriminating among CL values for protection of terrestrial resources in Adirondack watersheds against harm caused by S deposition. Nearly all of the modeled lake watersheds showed a simulated S CL to protect either Ca:Al or Bc:Al to 10 that was  $\leq 25 \text{ meq/m}^2/\text{yr}$ , which is lower than ambient levels of S deposition in these watersheds. A soil % BS target of 15%, like the soil solution ratio targets equal to 10, appears to be unattainable for many watersheds, based on the MAGIC simulations.

## **S.4 Sulfur versus Nitrogen Critical Loads**

For a given watershed and set of desired outcomes (e.g., CL criteria), the CLs for N were much higher than those for S. That is because Adirondack watersheds modeled for this study currently retain most of the N that is atmospherically deposited. The CL simulations with MAGIC assume that most of the future N loading will continue to be retained, causing limited influence of deposited N on the acid-base chemistry of the lake or soil. Some lakes and their watersheds were simulated by MAGIC to have low N CL, however, largely because they were calculated to have low lake ANC and soil % BS solely as a consequence of the baseline S deposition that was assumed for the N CL simulations. In addition, it is possible that N retention in some watersheds will decrease in the future under continued N loading. Such a change would decrease the N CLs for lakes in these watersheds.

## **S.5 Extrapolation of Modeled Critical Load Results**

Numeric extrapolation of model-simulated CL values for this project focused on estimating numbers and percentages of Adirondack lake watersheds predicted to exhibit various CL values. Several numeric extrapolation approaches were used in this study, and results varied depending on the frame of reference selected. The statistically selected watersheds were chosen to be representative of Adirondack watersheds containing lakes larger than 1 ha and deeper than 1 m. Model projections of aquatic and terrestrial CLs were numerically extrapolated to two EMAP lake watershed frames: 1) 1,320 low-ANC ( $\leq 200 \mu\text{eq/L}$ ) lakes, and 2) 1,829 lakes that generally span the spectrum of Adirondack lake ANC. This analysis yielded estimates of numbers and percentages of lakes and watersheds in various CL classes, without any information regarding where within the Adirondack Ecoregion those watersheds are located. To satisfy the need to discern where within the Adirondack Ecoregion these acid-sensitive lakes are located, model results for aquatic CLs were also spatially extrapolated to 1,136 lakes included in the Adirondack Lake Survey (ALS) and mapped throughout the study region. As an example, in Table S-1, CL results are compared among various groups of Adirondack lake watersheds based on protecting lake water to ANC =  $50 \mu\text{eq/L}$  in the year 2100. The modeled lakes were generally skewed toward lower CL values, as compared with the lake population distributions. The EMAP populations (especially the full EMAP population of all lakes, regardless of ANC) and the ALS population were more skewed towards relatively high CL values.

The percentage of lakes found to be within the various CL classes, where each class represents a range of CL values in  $\text{meq/m}^2/\text{yr}$ , varied substantially based on the group of lakes modeled or represented by the estimate (Table S-1). In extrapolating CL and/or exceedance results obtained by modeling individual lake watersheds to the broader population of Adirondack lakes and their associated watersheds, it is therefore important to specify the lake population or statistical frame to which the results are being applied.

**Table S-1. Estimated percentage of Adirondack lake watersheds having various critical load of sulfur deposition values**

These values protect against sulfur-driven lake acidification to ANC = 50 µeq/L in the year 2100, using different approaches and population frames.

*Source: Modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. Water Resour. Res. 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.*

Approach	Number of Watersheds	Percentage of Lakes in Critical Load Class					
		CL (S)	≤25	25-50	50-75	75-100	> 100 (meq/m <sup>2</sup> /yr)
MAGIC model simulations for all modeled lake watersheds	97		28.9	26.8	17.5	15.5	11.3
MAGIC model simulations for all modeled lake watersheds that were calibrated using watershed-specific soil chemistry data derived from the 2003 soil survey	70		27.1	22.9	21.4	17.1	11.4
Extrapolation of MAGIC model simulation results for 44 EMAP probability survey lakes to the EMAP frame of Adirondack lakes that are larger than 1 ha, deeper than 1 m, and that have ANC ≤ 200 µeq/L	1,320		19.5	21.8	22.2	10.1	26.4
Same as above, except assuming a high CL for all EMAP lakes that were not modeled using MAGIC because they had ANC > 200 µeq/L	1,829		14.1	15.7	16.0	7.3	46.8
Spatial extrapolation of MAGIC model simulation results to all lakes surveyed by the ALS that are larger than 1 ha	1,136		25.8	13.1	10.4	8.9	41.8

To protect lake ANC from negative values (below 0) in the year 2050 or 2100, the S deposition CL for each EMAP lake was higher than 25 meq/m<sup>2</sup>/yr. If, however, the critical lake ANC threshold value was set to 20 or 50 µeq/L, then between 12% and 23% of the EMAP lakes had calculated CLs below 25 meq/m<sup>2</sup>/yr, depending on the choice of critical threshold value (20 or 50 µeq/L) and endpoint year (2050 or 2100). More than half of the 1,320 EMAP lakes had relatively high CL (>100 meq/m<sup>2</sup>/yr) to protect lake ANC to 0 µeq/L, with 62% for the endpoint year 2050 and 56% for the endpoint year 2100. More than one fourth of the 1,320 EMAP lakes had relatively high CL (> 100 meq/m<sup>2</sup>/yr) to protect lake ANC to 50 µeq/L (31% for the endpoint year 2050, 26% for the endpoint year 2100).



Critical S load estimates for protecting soil BS were very sensitive to selection of the critical threshold value for BS. For example, BS 5% was rather readily achievable, with more than three-fourths of the EMAP lake watersheds having CL >100 meq/m<sup>2</sup>/yr to protect to this BS level. In marked contrast, in order to achieve BS of 10%, about two-thirds or more of the EMAP lake watersheds showed very low CL values (< 25 meq/m<sup>2</sup>/yr). Nearly all of the EMAP lakes (≥ 94%) had S CL < 25 meq/m<sup>2</sup>/yr to achieve BS of 15%.

MAGIC model simulations of the CL of S deposition needed to protect lake ANC from falling below designated critical criteria values could successfully be predicted using only lake water ANC as a predictor variable. More specifically, r<sup>2</sup> values ranged from 0.72 (to protect ANC to 0 µeq/L in the year 2050) to 0.92 (to protect ANC to 50 µeq/L in the year 2100). The most robust predictions were obtained for estimating the CL to protect lake ANC from going below 50 µeq/L in the years 2050 and 2100 (r<sup>2</sup> = 0.90 and 0.92, respectively). For reasons of simplicity, the final equations applied here only used ANC and a constant to predict each CL. Inclusion of watershed features such as elevation, slope, watershed area, and/or soil characteristics (pH, percent clay, depth) did not appreciably improve CL predictions beyond what was achieved based only on lake ANC.

The spatial patterns in acid sensitivity are readily apparent in map depictions of S CLs extrapolated to the population of ALS lakes. One example is shown in Map S-1, which is based on protecting the lakes to ANC = 50 µeq/L in the year 2100. For these simulations, the vast majority of the ALS lakes in the southwestern third of the Adirondack Ecoregion have S CL less than 50 meq/m<sup>2</sup>/yr, as do many lakes in the high peaks area.

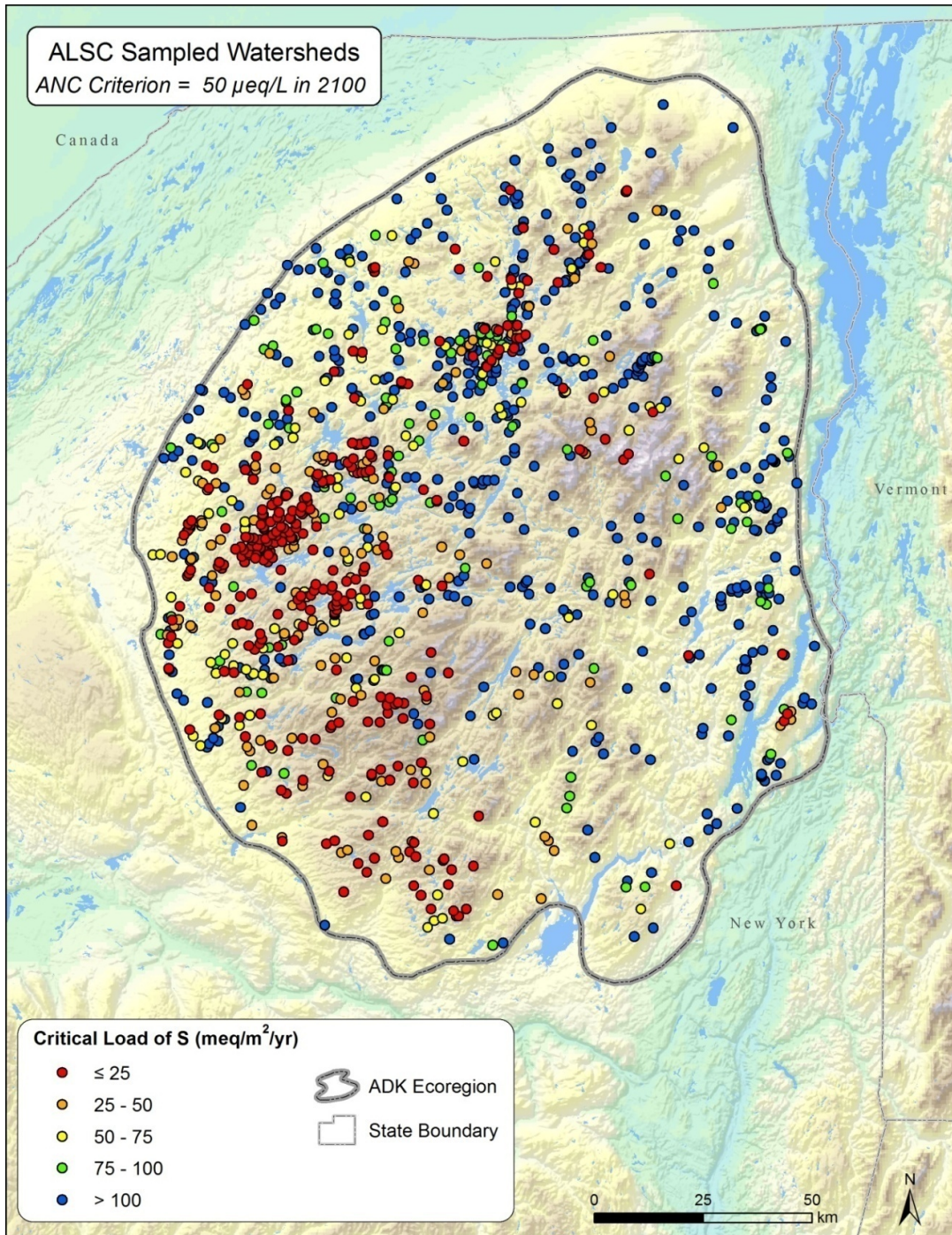
The population distributions of the S CLs for the low-ANC EMAP population of lakes and for the ALS surveyed lakes were generally similar. Median and quartile CL values were slightly lower for the ALS lake population than for the EMAP population, although the differences were small.

The modeled CL values varied with selection of endpoint year. For the most acid-sensitive lake watersheds (i.e., those having CL less than about 25 to 50 meq/m<sup>2</sup>/yr), the CL to protect sensitive resources was higher for protection to the year 2100, as compared with 2050. In other words, these most acid-sensitive watersheds were simulated to be able to tolerate slightly higher S loading if one was willing to wait an additional 50 years for the resources to recover. For the majority of the modeled watersheds, however, the S CL was higher than 50 meq/m<sup>2</sup>/yr, and for these less acid-sensitive watersheds the S CL was lower using an endpoint year of 2100. Thus, these watersheds are not able to tolerate as much S loading if the resource protection is intended to extend all the way to 2100, as opposed to only protecting the resources to 2050. The effect of endpoint year designation therefore depends on the current acid-base status of the watershed.

**Map S-1. Estimated critical load of sulfur deposition to protect lake ANC to 50  $\mu\text{eq/L}$  in the year 2100**

Based on extrapolation of MAGIC modeling results to the population of lakes surveyed by the Adirondack Lakes Survey (ALS).

Source: Modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.



## S.6 Exceedance

Regional estimates of ambient (average centered on the year 2002) total wet plus dry S and N deposition were highest in the southwestern portion of the Adirondack Ecoregion and lowest to the northeast. These deposition estimates were used to calculate CL exceedance by comparing ambient deposition with CL estimates. The number and percentage of Adirondack lakes that receive ambient S deposition above their respective CL are reported. For protecting lake water ANC in the year 2100, the percent of 1,320 low-ANC EMAP lakes projected to be in exceedance ranged from 15.5% for protecting to ANC = 0  $\mu\text{eq/L}$  to 46.3% for protecting to ANC = 50  $\mu\text{eq/L}$ ; the estimate for protection to ANC = 20  $\mu\text{eq/L}$  was intermediate (22.7%). Comparable calculations for the year 2050 yielded slightly lower estimates of the numbers and percentages of lakes in exceedance.

Only 13.5% (for the year 2100) and 11.6% (year 2050) of these low-ANC EMAP Adirondack lake watersheds were simulated to be in exceedance of the S CL to protect soil BS to 5%. If a more protective critical threshold value for BS is assumed (BS = 10% or 15%), many additional watersheds are estimated to be in exceedance for protecting soil condition.

Roughly half (44.1% for the year 2050 and 58.2% for the year 2100) of the low-ANC EMAP lake watersheds were estimated to be in exceedance to protect soil solution Ca:Al to 1, whereas nearly all of the watersheds (98.1%) were in exceedance to protect to a ratio value of 10. Similarly, nearly all (98.1%) of the EMAP low-ANC lake watersheds were calculated to be in exceedance to protect soil solution Bc:Al = 10. Very few (7.8% of the EMAP low-ANC lake watersheds) were in exceedance to protect to Bc:Al = 1. Thus, the Bc:Al ratios of 1 and 10 are not effective thresholds for discriminating exceedance of S CLs. If a critical threshold criterion of 1 is assumed, very few watersheds are estimated to currently be in exceedance; if a critical threshold criterion of 10 is assumed, nearly all watersheds are estimated to be in exceedance under ambient deposition loading rates.

Selection of the sensitive criterion and its associated critical threshold value have considerable influence on the resulting CL exceedance calculations. Some lakes and their watersheds receive ambient S deposition that is more than double the respective CL, especially for protecting soil BS to 15%, soil solution Ca:Al ratio to 10, and soil solution Bc:Al ratio to 10.

## S.7 “Can’t Get There from Here” Lakes and Watersheds

Depending on the critical threshold value of the sensitive criterion and the endpoint year that was selected for a particular CL analysis, some receptors are unable to attain the critical threshold value by the specified endpoint year even if S or N deposition is reduced to zero and maintained at zero throughout the duration of the simulation (to the endpoint year). These lakes, soils, or soil solution receptors are sometimes called “can’t get there from here” receptors.

All of the EMAP lakes were simulated to be able to achieve  $ANC = 0 \mu\text{eq/L}$ , regardless of endpoint year. Most (93%) EMAP lakes could achieve  $ANC = 20 \mu\text{eq/L}$  by the years 2050 or 2100. Somewhat fewer (84%) could attain  $ANC = 50 \mu\text{eq/L}$  by either of these endpoint years. For the lakes that were judged to be unable to achieve the specified ANC threshold, that inability to achieve the target was attributed primarily to low ANC during pre-industrial time, and secondarily to delayed recovery response due to effects of acidic deposition on watershed soils.

## S.8 Uncertainty

MAGIC CL simulation uncertainty, reflected in the fuzzy calibration procedure, is relatively small across the distribution of CL values for protecting or restoring lake ANC. In general, the difference between the maximum and minimum simulated CL values was less than about 10 to 20  $\text{meq/m}^2/\text{yr}$ . Similarly, the uncertainty in simulating the CL to protect or restore soil BS was relatively low (generally less than about 10  $\text{meq/m}^2/\text{yr}$ ) for the watersheds that were most acid-sensitive (those that have CL to protect soil BS less than about 75  $\text{meq/m}^2/\text{yr}$ ). For the less acid-sensitive watersheds, however, the CL to protect soil BS was much more uncertain. However, added uncertainty is not of great concern for watersheds that have relatively high CL. A major modeling objective is to minimize uncertainty (increase precision) for the watersheds that are especially acid-sensitive (those having low CL).

Uncertainty was high across the range of CL values for simulations of S CL to protect soil solution base cation to Al ratios. These results suggest that the model performs best for simulating the CL to protect lake ANC, and for protecting soil BS for the most acid-sensitive watersheds. Model performance is more uncertain for protecting soil BS in the more base cation-rich watersheds and for protecting soil solution base cation to Al ratios across the spectrum of acid sensitivity.

## **S.9 Importance**

The results of this research are important for management of the ecosystems in New York State that have been highly impacted by acidic deposition. Model simulations developed in this project indicate the CLs of atmospheric deposition needed to affect resource recovery to a range of chemical indicator values, and at several different future time periods. In addition, the estimated aquatic CLs at discrete dynamic modeling sites were extrapolated to 1,136 lake locations in the region that have been surveyed for lake chemistry. We identified locations in the Adirondacks where acidified lakes and forest soils receive ambient acidic deposition in exceedance of their CL, where they are most likely to recover, and the long-term sustained deposition loads that would be required to affect such recovery.

## **S.10 References**

- Sullivan, T.J., C.T. Driscoll, B.J. Cosby, I.J. Fernandez, A.T. Herlihy, J. Zhai, R. Stemberger, K.U. Snyder, J.W. Sutherland, S.A. Nierzwicki-Bauer, C.W. Boylen, T.C. McDonnell, and N.A. Nowicki. 2006a. Assessment of the Extent to Which Intensively-Studied Lakes Are Representative of the Adirondack Mountain Region. Final Report 06-17. New York State Energy Research and Development Authority, Albany, NY.
- U.S. Environmental Protection Agency. 2008a. Integrated Science Assessment for Oxides of Nitrogen and Sulfur – Ecological Criteria. EPA/600/R-08/082F. National Center for Environmental Assessment, Office of Research and Development, Research Triangle Park, NC.

# 1 Introduction

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Atmospheric deposition of sulfur (S) and nitrogen (N), derived from utility, industrial, and area air pollution sources has caused acidification of soils, soil water, and drainage water across broad areas of the eastern United States (U.S. EPA 2008a). Such acidification has been associated with enhanced leaching of sulfate ( $\text{SO}_4^{2-}$ ) and nitrate ( $\text{NO}_3^-$ ) to drainage waters, depletion of available calcium ( $\text{Ca}^{2+}$ ) and other nutrient cations from soil, decreases in pH and acid neutralizing capacity (ANC) in surface waters, and increased mobilization of potentially toxic dissolved inorganic aluminum from soil to drainage waters ( $\text{Al}_i$ ; Charles 1991, Sullivan 2000). Resulting biological effects have included toxicity to fish and aquatic invertebrates and adverse impacts on forest vegetation, especially red spruce and sugar maple trees (U.S. EPA 2009).

Aquatic and terrestrial ecosystem damage from air pollution in the Adirondack Mountains in New York State has been substantial, mainly from atmospheric deposition of S (Driscoll et al. 1991, Sullivan 2000, Driscoll et al. 2003a, Sullivan et al. 2006a, Lawrence et al. 2008). There is also evidence that historic increases in S deposition resulted in large increases in base cation concentrations of many Adirondack lakes, resulting in modest (compared with historic increases in  $\text{SO}_4^{2-}$  concentration) decreases in ANC during the past century (cf., Sullivan 1990, Cumming et al. 1992). Thus, it might be reasonable to expect that subsequent decreases in solution  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations and mobility caused by decreases in S and  $\text{NO}_3^-$  deposition would decrease base cation concentrations in runoff. Most efforts to quantify damages, and to examine more recent ecosystem recovery in response to emissions controls, have focused on lake water chemistry.

Relatively large decreases in regional S and more modest decreases in nitrogen oxide emissions, and generally similar decreases in S and  $\text{NO}_3^-$  deposition in the Adirondack Mountains over the past two to three decades have resulted in limited recovery of lake water acid-base chemistry (Stoddard et al. 1998, Driscoll et al. 2003b, Stoddard et al. 2003, Driscoll et al. 2007). Sulfate concentrations in lake water have decreased markedly. Also, since the 1990s there have been widespread decreases in lake  $\text{NO}_3^-$  concentrations. Decreases in surface water  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  have coincided with decreases in concentrations of base cations. Therefore, measured increases in lake water pH and ANC have generally been small.

This limited chemical recovery of surface waters in the Adirondack Mountains and elsewhere in the northeastern United States has been attributed in part to base cation, especially  $\text{Ca}^{2+}$ , depletion of watershed soils (Lawrence et al. 1995, Likens et al. 1996, Warby et al. 2009) in response to long-term elevated levels of S and  $\text{NO}_3^-$  deposition. Changes in atmospheric deposition of base cations have also likely contributed to this response (Gbondo-Tugbawa and Driscoll 2003). The relatively small recent increases in lake water pH and ANC in response to substantial (> 40%) reductions in S and  $\text{NO}_3^-$  deposition in the 1980s and 1990s (Stoddard et al. 2003) might be attributable to remaining base cation exchange buffering in these watershed soils.

Much of the acidic deposition effects research in the Adirondacks has focused on individual lakes and their watersheds. There is a great deal of information available for a relatively small number of watersheds (c.f., Driscoll et al. 1991, Sullivan 2000), including intensive chemical, and in many cases biological, monitoring data collected during the past one to three decades. Despite substantial variability, there are recognizable patterns with respect to watershed characteristics associated with various acidification processes (c.f., Driscoll and Newton 1985, Driscoll et al. 1991, Sullivan et al. 1999). However, knowledge of acidification and recovery processes for individual watersheds is of limited value as a basis for natural resource management and public policy. Management decisions require information regarding numbers and percentages of the population of lakes for a region that have responded, or in the future will be expected to respond, in various ways. Sullivan et al. (2006a) determined the extent to which these well-studied watersheds represent larger units. Clearly, chemical, and likely biological, recovery from elevated inputs of acidic deposition is now occurring in some Adirondack lakes. We now need to know the extent to which additional reductions in atmospheric deposition will be required to affect more complete or full recovery.

The critical load (CL) is the level of sustained atmospheric deposition of S, N, or acidity below which harmful effects to sensitive ecosystems do not occur according to current scientific understanding. The CL is typically estimated using one or more process-based or empirical models. Databases developed by seven major research programs, coupled with process-based model simulations, offer an opportunity to evaluate the CL of acidic deposition of acid-sensitive Adirondack watersheds:

- Eastern Lakes Survey (ELS; Linthurst et al. 1986).
- Direct/Delayed Response Project (DDRP; Church et al. 1989).
- Environmental Monitoring and Assessment Program (EMAP; Larsen et al. 1994).
- Adirondack Long-Term Monitoring Project (ALTM; Driscoll et al. 2003b).
- Adirondack Effects Assessment Program (AEAP; Nierzwicki-Bauer et al. 2010).
- Adirondack Lakes Survey Corporation's (ALSC) survey (Kretser et al. 1989).
- The model-based assessment of Sullivan et al. (2006a).

The ELS, DDRP, EMAP, and Sullivan et al. (2006a) studies were all statistically based, thereby allowing population estimates to be developed. ALTM and AEAP involve on-going long-term lake monitoring efforts, but are not statistically based. DDRP contained a soils sampling component, but the soils data were regionally aggregated and were not specific to the watersheds under study. Sullivan et al. (2006b) developed a database of soil chemistry for the EMAP, ALTM, and AEAP lake watersheds selected for study here. Such soil data are needed for estimating CL using a process-based modeling approach.

Long-term monitoring programs revealed that some lakes show evidence of recent recovery of resources from acidification. The ALTM project has been conducting monthly monitoring of the water chemistry of 17 lakes, mostly in the southwestern Adirondack Mountains, since 1982 (Driscoll et al. 2003b). In 1992, the program was expanded by the addition of 35 lakes to the monitoring effort. The AEAP sampled the zooplankton of 30 lakes approximately twice per summer over a 10-year period beginning in 1994. Most of those lakes are also included within ALTM. The ALTM program has also sampled fish communities in many study lakes in recent years.

ALSC's survey was the most extensive survey of lake water chemistry in the Adirondack Mountains (Kretser et al. 1989, Baker et al. 1990). Over a four-year period, ALSC surveyed the chemistry and fisheries of 1,469 lakes. Despite the large number of lakes included, however, they were not drawn from a statistical frame, and therefore, the results cannot be used directly for population estimates. Also, some were smaller than 1 ha, which was the lower size cutoff for the EMAP survey.

The best statistical frame for assessing acidification and recovery responses of Adirondack lakes was developed by the U.S. EPA's EMAP (Larsen et al. 1994). The EMAP included lakes as small as 1 ha, involved both chemical and more limited biological characterization, and it was based on more accurate maps than the DDRP. The EMAP was designed to provide unbiased regional characterization of the entire population of Adirondack Mountain lakes larger than 1 ha.

Rates and extent of future ANC increase in Adirondack lakes in response to decreases in acidic deposition and associated CL values are of considerable policy interest. However, making regional assessments from sites such as the ALTM sites that were not statistically selected can lead to incorrect evaluations. Paulsen et al. (1998) reported on a number of inaccurate assessments made as a result of extrapolating such data. In all cases, a statistically-based probability survey showed markedly different regional conditions than did an evaluation that assumed that available data adequately represented the regional population of interest. The data record for the EMAP probability lakes is insufficient for measurement of recovery responses. Although these lakes can be used as a basis for model forecasts of recovery, they provide limited ability to determine whether or not predicted responses actually occur. Similarly, the scarcity of seasonal water chemistry and biological response data for the probability lakes limit their utility for assessment purposes. Nevertheless, the lakes that are surveyed based on probabilistic sampling are essential in order to place results and conclusions from the intensively-monitored lakes into the regional context.

Future changes in the structure and function of aquatic communities will be required to restore ecosystem health. Biological response data, especially for zooplankton and to a lesser extent for fish and phytoplankton, are available for Adirondack lakes from the AEAP, EMAP, ALTM, and ALSC programs. Data recently collected within these programs can provide important information regarding the biological resources at greatest risk of adverse impacts from acidic deposition, and the spatial and temporal patterns of biological recovery as deposition continues to decline (Nierzwicki-Bauer et al. 2010).



There is interest in techniques, such as liming, to accelerate the recovery of aquatic and terrestrial organisms in acid-sensitive regions of New York State as acidic deposition continues to be controlled. If the rate of chemical recovery can be enhanced, it may be possible to more rapidly restore biological community structure and function. However, much can be gained by examining the rate at which aquatic ecosystems are recovering now in response to recent decreases in emissions, and estimating the CL that would be required to achieve further or full recovery. Knowledge of the rate of ongoing responses and of the characteristics associated with those lakes that are actually recovering is critical to development of sound management policies. Such information will provide the basis for evaluating the efficacy of current and possibly expanded accelerated-recovery programs.

The research reported here was undertaken to determine the CL values that will promote resource recovery in aquatic and forest Adirondack ecosystems. The focus is on the Adirondack Ecoregion, and in particular the intensively-monitored watersheds, many of which are in the southwestern portion of the Adirondack Ecoregion. The latter group of watersheds include many that contain shallow surficial deposits (thin-till) that are highly sensitive to acidification and that are expected to be highly responsive to reductions in atmospheric S and N emissions and acidic deposition.

Resource managers are now confronted with questions regarding the extent to which air pollution emissions need to be further controlled in order to allow damaged resources to fully recover. To inform public policy regarding air pollutant emissions controls, it is important to determine 1) the level of emissions, and associated atmospheric deposition, that are associated with varying degrees of chemical effects and 2) the associations among water and soil chemistry and consequent biological impacts. Among the most important tools available to natural resource managers in this context are model calculations of critical and target loads.

The CL process typically involves selection of one or more sensitive receptor (s), one or more chemical indicator (s) of biological response for the sensitive receptor (s) of concern, and one or more critical chemical indicator threshold criteria values that have been shown to be associated with adverse biological impacts. For the sensitive receptor lake water, the most commonly selected chemical indicator is ANC. A number of critical criteria values of ANC have been used as the basis for CL calculations, the most common of which have been 0, 20, 50, and occasionally 100  $\mu\text{eq/L}$ . These levels appear to approximately correspond to chronic effects on brook trout (ANC = 0  $\mu\text{eq/L}$ ), episodic effects on brook trout and chronic effects on more sensitive fish species (ANC = 20  $\mu\text{eq/L}$ ), and effects on other more sensitive aquatic species (ANC = 50 to 100  $\mu\text{eq/L}$ ; cf., Cosby et al. 2006).

For protecting terrestrial resources, the  $\text{Ca}^{2+}$  or nutrient base cation ( $\text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+$ ) to inorganic Al molar ratio in soil solution is most commonly used, with critical thresholds usually set to 1.0 and sometimes 10.0. It might also be possible to use soil percent BS or exchangeable  $\text{Ca}^{2+}$  or a variable based on foliar chemistry, but critical thresholds for such indicators have not been well established.

The CL is generally calculated as a long-term steady state condition. Under constant atmospheric deposition at the determined CL, it may take many decades, centuries, or longer for the sensitive chemical indicator (i.e., lake ANC) to reach the designated critical criterion value. A dynamic CL, often called a target load (TL), can be calculated specific to a certain time period. For example, one might calculate the dynamic CL that would allow recovery of ANC to a level of 50  $\mu\text{eq/L}$  by the year 2050 for a particular lake or stream under constant loading at the CL level.

The exceedance is calculated by subtracting the ambient deposition loading from the CL. It reflects the extent to which the level of current ambient deposition exceeds the calculated CL. A target load (TL) can be set on the basis of the CL, also considering issues of recovery response times and other political or economic considerations. A TL can incorporate various management objectives. For example, if the CL for resource recovery has been estimated to be  $x$ , one may set a TL equal to  $1.5x$  (or some other value) as an interim target with the intention of reaching the TL within a certain number of years. This interim target, although higher than the CL, might be considerably lower than ambient deposition, thereby allowing for only partial resource recovery within a finite time period. Conversely, the TL could be set lower than the CL, for example if managers are unwilling to wait the decades or centuries that it might take to attain the critical criterion under constant loading at the CL level. The CL and the TL concepts have been used extensively in Europe for more than two decades to aid in air pollution abatement policy negotiations (Posch et al. 2001).

In this project, we integrate existing data from the AEAP, ALTM, EMAP, ALSC, and DDRP programs to more fully utilize available data and conduct a statistically representative assessment of the CLs of S and N deposition. The principal model for simulating aquatic, edaphic, and forest effects is called the Model of Acidification of Groundwater in Catchments (MAGIC), and it is used here as a CL simulation and integration tool. The MAGIC model provides the foundation for estimating the CL and for generating regional projections of ecosystem responses.

A previous NYSERDA project by Sullivan et al. (2006a) developed the technical foundation for the CL project reported here. In that earlier project, we:

- Surveyed soil chemistry.
- Compiled and collected lake water chemistry data.
- Developed input data for the MAGIC and PnET-BGC models.
- Developed biological dose/response relationships for fish and zooplankton.
- Simulated past and future lake and soil acid-base chemistry in 70 Adirondack watersheds in response to various emission control scenarios.
- Extrapolated model output to the region.
- Compared simulated output from the two models.
- Evaluated the representativeness of ALTM lakes with respect to extent of past acidification and future recovery responses for the Adirondack region. This new research project builds upon the previous project to determine long-term levels of acidic deposition that will be required to protect and restore acid-sensitive aquatic and terrestrial resources in the Adirondack Ecoregion.

Three sets of Adirondack lakes were available for CL modeling, each of which has previously been modeled using MAGIC. Sullivan et al. (2006a) applied MAGIC to 44 statistically-selected EMAP lakes and 32 long-term monitoring lakes, with an overlap of six lakes. MAGIC was previously applied to 35 Adirondack lakes within the USEPA's DDRP project. Population estimates of modeled CL values for the Adirondack region can be generated using just the 44 EMAP lakes. However, development of CL maps, based on modeled sensitivity classes and their relationship to mappable water chemistry and/or landscape features, benefit from access to dynamic CL model output for a larger number of lakes. In addition, it is desirable to ascertain the extent to which ALTM lakes represent the overall population of Adirondack lakes with respect to calculated CL amounts. Thus, there is value in modeling lakes from all three groups.

We developed a suite of CL values, each based on a specific combination of selected indicator, critical value, and endpoint years. These combinations included changes in S, and changes in N, atmospheric loading. For both sets of simulations, deposition of the strong acid anion not being varied for determination of CL (i.e., N loading for determination of critical S load) was set to follow future trajectories anticipated by the U.S. EPA in the Clean Air Interstate Rule (CAIR). Intermodel comparisons focus primarily on long-term CL estimates (year 2100) for lake water ANC = 50  $\mu\text{eq/L}$  and lake water  $\text{NO}_3^- = 10 \mu\text{eq/L}$ , based on deposition of S and  $\text{NO}_3^-$ . We investigate CL differences as a function of timeframe and as a function of endpoint criteria and critical threshold level.

Our goal was to establish CLs for S and N deposition to acid-sensitive Adirondack lake watersheds that are necessary to promote the continuation of ongoing aquatic recovery and to stimulate forest and soil recovery in the Adirondack Ecoregion. Major objectives include to:

1. Quantify, classify, and map lakes and their watersheds according to their CL to allow for resource recovery and to protect against further acidification.
2. Evaluate the sensitivity of CL calculations for Adirondack lake-watersheds to:
  - Watershed characteristics and associated biogeochemical processes.
  - The selection of critical chemical indicators (i.e., soil % base saturation, surface water ANC, and soil solution base cation-to-aluminum ratios) and their specific critical chemical limits (e.g., ANC 0  $\mu\text{eq/L}$ , 20  $\mu\text{eq/L}$ , 50  $\mu\text{eq/L}$ ).
3. Refine algorithms of biological response to acidic deposition for use in CL calculations for the Adirondack region.

Major findings of the research in this report were also published in two scientific journals:

- Sullivan, T.J., B.J. Cosby, C.T. Driscoll, T.C. McDonnell, and A.T. Herlihy. 2011. "Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification." *J. Environ. Stud. Sci.* 1(4):301-314. <http://dx.doi.org/10.1007/s13412-011-0062-8>.
- Sullivan, T.J., B.J. Cosby, C.T. Driscoll, T.C. McDonnell, A.T. Herlihy, and D.A. Burns. 2012. "Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York." *Water Resour. Res.* 48 doi:10.1029/2011WR011171.

## 2 Methods

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### 2.1 Site Selection

The primary sites selected for simulation of CL using the MAGIC model were those modeled by Sullivan et al. (2006a). In that study, an initial group of watersheds was selected based on the EMAP statistical design. An additional set of watersheds was selected from among those that were subjects of long-term chemical and biological monitoring efforts.

In the Adirondacks, the regional EMAP probability sample consisted of 115 lakes and their watersheds. The total number of target Adirondack lakes included in the EMAP frame was 1,829 (SE = 244). These included the lakes depicted on 1:100,000-scale USGS maps that were larger than 1 ha, deeper than 1 m, and that contained more than 1,000 m<sup>2</sup> of open water. Of those target lakes, an estimated 509 had summer index lake water ANC > 200 µeq/L; these were considered insensitive to acidic deposition effects and were not specifically modeled for the study reported here. The remaining 1,320 (SE = 102) low-ANC lakes constituted the frame for extrapolation of CL results. Details of the EMAP design were given by Larsen et al. (1994). Whittier et al. (2002) presented an overall assessment of the relative effects of various environmental stressors across northeastern lakes using EMAP probability survey data. Based on field measurements, 42% of the lakes in the EMAP statistical frame for the Adirondack region had summer index ANC ≤ 50 µeq/L and another 30% had ANC between 50 and 200 µeq/L. We focused this study on watersheds containing these two strata of low ANC lakes (≤ 50 and between 50 and 200 µeq/L) as they are thought to be most responsive to changes in air pollution. Lake water ANC provides an integrating watershed acid-base chemistry variable that reflects biotic, edaphic, geologic, and hydrologic conditions throughout the watershed. In this report, we present summer index ANC values. These approximately correspond to annual average values; ANC measurements during spring, especially in conjunction with snowmelt, would be expected to be lower.

Sullivan et al. (2006a) used a random selection process to choose candidate watersheds for soil sampling and modeling from among the 44 EMAP watersheds containing lakes with ANC ≤ 50 µeq/L and the 39 EMAP watersheds containing lakes with ANC between 50 and 200 µeq/L. Both primary and alternate sampling candidates were selected in the order they were to be included, in anticipation of the problem that we would be unable to sample soils in some of the selected watersheds (e.g., access difficulty or permission denied). The goal was to sample as least 30 EMAP watersheds containing lakes having ANC ≤ 50 µeq/L and 10 EMAP watersheds containing lakes having ANC between 50 and 200 µeq/L. To obtain a spatially balanced subsample, county was used as a spatial clustering variable in a manner identical to that used in the original EMAP probability design (Larsen et al. 1994). For lakes with ANC between 50 and 200 µeq/L, we used a variable probability factor based on lake ANC

class (50 to 100, 100 to 150, and 150 to 200  $\mu\text{eq/L}$ ) to obtain more samples in the lower ANC ranges. No variable probability factors were used for the  $\text{ANC} \leq 50 \mu\text{eq/L}$  lakes. Results of measurements or model projections for the selected EMAP watersheds can be extrapolated to the entire population of watersheds containing lakes with  $\text{ANC} \leq 50$  or  $\leq 200 \mu\text{eq/L}$ , using the original EMAP sample weights adjusted for this random subsampling procedure.

A total of 70 watersheds were sampled for soil chemistry and modeled with the MAGIC model by Sullivan et al. (2006a). Of those 70 watersheds, 44 were statistically selected to be representative of the wider regional lake population (estimated  $N = 1,320$ ). The EMAP study provides the best available base for statistical extrapolation of CL to the regional population of Adirondack lake-watersheds. This combined group of sites provided the basis for numerically extrapolating watershed-specific model estimates of CL region-wide.

Intensively-studied watersheds were drawn from the AEAP and ALTM databases, which included an overlap of 27 lakes. Six of the intensively-studied watersheds were also included within the selected EMAP lakes. We included in this study 29 of the 30 AEAP watersheds which have extensive databases for both chemical and biological lake monitoring. We also modeled 27 of the DDRP watersheds to aid in the spatial extrapolation of CL results across the Adirondack region. Selected EMAP, ALTM/AEAP, and DDRP study watersheds are listed in Table 2-1.

The spatial extent of the watersheds modeled for CLs using MAGIC is shown in Map 2-1 (left panel). Also shown is the spatial extent of the watersheds included in the ALS lake chemistry survey to which MAGIC S CL simulations were spatially extrapolated (right panel). In general, watersheds in the western Adirondacks were better represented in the modeling and extrapolation than were watersheds in the eastern Adirondacks.

**Table 2-1. Study watersheds**

Probability (EMAP) Sites				Intensively Monitored (ALTM) Sites			DDRP Sites			
Lake Name	EMAP ID	ALSC ID	Watershed Area (km <sup>2</sup> )	Lake Name	ALSC ID	Watershed Area (km <sup>2</sup> )	Lake Name	DDRP ID	ALSC ID	Watershed Area (km <sup>2</sup> )
<b>Low ANC (&lt;50 µeq/L)</b>				<b>Low ANC (&lt;50 µeq/L)</b>			<b>Low ANC (&lt;50 µeq/L)</b>			
Antediluvian Pond	NY287L	060126	1.5	Big Moose Lake	040752	92.7	Chub Lake	1A2-052	050264	0.6
Bennett Lake	NY256L	050182	2.7	Brook Trout Lake	040874	1.8	Fish Ponds	1A2-037	050288	8.7
Bickford Pond	NY297L	030273	0.7	Bubb Lake	040748	179	Gull Lakes (South)	1A1-073	040758	0.9
Big Alderbed	NY017L	070790	15.9	Carry Pond	050669	0.2	Hawk Pond	1A1-003	040504	1.0
Boottree Pond	NY284L	030374	0.1	Constable Pond	040777	9.7	Hitchcock Lake	1A1-057	040639	1.7
Canada Lake	NY292L	070717	101.5	Dart Lake	040750	14.8	Long Pond	1A3-046	050310	1.4
Dismal Pond	NY791L	040515	2.1	G Lake	070859	4.3	Middle Pond	1A1-029	020143	2.0
Dry Channel Pond	NY033L	030128	1.8	Grass Pond	040706	2.4	Mud Lake	1A2-041	050216	1.8
Effley Falls Pond	NY277L	040426	640.8	Helldiver Pond	040877	0.3	Nate Pond	1A3-001	050577	1.0
Hope Pond	NY012L	020059	0.5	Indian Lake <sup>a</sup>	040852	10.8	Nicks Pond	1A1-038	040292	0.6
Horseshoe Pond	NY285L	030373	0.9	Jockeybush Lake	050259	1.5	Nine Corner Lake	1A2-046	070719	2.0
Indian Lake <sup>a</sup>	NY015L	040852	10.8	Lake Rondaxe	040739	139.4	North Branch Pond	1A2-042	070825	2.3
Little Lilly Pond	NY029L	40566	2.2	Limekiln Lake	040826	10.4	Parlow Lake	1A1-046	040382	3.6
Long Lake	NY018L	070823	0.7	Long Pond	050649	0.1	St. John Lake	1A2-002	050192	1.5
Lower Beech Ridge	NY790L	040203	0.3	Middle Branch Lake	040707	3.6	Trout Lake	1A2-054	070793	2.4
Mccuen Pond	NY782L	060039	0.6	Middle Settlement	040704	1	Wilmurt Lake	1A1-014	070850	2.6
North Lake <sup>a</sup>	NY279L	041007	75.4	North Lake <sup>a</sup>	041007	75.4	Woodhull Lake	1A1-066	040982	18.8
Parmeter Pond	NY278L	030331	0.5	Queer Lake	060329	3.5	Woods Lake	1A2-045	050156	1.7
Payne Lake	NY794L	040620	1.7	Round Pond	040731	0.1	Zack Pond	1A3-040	050673	2.5
Razorback Pond	NY280L	040573	0.2	Sagamore Lake	060313	48.4				
Rock Pond	NY286L	060129	35.9	South Lake <sup>a</sup>	041004	14.5				
Rocky Lake	NY527L	040137	1.1	Squash Pond	040754	0.5				
Second Pond	NY013L	050298	4.4	Squaw Lake <sup>a</sup>	040850	1.2				
Seven Sisters Pond	NY288L	060074	0.7	West Pond	040753	1.2				
Snake Pond	NY281L	040579	0.5	Wheeler Lake	040731	0.2				
South Lake <sup>a</sup>	NY282L	041004	14.5	Willis Lake	050215	1.4				
Squaw Lake <sup>a</sup>	NY014L	040850	1.2	Wilys Lake <sup>a</sup>	040210	1.3				
Trout Pond	NY767L	060146	2.5							
Upper Sister Lake	NY030L	040769	13.9							
Whitney Lake	NY797L	070936	2.3							
Wilys Lake <sup>a</sup>	NY789L	040210	1.3							
Witchhopple Lake	NY788L	040528	19.7							
Wolf Pond	NY515L	030360	0.2							

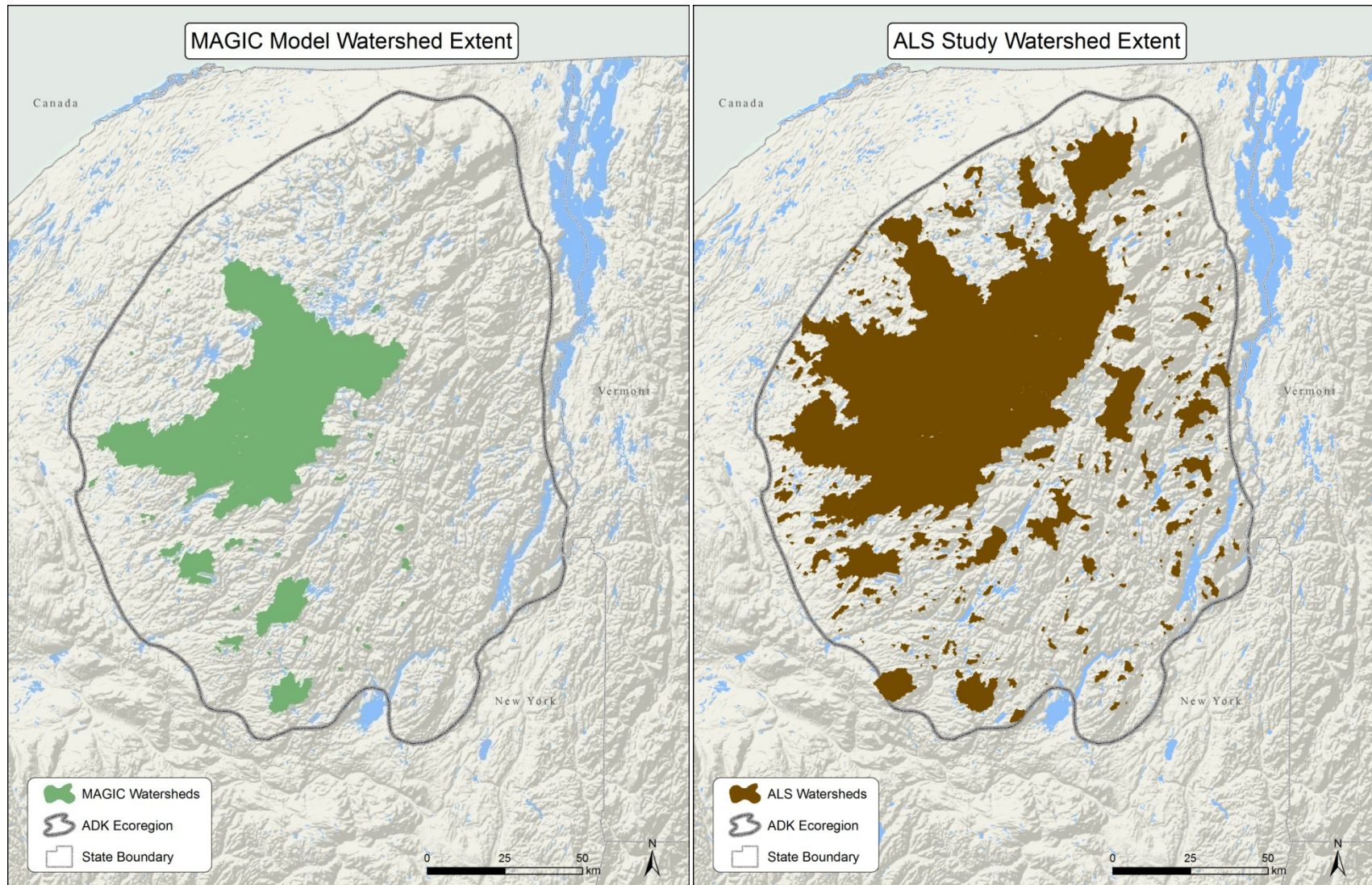
Table 2-1 continued

Probability (EMAP) Sites				Intensively Monitored (ALTM) Sites			DDRP Sites			
Lake Name	EMAP ID	ALSC ID	Watershed Area (km <sup>2</sup> )	Lake Name	ALSC ID	Watershed Area (km <sup>2</sup> )	Lake Name	DDRP ID	ALSC ID	Watershed Area (km <sup>2</sup> )
<i>Intermediate ANC (50-200 µeq/L)</i>				<i>Intermediate ANC (50-200 µeq/L)</i>			<i>Intermediate ANC (50-200 µeq/L)</i>			
Arbutus Pond <sup>a</sup>	NY786L	050684	3.2	Arbutus Pond <sup>a</sup>	050684	3.2	4th Bisby Lake	1A1-020	040973	2.7
Blue Mountain Lake	NY520L	060307	51.7	Cascade Lake	040747	4.8	Cheney Pond	1A3-042	050672	1.6
Bog Pond	NY528L	060175	27.3	Moss Lake	040746	12.5	John Pond	1A1-039	040321	1.9
Carry Falls Reservoir	NY522L	060035	2265.1	Raquette Lake	060315A	1.9	Kiwassa Lake	1A1-033	020100	5.2
Clear Pond	NY529L	020070	0.9	Windfall Pond	040750A	4	Mt. Arab Lake	1A1-064	060083	2.2
Clear Pond	NY005L	060176	1.6				No Name Pond	1A2-048	070712	0.4
Gull Pond	NY020L	050418	0.2				Wolf Pond	1A1-061	040873	2.1
Hitchins Pond	NY768L	060144	133.3							
Long Pond	NY010L	030170	0.6							
Piseco Lake	NY775L	050234	143.7							
Seventh Lake	NY533L	050631	43.1							
							<i>High ANC (&gt; 200) µeq/L</i>			
							Unknown pond	1A3-043	050658	4.5

<sup>a</sup> Occurs in both probability and intensively-monitored groups

## Map 2-1. Spatial extent of watersheds modeled for S critical loads

The left panel shows the extent of the watersheds modeled with MAGIC. The right panel shows the extent of the ALS watersheds included in the ALS lake chemistry survey to which MAGIC aquatic CL simulations were spatially extrapolated.





## 2.2 Data Compilation

Water quality data for this study were derived from the EMAP, ALTM, ELS, and Adirondack Lakes Survey Corporation (ALSC) survey and monitoring efforts, and from the study of Sullivan et al. (2006a). Soil chemistry data were derived from Sullivan et al. (2006b). The required lake water and soil composition data for the modeling efforts included the following measurements:

- Lake water composition: pH, ANC,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{Cl}^-$ .
- Soil properties: thickness and total cation exchange capacity, exchangeable bases ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ , and  $\text{Na}^+$ ), bulk density, porosity, and pH.

Where available, the water chemistry database also included dissolved organic carbon (DOC), dissolved inorganic carbon (DIC),  $\text{H}_4\text{SiO}_4$ , and inorganic monomeric Al ( $\text{Al}_i$ ).

Availability of lake water composition data for at least one sample occasion was a prerequisite for all candidate sites for model application. Typically, multiple lake water composition records were available. For most of the lakes included in the ALTM and/or AEAP studies, about 10 to 20 years of monthly monitoring data were available. For most other EMAP study lakes, data from approximately 3 to 10 water samples were available. For 13 study lakes, additional water samples were collected and analyzed at Syracuse University in 2003 by Sullivan et al. (2006a), following methods comparable to those of ALTM. For the 30 DDRP lakes, water chemistry data from the ELS study were used.

Soil data were derived from a survey of the 70 EMAP and ALSC/AEAP watersheds (Sullivan et al. 2006b). Soils data for model application to the DDRP watersheds in this study, other than those that overlapped with the EMAP or ALTM/AEAP studies, were borrowed from the watersheds surveyed by Sullivan et al. (2006b) using a nearest neighbor approach. A similar paired watershed (from among those having soil data) was identified for each of the DDRP watersheds considering location, drainage water chemistry (ANC,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$  concentrations), and elevation.

## 2.2.1 Effects of Soil Borrowing on CL Simulation Results

A series of CL simulations was conducted as a sensitivity analysis to evaluate the potential effects of borrowing soils data for the MAGIC model calibrations that were constructed for the 27 modeled DDRP sites that had not been included in the soil survey conducted by Sullivan et al. (2006b). For these 27 modeled sites, recent soils data were not available for use in model calibration. For each of these watersheds, a nearest-neighbor approach was used to identify, from among the 70 MAGIC modeling sites that did have recent soils data (the sites surveyed by Sullivan et al. 2006b), the watershed most similar to the DDRP site with respect to geographic location, elevation, ratio of watershed area to lake surface area (WA:SA), and key elements of lake water chemistry (ANC,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , DOC concentrations). Model soil input data were then borrowed from the nearest-neighbor site to provide the needed soil input for calibrating MAGIC to the DDRP site.

To evaluate the uncertainty contributed to modeling the various CL values by this soil data borrowing procedure, 10 watersheds from among the 70 sites surveyed by Sullivan et al. (2006b) were calibrated twice. The first calibration for each was based on the actual soil data obtained from that watershed; the second sensitivity calibration was based on borrowed soils data, using the same borrowing procedures as employed for the DDRP watersheds. The subject watersheds were selected to span a range of lake water ANC and simulated CL values. Of the 10 selected watersheds, 8 yielded successful model calibration pairs (calibrations based on measured watershed soils and borrowed soils data, respectively). The calibration pairs were then used as the basis for simulating paired CL values to protect against damages caused by S deposition. The difference in simulated CL for a given site, depending on whether measured or borrowed soils data were used in the calibration, is used as an indication of the additional uncertainty introduced to the CL modeling effort by the soils borrowing procedure used for the DDRP sites that lacked recent measured soils data.

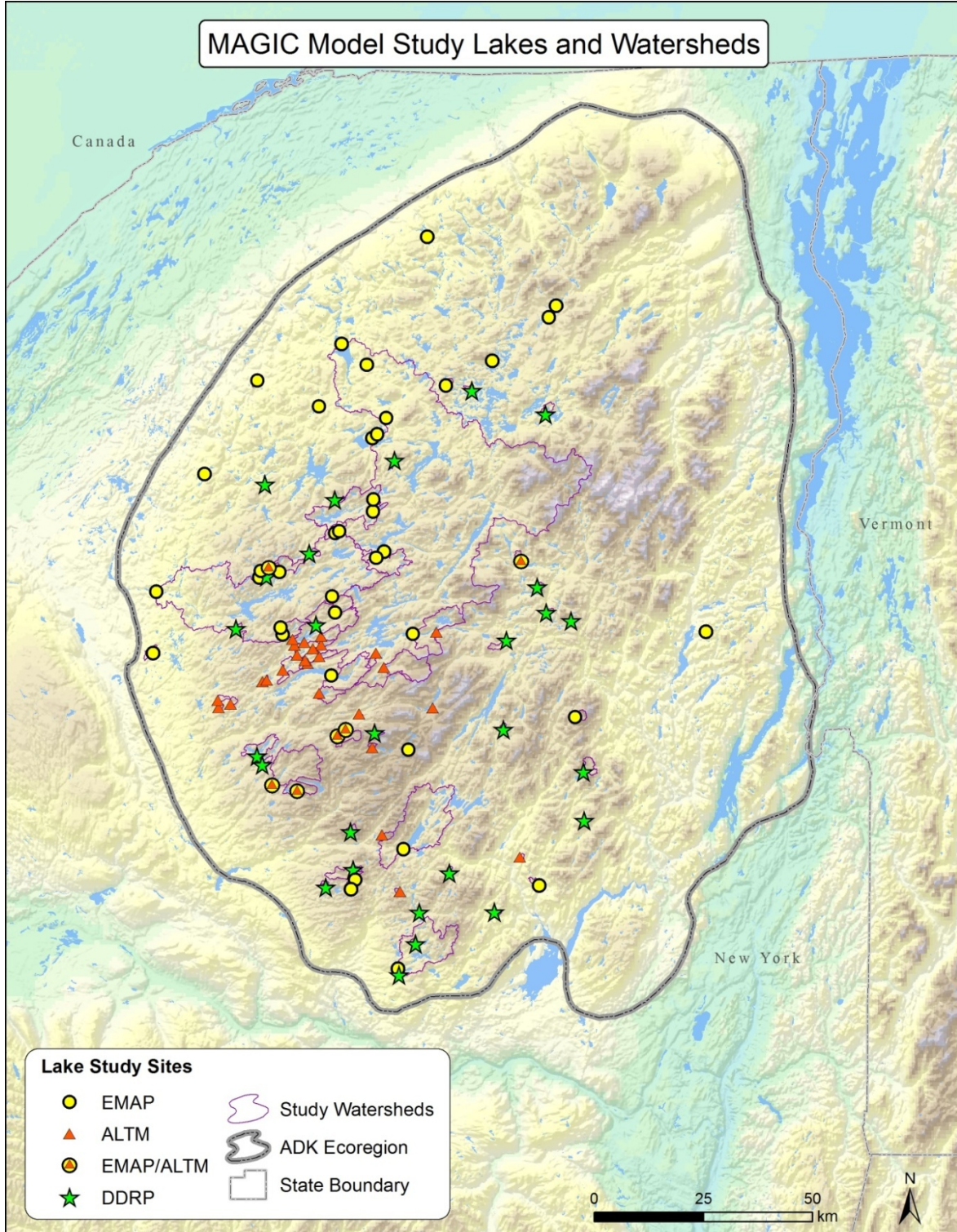
## 2.3 Modeling

### 2.3.1 Modeling Approach

The MAGIC model was used to simulate CL based on lake water and soil indicator values for 97 Adirondack lakes and their watersheds. The locations of the Adirondack lakes modeled with MAGIC are shown in Map 2-2. Map symbols indicate which of the modeled lakes were included in the EMAP probability sampling, the ALTM/AEAP monitoring programs, and the additional lakes included in the DDRP study. Six lakes were included in both EMAP and ALTM. Results were extrapolated to the regional population. Aquatic S CL values were mapped region-wide. MAGIC (Cosby et al. 1985b, a, Cosby et al. 1985c) was developed to predict the long-term effects of acidic deposition on surface water chemistry. A critical concept in MAGIC is the size of the pool of exchangeable base cations on the soil. As the fluxes to and from this pool changeover time owing to changes in atmospheric deposition, the chemical equilibria between soil and soil solution shift to give changes in surface water chemistry. MAGIC was

**Map 2-2. Locations of lake watersheds in the Adirondack Ecoregion that were modeled with the MAGIC model**

Watershed boundaries are also indicated. Many of the study watersheds are too small to be seen at this map scale.



the principal model used by the National Acid Precipitation Assessment Program (NAPAP) in assessment of potential future damage to lakes and streams in the eastern United States (Thornton et al. 1990, NAPAP 1991). The validity of the model has been confirmed by comparison with estimates of lake acidification inferred from paleolimnological reconstructions of historical changes in lake pH (Wright et al. 1986, Jenkins et al. 1990b, Sullivan et al. 1995) and with the results of several catchment-scale experimental acidification and de-acidification experiments (e.g., Wright and Cosby 1987, Cosby et al. 1995, Cosby et al. 1996, Moldan et al. 1998). MAGIC has been used to reconstruct the history of acidification and to simulate future trends on a regional basis and in a large number of individual catchments in both North America and Europe (e.g., Wright et al. 1986, Lepistö et al. 1988, Whitehead et al. 1988, Hornberger et al. 1989, Cosby et al. 1990, Jenkins et al. 1990a, Jenkins et al. 1990c, Norton et al. 1992, Wright et al. 1994, Ferrier et al. 1995, Sullivan et al. 1996, Wright et al. 1998, Cosby et al. 2001, Sullivan et al. 2002, Sullivan et al. 2008).

MAGIC is a lumped-parameter model of intermediate complexity that was developed to predict the long-term effects of acidic deposition on surface water chemistry (Cosby et al. 1985b, Cosby et al. 1985c). The model simulates soil solution chemistry and surface water chemistry to predict the monthly and annual average concentrations of major ions in these waters. MAGIC consists of: 1) a submodel in which the concentrations of major ions are assumed to be governed by simultaneous reactions involving  $\text{SO}_4^{2-}$  adsorption, cation exchange, dissolution-precipitation- speciation of Al and dissolution-speciation of inorganic C; and 2) a mass balance submodel in which the flux of major ions to and from the soil is assumed to be controlled by atmospheric inputs, chemical weathering, net uptake and loss in biomass, and losses to runoff. The degree and rate of change of surface water acidity depend both on flux factors and the inherent characteristics of the affected soils.

Cation exchange is modeled using equilibrium (Gaines-Thomas) equations with selectivity coefficients for each base cation and Al. Sulfate adsorption is represented by a Langmuir isotherm. Aluminum dissolution and precipitation are assumed to be controlled by equilibrium with a solid phase of  $\text{Al}(\text{OH})_3$ . Aluminum speciation is calculated by considering hydrolysis reactions as well as complexation with  $\text{SO}_4^{2-}$  and  $\text{F}^-$ . Effects of  $\text{CO}_2$  on pH and on the speciation of inorganic C are computed from equilibrium equations. Organic acids are represented in the model as tri-protic analogues. Element weathering and the uptake rate of N are assumed to be constant, based on model calibration. A set of mass balance equations for base (Cosby et al. 1989) cations and strong acid anions are included.

Given a description of the historical deposition at a site, the model equations are solved numerically to give long-term reconstructions of surface water chemistry. For more complete details of the model, see Cosby et al. (1985b, 1985c, 1989).

The aggregated nature of the model requires calibration to observed data from a system before it can be used to examine potential system response. Calibration is achieved by setting the values of certain parameters within the model that can be directly measured or observed in the system of interest (called fixed parameters). The model is then run (using observed and/or assumed atmospheric and hydrologic inputs) and the outputs (stream water and soil chemical variables, called criterion variables) are compared to observed values of these variables. If the observed and simulated values differ, the values of another set of parameters in the model (called optimized parameters) are adjusted to improve the fit. After a number of iterations, the simulated-minus-observed values of the criterion variables usually converge to zero (within some specified tolerance). The model is then considered calibrated.

The estimates of the fixed parameters and deposition inputs are subject to uncertainties so a "fuzzy optimization" procedure is implemented for calibrating the model. The optimization procedure consists of multiple calibrations using random values of the fixed parameters drawn from the observed possible range of values, and random values of deposition from a range including uncertainty about the estimated values. Each of the multiple calibrations begins with (1) a random selection of values of fixed parameters and deposition, and (2) a random selection of the starting values of the optimized parameters. The optimized parameters are then adjusted using the Rosenbrock (1960) algorithm to achieve a minimum error fit to the target variables. This procedure is undertaken 10 times. The final calibrated model is represented by the ensemble of parameter values and variable values of the 10 calibrations.

## **2.3.2 Input Data**

### **2.3.2.1 Deposition**

The acid-base chemistry modeling for this project was conducted using 2002 as the Base Year. The effects models were calibrated to the available atmospheric deposition and water chemistry data and then interpolated or extrapolated to yield Base Year estimates of lake water chemistry in the year 2002, which served as the starting point for specifying the CL values.

Wet deposition in the Adirondacks has been monitored by the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) since 1978 at two sites: Huntington Forest and Whiteface Mountain. Measured values of recent and current wet deposition throughout the eastern United States were derived from NADP/NTN through the year 2005. Wet deposition measurements by NADP/NTN for the five-year period centered on 2002 were interpolated to each study watershed and to the study region (cf., Grimm and Lynch 1997). Dry deposition was estimated using output from the CMAQ model for 2002 to establish dry to wet deposition ratios for S and N. For each study watershed, a really-weighted total wet plus dry S and N deposition values were calculated using the NADP wet deposition and the CMAQ dry to wet ratios.

Cloud deposition was not considered, because the study watersheds are almost completely lacking in land above 1,000-m elevation. Only three of the EMAP and ALTM/AEAP study lakes (Carry Falls Reservoir, Blue Mountain Lake, Sagamore Lake) included any land in the watershed that was higher than 1,000 m. The Sagamore Lake watershed included the highest percentage of such high-elevation land, but it only accounted for 2.4% of the watershed.

Empirical relationships between regional emissions and ionic concentrations in precipitation, coupled with historical regional emissions inventories, were used to estimate the time series of historical wet S and N deposition for each study watershed (c.f., Driscoll et al. 2001). For the base cations and chloride, background pre-industrial deposition was assumed to be 10% of current deposition. Deposition inputs after 1850 were assumed to increase linearly to estimated values obtained for 1950. Wet deposition estimates during 1950 to 1978 for  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ , and  $\text{Cl}^-$  were derived from empirical relationships between wet deposition and emissions of particulate matter (PM-10; Nizich et al. 1996). The model calculations assumed a fixed wet to dry deposition ratio. This value was assumed to be 0.3 for each of the base cations and 0.24 for  $\text{Cl}^-$ , based on a summary of dry-bucket deposition data by Baker (1991).

Past, current, and future total deposition amounts of major ions were interpolated to the midpoint of each study watershed. Although the absolute values of total deposition were variable from watershed to watershed, the patterns of change over time were similar.

### **2.3.2.2 Base Cation Uptake**

Forest uptake fluxes of the three nutrient base cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ;  $\text{Bc}_{\text{up}}$ ) were estimated from literature values summarized by McNulty et al. (2007). To estimate base cation removal from the watershed, estimates of annualized growth rate were used under the assumption that 65% of the tree volume is removed from the site during harvest. These uptake terms reflect uptake into woody materials that are removed from the watershed through timber harvest. Uptake into vegetation that subsequently dies on site represents within-watershed recycling; this is not a net watershed loss. Lands identified as designated wilderness and other protected areas were classified as “no harvest;”  $\text{Bc}_{\text{up}}$  was set to zero in such areas. These included areas identified in the Protected Areas database constructed by the Commission for Environmental Cooperation, corresponding to GAP codes 1 and 2 (Scott et al. 1993). The  $\text{Bc}_{\text{up}}$  parameter was determined for each of the 97 modeled watersheds as a spatially-weighted watershed average.

## 2.4 Regional Population Frame

In reporting numbers and percentages of Adirondack lakes above or below various CL or exceedance values from MAGIC simulations, it is of critical importance to consider the population of lakes to which these numbers and percentages pertain. Lakes can be defined in various ways, and the selected definition can have considerable influence on the results reported for a regional CL assessment.

Of particular importance in describing and defining a lake population is the lower size limit for what constitutes a lake. The EPA's Eastern Lakes Survey (ELS) surveyed lakes larger than about 4 ha within the Adirondack subregion of the Northeast region. The Adirondack subregion of the ELS included some landscape that is beyond the borders of the Adirondack Ecoregion. The EPA's EMAP study surveyed lakes larger than 1 ha. The ALS included some lakes that were considerably smaller than 1 ha. Because lake size distributions tend to be heavily skewed toward smaller size classes, choice of the minimum lake size included in the population of interest has a large influence on calculations of the number and percentage of acid-sensitive lakes (Sullivan 1990).

Analysis of the EMAP database indicates that there were an estimated 1,891 lakes larger than 1 ha within the Adirondack Ecoregion in the EMAP frame population based on a random sampling of lakes depicted at 1:100,000 map scale in the early 1990s. Of those lakes, about 13% were determined by EMAP field crews to be either non-lakes (not a water body) or non-target (<1 m depth, scarcity of open water; A.T. Herlihy, unpublished data). These criteria removed from consideration shallow and weedy areas in the transition zone between lake and wetland. At the time of the EMAP sampling, there were an estimated 1,645 target lakes in the Adirondack Ecoregion that were larger than 1 ha and deeper than 1 m. There was uncertainty associated with this estimate because it was based on a statistical sampling, rather than a count of known mapped lakes. In addition, there were unresolved EMAP issues related to wide areas along a stream system and the presence in the Adirondacks of chain lakes, which could be counted as one lake or as many.

The hydrography dataset has undergone improvement over the past 20 years. The current mapped population of lakes larger than 1 ha at a comparable scale to the EMAP frame (1:100,000) can be derived from the medium resolution National Hydrography Dataset (NHD). This more recent dataset shows 2,266 lakes in the Adirondack Ecoregion larger than 1 ha. There is no basis available, however, for subsetting the NHD to those lakes that are deeper than 1 m and have sufficient open water; this would require a statistical sampling with field visits.

The EMAP lake frame for the Adirondack Ecoregion was further subset by Sullivan et al. (2006a) to only include those lakes that had ANC <200 µeq/L. This was done to focus the research effort on the lakes of greatest interest with respect to acid sensitivity. There are an estimated 1,320 Adirondack lakes in the EMAP frame that have ANC below that threshold value.

The ALSC surveyed 1,469 Adirondack lakes during the period 1984 to 1990. Of those surveyed lakes, 1,136 were larger than 1 ha in area and located within the Adirondack Ecoregion. The lakes were not statistically selected from a mapped frame, but the ALS did survey a relatively high percentage of the lakes in the Adirondack Ecoregion that are larger than 1 ha.

For this CL assessment, a number of population frames and lake sets are considered. Results of CL calculations and extrapolations must always be evaluated in light of the population of lakes represented. All analyses reported here pertain only to lakes within the Adirondack Ecoregion, and to lakes larger than or equal to 1 ha in surface area.

## **2.5 Regional Extrapolation of Model Output**

Regional extrapolation of MAGIC model results focused on 1) numbers and percentages of lakes and their watersheds projected by the MAGIC model to have CL and exceedance values at certain levels, and 2) maps showing the locations of lake watersheds in various CL and exceedance classes. EMAP was the only statistically rigorous spatial frame available for quantitative extrapolation that covered the geographical extent of the Adirondack Ecoregion and included lakes as small as 1 ha. By applying modeling results from the 44 statistically-selected modeled watersheds to the EMAP statistical frame, we were able to estimate CL levels for all of the watersheds in the region that were represented by the statistical design. The best basis available for spatial extrapolation and CL mapping was the ALS lake chemistry survey in the 1980s.

The statistical extrapolation work performed for this study followed the procedures developed by Sullivan et al. (2006a). Population expansions were made using the adjusted sample weights from EMAP. Regional CL mapping was based on relationships between modeled critical loads and mappable water chemistry and/or landscape features that correlate with CL. Although spatial patterns of lake water acidification within the Adirondack region are well-known (c.f., Sullivan et al. 1990, Driscoll et al. 1991, Sullivan 2000), spatial patterns of CL and resource recovery in response to decreases in acidic deposition have been less thoroughly studied.

The acid-base chemistry of lakes is reflected in the lake water by ANC and the concentrations of strong acid anions and base cations in solution. Thus, candidate water chemistry independent variables selected for extrapolation in this analysis included the variables given in Table 2-2. MAGIC calculates ANC as the difference between the sum of the base cations (SBC) and the sum of the mineral acid anions (SAA) The calculated ANC is termed CALK.



**Table 2-2. Candidate variables for spatially extrapolating CL**

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<b>Landscape Characteristics</b>	<b>Water Chemistry</b>
<ul style="list-style-type: none"><li>• Watershed area</li><li>• WA:SA</li><li>• Elevation</li><li>• Slope</li><li>• % clay in soil</li><li>• Soil pH</li><li>• Soil depth to restricting layer</li></ul>	<ul style="list-style-type: none"><li>• ANC</li><li>• pH</li><li>• Sum of base cations</li><li>• Sum of base cations – chloride</li><li>• Sulfate</li><li>• Nitrate</li></ul>

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### **2.5.1 Development of Spatial Extrapolation Parameters**

Elevation data at a resolution of one arc-second (approximately 30 meters) were extracted from NHDPlus data as prepared for the National Elevation Dataset by the U.S. Geological Survey (USGS). Average elevation and percent slope for each watershed modeled with MAGIC were calculated from the elevation data.

Soils data from the Soil Survey Geographic (SSURGO) database were available for the majority of the study area (<http://www.nrcs.usda.gov/wps/portal/nrcs/main/soils/survey/geo/>). Where SSURGO data were not available, the coarser-scaled State Soil Geographic (STATSGO2; U.S. General Soils Map, ([http://www.nrcs.usda.gov/wps/portal/nrcs/detail/soils/survey/geo/?cid=nrcs142p2\\_053629](http://www.nrcs.usda.gov/wps/portal/nrcs/detail/soils/survey/geo/?cid=nrcs142p2_053629))) data were substituted. Soil parameters that were extracted from these databases for this study included depth to restricting layer, percent clay, and pH. SSURGO and STATSGO are spatially represented using map units. Each map unit is typically comprised of multiple components. The soils parameters were tabulated and coded to each soil map unit based on a component weighted average. The resulting tabular data were joined with the spatial polygon data and converted to a 30-m grid using the maximum area cell assignment option in ArcGIS. Soils data that coincided with lake locations according to medium resolution NHD data were set to null values.

Depth to restricting layer was defined as the depth to the first soil layer that prevents root penetration and water movement as represented in the soil databases. These depths were calculated for each component and then weighted and summed to generate a representative depth to restricting layer for each map unit. STATSGO2 data were used where SSURGO data contained no data or a value of 0. A limited portion of the study area was classified as open water. This step was required in order to maintain continuity during application of the continuous upslope averaging function (McDonnell et al. 2012). The no-data cells (corresponding with open water) were filled with an average of the nearby data cells (30 × 30 cell window) using the focal statistics function in ArcGIS.

Soil components in SSURGO and STATSGO2 are attributed with percent clay at multiple soil horizons. Therefore, percent clay was calculated as a soil horizon thickness weighted average for each component. The representative percent clay for each map unit was then calculated as a component weighted average. STATSGO2 data were used where SSURGO data contained no-data or a value of 0. The open water cells were treated as for soil depth calculations. The same methods as described for percent clay were followed for generating a representative pH value for each map unit.

## **2.5.2 Establishing CL Predictor Equations**

Regression techniques were used to establish equations to be used for spatial CL extrapolation using Statistix 8.0. Both landscape and water chemistry variables were used as candidate predictor variables in the regression analyses (Table 2-1). We attempted to establish two predictor equations using stepwise linear regression, one using both landscape characteristics and water chemistry parameters (for use in watersheds for which lake chemistry data are available) and another using landscape characteristics only. Watershed averages were used to represent the spatial variability within each watershed for the landscape characteristics.

The CL predictor equations were developed using MAGIC model CL outputs because they were available for the largest number of Adirondack lake-watersheds (n=97). Analyses focused on mappable factors known or suspected to influence watershed sensitivity to acidification in this region. Predictor equations were developed for CLs of both S and N deposition for protecting the soil BS and lake water ANC criteria for the years 2050 and 2100. The predictor equations were used to generate aquatic CL maps for the Adirondacks.

## **2.6 Uncertainty**

### **2.6.1 Uncertainty in MAGIC**

There are numerous uncertainties associated with conducting an assessment of this type, some of which are quantifiable, some not. The major sources of uncertainty in the assessment based on MAGIC model simulations of CL include input data quality; temporal variability in water chemistry; variability in biological response to water chemistry; model validity and accuracy; model calibration uncertainty; errors associated with missing model input data; and errors associated with regional extrapolation of modeling results from individual watersheds to the region. In this discussion, we focus on the elements of uncertainty arising from the MAGIC model simulations.

The aggregated nature of the MAGIC model requires that it be calibrated to observed data from a system before it can be used to examine potential system response. Calibration of MAGIC for each watershed is accomplished by specifying the model inputs (forcing functions), setting the values of those parameters for which measurements were available (fixed parameters), and then determining the values of the remaining parameters for which data were not available (adjustable parameters). The latter step is accomplished through an optimization procedure that selects

values of the adjustable parameters such that the squared errors between the simulated and observed values of important state variables in the model for which observed data are available (target variables) are minimized. Formal procedures for estimation of optimal values of the adjustable parameters (and their error variances) customarily ignore several sources of uncertainty in this calibration procedure, including:

- The initial estimate of the adjustable parameters used in the optimization algorithm (noisy response surface).
- The values of the fixed parameters.
- measured variables used to evaluate the squared errors (noisy target variables).
- Errors in the specified inputs used to drive the model.

The relative magnitude of the effects of each of these sources of uncertainty has been extensively evaluated for regional, long-term MAGIC simulations in a series of uncertainty analyses using Monte Carlo methods (see Cosby et al. 1989, Hornberger et al. 1989, Cosby et al. 1990, Sullivan and Cosby 2002, Sullivan et al. 2003, Sullivan et al. 2004). The results of those analyses implied that, while their relative effects may vary from application to application (with data quality and/or quantity), each of the four categories of uncertainty could have important effects on MAGIC simulations. Those studies led to the development of a multiple (also called “fuzzy”) optimization procedure for MAGIC that can be used for both regional and site specific applications (see Cosby et al. 1990, Wright et al. 1994). The multiple optimization procedure explicitly accounts for components of each of the four listed categories of uncertainty, and produces a time-variable measure of overall simulation uncertainty for each state variable. The procedures developed in these previous studies were applied to the MAGIC applications in this project. Results were consistent with previously referenced studies.

## 3 Results and Discussion

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### 3.1 Model Calibration Results

#### 3.1.1 Predicted versus Observed Chemistry

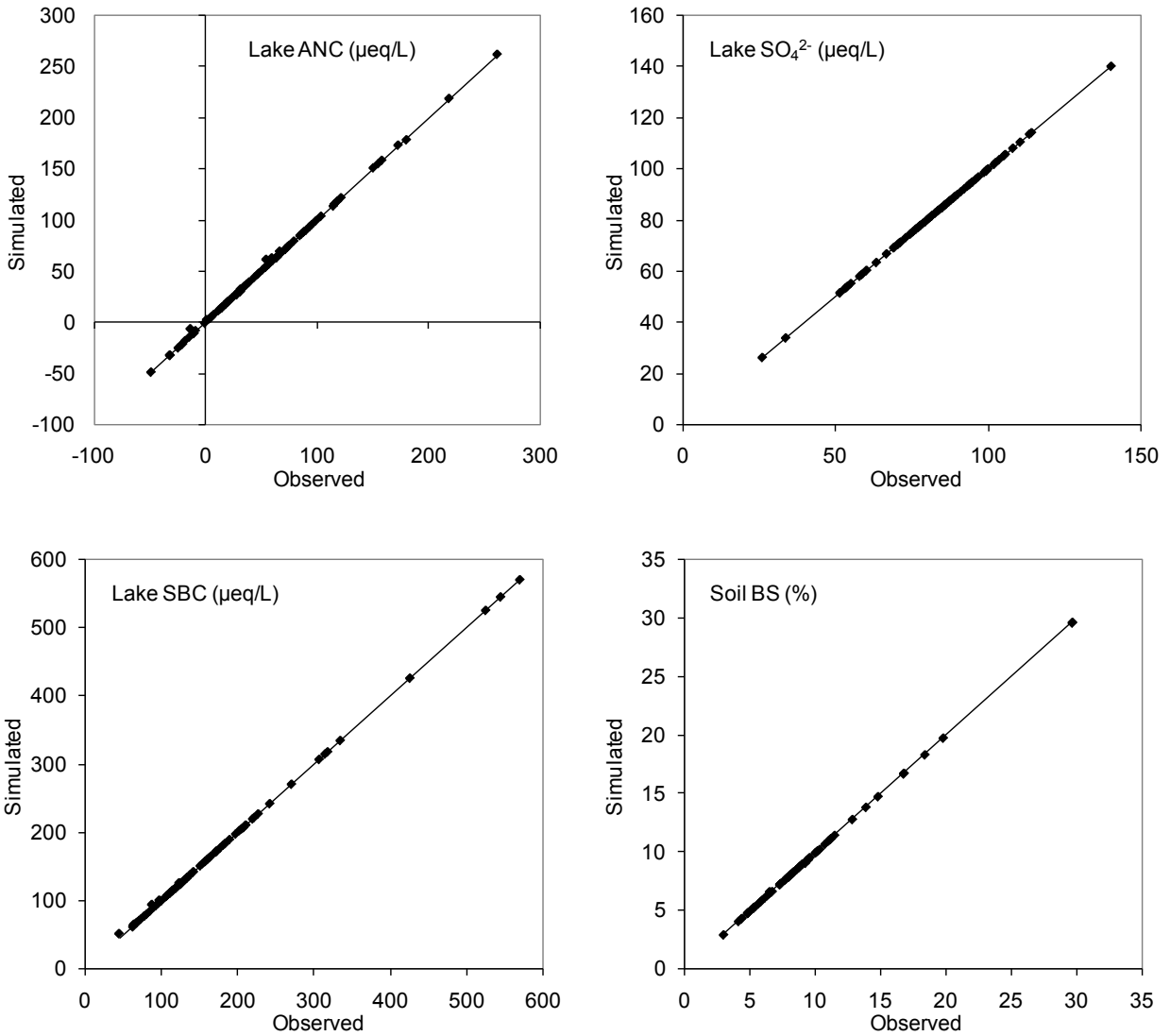
Results of predicted (using the MAGIC model) versus observed average lake water and soil chemistry during the calibration/evaluation period are given for selected variables in Figure 3-1. Results showed close agreement between simulated and measured values of key constituents at all sites. For the ANC simulations, the root mean squared error (RMSE) for predicted versus observed values, based on the average measured ANC value over a five-year period, was 1.2  $\mu\text{eq/L}$ . These results suggest that the model calibrations were unbiased across the modeled sites for simulating acid-base chemistry.

#### 3.1.2 Effects of Borrowing Soil Data for Calibrating DDRP Watersheds

Soil survey results obtained by Sullivan et al. (2006b) were used in calibrating each of the 70 study watersheds that comprise the EMAP and ALTM lake watershed datasets used in this study. Numeric extrapolations of CL to the full EMAP lake frame (presented in Section 3.4 of this report) are based only on watersheds for which soil samples were collected in 2003 and laboratory analyses performed. For spatial extrapolation, however, it is advantageous to simulate CL for the maximum number of Adirondack lake watersheds possible in order to provide a robust foundation for empirical extrapolation to the larger population of lake watersheds in the Adirondack Ecoregion. For that reason, 27 watersheds were modeled from the DDRP study in addition to the 70 that had been sampled for soils by Sullivan et al. (2006b) and modeled by Sullivan et al. (2006a). In order to calibrate these additional watersheds with the MAGIC model, soils data were borrowed from some of the 70 watersheds sampled by Sullivan et al. (2006b) using a nearest-neighbor approach.

Additional uncertainty was introduced to the CL calculations by the soil borrowing procedure, although this uncertainty only applied to model simulations for the 27 DDRP watersheds. This uncertainty was quantified by comparing simulated S CL values for a group of eight watersheds that were successfully calibrated twice: once based on measured soils data and a second time based on soils data that had been borrowed from the nearest neighbor watershed that also had recent measured soils data. Results of those sensitivity analyses are shown in Figure 3-2.

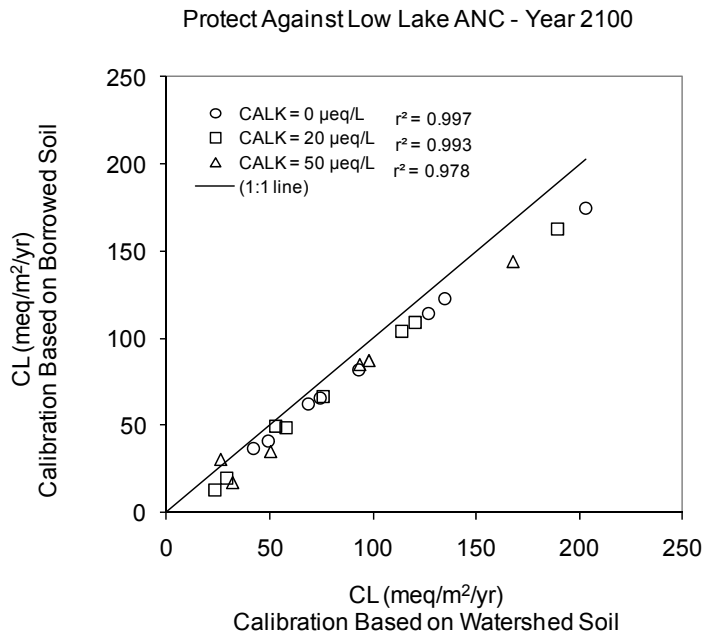
**Figure 3-1. MAGIC model simulated versus observed lake and soil chemistry in the model calibration year for the 70 modeled lake watersheds that had watershed-specific soils data**



**Figure 3-2. Results of sensitivity analyses to evaluate the effects on critical load (CL) simulations caused by borrowing soils data from a nearest neighbor watershed for calibrating the 27 DDRP watersheds used in spatial extrapolations**

All CL simulations conducted for this analysis use an end year of 2100, but multiple critical threshold criteria values are applied. Simulations are conducted to protect against: a) low lake ANC, b) low soil base saturation (BS), c) low soil solution  $\text{Ca}^{2+}$  to  $\text{Al}^{n+}$  ratio, and d) low soil solution nutrient base cation ( $\text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+$ ; Bc) to  $\text{Al}^{n+}$  ratio.

a)



b)

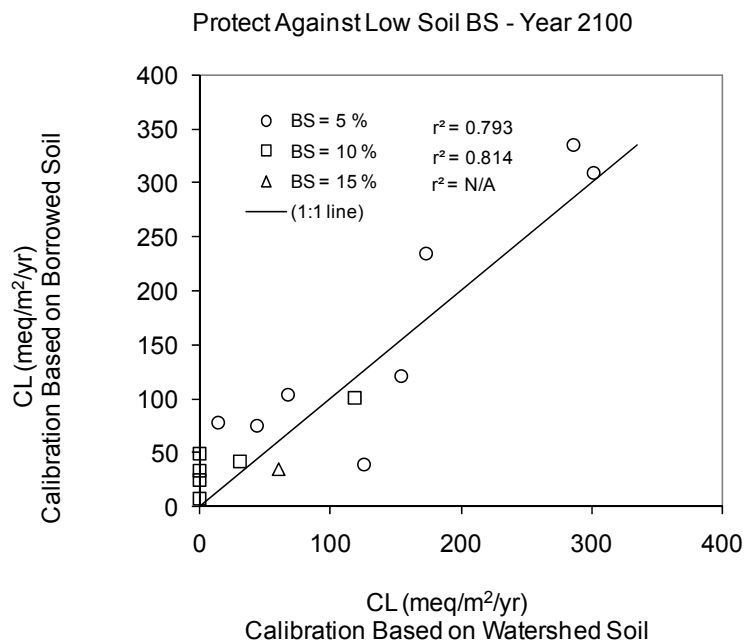
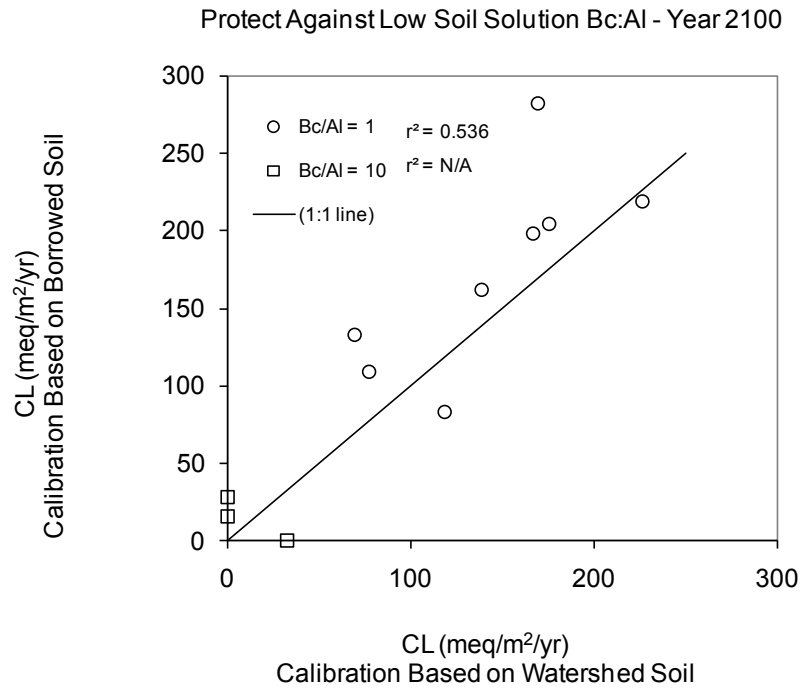
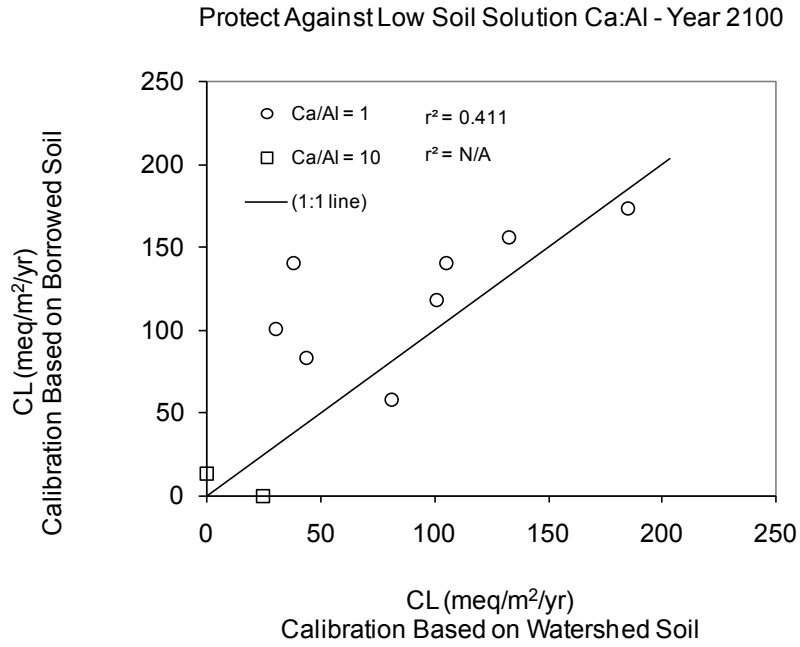


Figure 3-2 continued

c)



In general, little additional uncertainty was contributed by the soils borrowing procedure to the CL estimates for protecting lake water ANC. The relationship between CL based on measured soil data versus CL based on borrowed soil data was relatively strong ( $r^2=0.98-0.99$  for the various critical criteria values; Figure 3-2a). Most CL estimates to protect lake ANC that were based on borrowed soils were within 10-15 meq/m<sup>2</sup>/yr of the CL that was calculated using watershed-specific measured soils data in the model calibration. However, the soil borrowing procedure did introduce somewhat more error or uncertainty into the simulations of CL to protect soils against decreased BS, and especially to protect soil solution against low Ca:Al or Bc:Al ratio (Figures 3-2c and 3-2d).

It appears that the soil data borrowing procedure was an acceptable approach to use when modeling the CL to protect surface waters. In essence, the model calibration was successfully able to compensate for any error introduced in the model input soil data used to represent a given watershed. However, the substitution of borrowed soils data where measured data were lacking did appear to have an effect on CL estimates for protection of soil, and especially for protection of soil solution chemistry. We therefore only used the DDRP watersheds, for which soils data had to be borrowed in order to complete the model calibration, in extrapolating CL values to protect surface water quality. For extrapolating model results of soil and soil solution CL criteria in this study, only the 70 watersheds having measured soils data were used.

### **3.2 Sulfur Critical Loads**

A suite of CL calculations were performed using the MAGIC model (Table 3-1). Simulations were based on two acidic deposition drivers (S and N), three sensitive receptors (lake water, soil, and soil solution), one or more chemical indicators for each, two to three critical threshold levels for each indicator, and three to four endpoint years or periods. Selection of these various CL parameters had important influence on the resulting CL calculations. Decisions regarding the pollutant of interest, sensitive receptor, appropriate chemical indicator and associated threshold to protect against biological impact, and the timeframe of desired protection all influence the CL calculation. Thus, we present a large matrix of CL results. The calculated CL values must be interpreted in the context of these decisions.

MAGIC simulations of CL based on the endpoint year 2020 generally did not allow sufficient time for the model simulations to stabilize. Therefore, most analyses presented in this report focus on the endpoint years 2050 and, in particular, 2100.



**Table 3-1. Indicators, critical levels, and timeframes investigated for critical loads modeling**

Pollutants	Ecosystem Stress	Sensitive Receptor	Indicator	Critical Level	Timeframe
S, N	Acidification	Lake Water	Lake ANC	0, 20, 50 µeq/L	2020, 2050, 2100, steady-state
	Eutrophication	Lake Water	Lake NO <sub>3</sub> <sup>-</sup>	10, 20 µeq/L	2020, 2050, 2100
	Acidification	Soil	B-horizon BS	5, 10, 15%	2020, 2050, 2100
	Acidification	Soil Solution	B-horizon Ca:Al	1, 10	2020, 2050, 2100
	Acidification	Soil Solution	B-horizon Bc:Al	1, 10	2020, 2050, 2100

### 3.2.1 Sulfur Critical Load to Protect Lake ANC

The most commonly used combination of sensitive receptor and chemical indicator for acidification CL calculation is surface water ANC. For this analysis with MAGIC, ANC is defined by the charge balance as the difference between the sum of the concentrations of the base cations and the strong acid anions (Equation 3-1):

$$\text{ANC} = (\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^{+} + \text{K}^{+} + \text{NH}_4^{+}) - (\text{SO}_4^{2-} + \text{NO}_3^{-} + \text{Cl}^{-}) \quad (3-1)$$

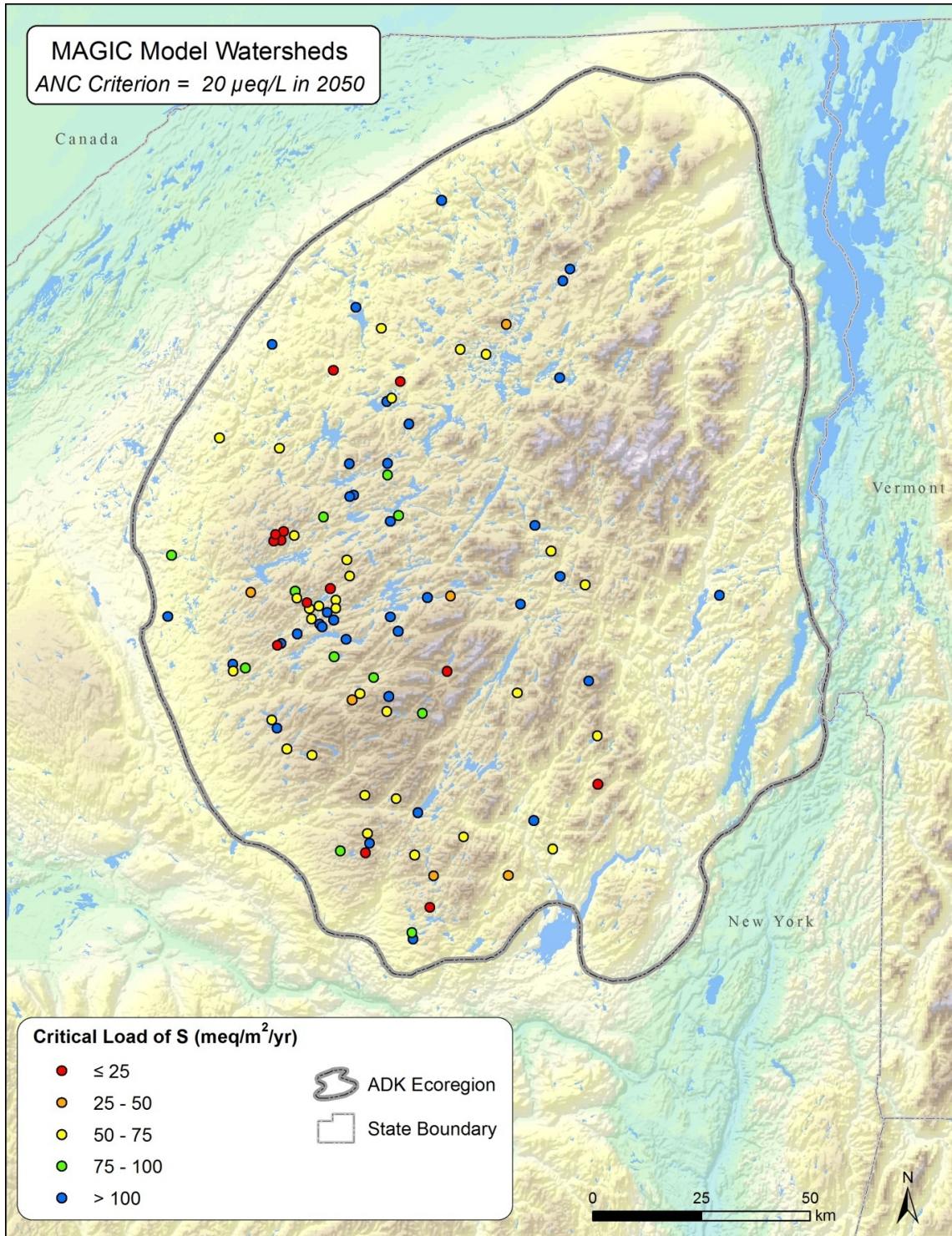
ANC calculated in this way is often termed CALK, or calculated ANC, as opposed to laboratory-titrated ANC.

Results of MAGIC model CL simulations to protect lake ANC are depicted in Maps 3-1 through 3-4, which show two critical threshold criteria (20 and 50 µeq/L) and two endpoint years (2050 and 2100). All 97 of the lake watersheds modeled with MAGIC are included on the maps. Modeled lakes having low to very low S CL values (less than about 50 meq/m<sup>2</sup>/yr) are scattered throughout much of the Adirondack Ecoregion.

The simulated critical S load to protect lake ANC varied with the lake ANC during the Reference Year (2002). Lower CL values (indicating greater acid sensitivity) were generally found for Adirondack lakes that had low ANC in 2002 (Figure 3-3). Furthermore, CL values tended to be lower if the objective was to protect lake water to a higher level (i.e., ANC = 50 µeq/L) as compared with a lower level of protection, such as protecting to ANC = 0 µeq/L.

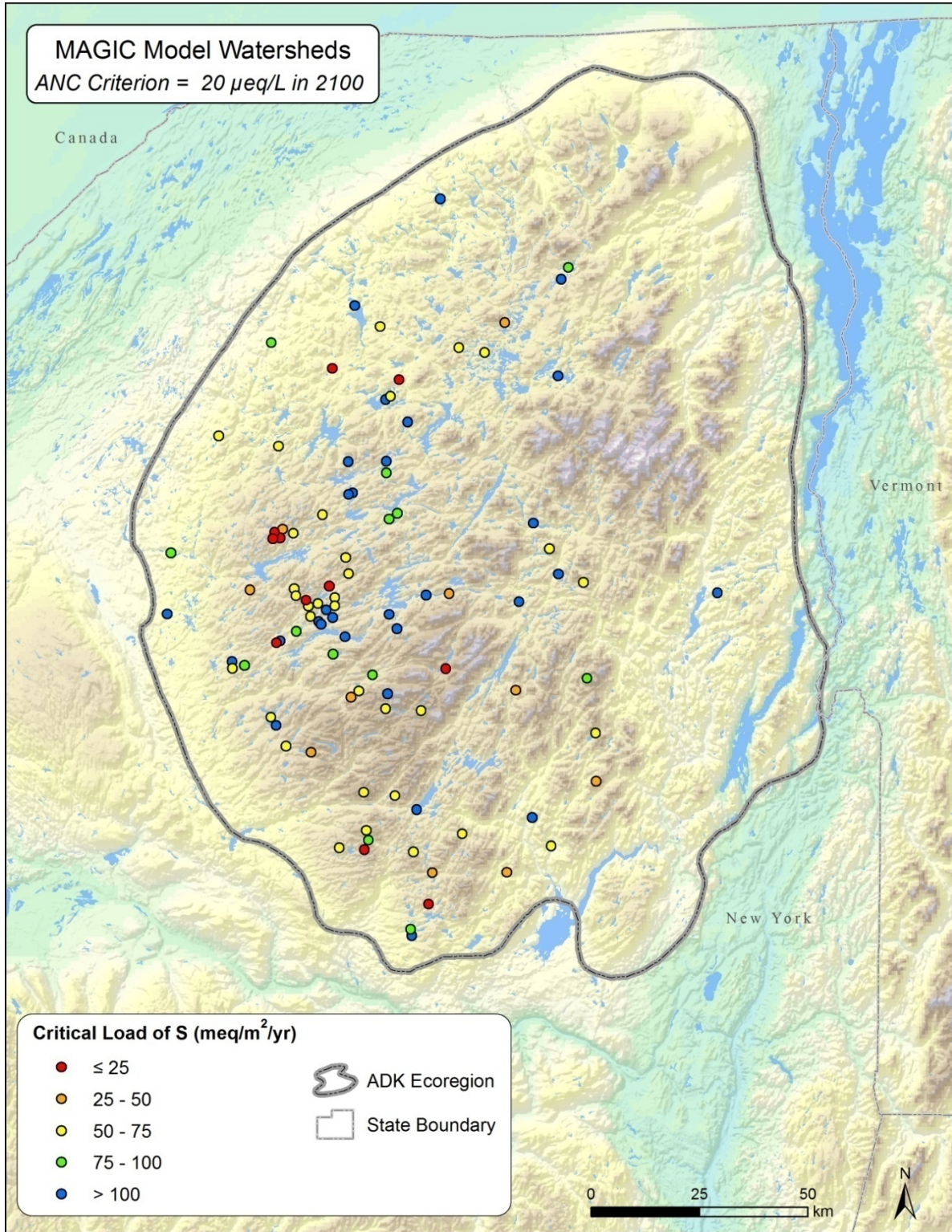
**Map 3-1. Simulated critical load of sulfur deposition to protect lake ANC to 20  $\mu\text{eq/L}$  in the year 2050**

Based on application of the MAGIC model to 97 lake watersheds



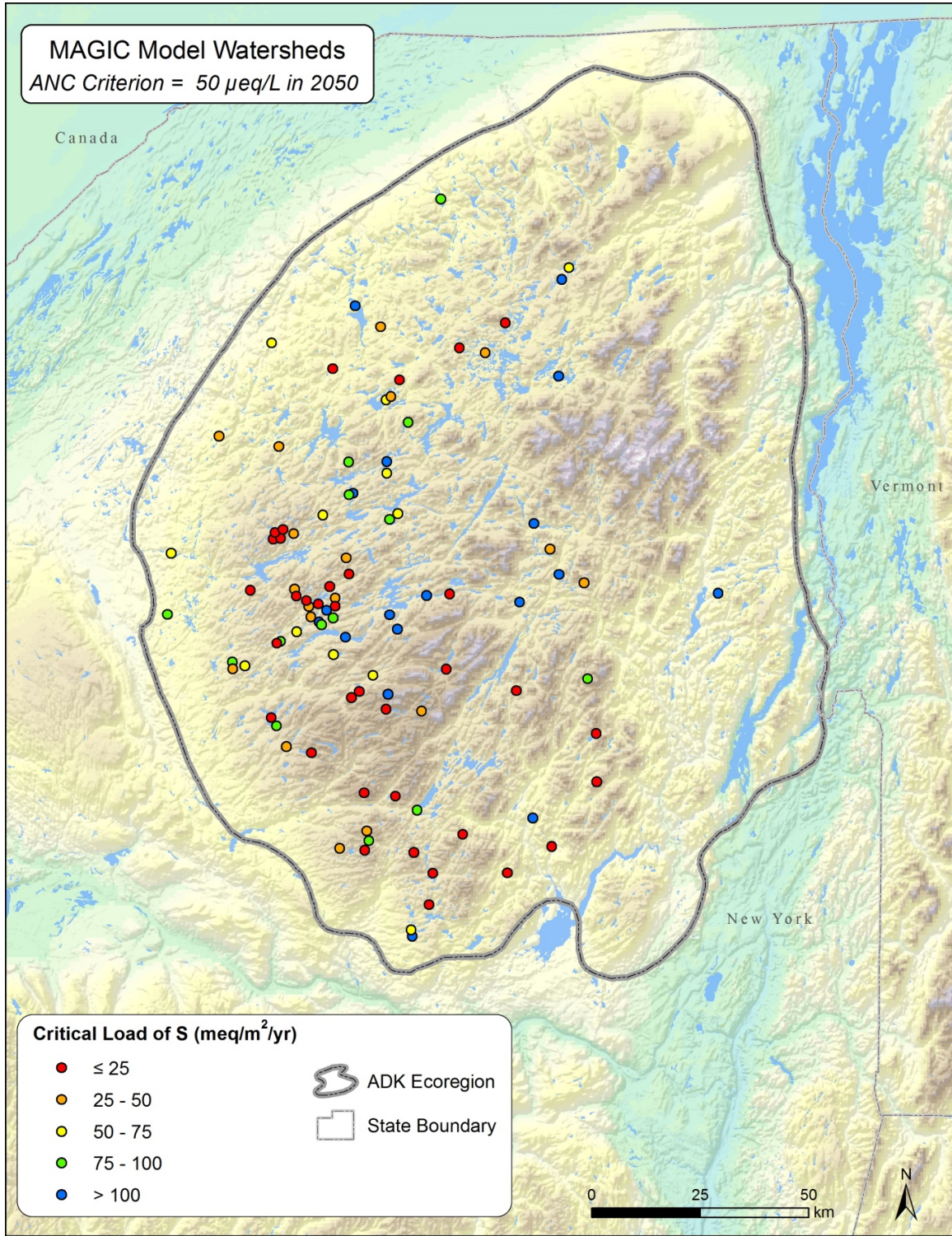
**Map 3-2. Map showing simulated critical load of sulfur deposition to protect lake ANC to 20  $\mu\text{eq/L}$  in the year 2100**

Based on application of the MAGIC model to 97 lake watersheds.



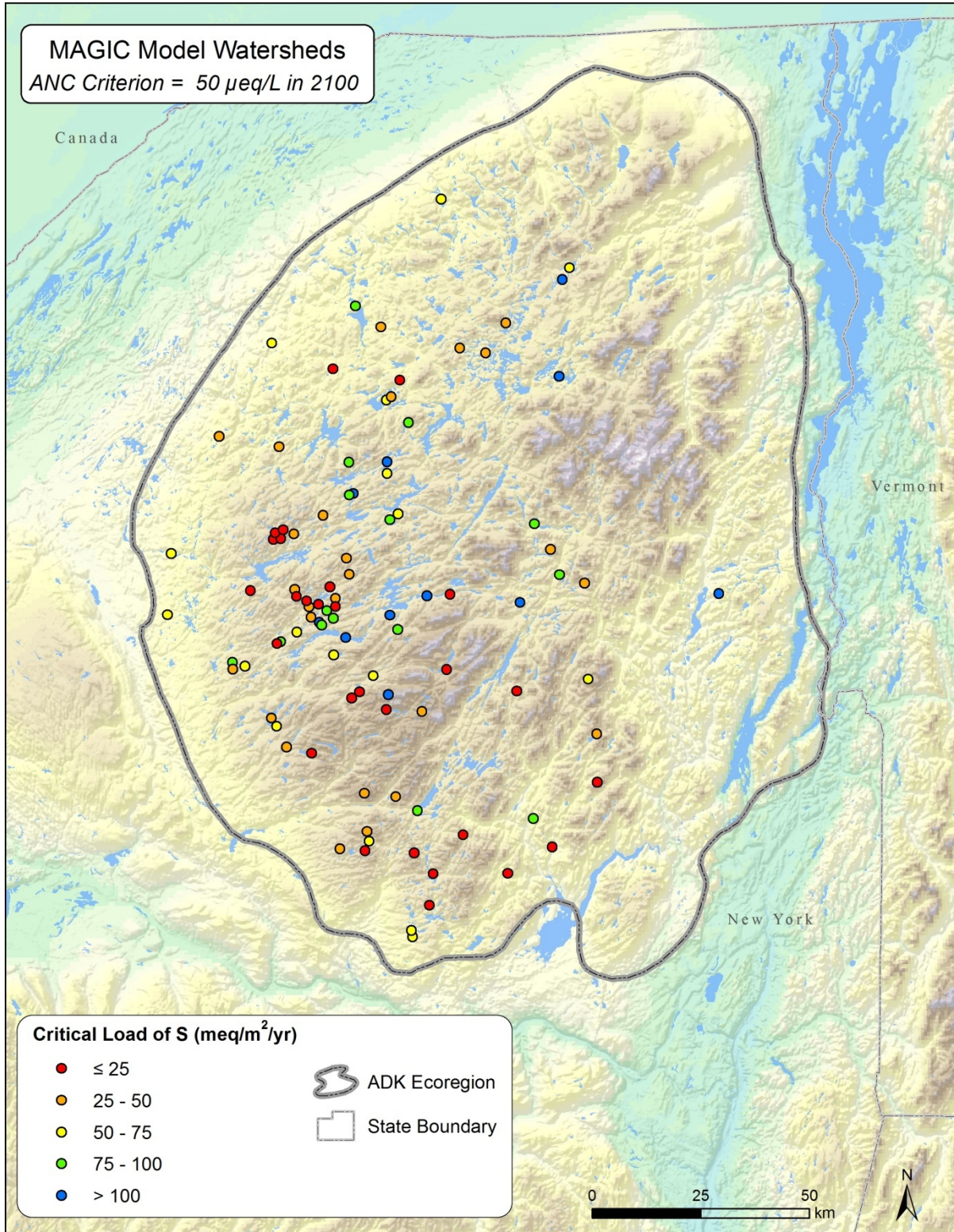
**Map 3-3. Map showing simulated critical load of sulfur deposition to protect lake ANC to 50  $\mu\text{eq/L}$  in the year 2050**

Based on application of the MAGIC model to 97 lake watersheds.



**Map 3-4. Map showing simulated critical load of sulfur deposition to protect lake ANC to 50  $\mu\text{eq/L}$  in the year 2100**

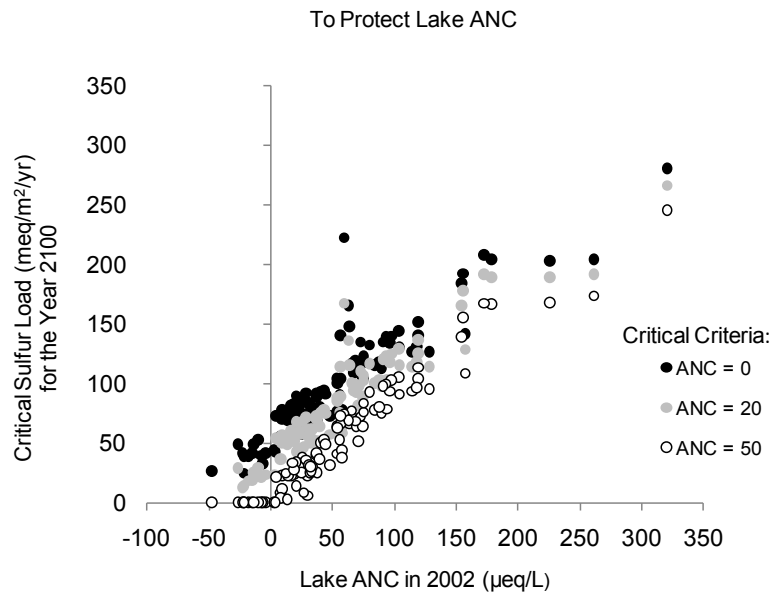
Based on application of the MAGIC model to 97 lake watersheds.



**Figure 3-3. Critical load of sulfur deposition for the year 2100 to protect lake ANC from going below various critical criteria thresholds versus lake ANC in the reference year (2002)**

The critical criteria thresholds examined include ANC 0, 20, and 50  $\mu\text{eq/L}$ ; all sites modeled using MAGIC are depicted.

*Source: Modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. Water Resour. Res. 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.*



### 3.2.2 Sulfur Critical Load to Protect Soil BS

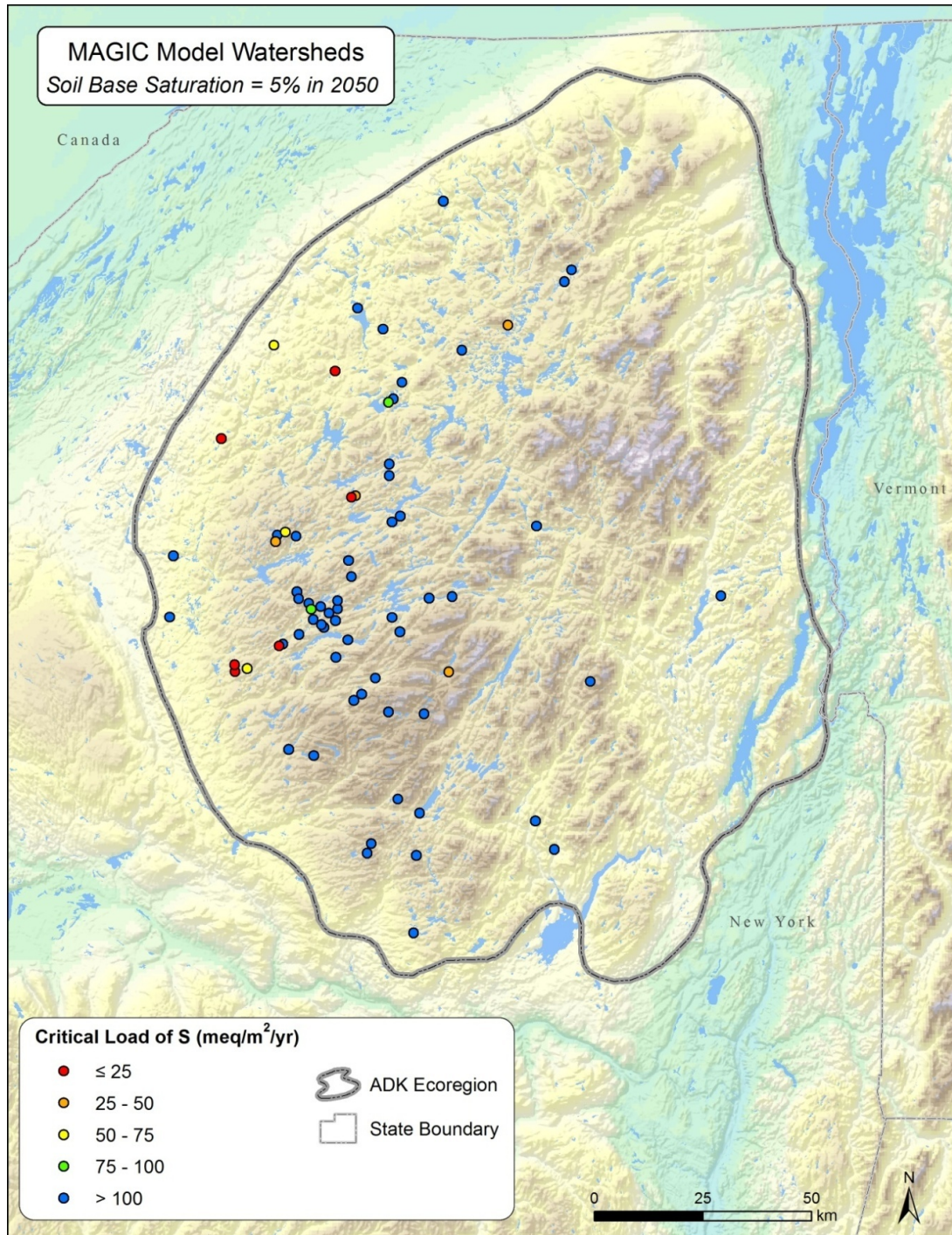
MAGIC model S CL simulations to protect soil BS are shown on Maps 3-5 through 3-9. Watersheds having low (i.e., less than about 50  $\text{meq/m}^2/\text{yr}$ ) simulated S CL to protect soil BS to 5% were generally located in the western portion of the Adirondack region (Map 3-5 and Map 3-6). To protect watershed soil BS to 10%, however, simulated S CL values were low throughout all of the Adirondack region except the southeastern corner (Map 3-7 and Map 3-8).

Spatial patterns in simulated S CL levels to protect soil BS to two critical threshold values (5% and 10%) are depicted in Map 3-9. Only the smaller 90% of the watersheds with respect to watershed area are shown so as to avoid representation of a few very large watersheds based on only a small number of sampled soil pits. The relatively few watersheds simulated to have CL less than 75  $\text{meq/m}^2/\text{yr}$  to protect soil BS to 5% in the year 2100 were all small in area. There was a wide distribution in watershed area for those watersheds simulated to have higher S CL values. Selection of the critical threshold value for BS as equal to 5% or 10% had a large influence on the spatial patterns in CL for protecting soil chemistry.

**Map 3-5. Simulated sulfur critical load to protect soil base saturation to 5% in the year 2050, based on application of the MAGIC model to 70 modeled lake watersheds**

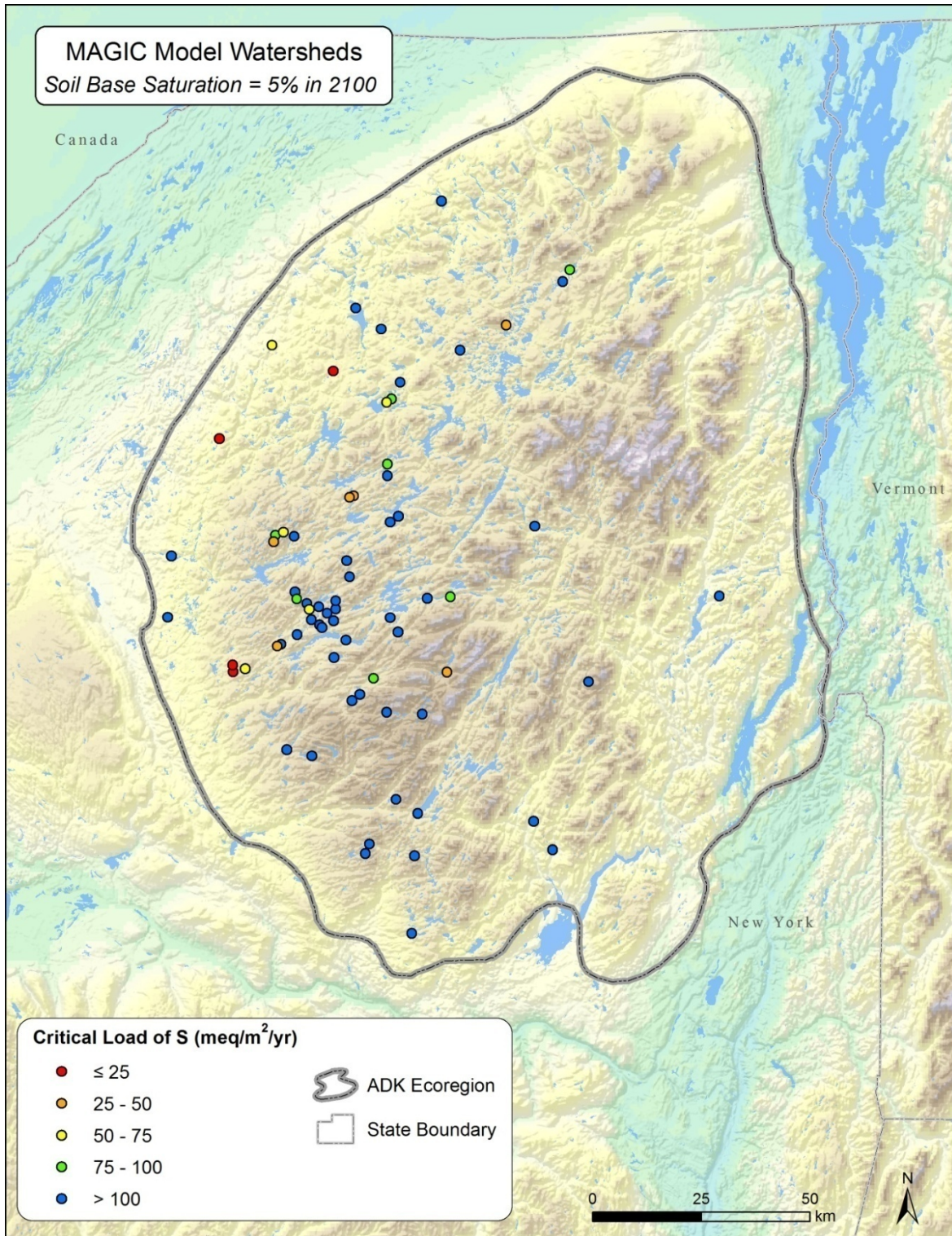
Only watersheds within which soil samples were collected and analyzed by Sullivan et al. (2006a) are included.

Source: Modified from Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. *J. Environ. Stud. Sci.* 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.



**Map 3-6. Simulated sulfur critical load to protect soil base saturation to 5% in the year 2100**

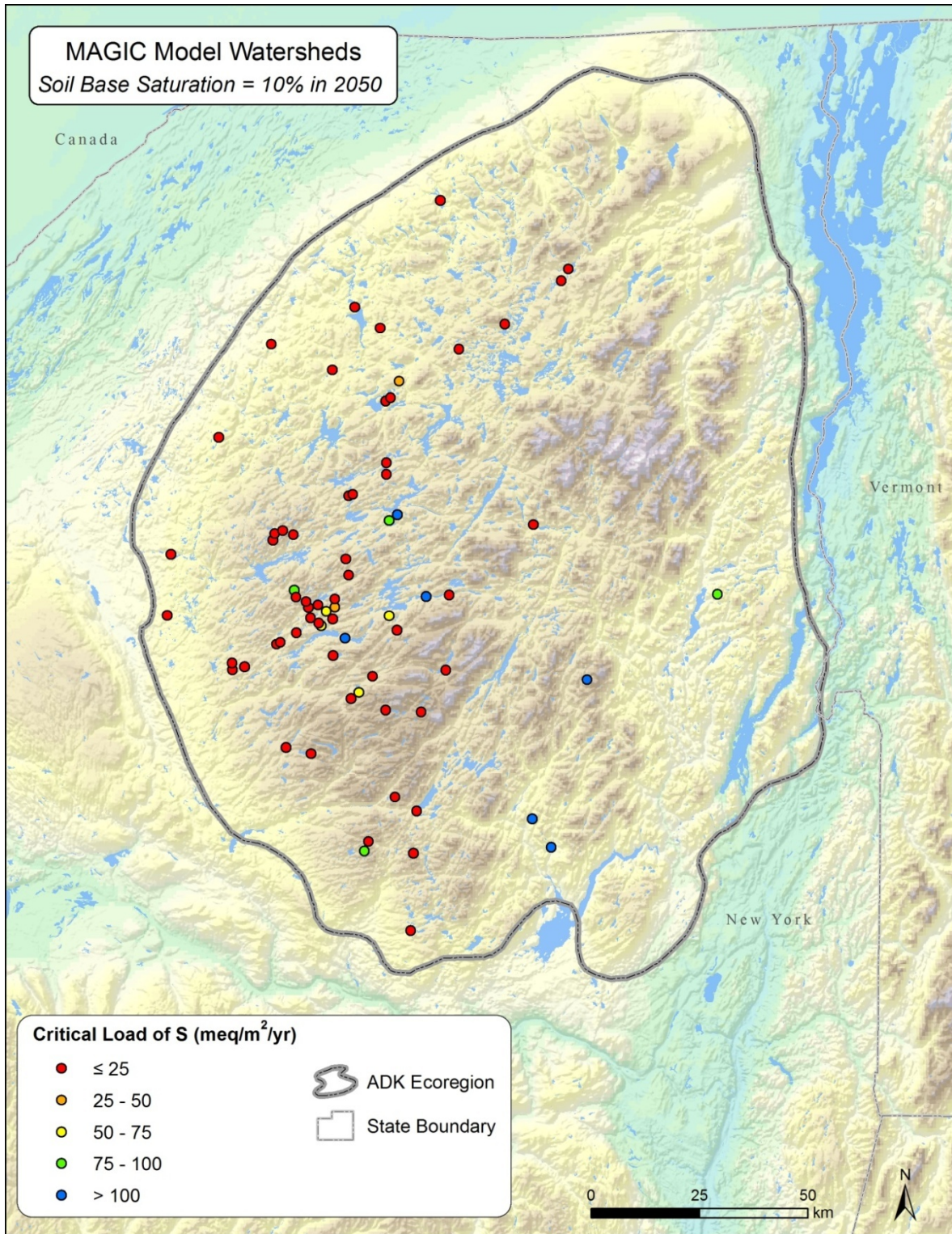
Based on application of the MAGIC model to 70 modeled lake watersheds. Only watersheds within which soil samples were collected and analyzed by Sullivan et al. (2006a) are included.





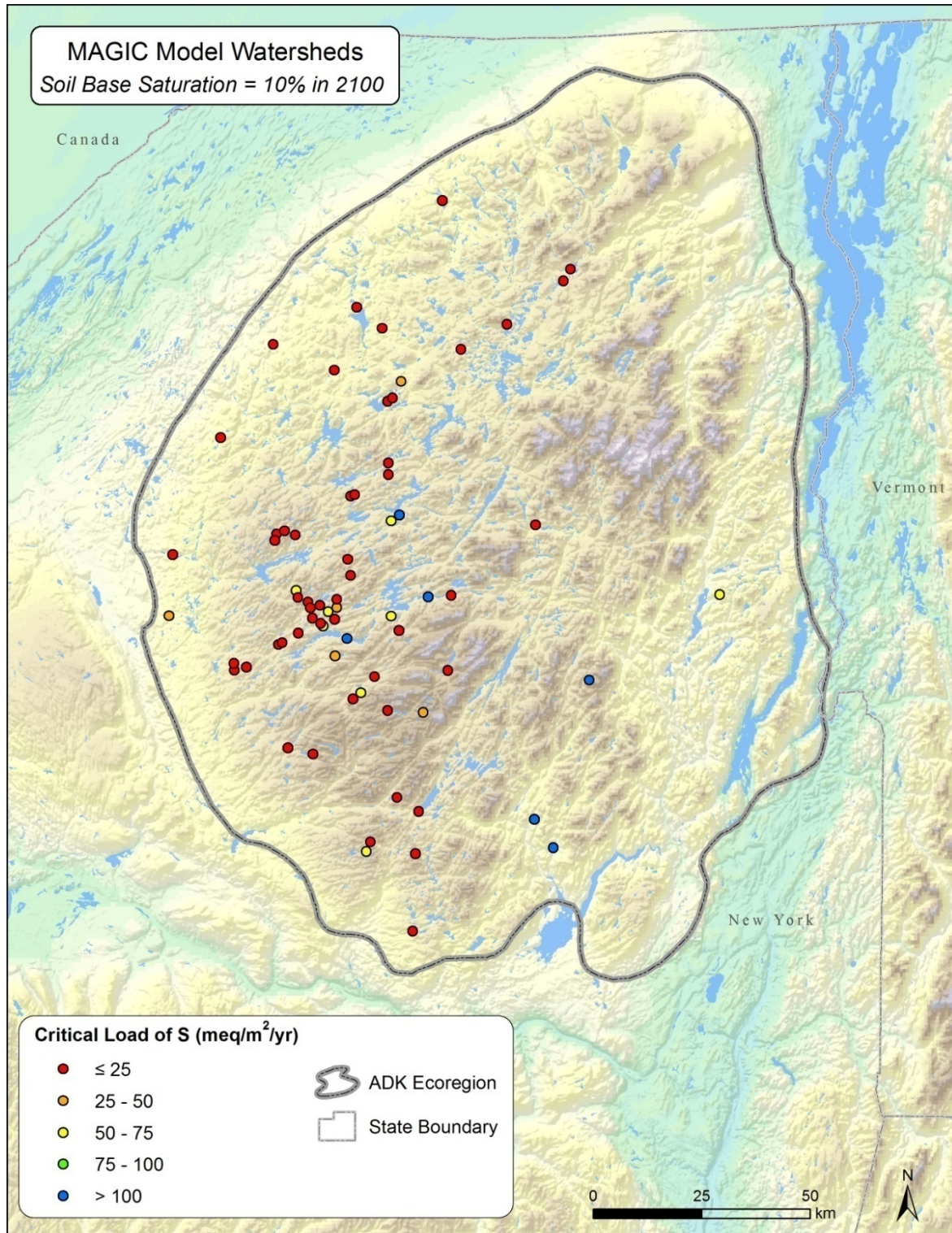
**Map 3-7. Simulated sulfur critical load to protect soil base saturation to 10% in the year 2050**

Based on application of the MAGIC model to 70 modeled lake watersheds. Only watersheds within which soil samples were collected and analyzed by Sullivan et al. (2006a) are included.



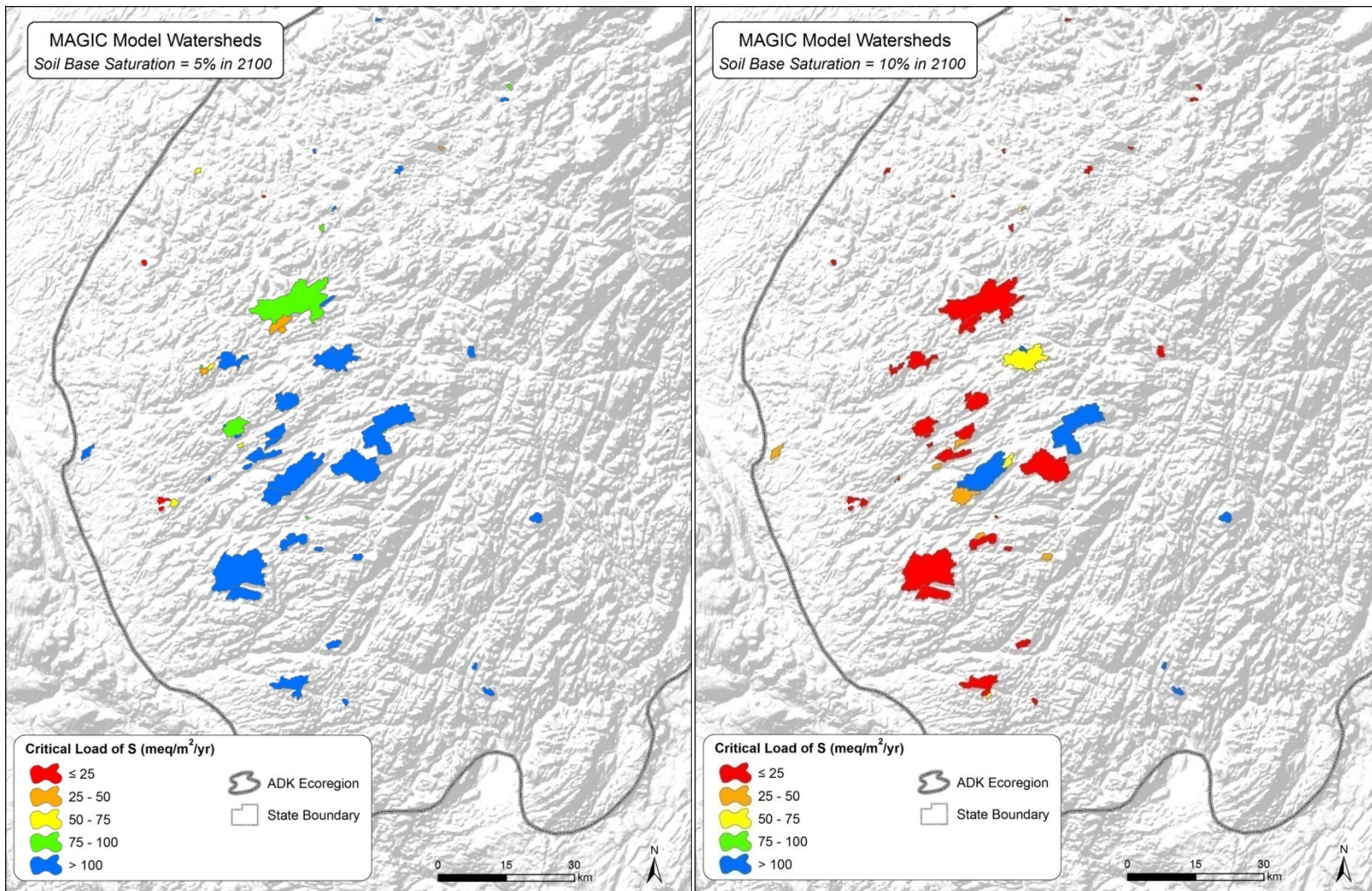
**Map 3-8. Simulated sulfur critical load to protect soil base saturation to 10% in the year 2100**

Based on application of the MAGIC model to 70 modeled lake watersheds. Only watersheds within which soil samples were collected and analyzed by Sullivan et al. (2006a) are included.



### Map 3-9. MAGIC model S CLs to protect soil BS to two critical threshold criteria in the year 2100

Watersheds that contribute drainage water to each of the modeled lakes are shown for BS protection to 5% (left panel) and 10% (right panel). MAGIC modeled watersheds having watershed-specific soil data are shown, but the 10% of the 70 modeled watersheds having the largest watershed area were deleted from the map presentations.



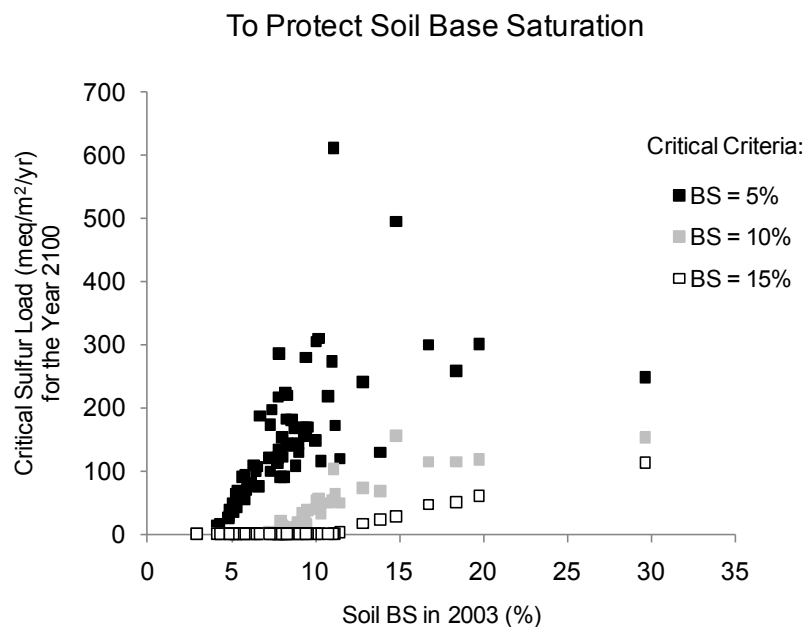
The CL to protect soil BS varied with soil BS measured in 2003 by Sullivan et al. (2006b). Critical loads to protect soil BS were generally lower in watersheds that had low measured values of BS in 2003 (Figure 3-4). Critical loads were lower to protect soil to a greater level (i.e., to protect to BS = 15%) as compared with CL to protect soil to BS = 10% and to BS = 5%. In fact, many of the modeled lake watersheds showed CL to be approximately equal to 0 for protecting BS to 10% and especially to 15%. In other words, the simulation results suggested that if the target is BS = 10% or 15%, in this case by the year 2100, then no amount of S deposition can be tolerated by that watershed. In contrast, most of the modeled watersheds showed positive values of CL to protect to BS = 5%. This result implies that some level of S deposition could be tolerated by those watersheds and still allow BS to be maintained at a level of 5% or more in the year 2100.

There was a very weak relationship between the simulated CL to protect soil BS and the ANC of the lake at the base of a given watershed. In general, watersheds having low CL to protect soil BS also tended to exhibit low lake ANC, but there was considerable scatter in that relationship, as shown in Figure 3-5.

**Figure 3-4. Critical load of sulfur deposition for the year 2100 to protect soil BS from going below various critical criteria thresholds versus average watershed soil BS in 2003**

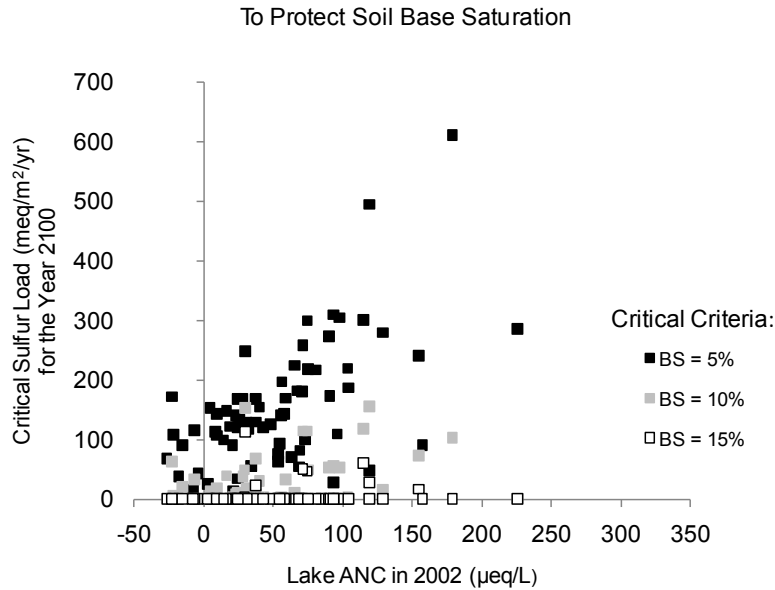
The critical criteria thresholds examined include BS 5, 10, and 15%; all sites (n=70) that were modeled using MAGIC based on watershed-specific soils data are depicted.

*Source: Modified from Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. J. Environ. Stud. Sci. 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.*



**Figure 3-5. Critical load of sulfur deposition for the year 2100 to protect soil BS from going below critical criteria versus lake ANC in the reference year (2002)**

The critical criteria thresholds examined include BS 5, 10, and 15%; all sites modeled using MAGIC are depicted.



### 3.2.3 Sulfur Critical Load to Protect Soil Solution Base Cation to Aluminum Ratios

For protection of terrestrial resources, a molar ratio of Ca to Al or Bc to Al in soil solution is commonly used as a chemical indicator, where the nutrient base cations Bc, include (Equation 3-2):

$$\text{Bc} = \text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+ \quad (3-2)$$

The critical threshold criterion value is commonly set to 1.0 (Cronan and Grigal 1995), although a threshold of 10 is also used (c.f., Sverdrup and Warfvinge 1993). Perhaps an intermediate threshold value might be more indicative of biological harm. Data with which to evaluate the efficacy of such a tipping point are not available for Adirondack watersheds.

In general, the threshold value of 10 for these two chemical indicators did not prove to be a useful criterion for discriminating among CL values for Adirondack watersheds. Nearly all of the modeled lake watersheds showed a simulated CL to protect either Ca:Al or Bc:Al to 10 that was  $\leq 25$  meq/m<sup>2</sup>/yr (Figure 3-6), which is substantially lower than ambient levels of S deposition in these watersheds.

The major reason for these low simulated CL values to protect base cation to Al ratios to a level of 10 was that most of the modeled lakes were simulated to have had ratios well below 10 even in pre-industrial times, prior to the onset of acidic deposition (Figure 3-7). In contrast, relatively few modeled watersheds were simulated to have had lake ANC < 50 µeq/L, soil BS < 5%, soil solution Ca:Al < 1, or soil solution Bc:Al < 1 during the pre-industrial period (Figure 3-7). For some lakes or watersheds, these latter target criteria values may be unreasonable because the criteria were apparently (based on the model simulations) not attained in the absence of acidic deposition. For most lakes and watersheds, however, these target criteria values do appear reasonable (potentially attainable). Soil BS targets of 10% and 15%, like the soil solution ratio targets equal to 10, appear to be unreasonable for many watersheds, based on the MAGIC simulations.

### **3.2.3.1 Selection of Sensitive Indicators for Sulfur Critical Load**

To protect sensitive Adirondack resources from adverse acidification effects that can be caused by atmospheric S deposition, any one or more of several sensitive chemical indicators (with associated threshold criteria) could be selected. The choice depends in large part on whether one is attempting to protect aquatic or terrestrial resources. If the intention is to protect aquatic resources, then ANC is the most commonly applied chemical indicator. Surface water NO<sub>3</sub><sup>-</sup> concentration is an alternative indicator that provides information regarding potential watershed N saturation and/or potential aquatic nutrient enrichment (eutrophication) effects from N deposition. Nitrogen CL approaches are presented in Section 3.3 of this report. If the intention is to protect terrestrial resources, such as the soil base cation supply or the health and growth of terrestrial vegetation, then either soil BS or one of the soil solution base cation to Al ratios could be used as a chemical indicator.

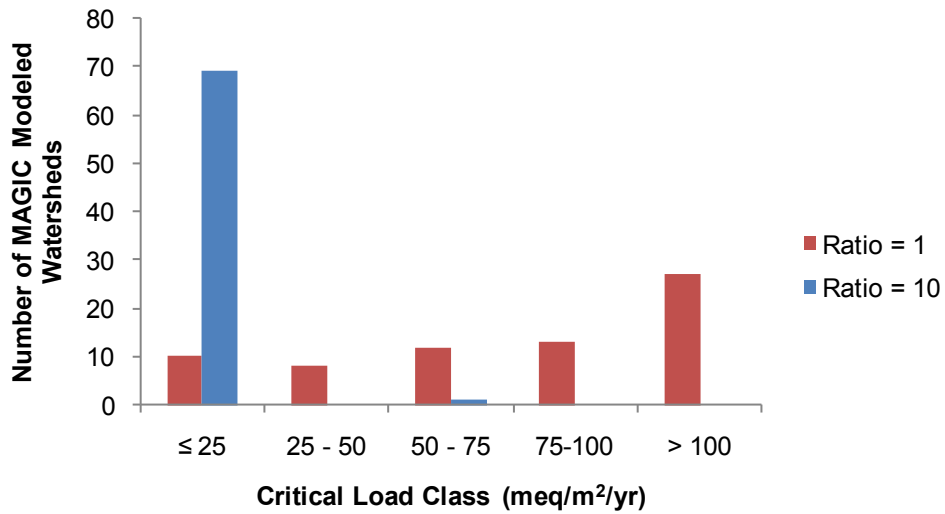
Lake chemistry, including ANC and NO<sub>3</sub><sup>-</sup> concentration, is measured in the field and these data are used in model calibration. Similarly, watershed soil BS is quantified based on soil parameters that are measured in the field. Soil % BS is averaged across a modeled watershed and is used in MAGIC model calibration. Thus, the MAGIC model CL applications are constrained by the measured lake and soil chemistry data. This provides an opportunity to compare simulated with observed lake and soil chemistry to examine model performance (Figure 3-1), and constrains the model projections of CL so that they fit the observations, at least during the calibration period. No such constraints are possible for the soil solution chemical indicators because soil solution chemistry is not commonly measured in the field as part of typical regional soil or water surveys. As a consequence, there is additional uncertainty introduced into the CL calculations for protecting soil solution base cation to Al ratios, as compared with lake chemistry or soil chemistry indicators. The model calibration is not constrained to measured soil solution data because such data are largely unavailable.

**Figure 3-6. Histograms showing the number of MAGIC modeled watersheds (n=70) simulated to be in various CL classes to protect the ratio of base cations to aluminum in soil solution to two thresholds of protection (1 and 10)**

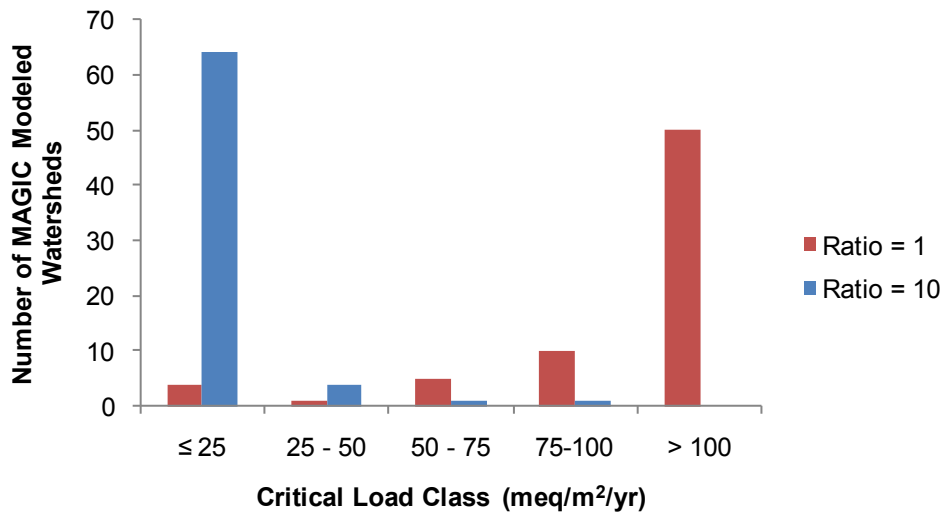
Panel A shows results for the Ca:Al ratio; Panel B shows results for the Bc:Al ratio.

Source: Modified from Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. *J. Environ. Stud. Sci.* 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.

**a) CL of S for Ca/Al ratio criterion for the year 2100**



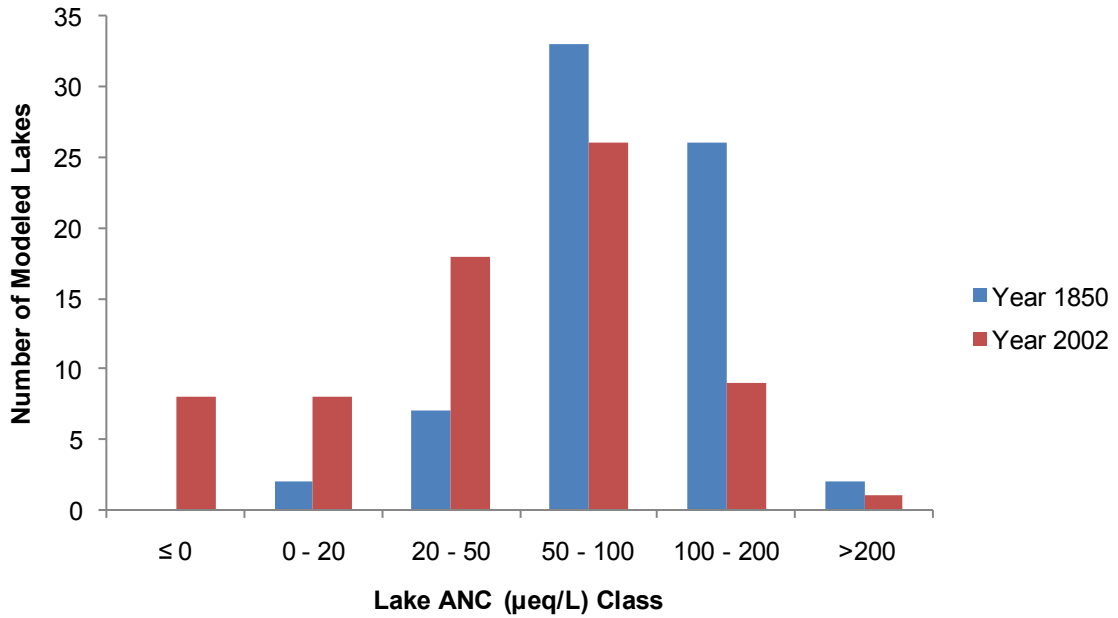
**b) CL of S for Bc/Al ratio criterion for the year 2100**



**Figure 3-7. Histograms showing MAGIC simulated lake, soil, and soil solution chemistry for the 70 modeled lake watersheds having watershed-specific soil chemistry data at two points in time: the pre-industrial period (1850) and the current period (2002)**

Simulation results are shown for a) lake ANC, b) soil BS, c) soil solution Ca:Al, and d) soil solution Bc:Al.

a)



b)

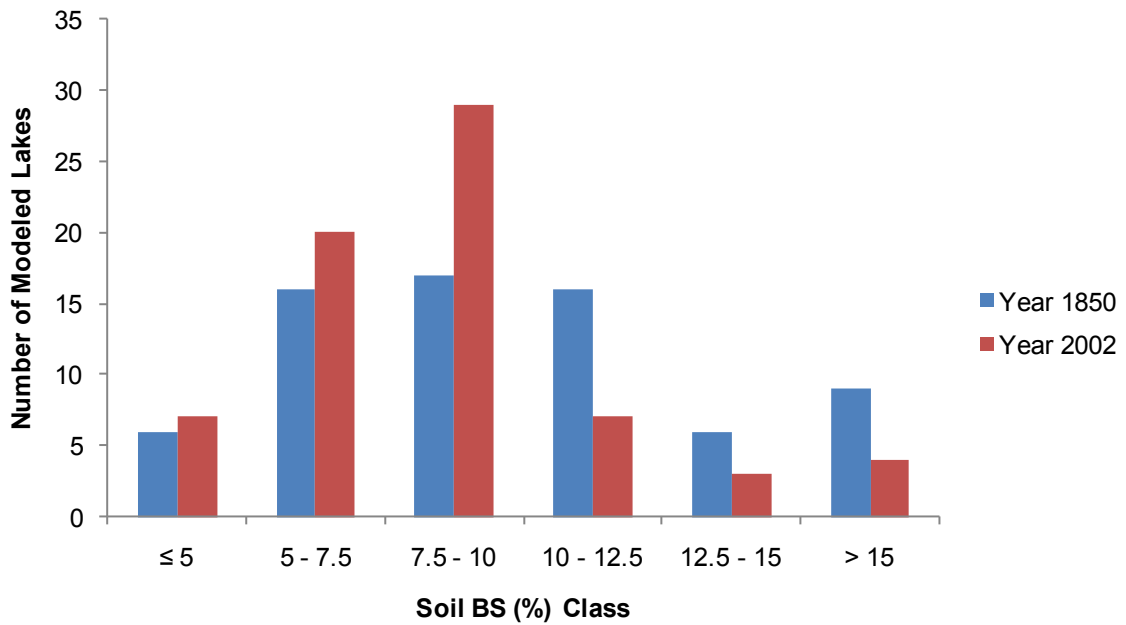
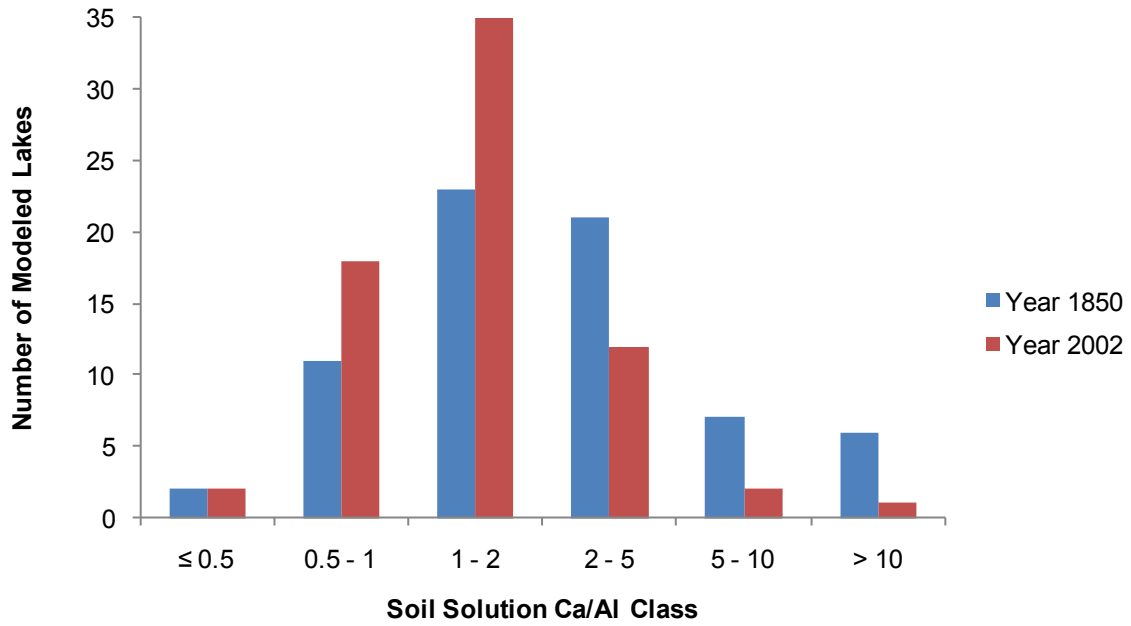


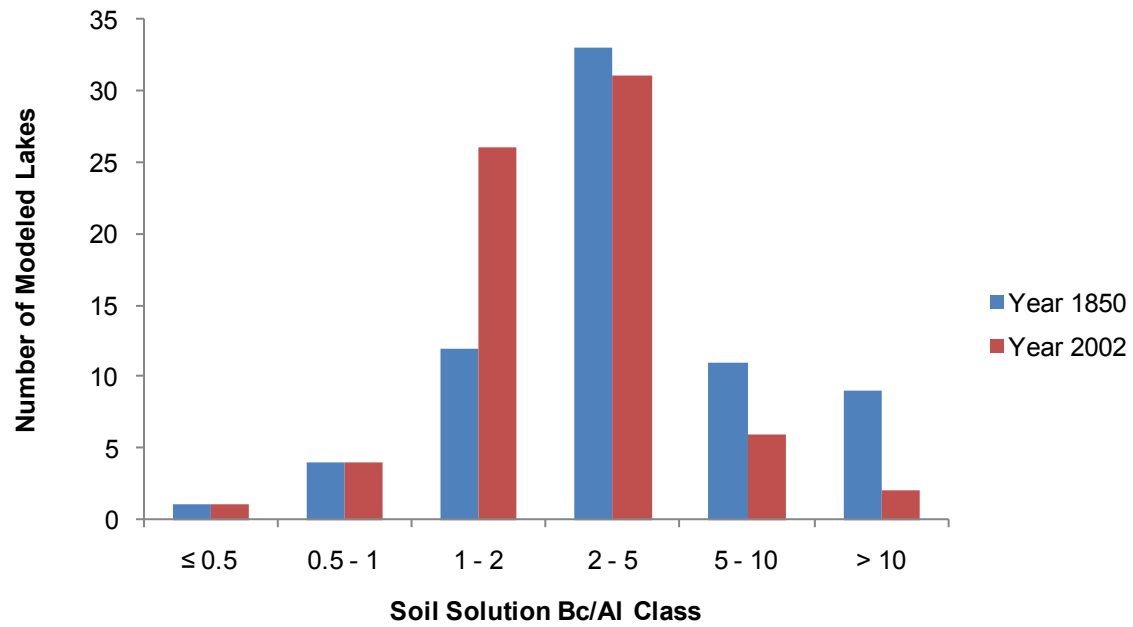


Figure 3-7 continued

c)



d)

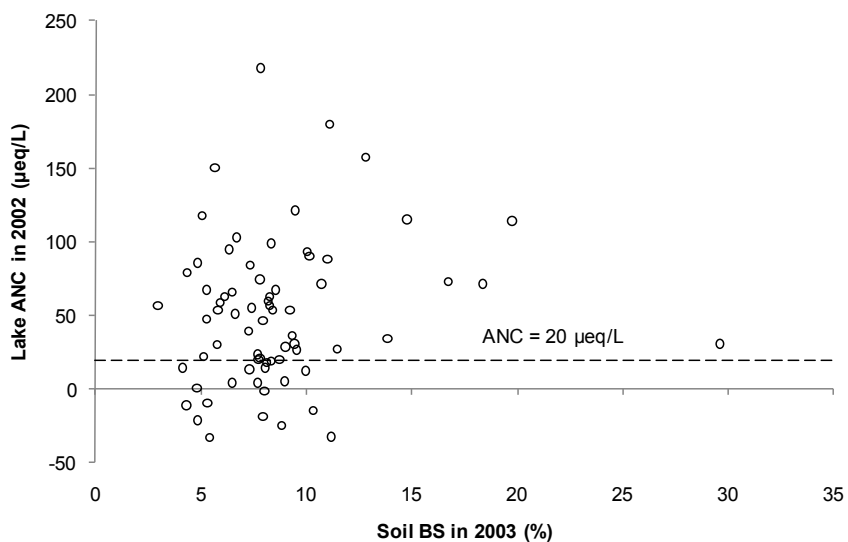


There are some connections to be expected between lake, soil, and soil solution chemistry. For example, lakes that have low ANC might be expected to occur within watersheds that have low soil BS. Although no clear relationship exists between watershed soil BS and lake ANC for the Adirondack study watersheds (Figure 3-8), lake ANC below 20  $\mu\text{eq/L}$  (an important aquatic acidification CL target criterion) only occurs in Adirondack watersheds that have soil BS less than about 12%. Sullivan et al. (2008) obtained similar results for stream watersheds in Virginia. A BS value of 12% also represents a tipping point for sugar maple tree regeneration in the Adirondack Mountains (Sullivan et al. 2013).

**Figure 3-8. Relationship between simulated lake ANC in 2002 and watershed averaged soil BS measured in the field in 2003 for the 70 MAGIC modeling lakes investigated by Sullivan et al. (2006a)**

A reference line is added at  $\text{ANC} = 20 \mu\text{eq/L}$ , illustrating that lake ANC below 20  $\mu\text{eq/L}$  is only found in Adirondack watersheds that have average BS below about 12%.

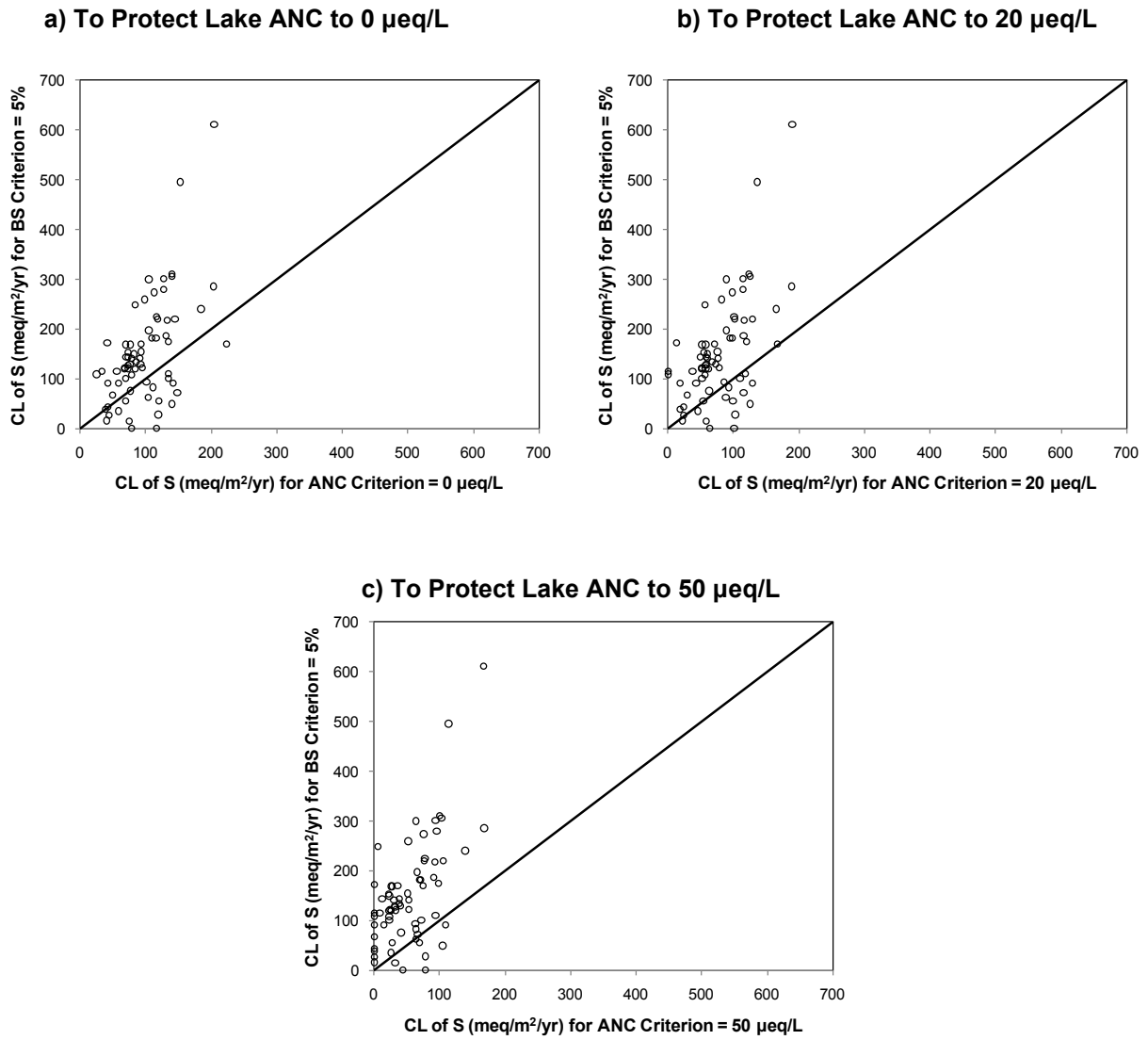
*Source: Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. J. Environ. Stud. Sci. 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.*



The S CLs simulated for the modeled watersheds calibrated using watershed-specific soil data varied with the selected chemical indicator and critical threshold criterion. In general, however, the S CL to protect soil BS to 5% in the year 2100 was similar to the S CL to protect lake ANC to 0 (Figure 3-9). Protecting lake ANC to 20  $\mu\text{eq/L}$ , and especially to 50  $\mu\text{eq/L}$ , required a lower S deposition level than protecting soil BS to 5% (Figure 3-9).

**Figure 3-9. Comparison between MAGIC model simulated CL of S deposition to protect soil BS to 5% and to protect lake ANC to a) 0  $\mu\text{eq/L}$ , b) 20  $\mu\text{eq/L}$ , and c) 50  $\mu\text{eq/L}$  in the year 2100**

A 1:1 line is provided on each plot for reference.



### 3.3 Nitrogen Critical Loads

#### 3.3.1 Nitrogen Critical Load to Protect Acid-Base Chemistry

In general, N CLs were substantially higher than S CLs for a given watershed and set of CL criteria (Figure 3-10). Adirondack watersheds modeled for this study currently retain most of the N that is atmospherically deposited (Figure 3-11). The MAGIC model CL simulations assume that the percent of atmospherically deposited N that is retained in the watershed will remain constant into the future under moderate changes in N deposition loading. Thus, the CL simulations assume that most of the future N loading will continue to be retained, causing limited influence of deposited N on the acid-base chemistry of the lake or soil. The degree to which this assumption will hold long-term will depend on the nature of the various N retention mechanisms, including assimilation and denitrification. It is possible that N retention will decrease in the future in some watersheds under continued N loading. Such a change would result in a lower N CL for lakes in the affected watersheds.

**Figure 3-10. Comparison between N CLs and S CLs for the year 2100 to protect lake ANC to 20  $\mu\text{eq/L}$  and soil BS to 5%**

MAGIC-simulated CL values are plotted on a log scale because N CLs tended to be very high. CL values equal to 0 were set to 1.0 for ease of graphing.

##### a) To Protect Lake ANC to 20 $\mu\text{eq/L}$

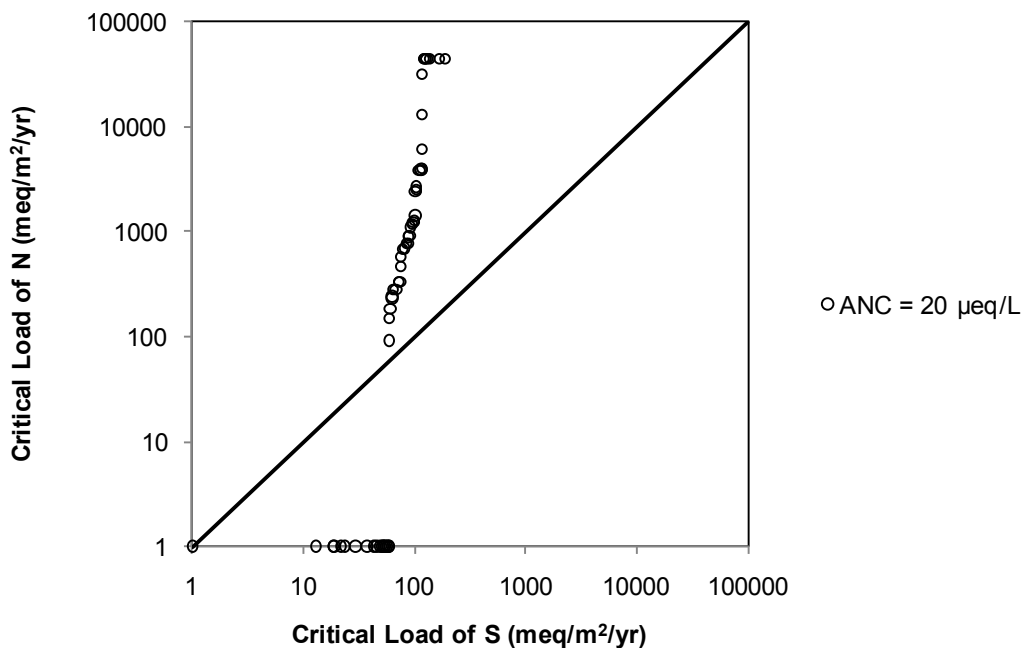


Figure 3-10 continued

b) To Protect Soil BS to 5%

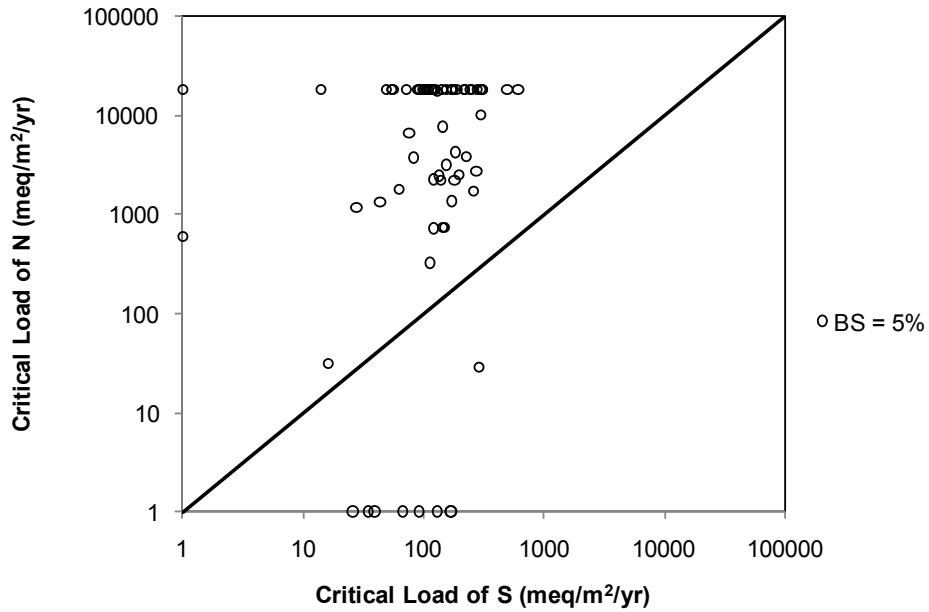
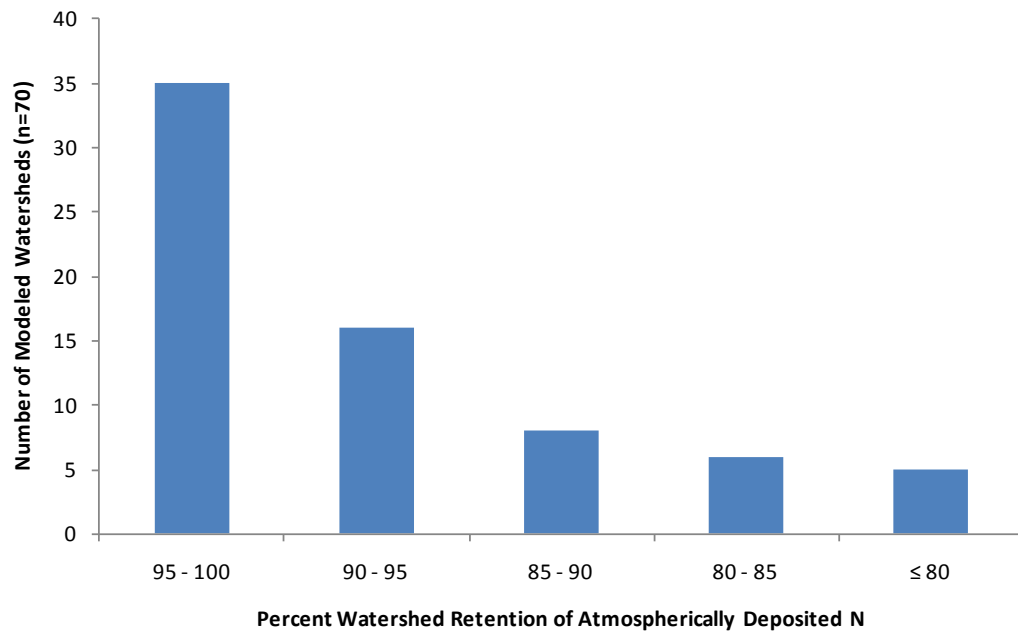


Figure 3-11. Histogram showing the modeled ambient percent watershed N retention for the 70 watersheds having watershed-specific soil chemistry data for MAGIC model calibration

Source: Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.



Note that there are some lakes that have simulated CL of N deposition equal to zero (Figure 3-10). There are multiple reasons for this result. First, the N CL simulations were conducted assuming future S deposition as expected in the CAIR scenario. For some lakes, the CL is exceeded based only on the S deposition level represented by CAIR. For such lakes, the N CL will be zero, because the ANC target criterion cannot be reached with or without added N deposition.

There are also some lakes that appear to be currently leaching greater quantities of  $\text{NH}_4^+$  than  $\text{NO}_3^-$  from watershed soil to lake water. As N deposition to such watersheds increases in the CL scenario, there is a net increase in lake ANC because  $\text{NH}_4^+$  contributes to ANC, defined as the difference between the sums of the base cations (which include  $\text{NH}_4^+$ ) and the mineral acid anions (which include  $\text{NO}_3^-$ ), for lakes that are exporting relatively large quantities of  $\text{NH}_4^+$ . To decrease lake ANC to a target criterion value (i.e., 20  $\mu\text{eq/L}$ ), the N deposition to such a lake watershed must decrease rather than increase. If N deposition cannot be decreased enough to achieve the target ANC, the CL for N deposition will be zero.

### **3.3.2 Nitrogen Critical Load to Protect Against Nutrient Enrichment**

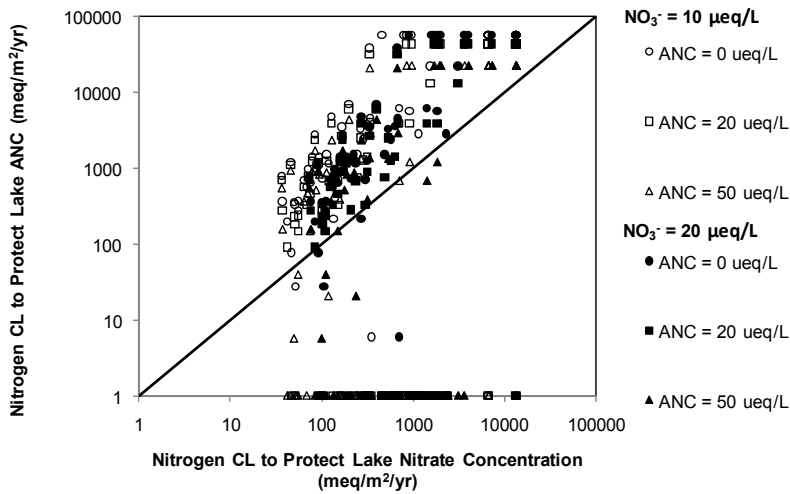
Nitrogen CLs were also calculated to protect lake  $\text{NO}_3^-$  concentration from exceeding two critical criteria values: 10  $\mu\text{eq/L}$  and 20  $\mu\text{eq/L}$ . One or both of these critical target criteria can be used as the basis for evaluating watershed N saturation and/or potential nutrient enrichment (eutrophication) of lake water. These criteria were used for illustrative purposes only. We have no basis for specifying ecological tipping points of lake  $\text{NO}_3^-$  concentration that would be indicative of impending N saturation or eutrophication. Also, note that the CL analyses reported here were limited to values needed to protect or restore resources sensitive to acidification damage. Sensitivity to nutrient enrichment (eutrophication) from N addition was not considered.

The N CL to protect lake  $\text{NO}_3^-$  concentration generally increased with the N CL to protect lake ANC, although there was considerable variability in that relationship. For most modeled lakes, the N CL to protect lake  $\text{NO}_3^-$  concentration to 10  $\mu\text{eq/L}$  or to 20  $\mu\text{eq/L}$  was somewhat lower than the N CL to protect ANC to the three critical threshold criteria of 0, 20, and 50  $\mu\text{eq/L}$  (Figure 3-12). There were, however, some lakes that had N CL values equal to zero because the CL target was unattainable solely as a consequence of S deposition inputs assumed in the CAIR emissions scenario.

The CL for attaining a given lake water  $\text{NO}_3^-$  concentration is the same for the years 2050 and 2100. This is because the MAGIC model specifies N retention characteristics that are constant over time. Therefore, the watersheds could retain the same amount of N deposition in 2100 as they can in 2050.

**Figure 3-12. Comparisons between nitrogen critical load to protect lake ANC and nitrogen critical load to protect lake nitrate concentration for the 70 MAGIC modeling watersheds that had watershed-specific soils data**

A 1:1 line is provided as reference.



### 3.4 Regional Extrapolation of Model Critical Load Results

Numeric extrapolation of model-simulated CL values for this project focused on estimating numbers and percentages of the population of Adirondack lake watersheds predicted to exhibit various CL and exceedance values. A CL for a particular suite of acidifying pollutant, sensitive criterion, critical threshold value and endpoint year specifications can be simulated and extrapolated to one or more (up to the full population) Adirondack lake watersheds using a variety of approaches. Several such approaches were used in this study, as outlined in Table 3-2. The CL and associated exceedance results might apply to individual lakes or to groups of lakes within the Adirondack region. Results can vary dramatically depending on what lake or group of lakes is selected for examination.

**Table 3-2. Outline of different approaches for estimating the critical sulfur load for a given lake watershed or group of watersheds**

Approach	Number of Lake Watersheds	Description
Modeled – all MAGIC sites	97	MAGIC simulation of CL to protect lake ANC or $\text{NO}_3^-$ concentration at all sites modeled using MAGIC.
Modeled – MAGIC sites included in soil survey	70	MAGIC simulation of CL to protect soil BS, soil solution Bc:Al, or soil solution Ca:Al at all sites modeled using MAGIC that had watershed-specific soils data (no soils data borrowing).
Numerically Extrapolated – all EMAP lakes having ANC < 200 $\mu\text{eq/L}$	1,320	Numeric expansion of MAGIC simulation results for 44 modeled EMAP lakes to the EMAP frame of Adirondack lakes larger than 1 ha and deeper than 1 m that had ANC < 200 $\mu\text{eq/L}$ .
Numerically Extrapolated – all EMAP lakes, irrespective of ANC	1,829	Same as above except including high ANC lakes by assuming a high CL for all EMAP lakes that were not modeled using MAGIC because they had ANC > 200 $\mu\text{eq/L}$ .
Spatially Extrapolated using ALS	1,136	Critical load extrapolated from MAGIC sites using a regression model based on site-specific lake chemistry. This approach was used for all lakes sampled in the Adirondack Lakes Survey that were larger than 1 ha.

As an example, in Table 3-3, CL results for S deposition are compared among various groups of Adirondack lake watersheds based on one suite of CL specifications and assumptions, the estimated CL of S deposition to protect lake water to ANC = 50  $\mu\text{eq/L}$  in the year 2100. The modeled lakes were generally skewed toward lower CL values, as compared with the lake population distributions. The EMAP populations (especially the full EMAP population of all lakes, regardless of ANC) and the ALS population were more skewed toward high CL values.

Numeric extrapolations were performed based on the 44 modeled lakes that were taken from the EMAP survey of low-ANC ( $\leq 200 \mu\text{eq/L}$ ) lake watersheds, representing a population of 1,320 Adirondack lakes. Even though EMAP lakes having ANC > 200  $\mu\text{eq/L}$  were not modeled for this study, we can assume that the CL values to protect the ANC of such lakes would be high (but were not actually quantified). This assumption allows extrapolation to the full population of EMAP target lakes (estimated N = 1,829). These numeric extrapolations provide information on numbers and percentages of lakes in different CL classes.



**Table 3-3. Estimated percentage of Adirondack lake watersheds having various critical load of sulfur deposition values to protect against sulfur-driven lake acidification to ANC = 50 µeq/L in the year 2100, using different approaches and population frames**

Approach	Number of Watersheds	Percentage of Lakes in Critical Load Class						
		CL (S)	≤25	25-50	50-75	75-100	> 100	≤50 (meq/m <sup>2</sup> /yr)
MAGIC model simulations for all modeled lake watersheds	97		28.9	26.8	17.5	15.5	11.3	55.7
MAGIC model simulations for all modeled lake watersheds that were calibrated using watershed-specific soil chemistry data derived from the 2003 soil survey	70		27.1	22.9	21.4	17.1	11.4	50.0
Extrapolation of MAGIC model simulation results for 44 EMAP probability survey lakes to the EMAP frame of Adirondack lakes that are larger than 1 ha, deeper than 1 m, and that have ANC ≤ 200 µeq/L	1,320		19.5	21.8	22.2	10.1	26.4	41.3
Same as above, except assuming a high CL for all EMAP lakes that were not modeled using MAGIC because they had ANC > 200 µeq/L	1,829		14.1	15.7	16.0	7.3	46.8	29.8
Spatial extrapolation of MAGIC model simulation results to all lakes surveyed by the ALS that are larger than 1 ha	1,136		25.8	13.1	10.4	8.9	41.8	38.9

To investigate the spatial patterns in CL values, it is necessary to use a spatial extrapolation, rather than the EMAP numeric extrapolation. This was accomplished using the ALS lake chemistry data for 1,136 surveyed lakes larger than 1 ha in the Adirondack Ecoregion (Table 3-2).

As discussed previously, CLs were modeled using MAGIC for 97 lakes in this project, but only 70 of those had watershed-specific soils data for use in the model calibration. Soils data for the remainder of the sites were borrowed from a neighboring watershed. As discussed in Section 2.6, this soils data borrowing introduced uncertainty into the CL estimates constructed to protect the soil or soil solution chemical indicators, but not appreciably for CL estimates to protect lake chemistry. Thus, CL values were calculated here for all 97 modeled watersheds to protect lake chemistry, but only for 70 watersheds to protect soil and soil solution chemistry.

The percentage of lakes found to be within the various CL classes varied substantially based on the group of lakes modeled or represented by the estimate (Table 3-3). In extrapolating CL and/or exceedance results obtained by modeling individual lake watersheds to the broader population of Adirondack lakes and their associated watersheds, it is therefore important to specify the lake population or statistical frame to which the results are being applied. There are multiple options for specifying an Adirondack lake population or statistical frame (Table 3-4). The 1991-1994 EMAP Northeast Lake Survey used the digital 1:100,000 scale map frame existing at that time to make population estimates. Generally, lakes larger than approximately 1 ha are mapped at this scale. A hexagon grid was used to select a systematic, randomized, probability sample of lakes from this frame. There were an estimated 2,180 lakes in the Adirondack Ecoregion based on a sample of 133 lakes that were present in this ecoregion in the frame. After field visits to these sample lakes, only an estimated 1,829 lakes (sample size=115) were considered to be target lakes by EMAP definitions (actual water bodies larger than 1 ha with maximum depth  $\geq 1$  m and  $\geq 1,000$  m<sup>2</sup> of open water). The EMAP frame can be further broken down by expected lake ANC into two classes that include lakes that are greater than or less than ANC = 200  $\mu\text{eq/L}$ . For example, Sullivan et al. (2006a) modeled the responses of Adirondack lakes to past and future acidic deposition, based on a frame that included only EMAP lakes having ANC  $\leq 200$   $\mu\text{eq/L}$ .

**Table 3-4. Population frames for Adirondack lakes that were considered for inclusion in this study**

SE=standard error of estimate, n=number of probability lakes sampled to make estimate.

Frame	Description	Number of Lakes
EMAP, All Mapped Lakes	Survey estimate of all lakes present on the digitized 1:100,000 scale maps circa 1990	2,180 (SE=275) n=133
EMAP, All Target Lakes	Mapped lakes identified in the EMAP study were considered target if they were water bodies $\geq 1$ ha in surface area, maximum depth $\geq 1$ m, and $\geq 1,000$ m <sup>2</sup> of open water based on a summer index period field visit.	1,829 (SE=244) n=115
EMAP, Low ANC Lakes	Target lakes identified in the EMAP study that had lake water ANC less than 200 $\mu\text{eq/L}$ based on a summer index period field visit. This is the frame used by Sullivan et al. (2006a) for statistical extrapolation of MAGIC.	1,320 (SE=102) n=83
National Hydrography Dataset (NHD)	Lakes identified in the medium resolution NHD (1:100,000 scale) that are larger than 1 ha.	2,266
NHD – High Resolution	Lakes identified in the high resolution NHD (1:24,000) that are larger than 1 ha	3,411

For the CL analyses reported here, it can be assumed that lakes having  $ANC > 200 \mu\text{eq/L}$  would also have high CL to protect lake water ANC to various levels (generally higher than the lakes that have ANC below  $200 \mu\text{eq/L}$ ) even though those CLs were not modeled. If we assume that those high-ANC lakes that were not modeled would have high CL, it is possible to specify an approximate CL distribution that includes all EMAP lakes, not just those having low ANC. This adjustment only applies to calculations of CL to protect the sensitive receptor lake water. For protecting soil or soil solution, it cannot be assumed that the CL for a watershed containing a high-ANC lake would necessarily be higher than the CL for a watershed containing a low-ANC lake. Therefore, it is possible to extrapolate CL results to the entire EMAP frame, irrespective of lake ANC, but only for CL values that are selected to protect lake chemistry, and not for CL values that are selected to protect soil or soil solution chemistry.

The EMAP frame identified a total of 2,180 lakes in the Adirondack ecoregion that are larger than 1 ha. Some of those lakes in the statistical frame were considered to be “non-target lakes” (Table 3-4) in the EMAP study because they were either map errors (non-water bodies),  $< 1$  ha in surface area, or determined in the field to be shallower than 1 m and/or contained limited open water (weedy, macrophyte-dominated, wetland-type systems).

A more recently developed lake population frame is available from the medium-resolution National Hydrography Dataset (NHD). It includes 2,266 lakes larger than 1 ha within the Adirondack Ecoregion. An unknown number of these lakes may be shallower than 1 m depth and/or lack open water, and therefore fall outside the limits of the set of EMAP target lakes. Even more lakes (approximately 3,411) larger than 1 ha are represented in the high-resolution NHD. It is likely that this high-resolution dataset contains many shallow and/or macrophyte-dominated lakes or smaller lakes with near-shore wetlands. No estimate is available of the numbers of such presumably non-target lakes. Critical load and exceedance modeling results generated in this study are not extrapolated to either of these larger NHD frames. This is because we have no basis for quantifying the lakes in either NHD frame that might be considered non-target due to depth or open water conditions and because we were unable to develop a robust method for extrapolating the results of MAGIC modeling at 97 modeled sites to either of these NHD lake populations. Available landscape variables were inadequate as the basis for extrapolation of CLs. Rather, spatial extrapolation of MAGIC modeling CL results was based on water chemistry survey data. See Section 3.4.2 of this report for discussion. Such data were available for 1,136 lakes that were surveyed by the ALS in the 1980s.

A breakdown of the kinds of analyses possible with these various groups of lake watersheds is given in Table 3-5. The data set that is most versatile in generating relevant model results for the larger population of Adirondack lake watersheds is the EMAP population of 1,320 lakes having  $ANC \leq 200 \mu\text{eq/L}$  and surface area  $\geq 1$  ha.

**Table 3-5. Analyses possible with the various population frames and data sets employed or considered for this study**

Analyses Possible with This Dataset	EMAP	EMAP Target	ALS	Medium	High
	All Target Lakes (N = 1,829)	Low ANC (≤200 µeq/L) (N = 1,320)	Adirondack Lakes Larger than 1 ha (N=1,136)	Resolution NHD (N = 2,266)	Resolution NHD (N = 3,411)
MAGIC model estimates of pre-industrial chemistry	No <sup>a</sup>	Yes	No	No	No
Estimates of recent (since 1980s) or current chemistry	Yes	Yes	Yes	No	No
Simulated and extrapolated critical load	No <sup>b</sup>	Yes	Yes	No	No

<sup>a</sup> For EMAP lakes having ANC > 200 µeq/L, it can be assumed that pre-industrial ANC was high, but it cannot be quantified. Therefore, the approximate distribution of pre-industrial ANC can be developed for all EMAP lakes, even though the absolute estimates are not available for those having recent ANC > 200 µeq/L.

<sup>b</sup> For EMAP lakes having ANC > 200 µeq/L, it can be assumed that the critical load would be high, but the actual critical load cannot be quantified. Therefore, the approximate distribution of CL values can be developed for all EMAP lakes, even though the absolute CL values are not available for those having ANC > 200 µeq/L.

### 3.4.1 Numerical Extrapolation to EMAP Population Frames

#### 3.4.1.1 Watersheds Having Lake ANC ≤ 200 µeq/L

The numbers and percentages of Adirondack lakes having CL of S or N deposition in various classes were calculated based on extrapolation of the MAGIC modeling results to the regional EMAP population of Adirondack lakes having ANC ≤ 200 µeq/L, lake area ≥ 1 ha, and lake depth ≥ 1 m. These results are summarized in Table 3-6 and Table 3-7. The results are organized by sensitive receptor (lake water, soil, soil solution), sensitive chemical criterion (ANC, %BS, Bc/Al, Ca/Al, two or three critical threshold values for each sensitive chemical criterion (ANC 0, 20, 50 µeq/L; BS 5, 10, 15%; Bc/Al and Ca/Al 1, 10), and two endpoint years (2050 and 2100).

**Table 3-6. Estimated number and percent of Adirondack lake watersheds having various critical load values to protect against sulfur-driven acidification within the EMAP population of 1,320 Adirondack lakes that have ANC less than 200 µeq/L**

Based on MAGIC model simulations for 44 statistically selected lakes.

Sources: Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.; and Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. *J. Environ. Stud. Sci.* 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.

Receptor	Sensitive Criterion	Critical Value	Endpoint Year	CL(S)	Number (and Percent) of Lakes in Critical Load Class										
					≤25		25-50		50-75		75-100		> 100		
					(meq/m <sup>2</sup> /yr)										
Lake water	ANC	0 µeq/L	2050	0	(0)	175	(13.3)	41	(3.1)	289	(21.9)	815	(61.7)		
			2100	0	(0)	175	(13.3)	208	(15.8)	197	(14.9)	740	(56.0)		
		20 µeq/L	2050	175	(13.3)	28	(2.1)	313	(23.7)	77	(5.8)	727	(55.1)		
			2100	159	(12.0)	58	(4.4)	329	(24.9)	176	(13.3)	599	(45.4)		
		50 µeq/L	2050	301	(22.8)	244	(18.5)	115	(8.7)	252	(19.1)	408	(30.9)		
			2100	257	(19.5)	288	(21.8)	293	(22.2)	134	(10.1)	348	(26.4)		
Soil	BS	5%	2050	99	(7.5)	54	(4.1)	41	(3.1)	12	(0.9)	1113	(84.3)		
			2100	54	(4.1)	99	(7.5)	54	(4.1)	109	(8.3)	1004	(76.1)		
		10%	2050	927	(70.2)	93	(7.1)	16	(1.2)	129	(9.7)	156	(11.8)		
			2100	873	(66.2)	163	(12.3)	129	(9.7)	0	(0)	156	(11.8)		
		15%	2050	1253	(94.9)	0	(0)	30	(2.3)	12	(0.9)	25	(1.9)		
			2100	1237	(93.7)	46	(3.5)	12	(0.9)	0	(0)	25	(1.9)		
		Soil Solution	B <sub>c</sub> :Al	1	2050	78	(5.9)	25	(1.9)	41	(3.1)	43	(3.2)	1133	(85.8)
					2100	78	(5.9)	25	(1.9)	71	(5.4)	355	(26.9)	791	(59.9)
10	2050			1252	(94.8)	28	(2.1)	16	(1.2)	0	(0)	25	(1.9)		
	2100			1235	(93.5)	45	(3.4)	16	(1.2)	25	(1.9)	0	(0)		
Soil Solution	Ca:Al			1	2050	268	(20.3)	204	(15.5)	308	(23.4)	62	(4.7)	478	(36.2)
					2100	252	(19.0)	221	(16.7)	370	(28.0)	112	(8.5)	366	(27.7)
		10	2050	1296	(98.1)	0	(0)	0	(0)	25	(1.9)	0	(0)		
			2100	1296	(98.1)	0	(0)	25	(1.9)	0	(0)	0	(0)		

**Table 3-7. Estimated number and percent of Adirondack lake watersheds having various critical load values to protect against nitrogen-driven acidification within the EMAP population of 1,320 Adirondack lakes that have ANC less than 200 µeq/L**

Based on MAGIC model simulations for 44 statistically selected lakes.

Receptor	Sensitive Criterion	Critical Value	Endpoint Year	CL(N)	Number (and Percent) of Lakes in Critical Load Class										
					≤25	25-50	50-75	75-100	> 100 (meq/m <sup>2</sup> /yr)						
Lake water	ANC	0 µeq/L	2050	465	(35.2)	.	.	17	(1.3)	.	.	839	(63.5)		
			2100	449	(34.0)	16	(1.2)	.	.	17	(1.3)	839	(63.5)		
		20 µeq/L	2050	515	(39.0)	.	.	.	.	14	(1.0)	792	(60.0)		
			2100	515	(39.0)	.	.	.	.	14	(1.0)	792	(60.0)		
		50 µeq/L	2050	599	(45.4)	12	(0.9)	.	.	.	.	709	(53.7)		
			2100	599	(45.4)	12	(0.9)	.	.	.	.	709	(53.7)		
Lake water	NO <sub>3</sub> <sup>-</sup>	10 µeq/L	2050	.	.	131	(9.9)	42	(3.2)	28	(2.1)	1120	(84.8)		
			2100	.	.	131	(9.9)	42	(3.2)	28	(2.1)	1120	(84.8)		
		20 µeq/L	2050	.	.	.	.	15	(1.2)	116	(8.8)	1189	(90.1)		
			2100	.	.	.	.	15	(1.2)	116	(8.8)	1189	(90.1)		
		Soil	BS	5%	2050	104	(7.9)	45	(3.4)	.	.	.	.	1171	(88.7)
					2100	104	(7.9)	45	(3.4)	.	.	.	.	1171	(88.7)
10%	2050			388	(29.4)	93	(7.1)	.	.	.	.	839	(63.5)		
	2100			402	(30.4)	.	.	17	(1.3)	.	.	902	(68.3)		
15%	2050			502	(38.0)	.	.	.	.	.	.	818	(62.0)		
	2100			502	(38.0)	16	(1.2)	.	.	.	.	802	(60.7)		

The percent of low-ANC Adirondack lakes estimated to have low CL ( $\leq 50 \text{ meq/m}^2/\text{yr}$ ) differed by more than a factor of two depending on threshold selection of 20 versus 50  $\mu\text{eq/L}$  (Figure 3-13a). Over 40% of the low-ANC lakes in the EMAP frame had CL  $\leq 50 \text{ meq/m}^2/\text{yr}$  for protecting lake ANC to 50  $\mu\text{eq/L}$  in the year 2100. That percentages was only about 17% using the critical ANC threshold of 20  $\mu\text{eq/L}$ . Even larger differences were observed for the two BS critical thresholds (Figure 3-13b). The numbers and percentages of lakes having intermediate CL values (50 to 100  $\text{meq/m}^2/\text{yr}$ ) were similar using the contrasting threshold ANC or BS values (Figure 3-13a and 3-13b).

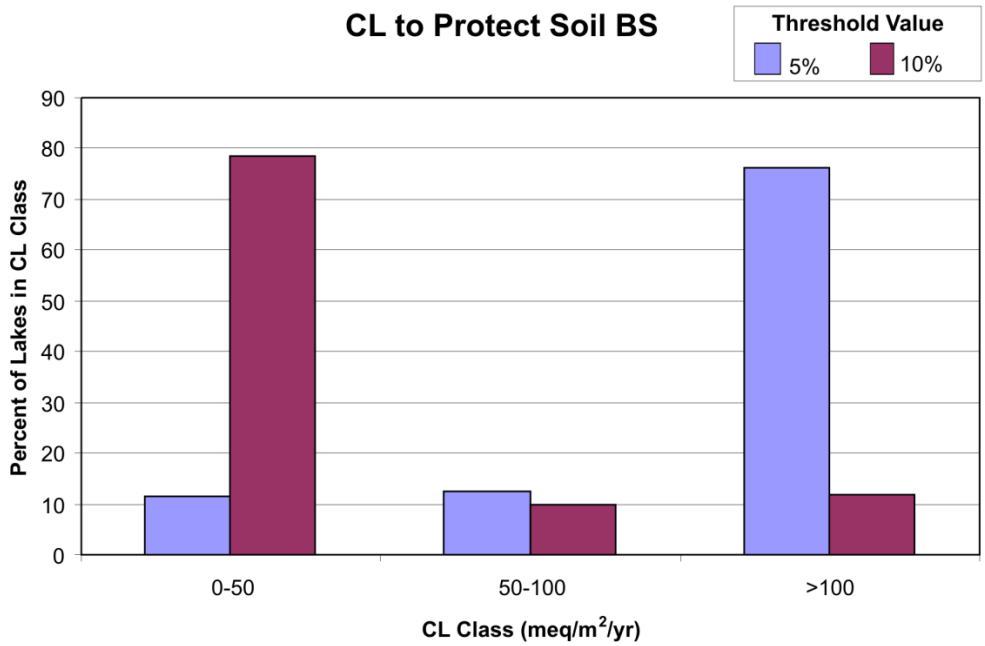
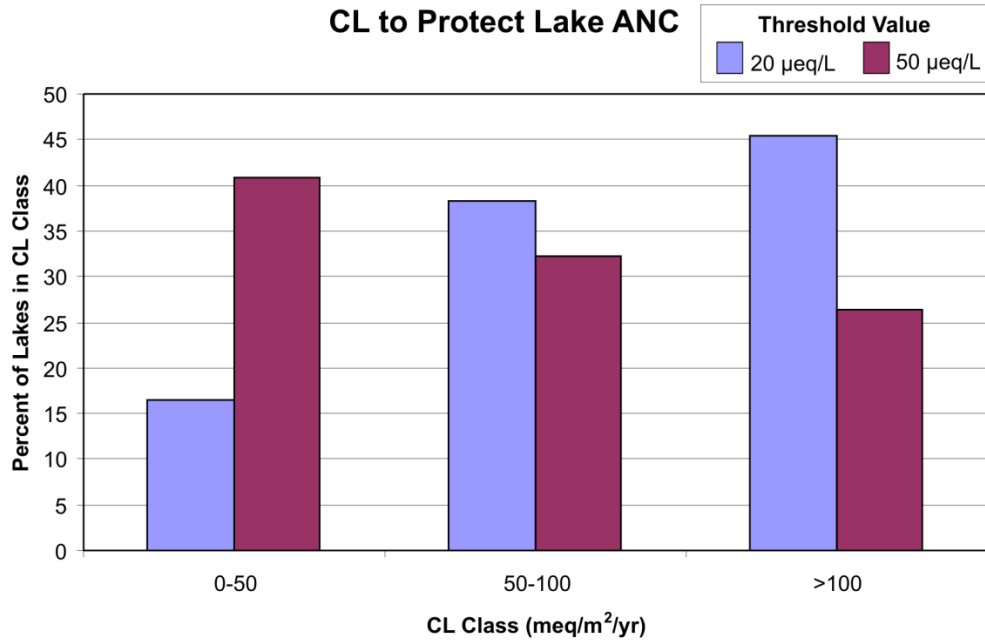
In evaluating CL results for resource protection against acidification, it can be helpful to examine the simulated historical and current lake water and soil chemistry. This is done by considering the simulated distributions of lake ANC and soil % BS in the years 1850 (pre-industrial period) and 2002 (calibration period; Figure 3-14). MAGIC model simulations suggested that there were no acidic (ANC  $\leq 0 \text{ } \mu\text{eq/L}$ ) lakes in the EMAP lake population in 1850 but about 175 such lakes (13% of the low-ANC EMAP population) in 2002. In addition, the simulated numbers of lakes having ANC  $\leq 20 \text{ } \mu\text{eq/L}$  and  $\leq 50 \text{ } \mu\text{eq/L}$  increased markedly from 1850 to 2002 (Figure 3-14a). Simulated effects on soil % BS since the pre-industrial time suggest that an estimated 79 lake watersheds (6% of the low-ANC population) had BS  $\leq 5\%$  in 1850. That number increased by about 50% between 1850 and 2002. The numbers of lake watersheds in all of the lower soil BS classes shown in the figure ( $\leq 5\%$ , 5-7.5%, and 7.5-10%) increased from the pre-industrial period to the present time (Figure 3-14b), although the differences were less pronounced as compared with simulated changes in lake ANC.

To protect lake ANC from negative values ( $< 0 \text{ } \mu\text{eq/L}$ ) in the year 2050 or 2100, the S deposition CL for each EMAP lake was above 25  $\text{meq/m}^2/\text{yr}$ . If, however, the critical lake ANC threshold value was set higher, between 12% and 23% of the EMAP lakes had calculated CL below 25  $\text{meq/m}^2/\text{yr}$ , depending on the choice of critical threshold value (20 or 50  $\mu\text{eq/L}$ ) and endpoint year (2050 or 2100). More than half of the 1,320 EMAP lakes had relatively high CL ( $> 100 \text{ meq/m}^2/\text{yr}$ ) to protect lake ANC to 0  $\mu\text{eq/L}$  (62% for the endpoint year 2050, 56% for the endpoint year 2100). More than one fourth of the 1,320 EMAP lakes had relatively high CL ( $> 100 \text{ meq/m}^2/\text{yr}$ ) to protect lake ANC to 50  $\mu\text{eq/L}$  (31% for the endpoint year 2050, 26% for the endpoint year 2100; Table 3-6).

Critical S load estimates for protecting soil BS were very sensitive to selection of the critical threshold value for BS (Table 3-6). For example, BS 5% was rather readily achievable, with more than three-fourths of the EMAP lakes having CL  $> 100 \text{ meq/m}^2/\text{yr}$  to protect to this BS level. In marked contrast, to achieve BS of 10%, about two-thirds or more of the EMAP lakes showed very low CL values ( $< 25 \text{ meq/m}^2/\text{yr}$ ). Nearly all of the EMAP lakes ( $\geq 94\%$ ) had S CL  $< 25 \text{ meq/m}^2/\text{yr}$  to achieve BS of 15% (Table 3-6).

Figure 3-13. Histogram giving results of numeric extrapolations of CL to protect a) lake ANC and b) soil BS, each to two critical thresholds for 1,320 low-ANC ( $\leq 200 \mu\text{eq/L}$ ) EMAP lakes

a)

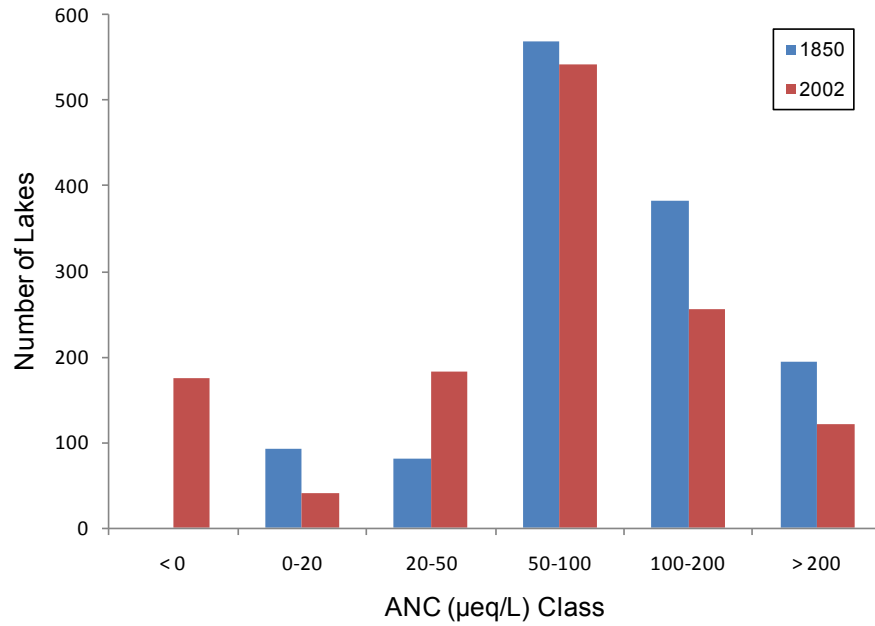




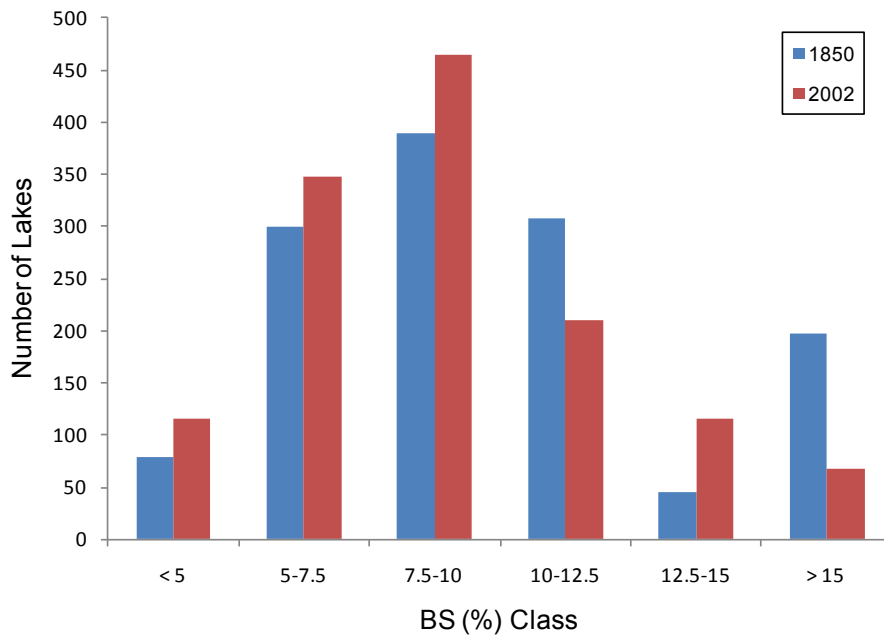
**Figure 3-14. Histograms of MAGIC simulations of A) lake water ANC and B) soil BS in 1850 and 2002 for the population of 1,320 Adirondack lakes included in the EMAP frame**

Source: Panel A, Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.; Panel B, modified from Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. *J. Environ. Stud. Sci.* 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.

a)



b)



The CL to protect soil solution ratios of Bc:Al and Ca:Al showed substantial differences depending on selection of the critical threshold value (1 or 10). Nearly all (>93%) of the EMAP lakes had CL for both of these variables that was very low (<25 meq/m<sup>2</sup>/yr) when the protection threshold was set to a ratio of 10. In marked contrast, the majority (≥60%) of the EMAP lakes had relatively high CL (>100 meq/m<sup>2</sup>/yr) to achieve Bc:Al = 1. Assuming a critical value of 1 yielded more diversity of response among the EMAP lake watersheds for the critical target criterion Ca:Al. For example, based on either endpoint year (2050 or 2100), there were large numbers of lakes estimated to have both high and low S CL values to protect the Ca:Al ratio from going below 1 (Table 3-6).

There were not particularly large differences in the simulated CL values to protect lake ANC against S loading for different critical threshold criteria (ANC = 20 or 50 µeq/L) and/or endpoint years (2050, 2100; Figure 3-15a). Differences in response to variation in the selected CL endpoint year were slightly more pronounced for the high CL values, with lower CLs required to achieve protection by the year 2050 as compared with 2100. For protecting soil BS, however, the choice of critical threshold criterion (5% or 10%) made a larger difference in the resulting CL (Figure 3-15b). In addition, the selection of endpoint year made a substantial difference in CL results to protect soil BS, but only for the relatively high (higher than about 75 meq/m<sup>2</sup>/yr) CL values (Figure 3-15b). Those CL values above 75 meq/m<sup>2</sup>/yr are higher than ambient S loading (Figure 3-15c).

Based on MAGIC model outputs extrapolated to the regional lake population, CL population statistics were generated for various endpoint years (Table 3-8). Results suggested that the median (50th percentile) lake, from among the 1,320 Adirondack lakes larger than 1 ha that have ANC <200 µeq/L, had CL equal to 67 meq/m<sup>2</sup>/yr to protect lake ANC to 50 µeq/L in the year 2100. An estimated one-fourth of those lakes had had CL less than 32 meq/m<sup>2</sup>/yr to achieve that same level of protection (Table 3-8).

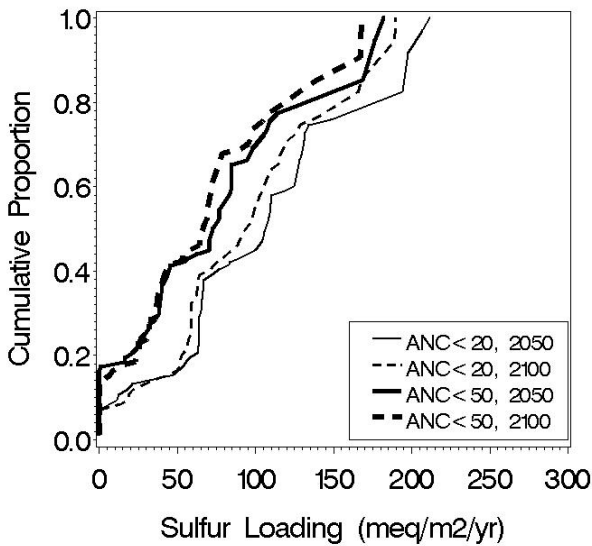
Results of MAGIC model simulations of N critical loads extrapolated to the EMAP population of 1,320 low-ANC (≤ 200 µeq/L) Adirondack lakes are shown in Table 3-7. In general, the model estimates suggested that most Adirondack lakes are considerably less sensitive to acidification from N deposition (Table 3-7), as compared with S deposition (Table 3-6). As a consequence, estimated N CL values tended to be relatively high. This is largely because most of the modeled lakes are currently retaining within watershed soils most of the deposited N.

**Figure 3-15. Cumulative distributions of MAGIC simulated critical sulfur load to protect against a) low lake ANC and b) low soil BS in the years 2050 and 2100 for the EMAP population of 1,320 Adirondack lakes having ANC less than 200  $\mu\text{eq/L}$**

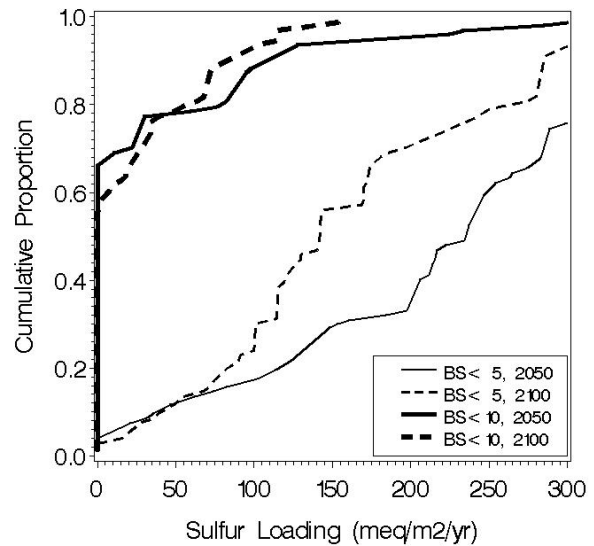
Current (2002) sulfur loading is also shown as a point of comparison (Panel c).

Source: Panel B, from Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. *J. Environ. Stud. Sci.* 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.

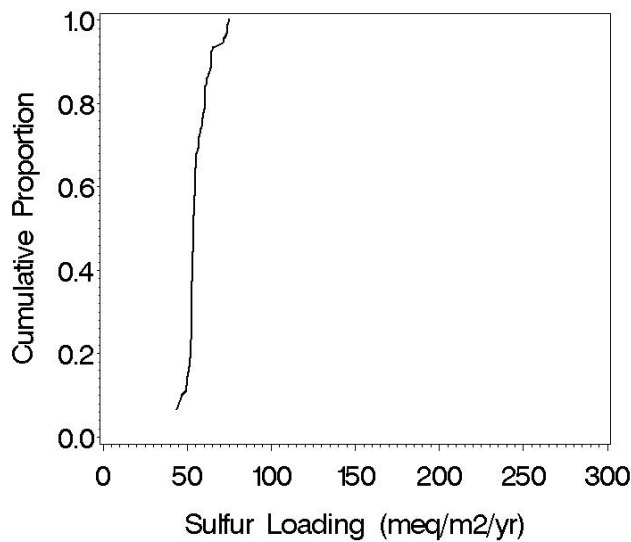
**a. Critical Load for Protecting Lake ANC**



**b. Critical Load for Protecting Soil Base Saturation**



**c. 2002 Ambient Sulfur Deposition Loading**



**Table 3-8. Critical load percentile values for three population estimates**

The three population estimates are the EMAP frame (numerical extrapolation from 44 modeled lake watersheds), for all lakes and for low-ANC lakes, and the ALS dataset (spatial extrapolation of CL based on ALS survey lake chemistry data for 1,136 lakes).

*Source: Modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. Water Resour. Res. 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.*

			Critical Sulfur Load (meq/m <sup>2</sup> /yr) to Protect Against Low Lake ANC					
Population Frame	Number	Percentile	ANC = 0 µeq/L		ANC = 20 µeq/L		ANC = 50 µeq/L	
			2050	2100	2050	2100	2050	2100
EMAP Low ANC Lakes <sup>a</sup>	1,320	10th	40	41	13	19	0	0
		25th	79	72	64	59	29	32
		50th	126	113	109	98	72	67
		75th	165	152	149	137	109	104
		90th	212	203	197	189	176	167
EMAP All Lakes <sup>b</sup>	1,829	10th	59	55	38	37	0	6
		25th	84	78	66	63	40	38
		50th	155	135	131	120	106	96
		75th	>165	>152	>149	>137	>109	>104
		90th	>212	>203	>197	>189	>176	>167
ALS Surveyed Lakes <sup>c</sup>	1,136	10th	47	43	21	21	0	0
		25th	78	71	54	51	23	23
		50th	133	121	113	104	82	77
		75th	211	192	197	179	166	152
		90th	316	287	309	279	279	254

**Table 3-8 continued**

			Critical Sulfur Load (meq/m <sup>2</sup> /yr) to Protect Against Low Soil Base Saturation					
			BS = 5%		BS = 10%		BS = 15%	
			2050	2100	2050	2100	2050	2100
EMAP Low ANC Lakes <sup>a</sup>	1,320	10th	46	49	0	0	0	0
		25th	149	101	0	0	0	0
		50th	237	143	0	0	0	0
		75th	298	240	30	34	0	0
		90th	571	286	128	103	0	17
EMAP All Lakes <sup>b</sup>	1,829	10th	NA	NA	NA	NA	NA	NA
		25th	NA	NA	NA	NA	NA	NA
		50th	NA	NA	NA	NA	NA	NA
ALS Surveyed Lakes <sup>c</sup>	1,136	75th	NA	NA	NA	NA	NA	NA
		90th	NA	NA	NA	NA	NA	NA
		10th	NA	NA	NA	NA	NA	NA
		25th	NA	NA	NA	NA	NA	NA
		50th	NA	NA	NA	NA	NA	NA
		75th	NA	NA	NA	NA	NA	NA
		90th	NA	NA	NA	NA	NA	NA

**Table 3-8 continued**

			Critical Nitrogen Load (meq/m <sup>2</sup> /yr) to Protect Against High Lake NO <sub>3</sub> <sup>-</sup> Concentration			
			NO <sub>3</sub> <sup>-</sup> = 10 µeq/L		NO <sub>3</sub> <sup>-</sup> = 20 µeq/L	
			2050	2100	2050	2100
EMAP Low ANC Lakes <sup>a</sup>	1,320	10th	52	52	105	105
		25th	322	322	644	644
		50th	964	964	1929	1929
		75th	ND <sup>d</sup>	ND	ND	ND
		90th	ND	ND	ND	ND
EMAP All Lakes <sup>b</sup>	1,829	10th	NA	NA	NA	NA
		25th	NA	NA	NA	NA
		50th	NA	NA	NA	NA
		75th	NA	NA	NA	NA
		90th	NA	NA	NA	NA
ALS Surveyed Lakes <sup>c</sup>	1,136	10th	NA	NA	NA	NA
		25th	NA	NA	NA	NA
		50th	NA	NA	NA	NA
		75th	NA	NA	NA	NA
		90th	NA	NA	NA	NA

<sup>a</sup> EMAP frame as modified by Sullivan et al. (2006a) to include only lakes having ANC ≤ 200 µeq/L.

<sup>b</sup> EMAP frame, including all lakes regardless of ANC. For lakes having ANC > 200 µeq/L, it was assumed that the CL would be high; it was set for assessing the CL to protect lake ANC in this analysis to the maximum value modeled for any lake by MAGIC or 1,000 meq/m<sup>2</sup>/yr, whichever was higher. For assessing the CL to protect soil or soil solution chemistry, modeling results were not extrapolated to the watersheds of high-ANC lakes (indicated as Not Analyzed, NA).

<sup>c</sup> CL to protect against low soil BS and CL to protect against low base cation to Al ratios could not be effectively extrapolated from the MAGIC modeled sites to the ALS population using available lake chemistry and mappable watershed features. These CL extrapolations are reported as not analyzed (NA).

<sup>d</sup> ND (no data) indicates that modeled CL values could not be determined for lake watersheds that currently show essentially 100% retention of atmospheric N deposition. More than 25% of the population of EMAP low-ANC lakes were in this class for the CL to protect against high lake water NO<sub>3</sub><sup>-</sup> concentration.

### 3.4.1.3 All Watersheds in EMAP Frame

Results of CL extrapolation to the entire EMAP lake population frame, irrespective of ANC, are shown in Table 3-9 for protecting lake water. Extrapolation to this larger lake population of CL values to protect soil and soil solution chemistry could not be conducted. For conducting the extrapolation of the CL to protect lake chemistry, it was assumed that the CL of high-ANC (>200 µeq/L) lakes would be high. Such an assumption could not necessarily be made for calculating the CL to protect soil or soil solution chemistry. The number of lakes simulated to be in the lower CL classes (below 100 meq/m<sup>2</sup>/yr) were the same regardless of which EMAP population frame was selected: the low-ANC frame (Table 3-6) or the entire frame (Table 3-9). However, there were many more lakes estimated to be in the highest CL class (>100 meq/m<sup>2</sup>/yr) when referenced to all EMAP lakes (Table 3-9). Percentages of lakes in the various CL classes differed depending on the population frame selected, especially the percentage of lakes estimated to have CL above 100 meq/m<sup>2</sup>/yr. Higher percentages of lakes were estimated to be in the highest CL class when the point of reference was the full EMAP population, as compared with the EMAP population of low-ANC lakes.

**Table 3-9. Estimated number and percent of Adirondack lake watersheds having various critical load values to protect lake ANC against sulfur-driven acidification for the EMAP population of 1,829 Adirondack lakes**

Based on MAGIC model simulations for 44 statistically selected lakes.

*Source: Modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. Water Resour. Res. 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.*

Receptor	Sensitive Criterion	Critical Value	Endpoint Year	CL(S)	Number (and Percent) of Lakes in Critical Load Class									
					≤25	25-50	50-75	75-100	> 100	(meq/m <sup>2</sup> /yr)				
Lake water	ANC	0 µeq/L	2050	0	(0)	175	(9.6)	41	(2.2)	289	(15.8)	1324	(72.4)	
			2100	0	(0)	175	(9.6)	208	(11.4)	197	(10.8)	1249	(68.3)	
		20 µeq/L	2050	175	(9.6)	28	(1.5)	313	(17.1)	77	(4.2)	1236	(67.6)	
			2100	159	(8.7)	58	(3.2)	329	(18.0)	176	(9.6)	1108	(60.6)	
		50 µeq/L	2050	301	(16.5)	244	(13.3)	115	(6.3)	252	(13.8)	917	(50.1)	
			2100	257	(14.1)	288	(15.7)	293	(16.0)	134	(7.3)	857	(46.8)	

### 3.4.2 Spatial Extrapolation to Lake Watersheds Surveyed by the Adirondack Lakes Survey

Numeric extrapolation results were summarized in the previous section. These give estimates of numbers and percentages of watersheds in various CL classes, but give no information regarding where these lakes or watersheds are located. Locational information is provided by a spatial extrapolation.

A suite of candidate predictor variables were considered as the basis for spatial extrapolation of MAGIC modeled results. These candidate predictor variables included elements of lake chemistry and landscape characteristics (Table 2-1). Candidate regression landscape variables that might be associated with estimated CL values were available at different spatial scales. These variables are given in Table 3-10. The scales are generally comparable except in cases where coarser STATSGO soils data were used at locations that lacked the higher resolution SSURGO data.

**Table 3-10. Source and scale of data representing landscape variables for comparison with critical loads modeling results**

Dataset	Source	Scale or Resolution
Elevation	National Elevation Dataset (NED)	1 arc-second (30 m) <sup>a</sup>
Slope	Derived from NED	1 arc-second (30 m) <sup>a</sup>
Soils <sup>b</sup>	SSURGO	1:24,000
	STATSGO	1:250,000
Watershed Area	Derived from NED	1 arc-second (30 m) <sup>a</sup>

<sup>a</sup> Approximately equivalent to 1:24,000 scale

<sup>b</sup> Soils variables include pH, depth, and percent clay

Pearson correlation coefficients are given in Table 3-11 to illustrate the correlations among the candidate CL predictor variables and the MAGIC model estimates of S-driven CLs for the modeled lake watersheds. Relatively high correlations are shaded in the table; correlations between 0.5 and 0.75 are shaded light grey and correlations above 0.75 are shaded dark grey. Some relatively strong correlations were observed among lake chemistry variables (i.e., among ANC, pH, and sum of base cations) and soil variables (i.e., between soil pH and soil % clay). Critical load estimates to protect lake water ANC at various levels were consistently most highly correlated ( $r = 0.85$  to  $0.96$ ) with lake ANC, and secondarily with lake sum of base cations (with and without  $Cl^-$  correction) and pH.



**Table 3-11. Pearson correlation coefficient matrix for candidate CL predictor values and MAGIC model estimates of the CL of sulfur deposition**

The selected candidate predictor variables reflect aspects of watershed morphology, soils, and lake chemistry that are expected to correlate to some degree with watershed and lake acid sensitivity. The model estimates of CL are based on different critical threshold criteria values and endpoint years. Correlations between 0.5 and 0.75 are coded light grey; correlations above 0.75 are coded darker grey.

Variable Type		Variable Type															
		Morphology				Soils			Lake Chemistry								
		WS Area	Elevation	Slope	WA:LA <sup>a</sup>	Soil % Clay	Soil pH	Soil Depth <sup>b</sup>	ANC	pH	SBC	SBC - CI	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>			
Morphology	WS Area	1.00															
	Elevation	-0.01	1.00														
	Slope	-0.04	0.34	1.00													
	WA:LA	0.49	0.04	-0.04	1.00												
Soils	Soil % Clay	0.01	0.39	0.19	0.05	1.00											
	Soil pH	0.01	-0.29	-0.40	-0.02	-0.66	1.00										
	Soil Depth	0.01	-0.28	-0.31	-0.08	0.03	0.01	1.00									
Lake Chemistry	ANC	0.15	-0.22	0.02	0.12	-0.05	-0.08	-0.01	1.00								
	pH	0.15	-0.26	0.23	0.14	-0.05	-0.17	-0.01	0.62	1.00							
	SBC	0.16	-0.05	0.13	0.08	0.06	-0.19	-0.03	0.82	0.57	1.00						
	SBC - CI	0.19	-0.07	0.13	0.17	0.01	-0.15	-0.06	0.94	0.67	0.88	1.00					
	SO <sub>4</sub> <sup>2-</sup>	0.10	0.33	0.29	0.13	0.19	-0.22	-0.14	0.07	0.25	0.36	0.40	1.00				
	NO <sub>3</sub> <sup>-</sup>	0.17	0.34	0.24	0.16	0.07	-0.17	-0.10	-0.20	0.02	0.06	0.02	0.38	1.00			
Critical Load	ANC = 0, 2050	0.08	-0.24	-0.02	0.06	-0.15	-0.08	0.01	0.85	0.58	0.66	0.77	-0.03	-0.18			
	ANC = 0, 2100	0.08	-0.22	-0.01	0.06	-0.13	-0.07	-0.01	0.89	0.59	0.72	0.82	0.01	-0.17			
	ANC = 20, 2050	0.10	-0.23	-0.01	0.08	-0.12	-0.08	0.01	0.90	0.61	0.71	0.83	0.03	-0.18			
	ANC = 20, 2100	0.10	-0.20	0.01	0.08	-0.10	-0.07	0.00	0.93	0.62	0.76	0.87	0.07	-0.17			
	ANC = 50, 2050	0.13	-0.22	0.01	0.10	-0.11	-0.06	0.01	0.95	0.60	0.79	0.91	0.13	-0.18			
	ANC = 50, 2100	0.12	-0.17	0.03	0.11	-0.08	-0.07	-0.01	0.96	0.61	0.82	0.93	0.18	-0.16			
	BS = 5, 2050	0.08	0.10	0.16	0.05	0.12	-0.38	0.02	0.29	0.15	0.40	0.31	0.11	0.14			

Table 3-11 continued

Variable Type	Variable Type												
	Morphology				Soils			Lake Chemistry					
	WS Area	Elevation	Slope	WA:LA <sup>a</sup>	Soil % Clay	Soil pH	Soil Depth <sup>b</sup>	ANC	pH	SBC	SBC - CI	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> -
BS = 5, 2100	0.11	0.14	0.21	0.08	0.12	-0.38	-0.02	0.47	0.24	0.58	0.52	0.22	0.17
BS = 10, 2050	-0.06	-0.08	0.04	-0.06	0.04	-0.26	0.05	0.10	0.07	0.14	0.08	-0.05	0.03
BS = 10, 2100	-0.05	-0.01	0.10	-0.07	0.07	-0.31	0.03	0.11	0.09	0.17	0.11	0.01	0.10
BS = 15, 2050	-0.04	-0.18	-0.04	-0.04	0.03	-0.18	0.09	0.04	0.00	-0.03	-0.02	-0.15	-0.09
BS = 15, 2100	-0.04	-0.17	0.01	-0.05	0.02	-0.19	0.06	0.06	0.05	0.02	0.01	-0.12	-0.07
Bc/Al = 1, 2050	-0.01	0.33	0.36	0.04	0.16	-0.37	-0.05	-0.13	-0.01	-0.03	-0.01	0.26	0.38
Bc/Al = 1, 2100	0.00	0.36	0.38	0.04	0.13	-0.35	-0.05	-0.06	0.02	0.04	0.08	0.32	0.39
Bc/Al = 10, 2050	-0.04	-0.10	0.09	-0.08	0.09	-0.17	-0.02	-0.10	0.02	-0.07	-0.06	0.07	0.06
Bc/Al = 10, 2100	-0.06	-0.03	0.09	-0.10	0.08	-0.19	-0.01	-0.17	-0.05	-0.12	-0.12	0.09	0.18
Ca/Al = 1, 2050	-0.04	0.29	0.33	0.04	0.16	-0.32	-0.10	-0.25	-0.04	-0.19	-0.14	0.21	0.33
Ca/Al = 1, 2100	-0.02	0.33	0.34	0.05	0.14	-0.31	-0.09	-0.27	-0.07	-0.19	-0.14	0.25	0.34
Ca/Al = 10, 2050	-0.02	-0.22	0.08	-0.04	0.06	-0.14	0.01	0.00	0.06	-0.02	-0.01	0.02	-0.10
Ca/Al = 10, 2100	-0.03	-0.15	0.08	-0.06	0.09	-0.15	0.00	-0.06	0.02	-0.05	-0.04	0.05	-0.04

<sup>a</sup> WA:LA – watershed area to lake area ratio.

<sup>b</sup> Depth data were not available for 4 of the 97 lake watersheds.

Only one of the CL estimates for soil chemistry protection (to protect to BS = 5% in the year 2100) was strongly correlated with any of the candidate predictor variables (in this case, sum of base cations). None of the CL estimates for soil solution protection were strongly correlated with any of the candidate predictor variables; the strongest correlation coefficient was only 0.39. None of the CL estimates were strongly correlated with any of the landscape morphology or soil chemistry variables. This latter result precludes or obfuscates using such variables to predict S CLs at Adirondack locations that lack lake water chemistry.

Table 3-12 provides a similar Pearson correlation coefficient matrix for N CLs. None of the N-based CLs were strongly correlated with any of the candidate predictor variables. This precludes or obfuscates extrapolating any of the N-based CL values to the broader population of Adirondack lakes using any of the candidate predictor variables.

MAGIC model simulations of the CL of S deposition needed to protect lake ANC from falling below designated critical criteria values could successfully be predicted using only lake water ANC as a predictor variable (Table 3-13). Each of the equations given in Table 3-13 explains the majority of the variation in modeled CL, with  $r^2$  values ranging from 0.72 (to protect ANC to 0  $\mu\text{eq/L}$  in the year 2050) to 0.92 (to protect ANC to 50  $\mu\text{eq/L}$  in the year 2100). The most robust predictions were obtained for estimating the CL to protect lake ANC from decreasing below 50  $\mu\text{eq/L}$  in the years 2050 and 2100 ( $r^2 = 0.90$  and  $0.92$ , respectively). The least robust predictions ( $r^2 = 0.72$  and  $0.80$ ) were obtained for predicting the CL to protect lake ANC from decreasing below 0  $\mu\text{eq/L}$ . Predictions for protecting to lake ANC of 20  $\mu\text{eq/L}$  were intermediate ( $r^2 = 0.81$  and  $0.86$ ; Table 3-13). Inclusion of other water chemistry attributes as predictor variables, in addition to lake ANC, had little effect on the predictions generated. Inclusion of lake  $\text{SO}_4^{2-}$  yielded a very minor improvement for the two equations that used a critical ANC criterion of 50  $\mu\text{eq/L}$ . For reasons of simplicity, the final equations reported here (Table 3-13) only use ANC and a constant to predict each CL.

**Table 3-12. Pearson correlation coefficient matrix for candidate critical predictor values and MAGIC model estimates of the CL of nitrogen deposition**

The selected candidate predictor variables reflect aspects of watershed morphology, soils, and lake chemistry that are expected to correlate to some degree with watershed and lake acid sensitivity. The model estimates of CL are based on different critical threshold criteria values and endpoint years. Correlations between 0.5 and 0.75 are coded light grey; correlations above 0.75 are coded darker grey.

Variable Type		Variable Type												
		Morphology				Soils			Lake Chemistry					
		WS Area	Elevation	Slope	WA:LA <sup>a</sup>	Soil % Clay	Soil pH	Soil Depth <sup>b</sup>	ANC	pH	SBC	SBC - CI	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>
Morphology	WS Area	1.00												
	Elevation	-0.01	1.00											
	Slope	-0.04	0.34	1.00										
	WA:SA	0.49	0.04	-0.04	1.00									
Soils	Soil % Clay	0.01	0.39	0.19	0.05	1.00								
	Soil pH	0.01	-0.29	-0.40	-0.02	-0.66	1.00							
	Soil Depth	0.01	-0.28	-0.31	-0.08	0.03	0.01	1.00						
Lake Chemistry	ANC	0.15	-0.22	0.02	0.12	-0.05	-0.08	-0.01	1.00					
	pH	0.15	-0.26	0.23	0.14	-0.05	-0.17	-0.01	0.62	1.00				
	SBC	0.16	-0.05	0.13	0.08	0.06	-0.19	-0.03	0.82	0.57	1.00			
	SBC - CI	0.19	-0.07	0.13	0.17	0.01	-0.15	-0.06	0.94	0.67	0.88	1.00		
	SO <sub>4</sub> <sup>2-</sup>	0.10	0.33	0.29	0.13	0.19	-0.22	-0.14	0.07	0.25	0.36	0.40	1.00	
	NO <sub>3</sub> <sup>-</sup>	0.17	0.34	0.24	0.16	0.07	-0.17	-0.10	-0.20	0.02	0.06	0.02	0.38	1.00
Critical Load	ANC = 0, 2050	-0.08	-0.36	-0.17	-0.17	-0.27	0.22	0.22	0.18	0.18	0.05	0.11	-0.12	-0.39
	ANC = 0, 2100	-0.08	-0.36	-0.17	-0.17	-0.27	0.22	0.22	0.18	0.18	0.05	0.10	-0.12	-0.40
	ANC = 20, 2050	-0.07	-0.35	-0.10	-0.14	-0.29	0.16	0.14	0.31	0.26	0.15	0.24	-0.06	-0.36
	ANC = 20, 2100	-0.07	-0.35	-0.10	-0.15	-0.29	0.16	0.14	0.30	0.26	0.15	0.24	-0.07	-0.36
	ANC = 50, 2050	-0.06	-0.42	-0.10	-0.13	-0.34	0.19	0.15	0.35	0.28	0.20	0.29	-0.04	-0.33
	ANC = 50, 2100	-0.06	-0.42	-0.10	-0.13	-0.34	0.19	0.15	0.34	0.28	0.19	0.28	-0.05	-0.33
	BS = 5, 2050	-0.17	-0.15	-0.07	-0.19	-0.07	0.02	0.10	0.01	-0.15	-0.10	-0.11	-0.23	-0.56
	BS = 5, 2100	-0.15	-0.20	-0.07	-0.18	-0.02	0.01	0.14	-0.03	-0.03	-0.10	-0.12	-0.18	-0.52

**Table 3-12 Continued**

Variable Type	Variable Type													
	Morphology				Soils			Lake Chemistry						
	WS Area	Elevation	Slope	WA:LA <sup>a</sup>	Soil % Clay	Soil pH	Soil Depth <sup>b</sup>	ANC	pH	SBC	SBC -		SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>
										CI				
BS = 10, 2050	-0.13	-0.27	-0.12	-0.25	-0.14	0.06	0.09	0.06	0.13	-0.03	-0.03	-0.20	-0.41	
BS = 10, 2100	-0.13	-0.35	-0.13	-0.23	-0.22	0.13	0.08	0.05	0.09	-0.03	-0.05	-0.24	-0.42	
BS = 15, 2050	-0.12	-0.33	-0.16	-0.24	-0.17	0.08	0.10	0.03	0.06	-0.06	-0.08	-0.27	-0.47	
BS = 15, 2100	-0.12	-0.35	-0.18	-0.23	-0.19	0.11	0.11	0.01	0.03	-0.15	-0.11	-0.30	-0.47	
NO3- = 10, 2050	-0.07	-0.28	-0.09	-0.07	-0.21	0.05	-0.01	0.22	0.16	0.01	0.12	-0.20	-0.44	
NO3- = 10, 2100	-0.07	-0.28	-0.09	-0.07	-0.21	0.05	-0.01	0.22	0.16	0.01	0.12	-0.20	-0.44	
NO3- = 20, 2050	-0.07	-0.28	-0.09	-0.07	-0.21	0.05	-0.01	0.22	0.16	0.01	0.12	-0.20	-0.44	
NO3- = 20, 2100	-0.07	-0.28	-0.09	-0.07	-0.21	0.05	-0.01	0.22	0.16	0.01	0.12	-0.20	-0.44	

<sup>a</sup> WA:LA – watershed area to lake area ratio.

<sup>b</sup> Depth data were not available for 4 of the 97 lake watersheds.

**Table 3-13. Regression equations to estimate the critical load of sulfur deposition to protect lake water ANC from decreasing below designated critical criteria values in designated future years**

Regressions are based on charge balance ANC determined by the ALS during the 1980s.

Source: Modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.

Critical ANC Criterion ( $\mu\text{eq/L}$ )	Endpoint Year	Equation to Predict Critical Load ( $\text{meq/m}^2/\text{yr}$ )	$r^2$
0	2050	$\text{CL} = 67.9 + 0.79 \text{ ANC}$	0.72
	2100	$\text{CL} = 61.7 + 0.72 \text{ ANC}$	0.80
20	2050	$\text{CL} = 43.4 + 0.85 \text{ ANC}$	0.81
	2100	$\text{CL} = 41.1 + 0.76 \text{ ANC}$	0.86
50	2050	$\text{CL} = 11.8 + 0.85 \text{ ANC}$	0.90
	2100	$\text{CL} = 13.7 + 0.77 \text{ ANC}$	0.92

Inclusion of landscape variables, along with candidate water chemistry variables, did not improve the predictive relationships given in Table 3-13. None of the candidate landscape predictor variables listed in Table 2-1 entered into any of the final regression equations. Standard errors of the equation coefficients are given in Table 3-14. Thus, the MAGIC-simulated CL values could be predicted fairly consistently ( $r^2 = 0.72$  to  $0.92$ ) using only lake water ANC as a predictor variable; inclusion of other water chemistry variables made only inconsequential improvement in the predictive ability of the resulting equations. Inclusion of watershed features such as elevation, slope, watershed area, and/or soil characteristics (pH, percent clay, depth) did not improve CL predictions beyond what was achieved based only on lake ANC.

We also attempted to develop, using only watershed variables, regression equations to predict the MAGIC-simulated CL of S deposition needed to protect lake ANC from decreasing below critical criteria values. Such equations would be useful for estimating CL values at Adirondack locations where measurements of water chemistry are not available. The resulting predictive relationships, with which to predict CL across the landscape based only on landscape variables, were judged to be inadequate. As a consequence, MAGIC model simulations of the CL to protect lake ANC were extrapolated to the population of Adirondack lakes for which there exists water chemistry data.

**Table 3-14. Standard errors of regression coefficients used to predict CL (meq/m<sup>2</sup>/yr) from lake ANC (µeq/L) measured during the 1980s**

Critical ANC Criterion (µeq/L)	Endpoint Year	Variable	Coefficient	Standard Error
0	2050	Constant	67.93	3.68
		ANC	0.79	0.05
	2100	Constant	61.70	2.74
		ANC	0.72	0.04
20	2050	Constant	43.45	3.07
		ANC	0.85	0.04
	2100	Constant	41.06	2.27
		ANC	0.76	0.03
50	2050	Constant	11.82	2.15
		ANC	0.85	0.03
	2100	Constant	13.68	1.69
		ANC	0.77	0.02

We were not able to spatially extrapolate the modeled CLs to watersheds lacking water chemistry data. Numeric extrapolations pertaining to the EMAP population frame were presented in Section 3.4.1. The numeric extrapolations provide estimates of numbers and percentages of lakes in various CL classes, but provide no information regarding where those lakes are located. The spatial extrapolation results shown here allow mapping of CL class locations, but only include the watersheds having water chemistry data that enabled the CL spatial extrapolation. Predicted versus MAGIC-simulated CL values are given in Figure 3-16 for two example critical threshold criteria (ANC = 20 and 50 µeq/L), both for the year 2100. In general, predicted values were within about 20 meq/m<sup>2</sup>/yr of MAGIC modeled values.

The spatial patterns in acid sensitivity are readily apparent in the map depictions of S CLs extrapolated to the population of ALS lakes. These patterns are shown as Maps 3-10 through 3-15, including three critical threshold criteria (ANC = 0, 20, 50 µeq/L) and two endpoint years (2050, 2100). Based on a threshold criterion of ANC = 0 µeq/L (Map 3-10 and Map 3-11), there are relatively few lakes having S CL < 50 meq/m<sup>2</sup>/yr, and those are primarily located in the southwestern portion of the Adirondack Ecoregion, with a second smaller cluster in the high peaks area in the north central region of the park. If the threshold ANC criterion is increased to 20 µeq/L (Map 3-12 and Map 3-13), and especially if it is increased to 50 µeq/L (Maps 3-14 and 3-15), this spatial pattern becomes

even more pronounced. To protect lake ANC to 50  $\mu\text{eq/L}$ , the vast majority of the ALS lakes in the southwestern portion of the Adirondack Ecoregion have S CL less than 50  $\text{meq/m}^2/\text{yr}$ , as do many lakes in the high peaks area (Map 3-14 and Map 3-15).

**Figure 3-16. Comparison between critical load (CL) of sulfur to protect lake ANC, estimated using the regression equations given in Table 3-13, and CL calculated by the MAGIC model for the 97 modeled Adirondack lakes**

Results are shown for the year 2100 based on ANC critical criteria thresholds equal to a) 20  $\mu\text{eq/L}$  and b) 50  $\mu\text{eq/L}$ . Lines indicating 1:1 correspondence are provided for reference.

*Source: Panel B from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. Water Resour. Res. 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.*

**a) ANC = 20  $\mu\text{eq/L}$ , Year 2100**

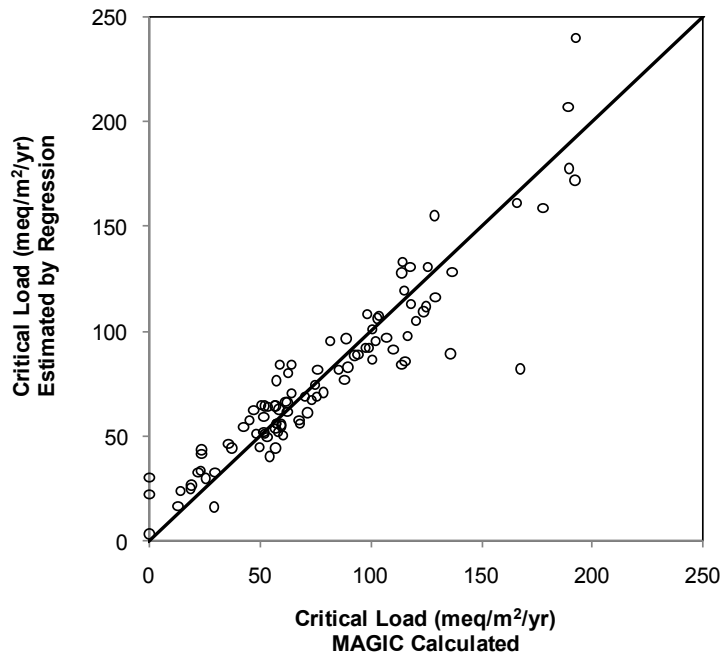
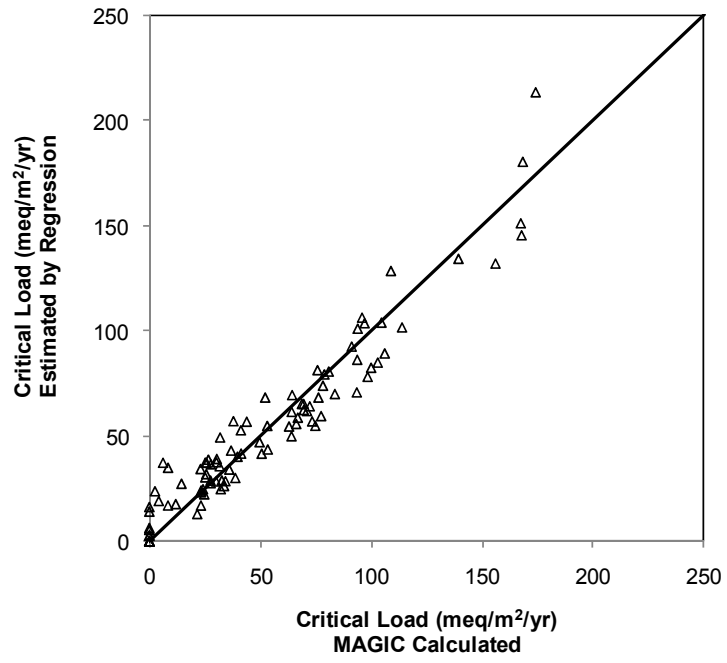


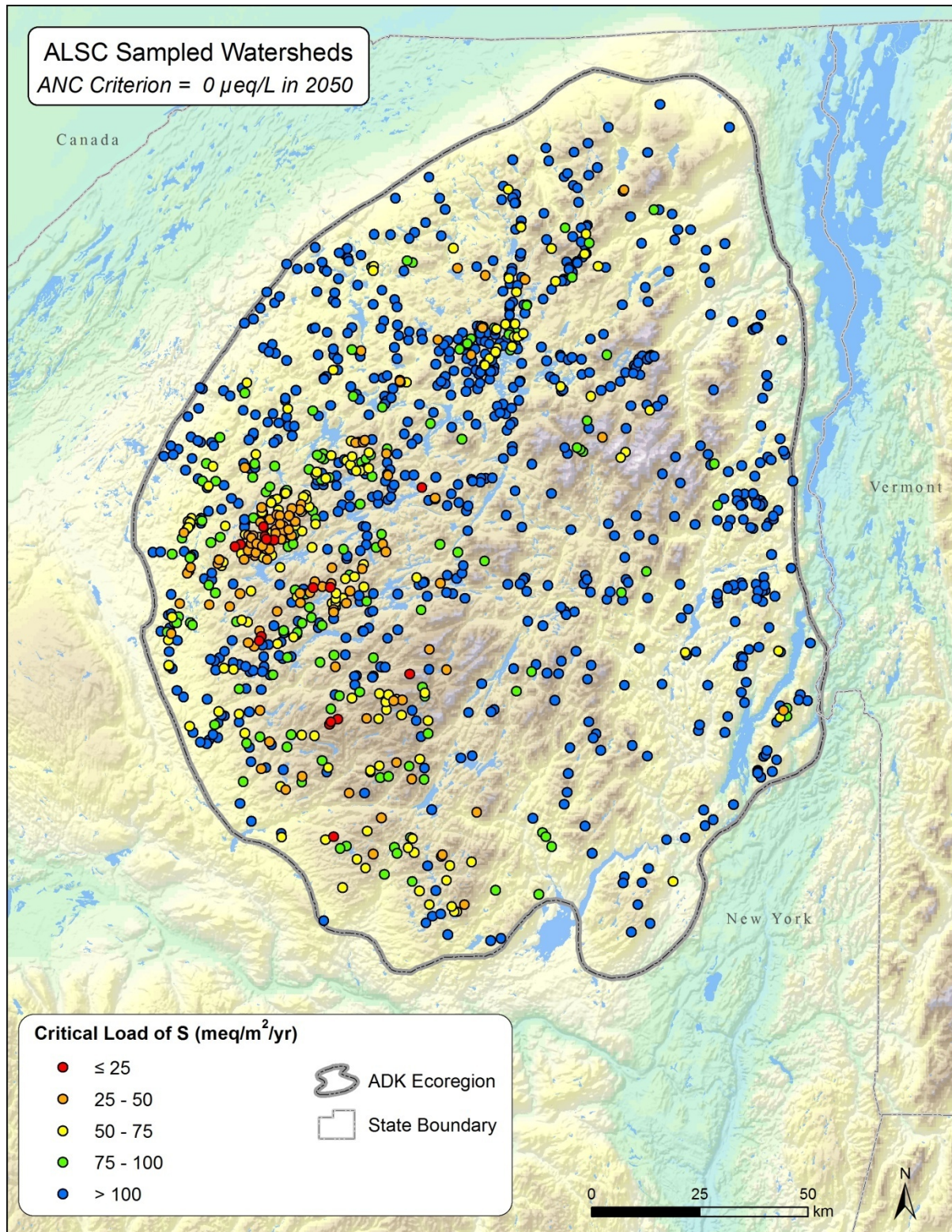


Figure 3-16 continued

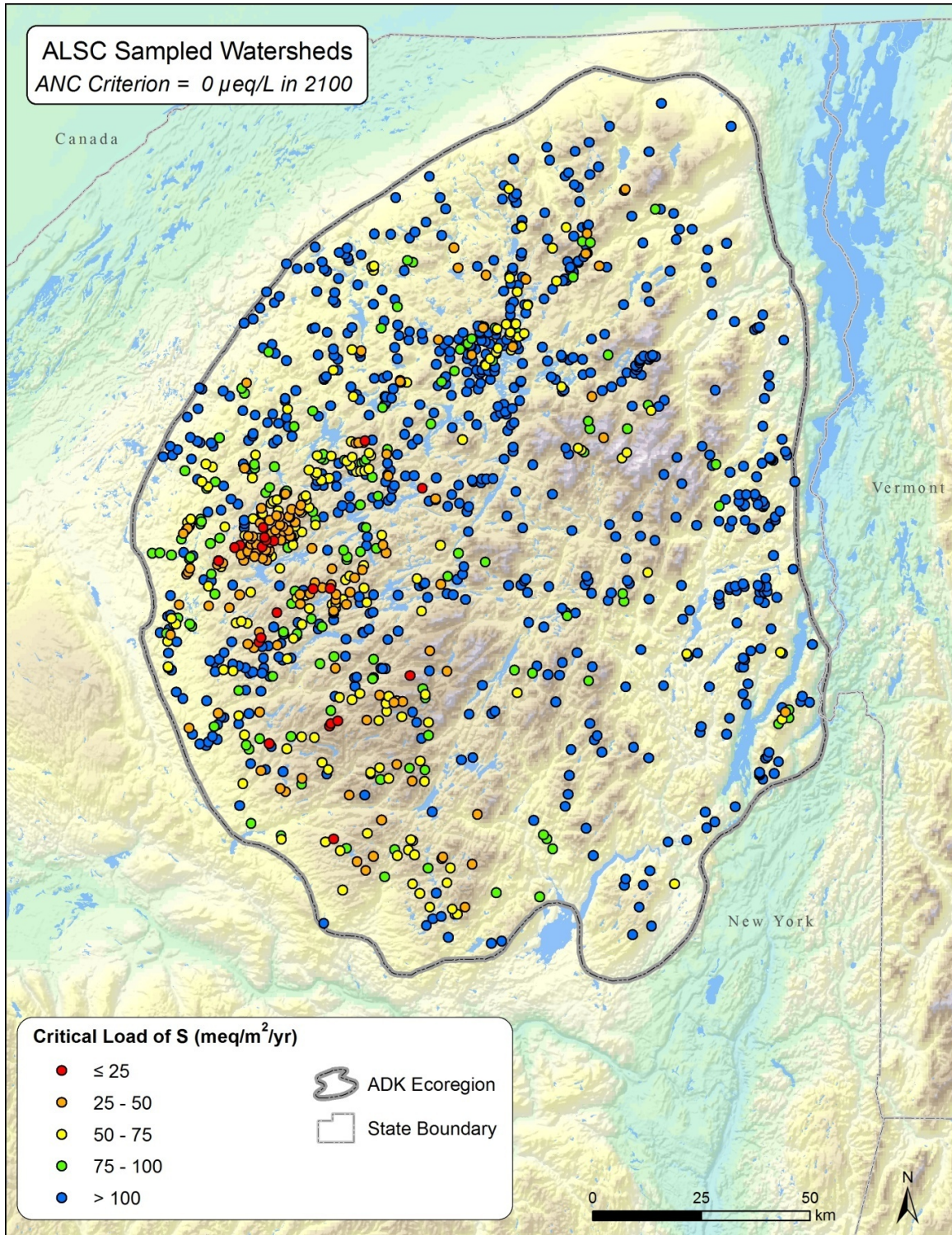
b) ANC = 50  $\mu\text{eq/L}$ , Year 2100



Map 3-10. Estimated critical load of sulfur deposition to protect lake ANC to 0  $\mu\text{eq/L}$  in the year 2050, based on extrapolation of MAGIC modeling results to the population of lakes surveyed in the ALS

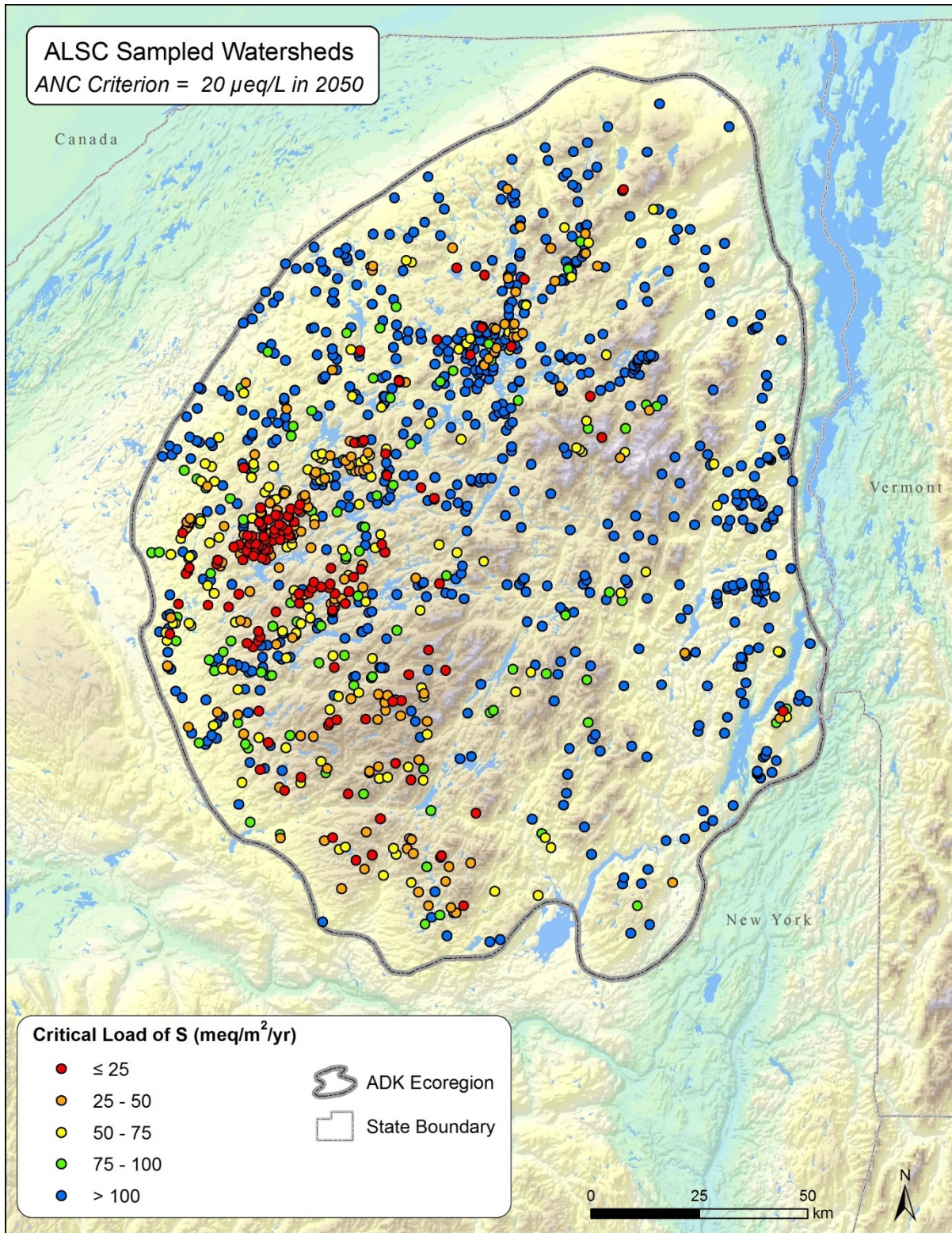


Map 3-11. Estimated critical load of sulfur deposition to protect lake ANC to 0  $\mu\text{eq/L}$  in the year 2100, based on extrapolation of MAGIC modeling results to the population of lakes surveyed in the ALS



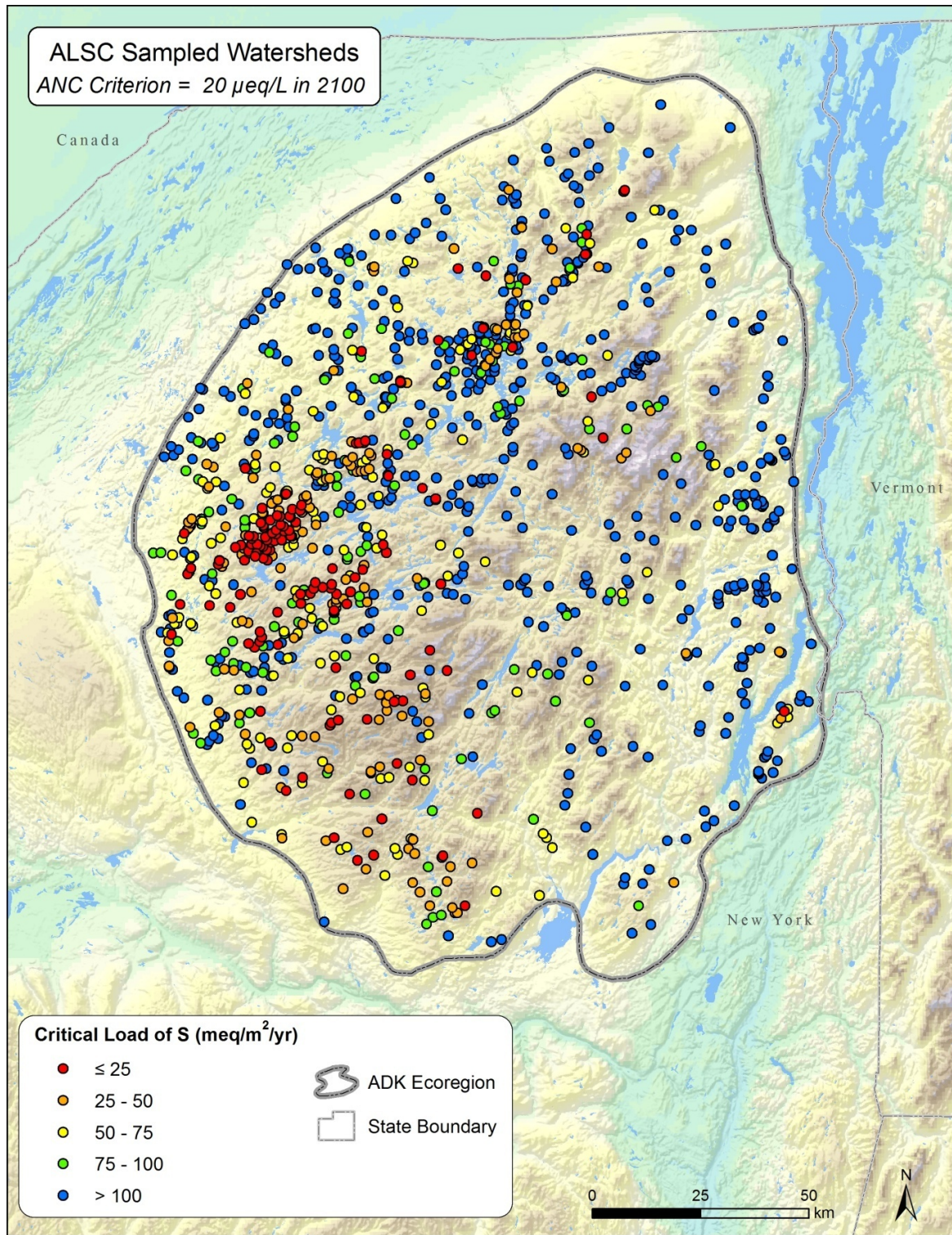
**Map 3-12. Estimated critical load of sulfur deposition to protect lake ANC to 20  $\mu\text{eq/L}$  in the year 2050**

Based on extrapolation of MAGIC modeling results to the population of lakes surveyed in the ALS.



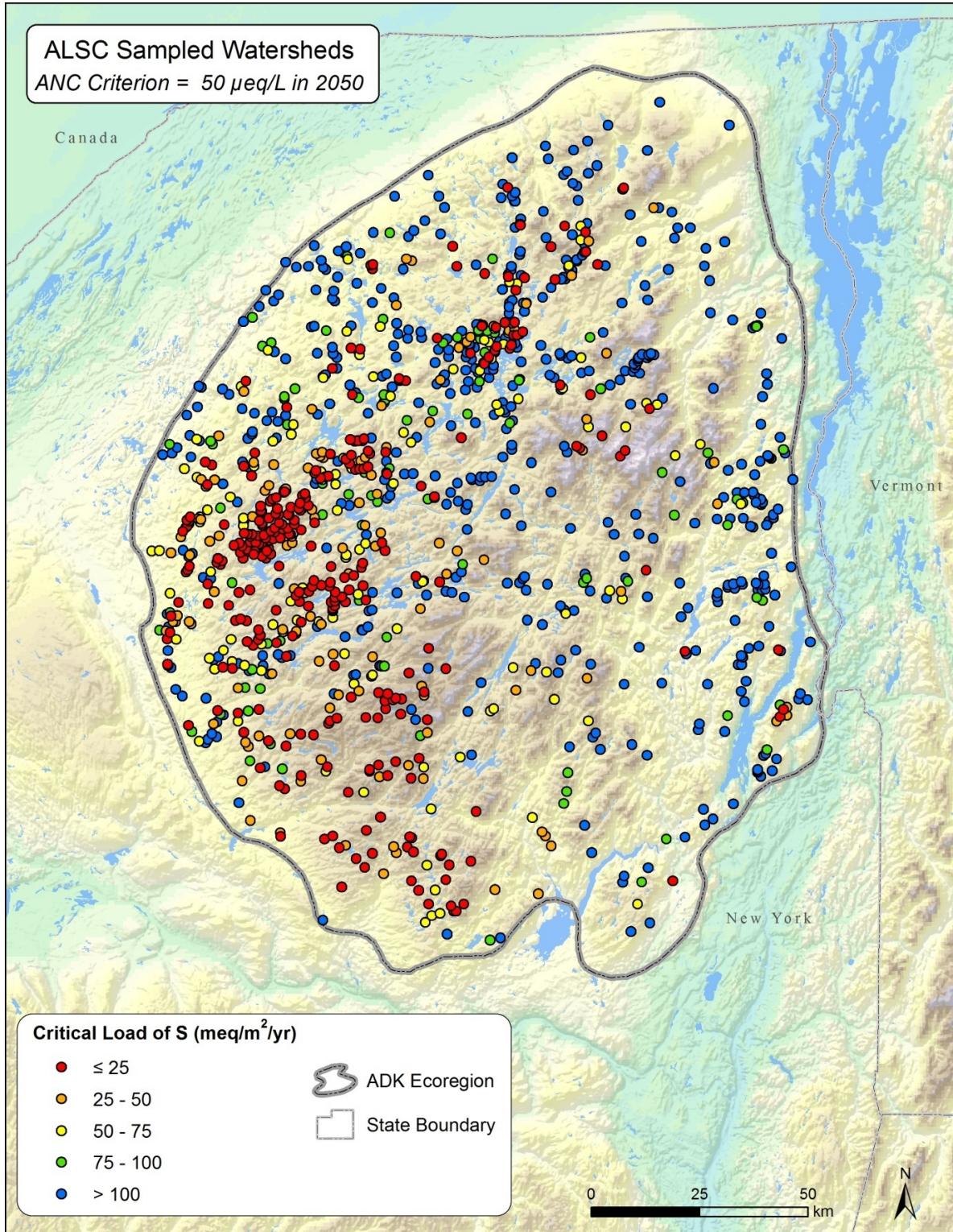
**Map 3-13. Estimated critical load of sulfur deposition to protect lake ANC to 20  $\mu\text{eq/L}$  in the year 2100**

Based on extrapolation of MAGIC modeling results to the population of lakes surveyed in the ALS.



**Map 3-14. Estimated critical load of sulfur deposition to protect lake ANC to 50  $\mu\text{eq/L}$  in the year 2050**

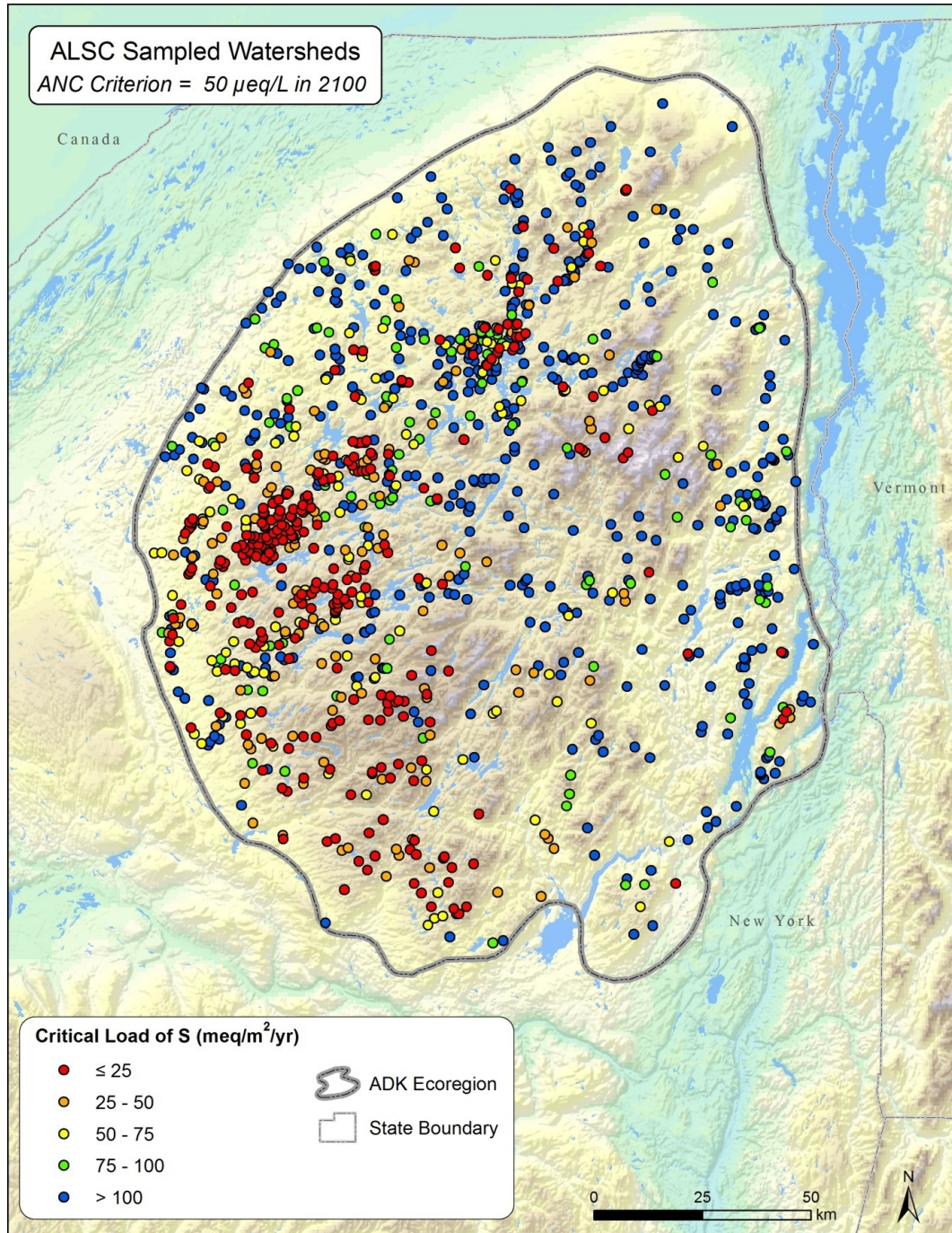
Based on extrapolation of MAGIC modeling results to the population of lakes surveyed in the ALS.



**Map 3-15. Estimated critical load of sulfur deposition to protect lake ANC to 50  $\mu\text{eq/L}$  in the year 2100**

Based on extrapolation of MAGIC modeling results to the population of lakes surveyed in the ALS.

Source: Modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.



The population distributions of the S CLs for the low-ANC EMAP population of lakes and for the ALS surveyed lakes were similar (Figure 3-17). In general, median and quartile CL values were slightly lower for the ALS lake population than for the EMAP population, although the differences were small. Median values of CL to protect lake water to ANC = 50 µeq/L in the year 2100 were similar to ambient deposition loadings, especially for the EMAP lake population.

The lower quartile values of the CL to protect lake ANC to 20 µeq/L for these two large lake populations were also generally similar to ambient S deposition loadings. Efforts to spatially extrapolate to the regional population of lake watersheds the MAGIC-simulated CL needed to protect soil base saturation to critical criteria values were unsuccessful. Neither the lake chemistry nor watershed attributes given in Table 2-1, alone or combined, were sufficient to provide a foundation for spatial extrapolation of soil base saturation CL results. The most robust relationship developed for spatially extrapolating the BS-based CL of S deposition was for protecting BS from going below 5% in the year 2100 (Equation 3-3):

$$\text{CL (BS = 5\%, 2100)} = 922.2 - 167 \times \text{soil pH} + 0.658 \times \text{lake SBC} \quad (r^2 = 0.41) \quad (3-3)$$

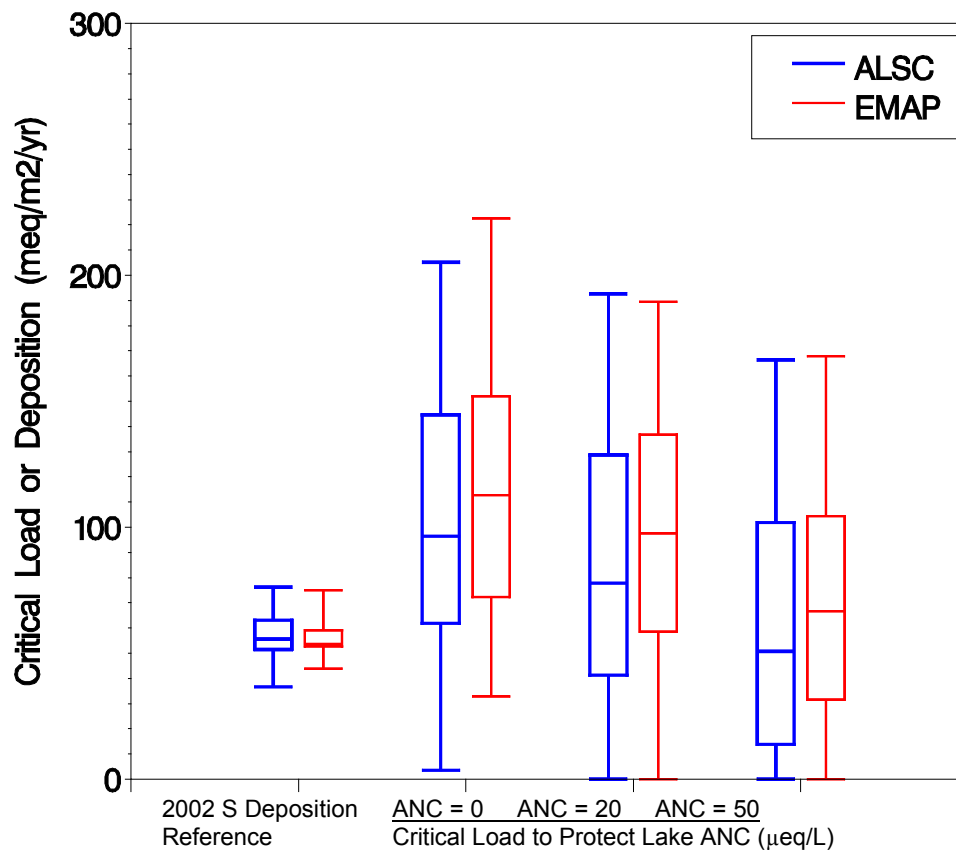
Equation 3-3 explained less than half of the variation in the CL to protect soil BS.

Stepwise linear regression was used in an attempt to establish relationships between CLs to protect soil solution nutrient ratios and landscape/water chemistry predictor variables. Weak relationships with a small set of individual predictor variables exist. The strongest correlation found with any of the soil solution CL endpoints was for Ca/Al = 1 in the year 2100, which could be predicted based on a combination of slope, soil pH, lake NO<sub>3</sub><sup>-</sup> concentration, and lake SBC concentration. Only 29% of the variation in CL was explained by this equation.



**Figure 3-17. Box and whisker plot showing the distribution of MAGIC simulated CL of Sde position in the year 2100 to protect lake ANC to three critical threshold values for two populations of low-ANC ( $\leq 200 \mu\text{eq/L}$ ) Adirondack lakes**

The two populations were the lakes surveyed in the ALS and the EMAP lake population. The box represents the interquartile range (25<sup>th</sup> to 75<sup>th</sup> percentiles), with a horizontal line indicating the median value. Maximum and minimum CL values are represented as upper and lower whiskers. Total ambient S deposition in 2002 is shown for reference perspective.



### 3.4.3 Regional Comparisons

Critical S load percentile values are provided in Table 3-8 for three populations of Adirondack lakes: low-ANC ( $\leq 200 \mu\text{eq/L}$ ) EMAP lakes, all EMAP lakes, and lakes surveyed by the ALSC. The CL values given in Table 3-8 pertain to three different sensitive receptors and criteria: lake ANC (top of table), soil BS (middle of table), and soil solution ratios of base cations to Al (bottom of table). The CL percentile estimates for protecting lake ANC differ among the three lake populations. Results for the population of all EMAP lakes are generally similar, however, to percentiles obtained from the spatial extrapolation of MAGIC results to the ALS surveyed lakes. These data are shown schematically in Figure 3-18 for the three selected ANC threshold criteria applied to the year 2100.

Critical load results to protect soil BS are only available for the two EMAP lake watershed frames; we were unsuccessful in spatially extrapolating MAGIC model CL results for soil protection to the ALS population using surveyed water chemistry and/or landscape data. Although there is a relationship between lake ANC and the BS of the soil within a given lake watershed, that relationship is not sufficiently robust as to provide a mechanism for extrapolating a CL to protect soil chemistry based on measured lake chemistry. Rather, the observed relationship between lake ANC and soil BS suggests that lake ANC below 20  $\mu\text{eq/L}$  can only be achieved in a watershed that contains soil having B-horizon BS below about 12%. The presence of low BS is not sufficient, on its own, to guarantee low lake ANC (Figure 3-8). Thus, low soil BS is a necessary, but not sufficient, condition for obtaining low lake ANC. Critical load results to protect soil solution base cation to Al ratios (Ca:Al and Bc:Al) were similarly only available for the EMAP population frame.

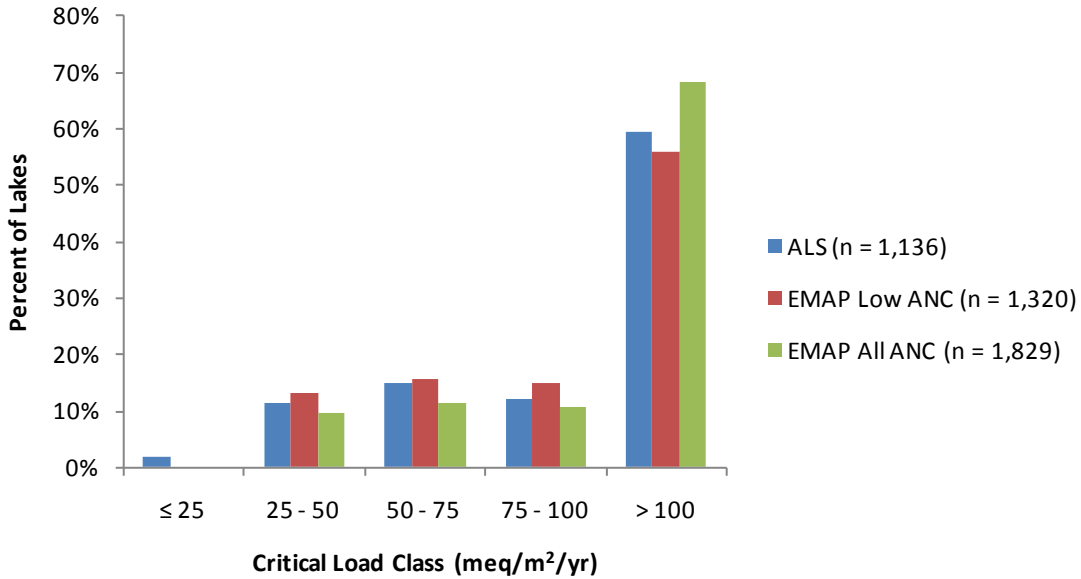
#### **3.4.3.1 Relationship between Critical Load and Watershed Area**

Estimated CL values for the 1,136 ALS lakes larger than 1 ha varied to some extent by watershed area, although not enough to use watershed area to successfully predict CL. Lakes estimated to have low CL tended to have smaller (especially  $< 1 \text{ km}^2$ ) watershed areas. This pattern was evident for the lowest CL class ( $< 25 \text{ meq/m}^2/\text{yr}$ ) for CL estimates based on the critical ANC criterion 50  $\mu\text{eq/L}$  (Figure 3-19a). It was also evident for the two lowest CL classes ( $< 25$ ,  $25\text{-}50 \text{ meq/m}^2/\text{yr}$ ) based on the critical ANC criterion 20  $\mu\text{eq/L}$  (Figure 3-19b). Lakes estimated to have relatively high CL ( $> 100 \text{ meq/m}^2/\text{yr}$ ) assuming both the 20 and 50  $\mu\text{eq/L}$  critical ANC criterion threshold values were disproportionately located in larger watersheds (Figure 3-19).

**Figure 3-18. Histograms showing the distributions of S critical load values for three lake populations (ALS lakes, all EMAP lakes, and EMAP low-ANC ( $\leq 200 \mu\text{eq/L}$ ) lakes**

CL values are given for critical ANC threshold criteria of a) 0  $\mu\text{eq/L}$ , b) 20  $\mu\text{eq/L}$ , and c) 50  $\mu\text{eq/L}$ , all in the year 2100.

**a) ANC = 0, Year 2100**



**b) ANC = 20, Year 2100**

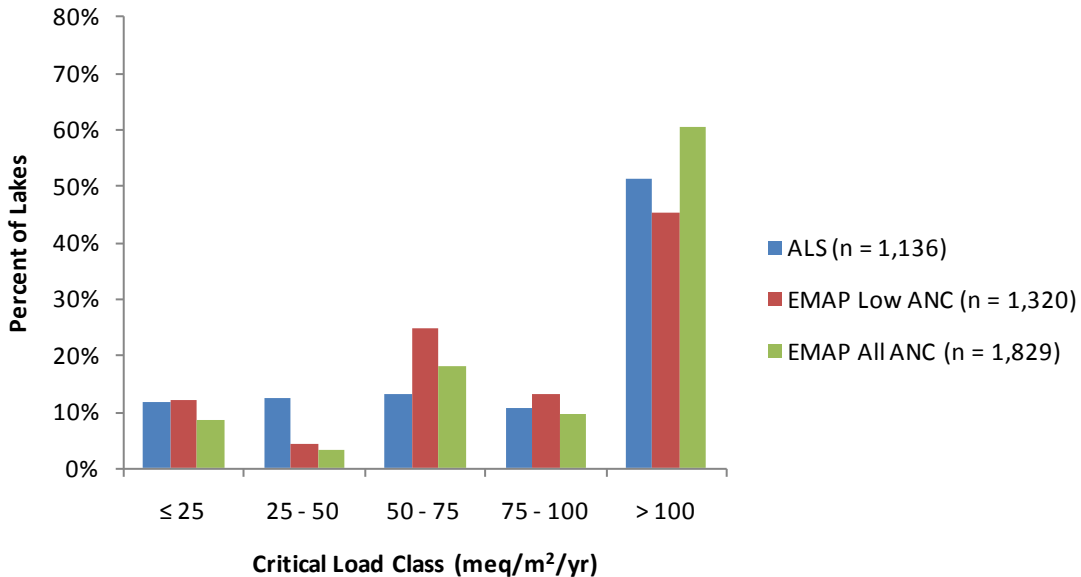
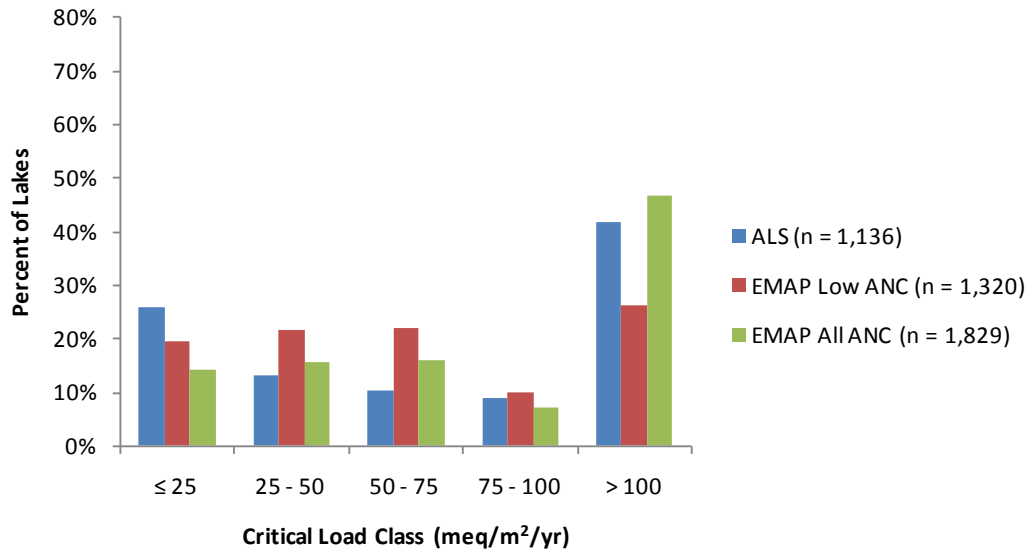


Figure 3-18 continued

c) ANC = 50, Year 2100

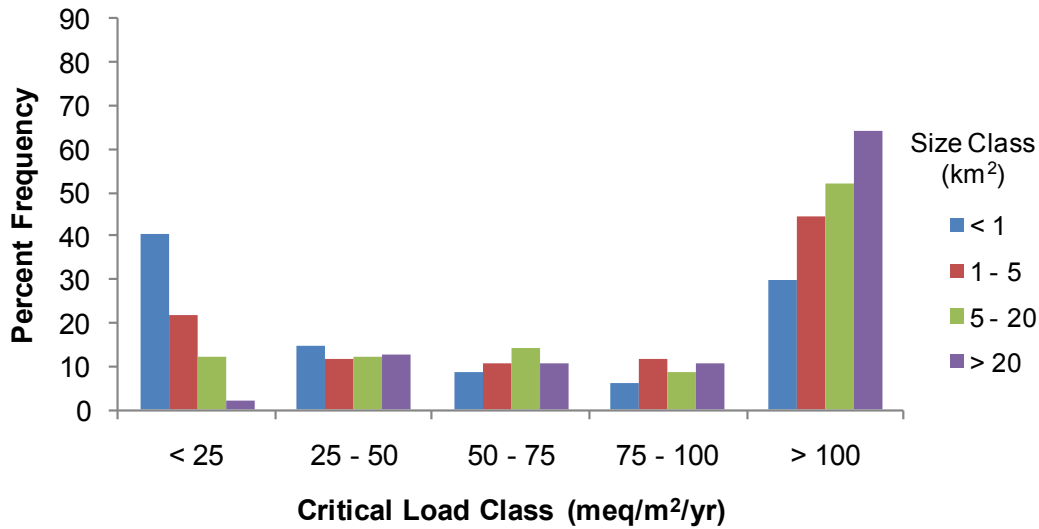


**Figure 3-19. Histogram showing the distribution of ALS lake (n=1,136) watershed areas within critical load classes.**

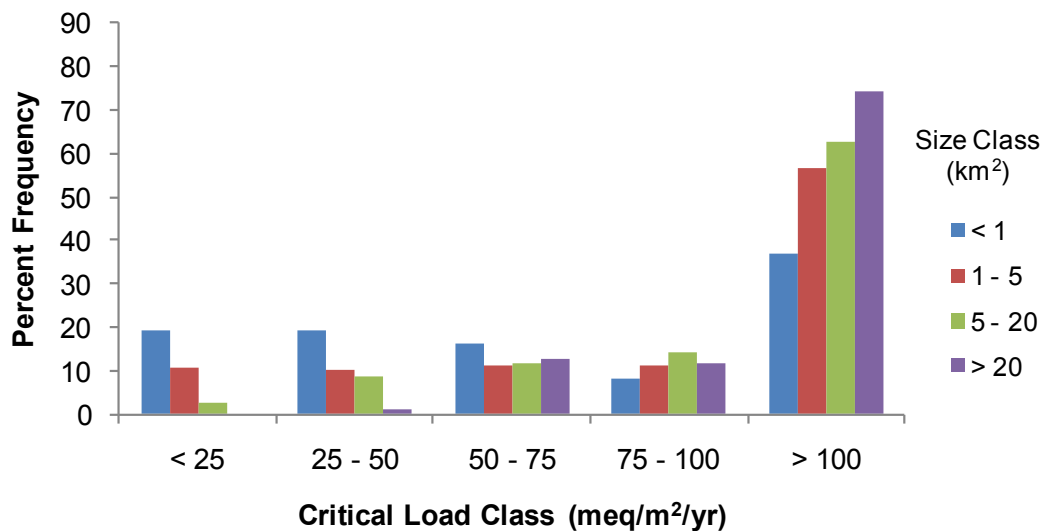
Panel a) shows results based on a critical ANC criterion equal to 50  $\mu\text{eq/L}$ ; Panel b) shows results based on 20  $\mu\text{eq/L}$ . Critical loads for both were estimated for the year 2100. Lakes that had low estimated (modeled and extrapolated) critical load values were disproportionately situated in smaller watersheds; lakes that had high estimated critical load values were disproportionately situated in larger watersheds.

Source: Panel A, modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.

**a) ANC 50  $\mu\text{eq/L}$ , Year 2100**



**b) ANC 20  $\mu\text{eq/L}$ , Year 2100**



### 3.5 Time Frame of Critical Load Responses

The modeled CL values varied with selection of endpoint year (Figure 3-20). For the most acid-sensitive lake watersheds (i.e., those having CL less than about 25 to 50 meq/m<sup>2</sup>/yr), the CL to protect sensitive resources was higher for protection to the year 2100, as compared with 2050. In other words, these most acid-sensitive watersheds were simulated to be able to tolerate slightly higher S loading if one was willing to wait an additional 50 years for the resources to recover. Lower deposition levels would be needed in order to achieve recovery in a shorter time period. For the majority of the modeled watersheds, however, the S CL was higher than 25 to 50 meq/m<sup>2</sup>/yr, and for these less acid-sensitive watersheds the S CL was lower using an endpoint year of 2100. Thus, these watersheds can tolerate lower S loading if the resource protection is intended to extend all the way to 2100, as opposed to only protecting the resources to 2050. The effect of endpoint year designation therefore depends on the current acid-base status of the watershed.

The effect of CL timeframe was more pronounced for protecting soil BS (Figure 3-20b) than it was for protecting lake ANC (Figure 3-20a). However, these larger differences were found for the watersheds having relatively high CL, especially those above about 200 meq/m<sup>2</sup>/yr. The pattern was similar to that found for CLs based on protecting lake ANC. For watershed soils having very low soil BS (left side of graph), a lower CL would be required to affect recovery within a shorter time period. For the more acid-sensitive watersheds having lower CL, the effect of waiting the additional 50 years, to the year 2100, for resource protection was fairly modest (left side of Figure 3-20b). For watershed soils having higher soil BS (right side of Figure 3-20b), a lower CL would be required to continue resource protection for a longer time period.

Selection of endpoint year had relatively little effect on the N CLs for most of the modeled lake watersheds (Figure 3-21). For a few modeled watersheds, however, the calculated CL was strongly influenced by the selection of endpoint year, especially for some of the calculations to protect soil BS.

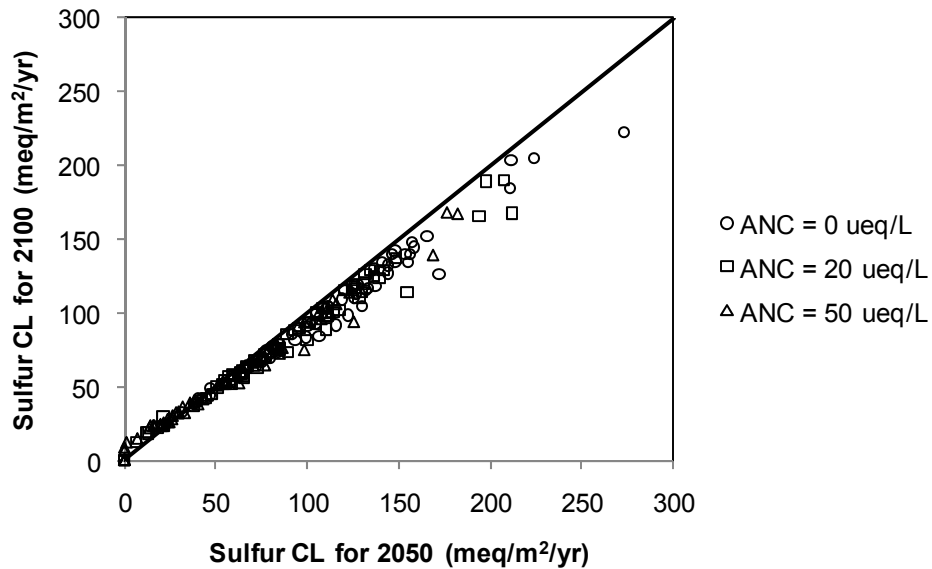
Critical loads of S deposition were also simulated for the steady state condition, based on the MAGIC input data files, and using the Steady State Water Chemistry (SSWC) model formulation. For the lakes having lowest CL values (less than about 25 meq/m<sup>2</sup>/yr) the long-term steady state CL was generally higher than the dynamic CL, or TL, that was formulated to protect lake ANC in 2100 (Figure 3-22). Those lakes showing such low CL values were generally the lakes that currently have lowest ANC. The longer-term steady state CL is higher than the shorter-term dynamic CL for these lakes; this indicates that these lake watersheds can tolerate more S deposition if one is willing to wait longer into the future (to the steady state condition) to affect recovery to the various ANC targets, mainly 20 and 50 µeq/L. In contrast, the lakes that show higher dynamic S CL to 2100, especially those higher than about 50 meq/m<sup>2</sup>/yr, show lower steady state CL as compared with dynamic CL. (Note that essentially all data points showing CL >50 meq/m<sup>2</sup>/yr are below the 1:1 line in Figure 3-22.) To protect those lakes to the critical criteria limits in the long term, deposition will have to be reduced below the values needed to only protect them to the year 2100.

**Figure 3-20. Comparison of critical sulfur loads to achieve resource protection in 2050 versus 2100**

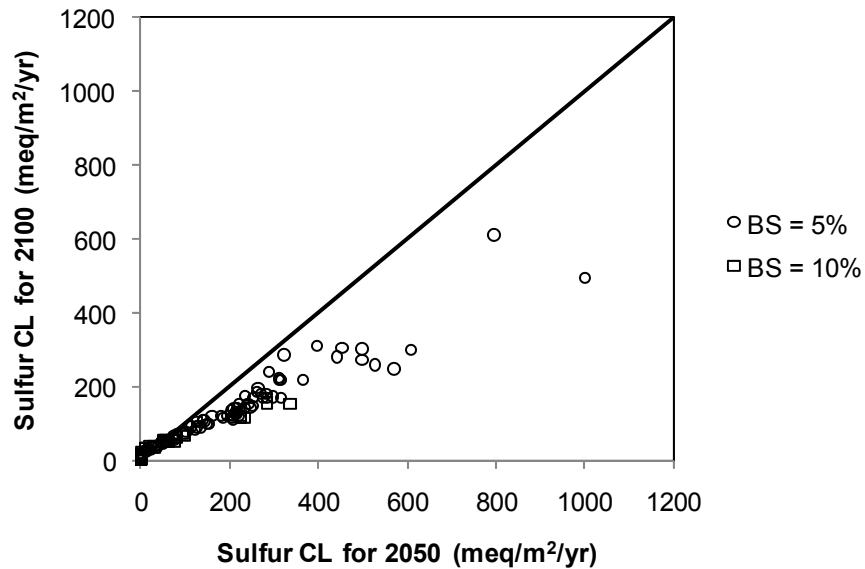
Critical loads were calculated with MAGIC for 70 lake watersheds to protect a) lake ANC and b) soil base saturation.

Source: Panel A, modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.; Panel B, modified from Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. *J. Environ. Stud. Sci.* 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.

**a) Critical Sulfur Load to Protect Lake ANC**



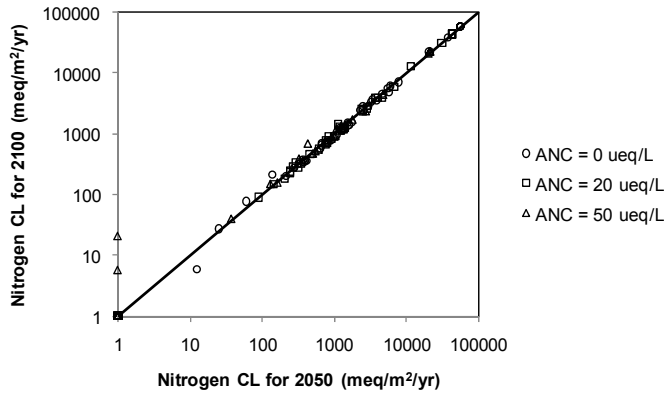
**b) Critical Sulfur Load to Protect Soil Base Saturation**



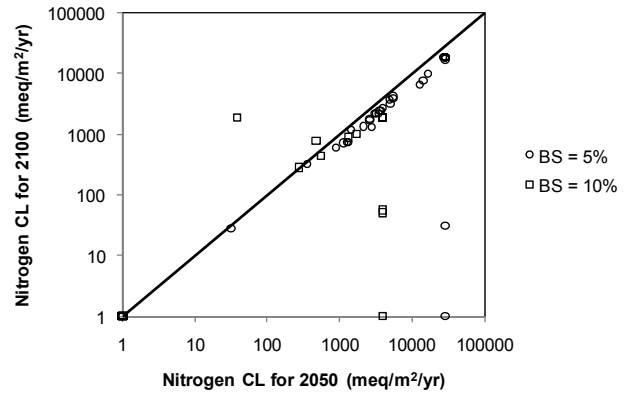
**Figure 3-21. Comparison of critical nitrogen loads to achieve resource protection in 2050 versus 2100**

Critical loads were calculated with MAGIC for 70 lake watersheds to protect a) lake ANC, b) soil base saturation, and c) lake nitrate concentration.

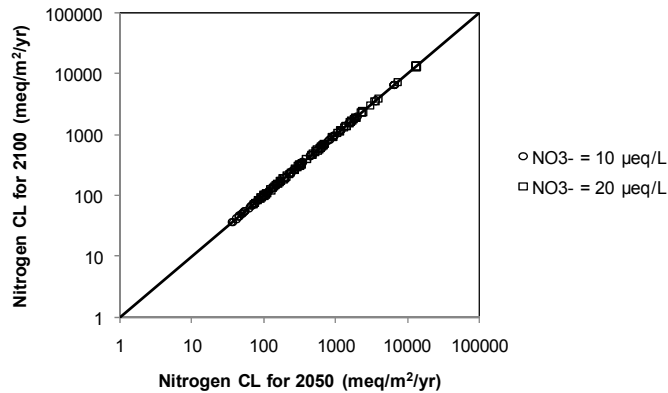
**a) Lake ANC**



**b) Soil Base Saturation**



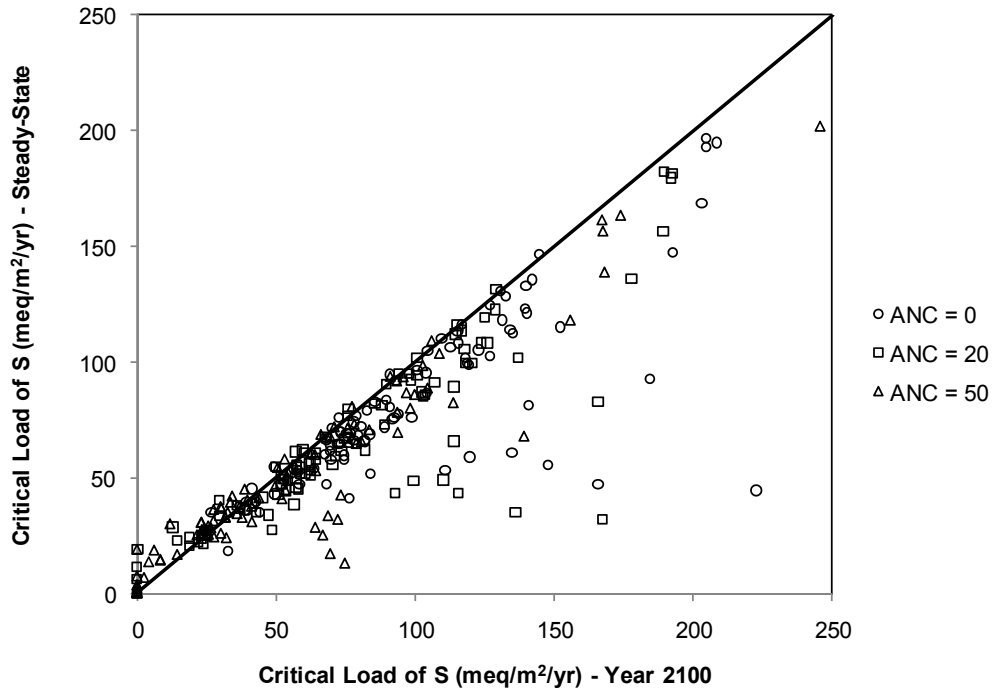
**c) Lake Nitrate Concentration**





**Figure 3-22. Critical loads of S deposition to achieve three critical ANC target values in the year 2100 (x-axis) versus upon reaching steady state conditions in the future (y-axis)**

Simulations to 2100 were constructed using MAGIC, and the steady state CL values were calculated using the SSWC model. A 1:1 reference line is provided



### 3.6 Critical Load Exceedance

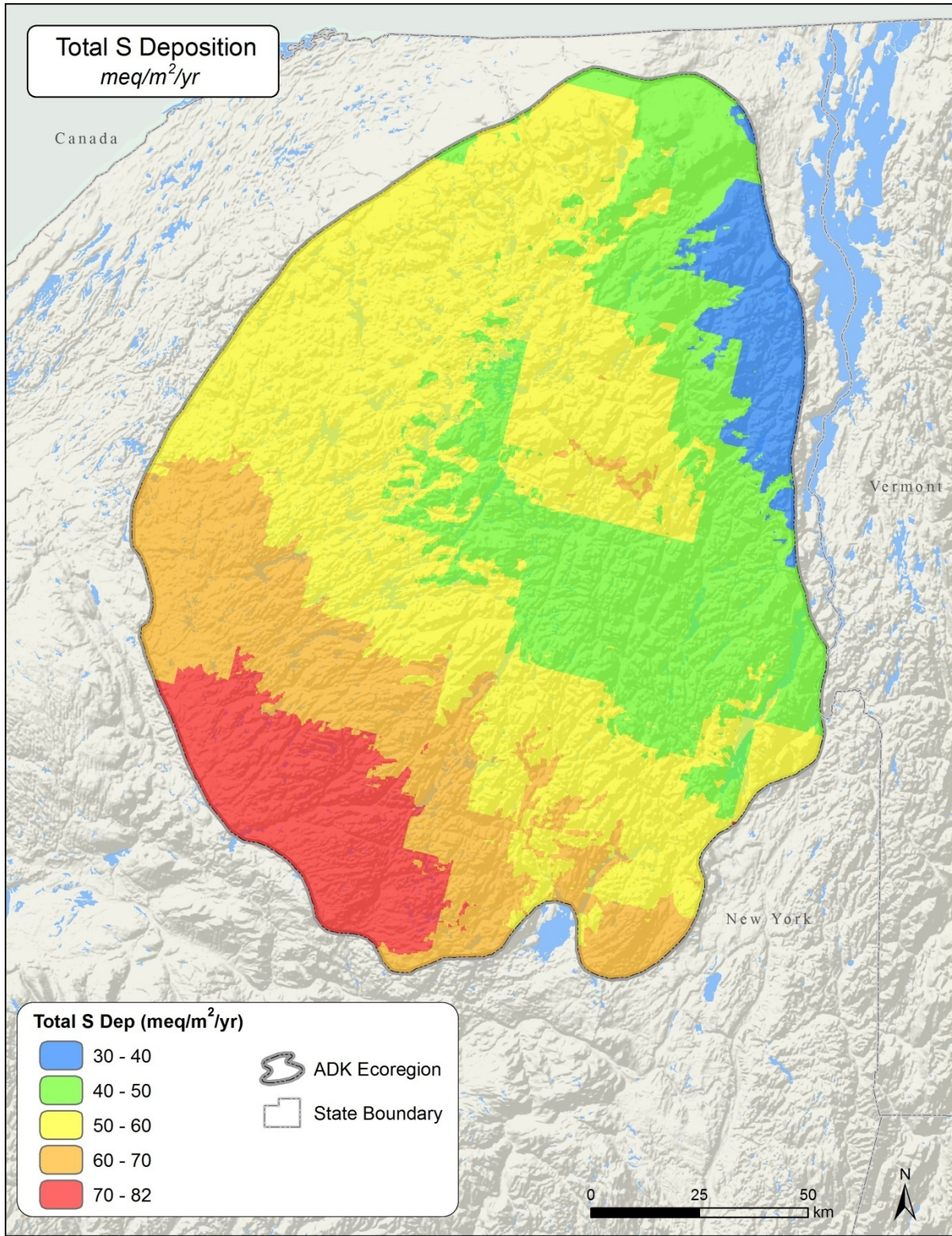
Model simulations of CL, and extrapolation of those simulation results to the regional EMAP and/or ALS populations of Adirondack lakes, provide important information regarding the atmospheric deposition levels of either S or N that various lake watersheds can tolerate without exceeding critical threshold criteria believed to be associated with biological harm. These simulated CL values on their own do not reveal whether or not ambient deposition levels are sufficiently high as to exceed those thresholds and actually cause biological harm. That determination is made using the exceedance calculation. Exceedance is calculated as the difference between the current ambient deposition of S or N and the CL for S or N. Thus, exceedance reflects the extent to which the current loading does or does not exceed the threshold criterion value at which biological harm might be expected.

Regional estimates of current (average centered on the year 2002) total wet plus dry S and N deposition are shown in Map 3-16 and Map 3-17. Current deposition levels of S and N are generally similar. Deposition values are highest in the southwestern portion of the Adirondack Ecoregion and lowest to the northeast. These deposition estimates were used to calculate CL exceedance by comparing ambient deposition with CL estimates.

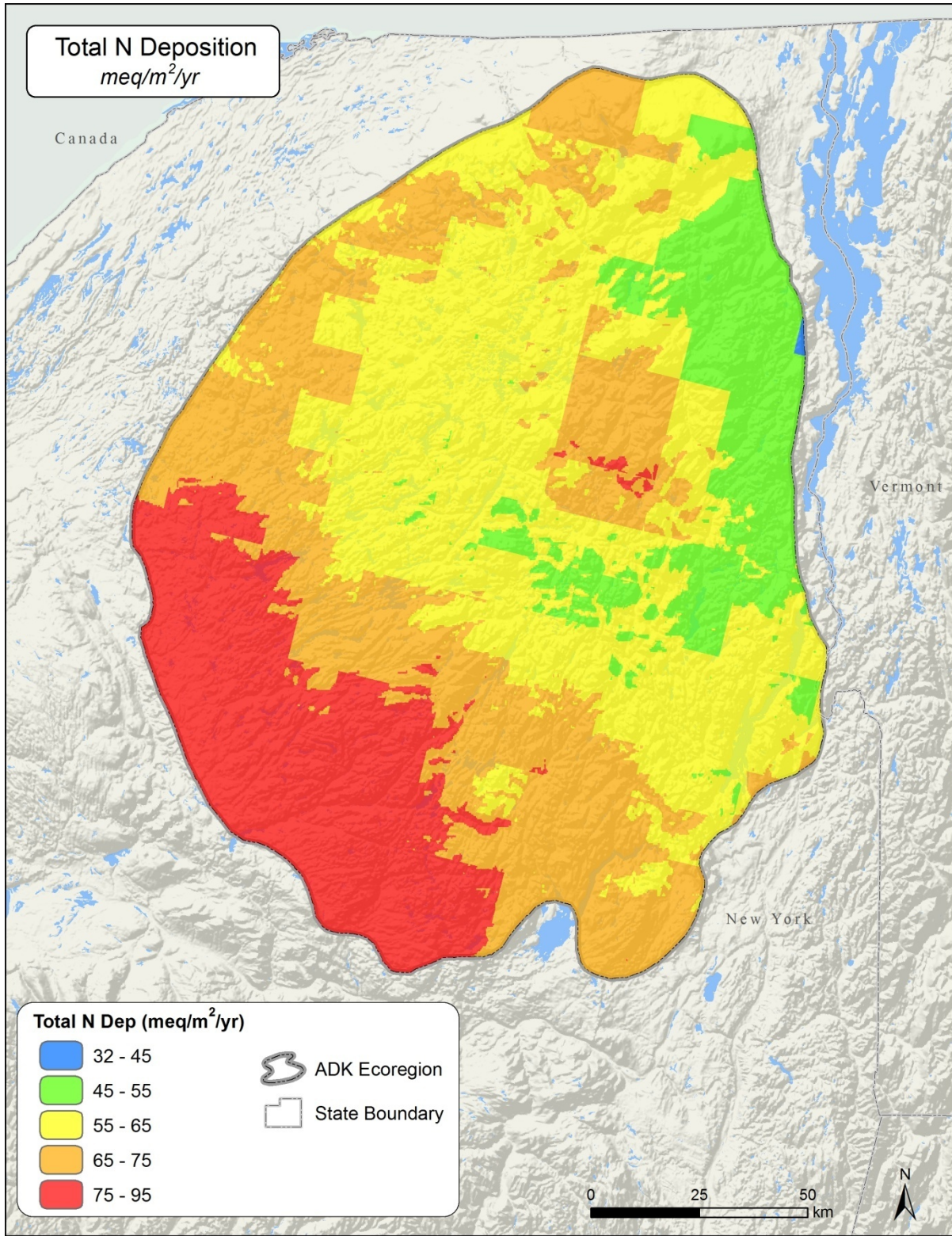
The number and percentage of Adirondack lakes that currently (estimated for the year 2002) receive ambient S deposition above their respective CL are reported in Table 3-15. Results are organized by sensitive criterion, assumed critical threshold value, and endpoint year and are referenced to the EMAP population of 1,320 low-ANC Adirondack lakes. For protecting lake water ANC, the percent of lakes projected to be in exceedance based on deposition in the year 2002 ranged from 15.5% to protect to ANC = 0  $\mu\text{eq/L}$  to 46.3% to protect to ANC = 50  $\mu\text{eq/L}$ ; the estimate for protection to ANC = 20  $\mu\text{eq/L}$  was intermediate (22.7%; Table 3-15). Comparable calculations for the year 2050 yielded slightly lower estimates of the number and percentage of lakes in exceedance.

Only 13.5% (for the year 2100) and 11.6% (year 2050) of these low-ANC EMAP Adirondack lake watersheds were simulated to be in exceedance of the S CL to protect soil BS to 5%. If a more protective critical threshold value for BS is assumed (BS = 10% or 15%), many additional watersheds are estimated to be in exceedance. Again, the simulated percent of watersheds in exceedance is slightly higher for the endpoint year 2100 as compared with 2050 and the percent in exceedance is highest for the more restrictive critical value of BS = 15% (95 to 98% of the low-ANC EMAP lakes currently in exceedance), as compared with a critical threshold value of 10% (78.5% to 79.7% currently in exceedance; Table 3-15).

**Map 3-16. Estimated total wet plus dry atmospheric deposition of sulfur for the five-year period centered on 2002 across the Adirondack Ecoregion**



Map 3-17. Estimated total wet plus dry atmospheric deposition of nitrogen for the five-year period centered on 2002 across the Adirondack Ecoregion



**Table 3-15. Estimated number of Adirondack lakes simulated by the MAGIC model to be in exceedance of the critical load of sulfur deposition**

Based on model simulations of 44 statistically selected lakes from the EMAP population of 1,320 Adirondack lakes that have ANC less than 200 µeq/L.

*Source: Modified from Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. J. Environ. Stud. Sci. 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.*

Receptor	Sensitive Criterion	Critical Value	Number (and percent) of Lakes in Exceedance in 2002 Based on Critical Load Endpoint Year	
			2050	2100
Lake water	ANC	0 µeq/L	191 (14.5)	204 (15.5)
		20 µeq/L	243 (18.4)	299 (22.7)
		50 µeq/L	581 (44.0)	611 (46.3)
Soil	BS	5%	153 (11.6)	178 (13.5)
		10%	1036 (78.5)	1052 (79.7)
		15%	1253 (94.9)	1296 (98.1)
Soil Solution	B <sub>c</sub> :Al	1	103 (7.8)	103 (7.8)
		10	1296 (98.1)	1296 (98.1)
Soil Solution	Ca:Al	1	582 (44.1)	768 (58.2)
		10	1296 (98.1)	1296 (98.1)

Roughly half (44.1% for the year 2050 and 58.2% for the year 2100) of the low-ANC EMAP lake watersheds were estimated to be in exceedance to protect soil solution Ca:Al to 1, whereas nearly all of the watersheds (98.1%) were in exceedance to protect to a ratio value of 10. Similarly, nearly all (98.1%) of the EMAP low-ANC lake watersheds were calculated to be in exceedance to protect soil solution B<sub>c</sub>:Al = 10. Very few (7.8% of the EMAP low-ANC lake watersheds) were in exceedance to protect to B<sub>c</sub>:Al = 1. Thus, the B<sub>c</sub>:Al ratios of 1 and 10 are not effective thresholds for discriminating exceedance of S CLs. If a critical threshold criterion of 1 is assumed, very few watersheds are estimated to be in exceedance; if a critical threshold criterion of 10 is assumed, nearly all watersheds are estimated to be in exceedance under ambient deposition loading rates.

A breakdown of exceedance classes for S deposition is provided in Table 3-16. The percent of low-ANC Adirondack lakes in the EMAP frame judged to not be in exceedance ranged from 1.9% (to protect soil BS to 15% and to protect either soil solution Ca:Al or Bc:Al to 10) to 92.2% (to protect soil solution Bc:Al to 1). Selection of the sensitive criterion and its associated critical threshold value have considerable influence on the resulting CL exceedance calculations. Some lakes and their watersheds receive ambient deposition that is more than double the respective CL (Table 3-16). This is especially true for protecting soil BS to 15%, soil solution Ca:Al ratio to 10, and soil solution Bc:Al ratio to 10.

**Table 3-16. Lake watersheds within the Adirondack study region in S deposition CL exceedance classes**

Based on MAGIC model simulations for 44 statistically selected lakes, using the endpoint year 2100.

*Source: Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. Water Resour. Res. 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.; and Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. J. Environ. Stud. Sci. 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.*

Receptor	Sensitive Criterion (µeq/L)	Critical Value	Number (and Percent) of Lakes within Exceedance Class			
			Not in Exceedance	1.0 to 1.5 Times the CL	1.5 to 2.0 Times the CL	> 2.0 <sup>a</sup> Times the CL
Lake water	ANC	0 µeq/L	1116 (84.5)	79 (6.0)	125 (9.5)	0 (0)
		20 µeq/L	1021 (77.4)	108 (8.2)	16 (1.2)	175 (13.3)
		50 µeq/L	710 (53.7)	221 (16.7)	103 (7.8)	287 (21.7)
Soil	BS	5%	1142 (86.5)	63 (4.7)	62 (4.7)	54 (4.1)
		10%	269 (20.3)	31 (2.4)	147 (11.1)	873 (66.2)
		15%	25 (1.9)	42 (3.2)	16 (1.2)	1237 (93.7)
Soil Solution	Bc:Al	1	1217 (92.2)	25 (1.9)	0 (0)	78 (5.9)
		10	25 (1.9)	16 (1.2)	28 (2.1)	1252 (94.8)
Soil Solution	Ca:Al	1	552 (41.8)	463 (35.1)	37 (2.8)	268 (20.3)
		10	25 (1.9)	0 (0)	0 (0)	1296 (98.1)

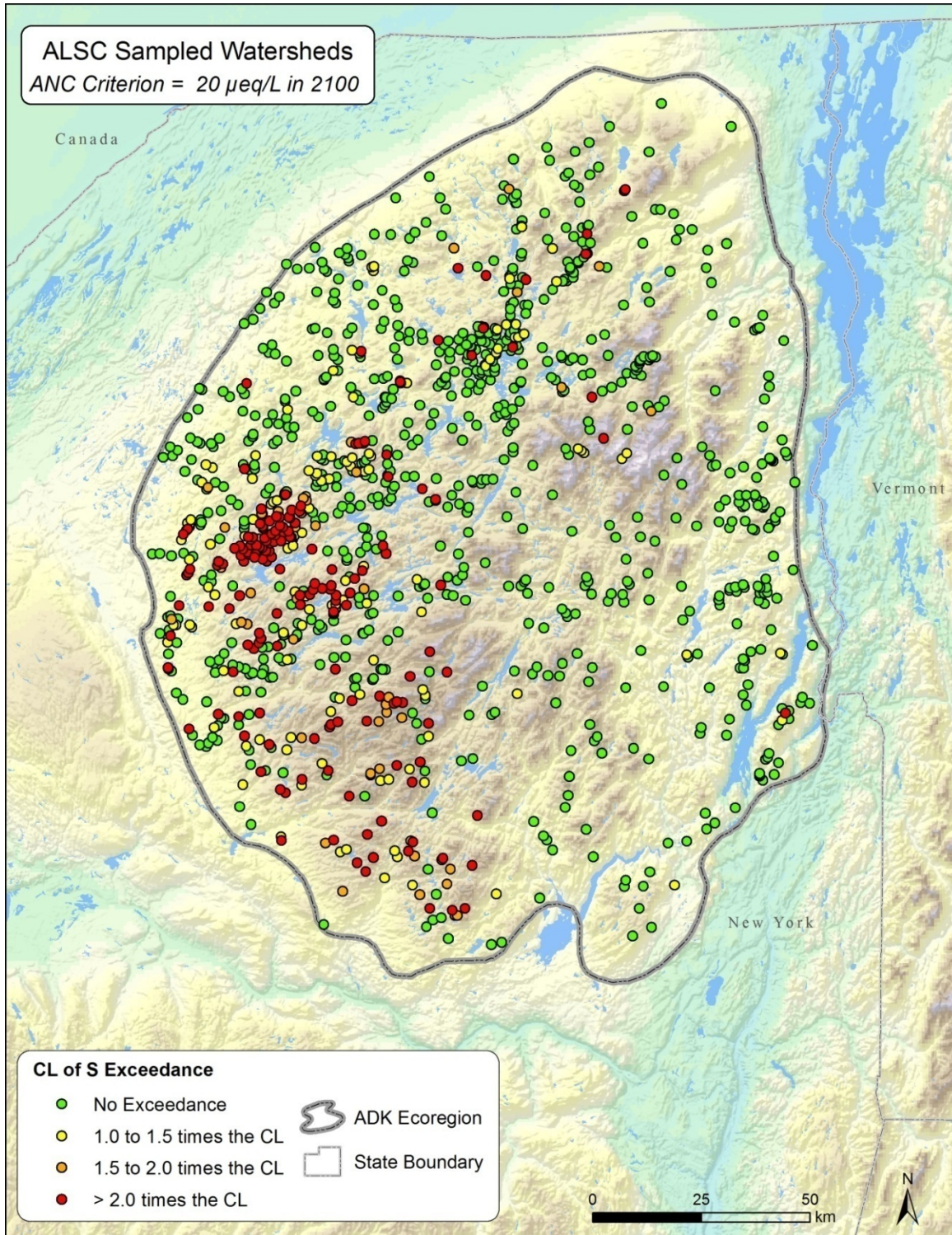
<sup>a</sup> For lakes simulated to have CL=0, the exceedance class was set to 2.0 times the CL

The number of low-ANCEMAP lakes estimated to receive ambient S deposition in exceedance of the CL to protect lake ANC varied with the critical ANC threshold criterion selected (Table 3-16). For protecting lake ANC to 0 µeq/L by the year 2100, 204 lakes (15.5%) are currently in exceedance based on estimated ambient S deposition in 2002. To protect lake ANC to 20 µeq/L, 299 lakes (22.7%) are in exceedance. To protect lake ANC to 50 µeq/L, 611 lakes (46.2%) are in exceedance, and nearly half of those receive ambient deposition that is greater than two times the CL.

Extrapolated exceedance classes are mapped in Map 3-18 and Map 3-19 for 1,136 ALS lakes based on ANC critical thresholds equal to 20 and 50 µeq/L for the year 2100. Lakes experiencing atmospheric S deposition more than double their respective CLs are broadly distributed throughout the southwestern Adirondack Mountains and the High Peaks region in the north central portion of the Adirondack Ecoregion. The number of ALS lakes (n=1,136) estimated to be in exceedance of their S CL for lake ANC show a similar pattern to the EMAP low-ANC population frame (Figure 3-23). To protect lake ANC to 0 µeq/L by the year 2100, 213 lakes (19%) are estimated to be in exceedance based on S deposition in 2002. To protect lake ANC to 20 µeq/L by 2100, 340 ALS lakes (30%) are estimated to be in exceedance. To protect lake ANC to 50 µeq/L, nearly one-third (326) of the ALS lakes are estimated to receive current S deposition greater than two times the CL, with a total of 482 (42%) lakes in exceedance. The numbers of ALS lakes estimated to be in exceedance were slightly lower for the endpoint year 2050 (Figure 3-23a), compared with 2100 (Figure 3-23b).

**Map 3-18. Exceedance classes for 1,136 Adirondack lakes based on extrapolation of MAGIC model results of S CLs to the ALS surveyed lakes**

Exceedances were calculated for the year 2100 using a critical threshold ANC value of 20  $\mu\text{eq/L}$

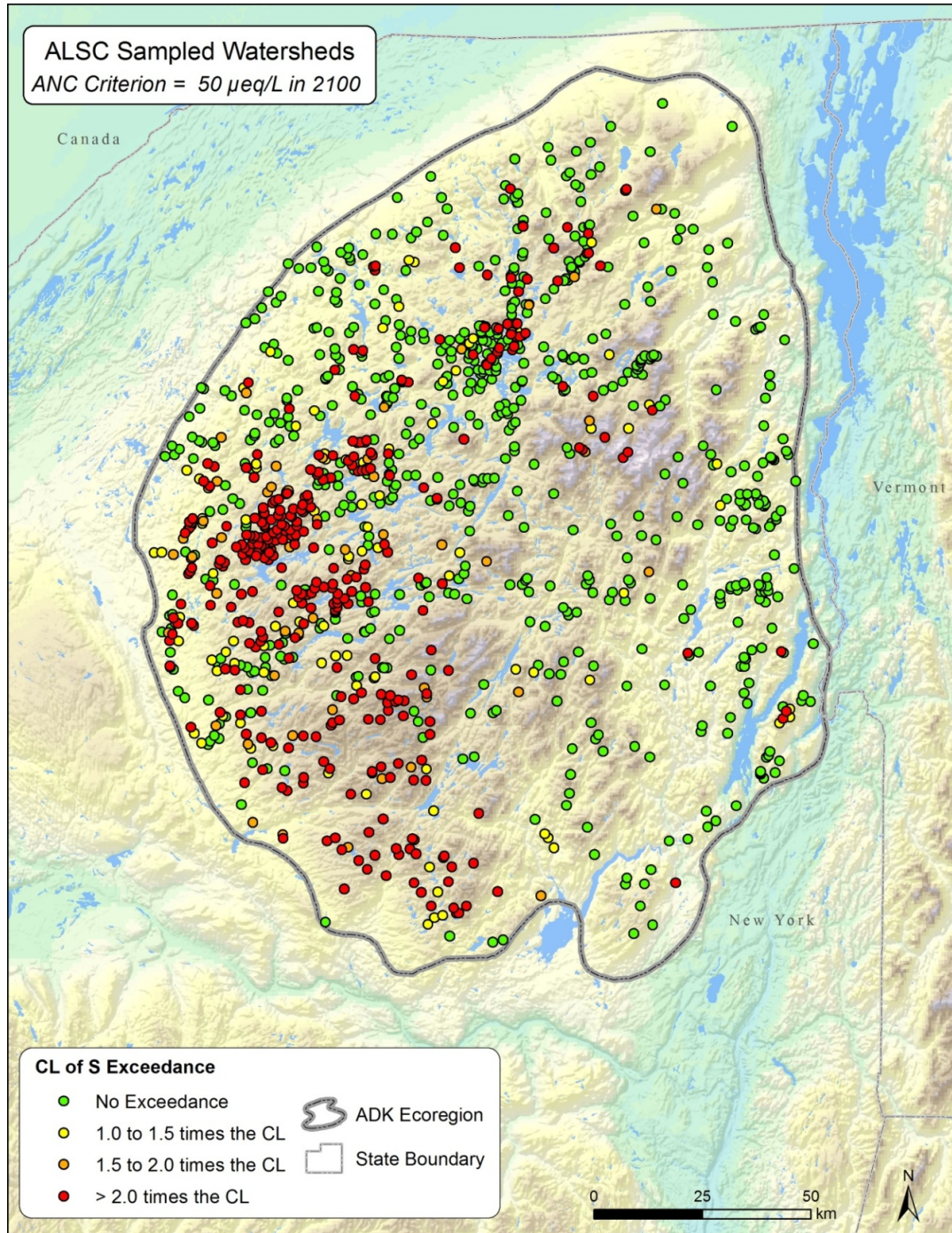




**Map 3-19. Exceedance classes for 1,136 Adirondack lakes based on extrapolation of MAGIC model results of S CLs to the ALS surveyed lakes**

Exceedances were calculated for the year 2100 using a critical threshold ANC value of 50  $\mu\text{eq/L}$ .

Source: Modified from Sullivan et al. (2012). Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171. Copyright © 2012, American Geophysical Union. Reprinted with permission of John Wiley and Sons, Inc.



Many of the modeled Adirondack lakes currently exhibit nearly 100% retention of N deposition inputs. For such lakes, it is not possible to calculate, using the MAGIC model, a CL for N. It can be assumed that such a CL would be very high. For the purpose of the analyses reported here, we assume for these lakes an N CL to protect lake water ANC that is equal to either 1,000 meq/m<sup>2</sup>/yr or equal to the highest simulated CL for any other Adirondack lake, whichever is higher.

The number and percentage of Adirondack lakes that receive ambient N deposition (estimated for the year 2005) above their respective CL are reported in Table 3-17. Results are organized by sensitive criterion, assumed critical threshold value, and endpoint year, and are referenced to the EMAP population of 1,320 low-ANC Adirondack lakes.

In many cases, the observed exceedance in response to assumed N deposition is determined largely by the assumed level of S deposition included in the CL simulation. For lakes and watershed soils that are already in exceedance based on S inputs, the N CL will essentially be zero. In other words, if the chemical indicator is already beyond the critical threshold criterion due to S inputs, then the ecosystem will not be able to tolerate additional acidifying inputs of N.

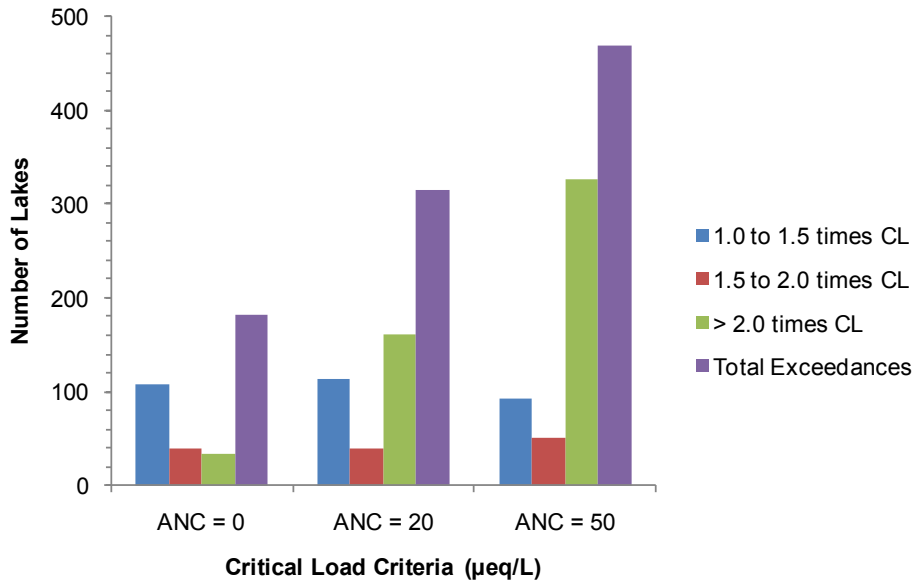
### **3.7 “Can’t Get There from Here” Lakes and Watersheds**

Depending on the critical threshold value of the sensitive criterion and the endpoint year selected for a particular CL analysis, some receptors are unable to attain the critical threshold value by the specified endpoint year even if S or N deposition is reduced to zero and maintained at zero throughout the duration of the simulation (to the endpoint year). These lakes, soils, or soil solution receptors are sometimes called “can’t get there from here” receptors.

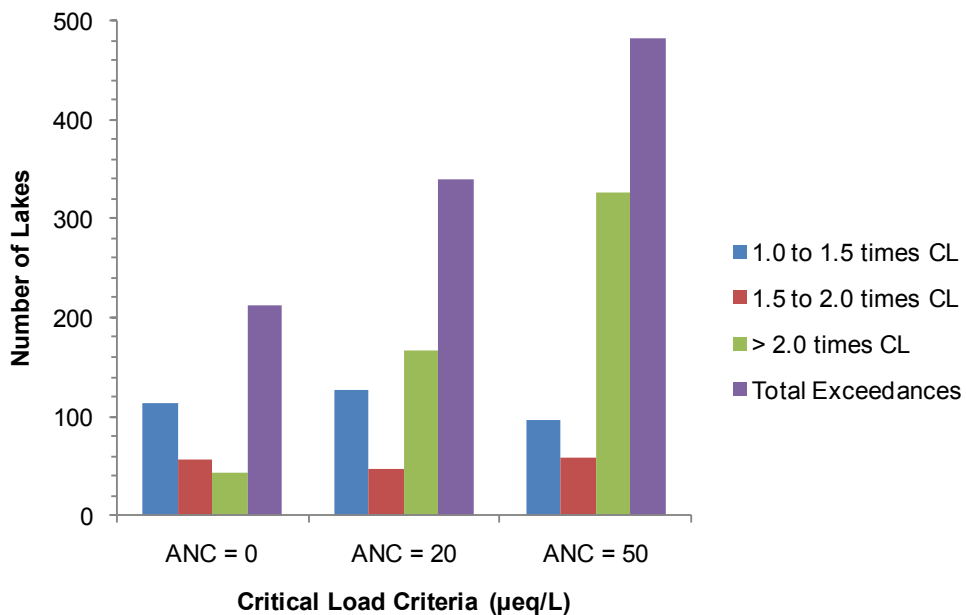
**Figure 3-23. Number of Adirondack lakes estimated to currently (2002) be in exceedance of the sulfur critical load to protect lake ANC to various levels in the years a) 2050 and b) 2100**

These estimates are based on the 1,136 ALS lake frame.

**a) Year 2050**



**b) Year 2100**



**Table 3-17. Adirondack lakes simulated by the MAGIC model to be in exceedance of the critical load of nitrogen deposition**

Based on model simulations of 44 statistically selected lakes from the EMAP population of 1,320 Adirondack lakes that have ANC less than 200 µeq/L.

Receptor	Sensitive Criterion	Critical Value	Number (and Percent) of Lakes in Exceedance in 2002 Based on Critical Load Endpoint Year	
			2050	2100
Lake water	ANC	0 µeq/L	481 (36.5)	465 (35.2)
		20 µeq/L	515 (39.0)	515 (39.0)
		50 µeq/L	611 (46.3)	611 (46.3)
Lake water	NO <sub>3</sub> <sup>-</sup>	10 µeq/L	173 (13.1)	173 (13.1)
		20 µeq/L	14 (1.0)	14 (1.0)
Soil	BS	5%	150 (11.3)	150 (11.3)
		10%	481 (36.5)	418 (31.7)
		15%	502 (38.0)	518 (39.3)

There are three reasons why a specified critical threshold value and endpoint year combination is unattainable: 1) the timeframe is too short to allow recovery to occur; 2) the chemical characteristics of the lake or watershed was such that target indicator values were not achieved (i.e., sufficiently high in ANC, BS, etc.) during pre-industrial times prior to the advent of acidic deposition; or 3) changes to the watershed soils in response to acidic deposition have been such that recovery is substantially delayed, for example due to depletion of exchangeable base cations.

All of the EMAP lakes were simulated to be able to achieve ANC = 0 µeq/L, regardless of endpoint year. Most (93%) EMAP lakes could achieve ANC = 20 µeq/L by the year 2050 or 2100. Somewhat fewer (84%) could attain ANC = 50 µeq/L by either of these endpoint years (Table 3-18). Results for the number of lakes able to attain ANC targets by the year 2020 were slightly lower.

For CLs intended to recover soil BS, the vast majority (94%) of the EMAP watersheds were simulated to be able to attain BS = 5% by the year 2050. In contrast, relatively few (34% by 2050; 41% by 2100) watersheds were simulated to recover to BS = 10%. Even fewer (6% by 2050; 15% by 2100) watersheds were simulated to attain BS = 15%. The base cation to Al ratio target equal to 10 was not an effective CL target. In contrast, the majority (71 to 92%) of those same watersheds were simulated to be able to achieve the ratio target of 1.

**Table 3-18. Estimated number (and percent) of Adirondack lake watersheds simulated to be able or unable to attain the selected critical value of the sensitive criterion by the various sulfur deposition critical endpoint years**

Based on the EMAP statistical frame of lakes having ANC  $\leq$  200  $\mu\text{eq/L}$ .

Receptor	Sensitive Criterion	Critical Value	Endpoint Year	Number (Percent) of Lakes Able to Attain Critical Value
Lake water	ANC	0 $\mu\text{eq/L}$	2020	1,320 (100)
			2050	1,320 (100)
			2100	1,320 (100)
		20 $\mu\text{eq/L}$	2020	1,145 (86.7)
			2050	1,227 (93.2)
			2100	1,227 (93.2)
		50 $\mu\text{eq/L}$	2020	1,037 (78.5)
			2050	1,105 (83.7)
			2100	1,105 (83.7)
Soil	BS	5%	2020	1,180 (89.3)
			2050	1,241 (94.0)
			2100	1,241 (94.0)
		10%	2020	393 (29.8)
			2050	447 (33.9)
			2100	536 (40.6)
		15%	2020	67 (5.1)
			2050	83 (6.3)
			2100	198 (15.0)
Soil Solution	Bc:Al	1	2020	1,217 (92.2)
			2050	1,217 (92.2)
			2100	1,217 (92.2)
		10	2020	69 (5.2)
			2050	85 (6.5)
			2100	172 (13.0)
Soil Solution	Ca:Al	1	2020	935 (70.8)
			2050	935 (70.8)
			2100	935 (70.8)
		10	2020	25 (1.9)
			2050	41 (3.1)
			2100	85 (6.5)

### 3.8 Results for Long-Term Monitoring Lake Watersheds

The ALTM/AEAP lakes are important because of their long-term record of water chemistry monitoring and rich biological response data. In general, these long-term monitoring lakes were selected for monitoring because they are sensitive to acidification effects. They do not represent the spectrum of Adirondack lake acid sensitivity, but rather are skewed towards the high end of acid sensitivity compared with the broader population of Adirondack lake watersheds (Sullivan et al. 2006a). It is therefore of interest to determine how each of the modeled ALTM/AEAP lake watersheds compares to the population of Adirondack lakes with respect to CL. This comparison is provided in Table 3-19 for two CL types: to protect lake water ANC and to protect soil BS. Three critical threshold criteria values are provided for each. Results are summarized as the estimated percentage of lakes in the EMAP low-ANC population that have lower CL than the selected ALTM/AEAP lake. For example, Willys Lake, Squash Pond, and Carry Pond (shown at the top of the table) had CL to protect lake ANC to 50  $\mu\text{eq/L}$  at the extreme end of the sensitivity spectrum relative to the EMAP statistical frame; there were no lakes in the EMAP frame that showed lower CL than these three ALTM/AEAP lakes for the ANC = 50  $\mu\text{eq/L}$  critical threshold value. Similarly, few or no EMAP lakes showed lower CL than these three lakes to protect to ANC 0 or 20  $\mu\text{eq/L}$  in the year 2100. In contrast, the ALTM/AEAP lakes shown at the bottom of the table have much higher CL values to protect lake water ANC. The majority of the EMAP lakes have lower CL values for ANC protection than these ALTM/AEAP lakes listed at the bottom of the table.

There is not a clear correspondence between the CL position of a given ALTM/AEAP lake relative to the EMAP lake population for protecting lake ANC versus protecting soil BS. The CL results for protecting soil BS to 10%, and especially to 15%, show many ALTM/AEAP lakes at the top of the acid sensitivity distribution for EMAP lakes (value in the table is 0). This is because the model simulations suggest that it is difficult or impossible to reach such high BS values in many of the modeled lakes by the year 2100, even if S deposition is reduced to zero. The relative sensitivity of the ALTM/AEAP lake watersheds compared to the population of Adirondack lake watersheds, for the purpose of protecting soil acid-base status, is better evaluated using a critical threshold criterion of BS = 5%. However, some ALTM/AEAP lakes show relatively low CL to protect lake water, but nevertheless a high CL to protect soil (i.e., Brook Trout Lake, Jockeybush Lake) or vice versa (i.e., Bubb Lake, Wheeler Lake).

Simulated CL values for each of the ALTM/AEAP lakes, compared with the EMAP CL population distribution for low-ANC (<200  $\mu\text{eq/L}$ ) lakes, are illustrated in Figures 3-24 and 3-25. The EMAP distributions are presented in the figures as cumulative distribution functions (CDFs) for the population of 1,320 low-ANC Adirondack lakes in the year 2100. Critical loads to protect lake ANC (acidification) are shown in Figure 3-24), and CLs to protect lake  $\text{NO}_3^-$  concentration (nutrient-N enrichment) are shown in Figure 3-25.

**Table 3-19. Modeled relationship between each of the modeled ALTM/AEAP lakes and the EMAP population of low-ANC ( $\leq 200$   $\mu\text{eq/L}$ ) Adirondack lakes with respect to critical sulfur load**

Based on MAGIC model simulations.

ALTM Lake		Estimated Percentage of Lakes in the Population Having Lower CL						
Name	ID	Lower <sup>a</sup> 2002 ANC	Lake Water Protection in Year 2100			Soil Protection in Year 2100		
			ANC = 0	ANC = 20	ANC = 50	BS = 5	BS = 10	BS = 15
			$(\mu\text{eq/L})$			$(\%)$		
Willys Lake	40210	0.0	12.0	12.0	0.0	13.5	0.0	0.0
Squash Pond	40754	2.4	0.0	0.0	0.0	30.1	59.2	0.0
Carry Pond	50669	13.3	12.0	12.0	0.0	9.7	0.0	0.0
Round Pond	040731A	13.3	13.3	13.3	15.1	30.1	0.0	0.0
Jockeybush Lake	50259	13.3	29.1	19.7	17.4	56.0	61.5	0.0
Indian Lake	40852	14.5	12.0	12.0	0.0	4.1	0.0	0.0
Brook Trout Lake	40874	14.5	18.7	15.4	16.3	55.0	62.6	0.0
South Lake	41004	15.5	35.0	21.6	18.5	30.1	0.0	0.0
Big Moose Lake	40752	16.4	16.6	16.4	19.5	41.4	0.0	0.0
Queer Lake	60329	16.4	15.4	14.5	17.4	21.7	58.5	0.0
Middle Settlement Lake	40704	16.4	29.1	23.9	27.3	2.8	0.0	0.0
Long Pond	50649	16.4	35.0	32.2	22.8	46.9	59.2	0.0
North Lake	41007	17.4	36.0	33.2	18.5	56.0	77.3	0.0
G Lake	70859	17.4	29.1	23.9	27.3	40.7	0.0	0.0
Dart Lake	40750	17.4	30.3	21.6	22.8	56.0	61.5	0.0
Constable Pond	40777	18.4	39.1	39.1	35.7	45.8	58.5	0.0
Squaw Lake	40850	22.1	36.0	33.2	17.4	39.5	77.3	85.0
West Pond	40753	24.0	18.7	19.7	22.8	13.5	0.0	0.0
Limekiln Lake	40826	27.3	42.3	41.3	41.3	56.0	66.2	0.0
Grass Pond	40706	30.2	44.0	44.0	44.0	13.5	0.0	0.0
Helldiver Pond	40877	35.0	44.0	44.0	44.0	23.0	0.0	0.0
Lake Rondaxe	40739	47.8	47.2	49.5	54.5	68.1	55.3	0.0
Cascade Lake	40747	47.8	52.7	54.6	64.5	70.2	61.5	0.0

**Table 3-19 continued**

ALTM Lake		Estimated Percentage of Lakes in the Population Having Lower CL						
Name	ID	Lower <sup>a</sup> 2002 ANC	Lake Water Protection in Year 2100			Soil Protection in Year 2100		
			ANC = 0	ANC = 20	ANC = 50	BS = 5	BS = 10	BS = 15
			(µeq/L)			(%)		
			52.7	54.6	64.5	0.0	0.0	0.0
			60.3	67.4	69.1	70.2	0.0	0.0
Sagamore Lake	60313	63.5	52.7	54.6	64.5	70.2	78.5	0.0
Arbutus Pond	50684	66.9	66.5	67.4	70.2	62.4	0.0	0.0
Windfall Pond	040750A	71.3	69.9	70.8	73.6	93.3	79.5	0.0
Wheeler Lake	40731	71.3	60.3	67.4	69.1	30.1	0.0	0.0
Raquette Lake Reservoir	060315A	71.3	73.7	74.7	75.6	70.2	58.5	0.0
Moss Lake	40746	71.3	69.9	70.8	73.6	93.3	78.5	0.0
Willis Lake	50215	72.5	58.0	64.2	69.1	93.3	96.9	98.1

<sup>a</sup> Estimated percentage of Adirondack lakes, from the EMAP low-ANC ( $\leq 200$  µeq/L) frame, having ANC in the year 2000 lower than the ANC of the modeled lake reported here.

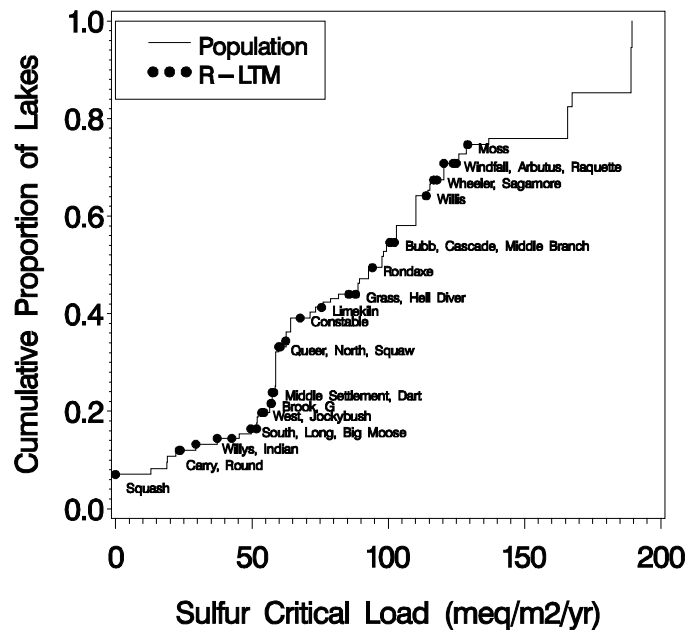
The data shown in Table 3-19 for protecting ANC to 20 µeq/L in the year 2100 are presented in Figure 3-24a as a cumulative distribution function of the S CL for protecting lake ANC to 20 µeq/L in the year 2100 for the EMAP low-ANC lake watershed population. Super-imposed on this distribution function is the location of each of the modeled ALTM/AEAP lakes. This shows graphically the relative position of each ALTM/AEAP lake compared to the broader population of Adirondack lakes. In general, the ALTM/AEAP lakes are well distributed across the lower 75% of the EMAP CL distribution for low-ANC lakes. These long-term monitoring lakes do not, however, represent those lakes in the population that have higher CL values. Figure 3-24 indicates that a number of lakes in the EMAP population, and several ALTM/AEAP lakes, were simulated to have CL of S deposition at or near zero to protect lake ANC by the year 2100 to 20 µeq/L, and especially to 50 µeq/L. All of the LTM/AEAP lakes and about 80% of the EMAP lakes had simulated CLs of S deposition that were less than about 120 meq/m<sup>2</sup>/yr to protect lake ANC to 20 or 50 µeq/L by 2100. Critical load values for N deposition to protect lake ANC tended to be much higher than CL values for S deposition. More than half of the EMAP population of lakes were simulated to have CL of N deposition higher than about 1,000 meq/m<sup>2</sup>/yr.



**Figure 3-24. Cumulative distribution functions (CDF) of critical load (CL) to protect lake water ANC concentration simulated with the MAGIC model for the EMAP population of Adirondack lakes having  $ANC \leq 200 \mu\text{eq/L}$**

Superimposed on each CDF is the location of each ALTM/AEAP study lake within the population distribution. The distributions represent a) CL of S deposition to attain lake water ANC = 20  $\mu\text{eq/L}$ , b) CL of S deposition to attain lake water ANC = 50  $\mu\text{eq/L}$ , c) CL of N deposition to attain lake water ANC = 20  $\mu\text{eq/L}$ , and d) CL of N deposition to attain lake water ANC = 50  $\mu\text{eq/L}$ . All CL simulations are projected to the year 2100. Lakes simulated to have N CL = 0  $\text{meq/m}^2/\text{yr}$  are represented on the log scale of panels c and d as having CL = 1  $\text{meq/m}^2/\text{yr}$  for ease of graphing.

**a) Sulfur Critical Load to Protect to ANC = 20  $\mu\text{eq/L}$**



**b) Sulfur Critical Load to Protect to ANC = 50  $\mu\text{eq/L}$**

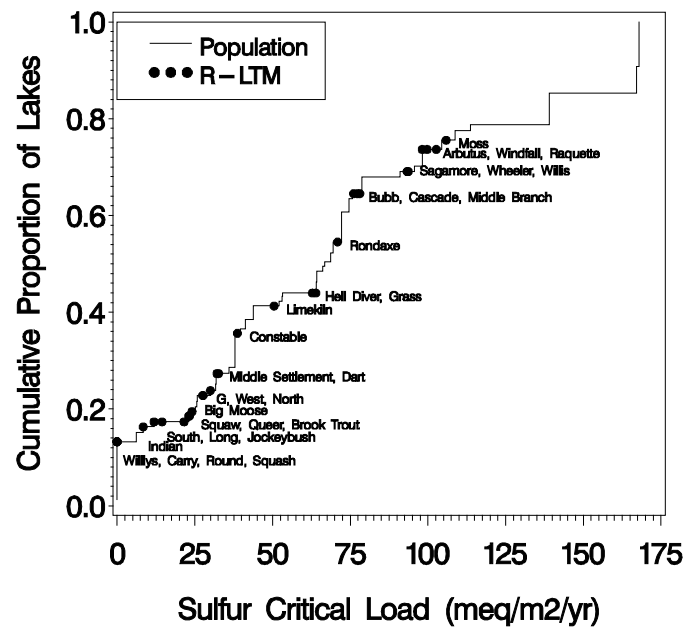
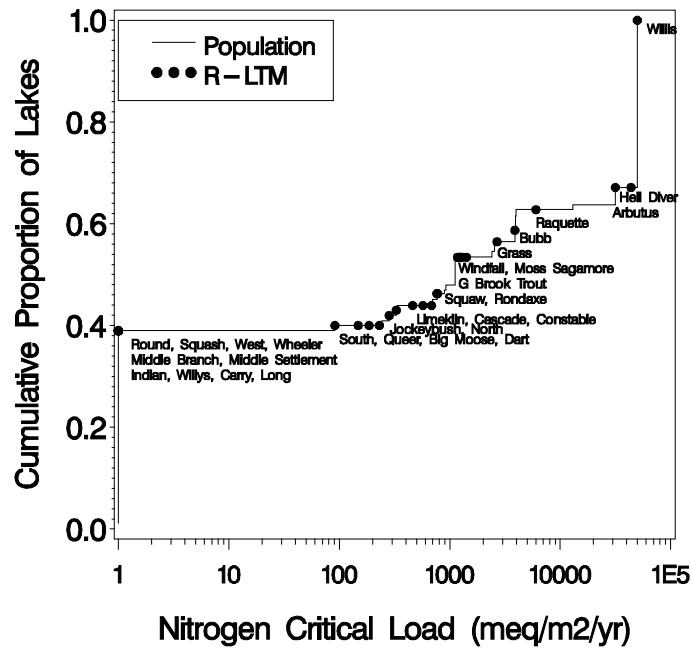
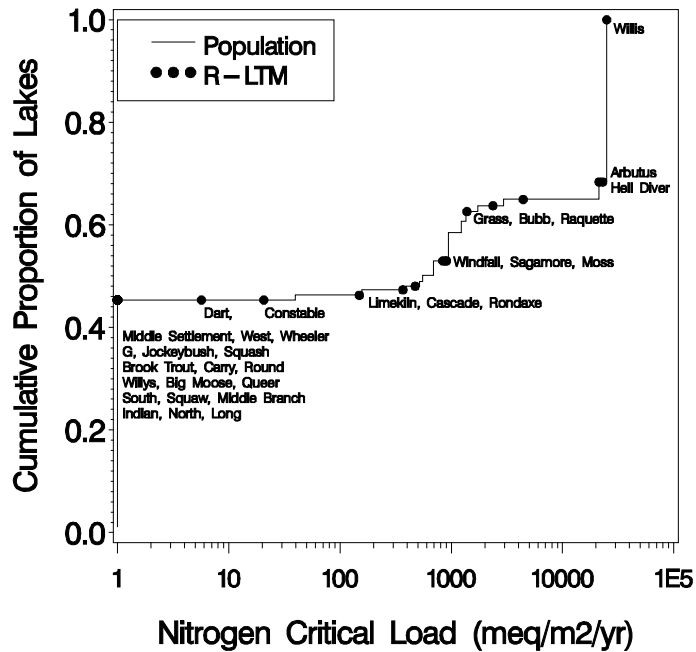


Figure 3-24 continued

c) Nitrogen Critical Load to Protect to ANC = 20 µeq/L



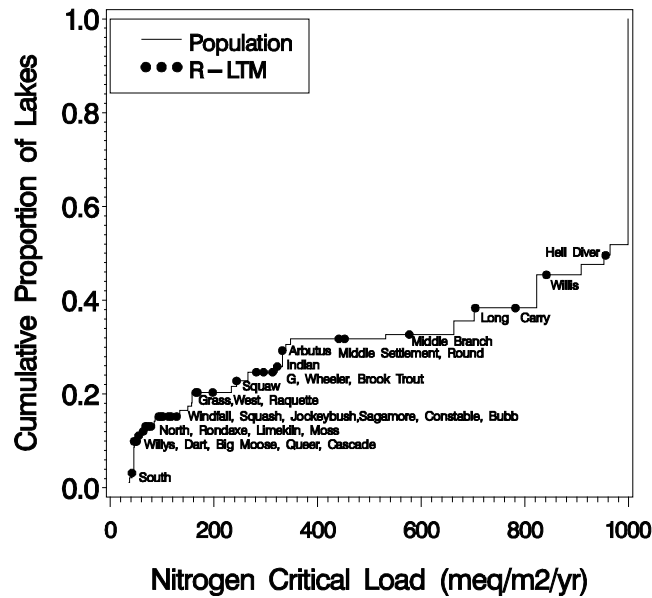
d) Nitrogen Critical Load to Protect to ANC = 50 µeq/L



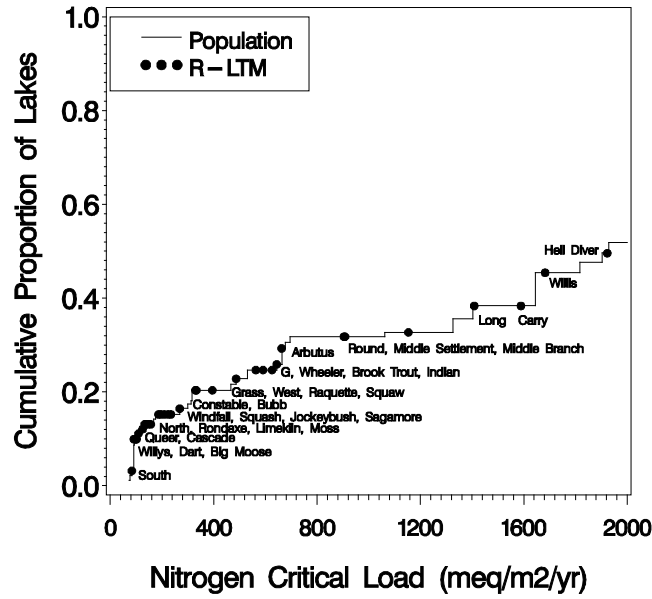
**Figure 3-25. Cumulative distribution functions (CDF) of critical load (CL) to protect lake water  $\text{NO}_3^-$  concentration simulated with the MAGIC model for the EMAP population of Adirondack lakes having  $\text{ANC} \leq 200 \mu\text{eq/L}$**

Superimposed on each CDF is the location of each ALT/EAEP study lake within the population distribution. The distributions represent a) CL of N deposition to attain lake water  $\text{NO}_3^-$  concentration equal to  $10 \mu\text{eq/L}$ , and b) CL of N deposition to attain lake water  $\text{NO}_3^-$  concentration equal to  $20 \mu\text{eq/L}$ . Both CL simulations are projected to the year 2100.

**a) Nitrogen Critical Load to Protect to Lake  $\text{NO}_3^- = 10 \mu\text{eq/L}$**



**b) Nitrogen Critical Load to Protect to Lake  $\text{NO}_3^- = 20 \mu\text{eq/L}$**



### 3.9 Uncertainty

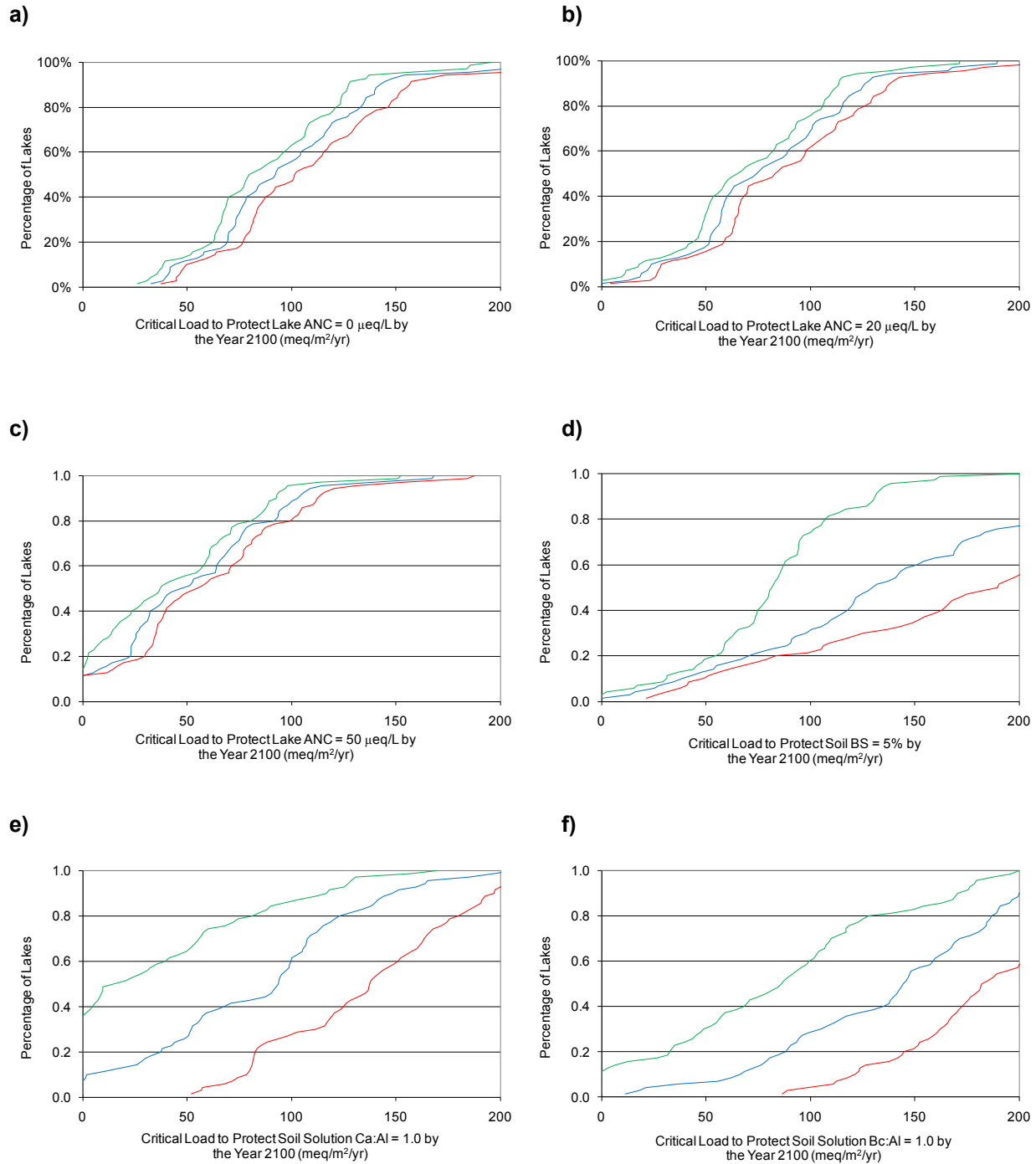
MAGIC CL simulation uncertainty, reflected in the fuzzy calibration procedure, is summarized in Figure 3-26.

The figure shows the CDFs of selected CL simulations, where the median value of each CL, from among the 8 to 10 successful calibrations, is represented as a thick solid blue line. The maximum and minimum simulated CL values are represented as thin green and red lines respectively. The uncertainty in simulating the CL to protect lake ANC is relatively small across the distribution of CL values (Figure 3-26a). In general, the difference between the maximum and minimum simulated CL values was less than about 10 to 20 meq/m<sup>2</sup>/yr. Similarly, the uncertainty in simulating the CL to protect soil BS was relatively low (generally less than about 10 meq/m<sup>2</sup>/yr) for the watersheds that were most acid-sensitive (those that have CL to protect soil BS less than about 75 meq/m<sup>2</sup>/yr). For the less acid-sensitive watersheds, however, the CL to protect soil BS was much more uncertain (right side of Figure 3-26d). However, added uncertainty is not of great concern for watersheds that have relatively high CL. A major modeling objective is to minimize uncertainty (increase precision) for the watersheds that are especially acid-sensitive (those having low CL). Uncertainty was high across the range of CL values for simulations of S CL to protect soil solution base cation to Al ratios (Figures 3-26 e,f). These results suggest that the model performs best for simulating the CL to protect lake ANC, and for protecting soil BS for the most acid-sensitive watersheds. Model performance is more uncertain for protecting soil BS in the more base cation-rich watersheds and for protecting soil solution base cation to Al ratios across the spectrum of acid sensitivity.

Figure 3-27 compares simulated CLs and associated uncertainties for only those lake watersheds that are highly acid-sensitive (simulated CL  $\leq$  50 meq/m<sup>2</sup>/yr). The uncertainty range determined by the fuzzy calibration procedure was smallest for CL simulations to protect lake ANC and only slightly higher for CL to protect soil BS. However, the CL simulations to protect soil solution base cation to Al ratios were much more uncertain. The estimated percent of modeled watersheds having maximum or minimum CL values below 50 meq/m<sup>2</sup>/yr changed by about a factor of two or more compared with median simulation CL values for the CL simulations designed to protect soil solution.

**Figure 3-26. Cumulative frequency distributions showing MAGIC model median (blue), maximum (green), and minimum (red) simulated values of CL to protect: a) lake ANC to 0  $\mu\text{eq/L}$ , b) lake ANC to 20  $\mu\text{eq/L}$ , c) lake ANC to 50  $\mu\text{eq/L}$ , d) soil BS to 5%, e) Bc:Al to 1.0, and f) Ca:Al to 1.0, all by the year 2100**

Source, Panels d, e, and f: Sullivan et al. (2011). Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. *J. Environ. Stud. Sci.* 1(4):301-314. Copyright © 2011, Springer. Reprinted with permission.



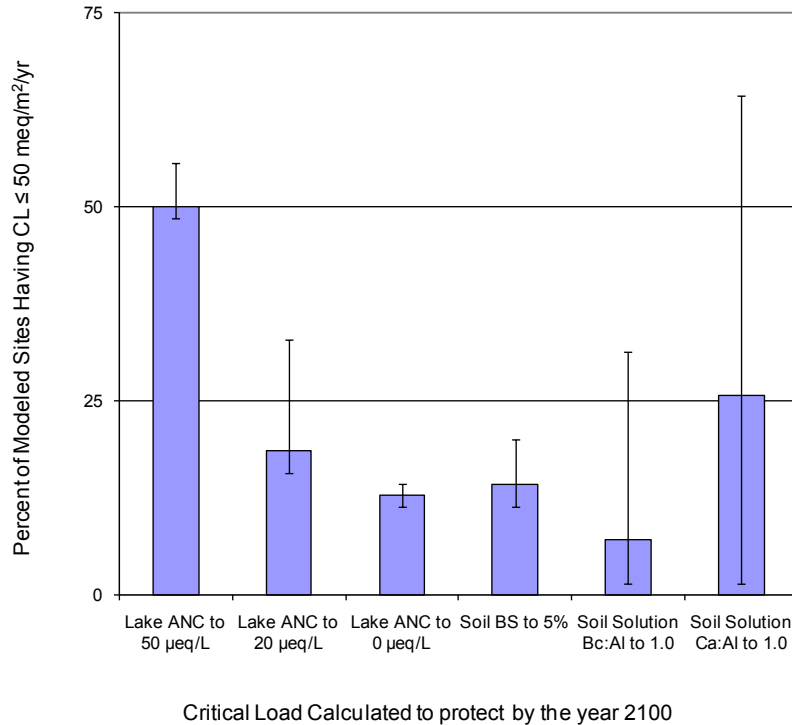
### 3.10 Next Steps

Additional work to further refine the TL and exceedance values presented in this report for Adirondack lakes and their watersheds might include some or all of the following:

- Develop additional information regarding tipping points of BS and/or mineral soil exchangeable Ca at which adverse impacts occur on terrestrial vegetation. Improved understanding of critical limits will improve and further substantiate terrestrial TLs.
- Reduce uncertainties in atmospheric S and N deposition to Adirondack watersheds. Improved estimates of deposition are needed to better document TL exceedance.
- Explore dose response functions for the effects of soil acidification on the common tree species in the Adirondack Mountains. Although some new data are available for sugar maple response (Sullivan et al. 2013), data are generally limited or lacking for other tree species.
- Expand terrestrial and aquatic TL calculations to include effects on biodiversity of nutrient N enrichment in addition to the effects of acidification discussed here.

**Figure 3-27. Histogram showing the percent of the 70 MAGIC model sites (i.e., from among those that had available watershed-specific soil data for calibration) that were simulated to have low CL ( $\leq 50$  meq/m<sup>2</sup>/yr)**

Results are presented to compare CLs to protect: lake ANC to three critical threshold criteria, soil BS to 5%, Bc:Al to 1.0, and Ca:Al to 1.0, all by the year 2100. Results are presented for the median simulations (represented by the histogram columns) among the 8 to 10 calibrations that were conducted at each site and the maximum and minimum simulated values (represented by error bars).



## 4 Conclusions

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Critical loads for atmospheric deposition of S and N were calculated using MAGIC, a dynamic model of watershed acid-base chemistry. A matrix of CL estimates was developed, based on differing air pollutants (e.g., S, N), sensitive resources and associated indicators (e.g., lake water ANC, soil % BS, soil solution Bc:Al or Ca:Al ratio), critical thresholds (e.g., lake ANC = 0, 20, or 50  $\mu\text{eq/L}$ ; soil % BS = 5, 10, or 15%), and evaluation year (e.g., 2050, 2100). Based on each of the estimated CLs, an exceedance was also calculated to reflect the extent to which the ambient atmospheric deposition loading exceeds the CL that would allow sensitive resources to recover from past damage or to be protected against future damage.

The CL and exceedance estimates simulated by MAGIC were extrapolated to several population frames of Adirondack lake watersheds. These analyses yielded estimates of the numbers and percentages of Adirondack lakes or watersheds that exhibit various levels of CL and exceedance. They also allowed aquatic CLs (and associated exceedances) for protecting and restoring lake chemistry to be mapped across the Adirondack Ecoregion. Results of these analyses will aid in the management of acid-sensitive Adirondack lakes and watershed soils by revealing the levels of acidic deposition that will allow damaged resources to recover and undamaged resources to remain protected in a sustainable fashion.

Specific conclusions of this research include:

1. Adirondack lakes that currently have low ANC also tend to have low CL.
2. Modeled CL values are lower if the intention is to protect chemical indicator parameters to higher levels as opposed to lower levels of protection. Thus, the CL to protect lake ANC to 50  $\mu\text{eq/L}$  is lower than the CL to protect to 0 or 20  $\mu\text{eq/L}$ .
3. The CL to protect and restore terrestrial resources can be estimated using a soil chemical indicator such as % BS or a soil solution chemical indicator such as Bc:Al. The MAGIC model uncertainty is greater for CLs based on protection of soil solution. This uncertainty is mainly because soil solution data are seldom available for regional assessment and are not used to constrain MAGIC model calibrations to observed current conditions.
4. Threshold values for the Bc:Al and Ca:Al soil solution ratios equal to 10 did not appear useful for CL calculation with MAGIC. Nearly all of the modeled watersheds showed simulated CL to protect these ratios to a value of 10 that was substantially lower than ambient deposition.
5. Critical loads for N deposition to protect against acidification were generally higher than corresponding CLs for S deposition. Most Adirondack watersheds retain the majority of deposited N, with relatively little  $\text{NO}_3^-$  leaching to surface waters. Critical loads of N deposition to protect against nutrient enrichment were not calculated in this study.
6. Extrapolation of simulated MAGIC CLs to the population of Adirondack lakes in EPA's EMAP frame suggested that about 41% of the low-ANC ( $\leq 200$   $\mu\text{eq/L}$ ) Adirondack lakes larger than 1 ha in area have a CL that is below 50  $\text{meq/m}^2/\text{yr}$  to protect ANC to 50  $\mu\text{eq/L}$  in the year 2100. The comparable statistic for all Adirondack lakes larger than 1 ha, irrespective of ANC, is about 30%.

7. The CLs to protect soil BS were very sensitive to selection of the critical threshold value for % BS. A threshold of 5% was readily achievable for the majority of Adirondack watersheds. In contrast, about two-thirds or more of the watersheds showed very low CL ( $< 25 \text{ meq/m}^2/\text{yr}$ ) to achieve % BS = 10%. More field and experimental information is needed regarding the relationships between % BS in soil and plant responses.
8. MAGIC model simulations of the CL to protect lake ANC were successfully extrapolated to the population of 1,136 ALS lakes using only ANC measurements made in the 1980s. The vast majority of the ALS lakes situated in the southwestern Adirondack Mountains had CL less than  $50 \text{ meq/m}^2/\text{yr}$ , as did many lakes in the high peaks area.
9. Modeled CL varied with selection of endpoint year and with starting point (above or below the threshold criterion). Lakes with the lowest CLs (most acid-sensitive) could tolerate higher S loading if one was willing to wait longer to achieve chemical recovery. Lakes with the higher CLs (less acid-sensitive) require more stringent controls (lower S loading) if the resource protection is intended to extend further into the future.
10. Regional S and N deposition estimates were combined with CLs to calculate exceedances. For protecting lake ANC to the year 2100, the percent of the 1,320 low-ANC ( $\leq 200 \text{ } \mu\text{eq/L}$ ) EMAP lakes projected to be in exceedance ranged from about 16% (to protect ANC to  $0 \text{ } \mu\text{eq/L}$ ) to 46% (to protect ANC to  $50 \text{ } \mu\text{eq/L}$ ). Some lakes and their watersheds had ambient S deposition that was more than double their respective CL.
11. Some critical threshold criteria were simulated to be unobtainable for given watersheds, even if deposition is reduced to zero and held at zero until the endpoint year.



## 5 References

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- Baker, J.P., S.A. Gherini, S.W. Christensen, C.T. Driscoll, J. Gallagher, R.K. Munson, R.M. Newton, K.H. Reckhow, and C.L. Schofield. 1990. Adirondack lakes survey: an interpretive analysis of fish communities and water chemistry, 1984-1987. Adirondack Lakes Survey Corporation, Ray Brook, NY.
- Baker, L.A. 1991. Regional estimates of dry deposition. Appendix B. In D.F. Charles (Ed.) *Acidic Deposition and Aquatic Ecosystems: Regional Case Studies*. Springer-Verlag, New York. pp. 645-652.
- Charles, D.F. (Ed.). 1991. *Acidic Deposition and Aquatic Ecosystems: Regional Case Studies*. Springer-Verlag, New York, NY. 747 pp.
- Church, M.R., K.W. Thorton, P.W. Shaffer, D.L. Stevens, B.P. Rochelle, R.G. Holdren, M.G. Johnson, J.J. Lee, R.S. Turner, D.L. Cassell, D.A. Lammers, W.G. Campbell, C.I. Liff, C.C. Brandt, L.H. Liegel, G.D. Bishop, D.C. Mortenson, and S.M. Pierson. 1989. Future effects of long-term sulfur deposition on surface water chemistry in the Northeast and Southern Blue Ridge Province (Results of the Direct/Delayed Response Project. U.S. Environmental Protection Agency, Corvallis, OR.
- Cosby, B.J., G.M. Hornberger, J.N. Galloway, and R.F. Wright. 1985a. Time scales of catchment acidification: a quantitative model for estimating freshwater acidification. *Environ. Sci. Technol.* 19:1144-1149.
- Cosby, B.J., G.M. Hornberger, J.N. Galloway, and R.F. Wright. 1985b. Modelling the effects of acid deposition: assessment of a lumped parameter model of soil water and streamwater chemistry. *Water Resour. Res.* 21(1):51-63.
- Cosby, B.J., R.F. Wright, G.M. Hornberger, and J.N. Galloway. 1985c. Modelling the effects of acid deposition: estimation of long-term water quality responses in a small forested catchment. *Water Resour. Res.* 21(11): 1591-1601.
- Cosby, B.J., G.M. Hornberger, P.F. Ryan, and D.M. Wolock. 1989. MAGIC/DDRP Final Report. U.S. Environmental Protection Agency, Corvallis, OR.
- Cosby, B.J., A. Jenkins, R.C. Ferrier, J.D. Miller, and T.A.B. Walker. 1990. Modelling stream acidification in afforested catchments: long-term reconstructions at two sites in central Scotland. *J. Hydrol.* 120:143-162.
- Cosby, B.J., R.F. Wright, and E. Gjessing. 1995. An acidification model (MAGIC) with organic acids evaluated using whole-catchment manipulations in Norway. *J. Hydrol.* 170:101-122.
- Cosby, B.J., S.A. Norton, and J.S. Kahl. 1996. Using a paired catchment manipulation experiment to evaluate a catchment-scale biogeochemical model. *Sci. Total Environ.* 183:49-66.
- Cosby, B.J., R.C. Ferrier, A. Jenkins, and R.F. Wright. 2001. Modeling the effects of acid deposition: refinements, adjustments and inclusion of nitrogen dynamics in the MAGIC model. *Hydrol. Earth Syst. Sci.* 5(3):499-517.

- Cosby, B.J., J.R. Webb, J.N. Galloway, and F.A. Deviney. 2006. Acidic Deposition Impacts on Natural Resources in Shenandoah National Park. NPS/NER/NRTR-2006/066. U.S. Department of the Interior, National Park Service, Northeast Region, Philadelphia, PA.
- Cronan, C.S. and D.F. Grigal. 1995. Use of calcium/aluminum ratios as indicators of stress in forest ecosystems. *J. Environ. Qual.* 24:209-226.
- Cumming, B.F., J.P. Smol, J.C. Kingston, D.F. Charles, H.J.B. Birks, K.E. Camburn, S.S. Dixit, A.J. Uutala, and A.R. Selle. 1992. How much acidification has occurred in Adirondack region lakes (New York, USA) since preindustrial times? *Can. J. Fish. Aquat. Sci.* 49(1):128-141.
- Driscoll, C.T. and R.M. Newton. 1985. Chemical characteristics of Adirondack lakes. *Environ. Sci. Technol.* 19:1018-1024.
- Driscoll, C.T., R.M. Newton, C.P. Gubala, J.P. Baker, and S.W. Christensen. 1991. Adirondack Mountains. In D.F. Charles (Ed.) *Acidic deposition and aquatic ecosystems: regional case studies*. Springer-Verlag, New York, NY. pp. 133-202.
- Driscoll, C.T., G.B. Lawrence, A.J. Bulger, T.J. Butler, C.S. Cronan, C. Eagar, K.F. Lambert, G.E. Likens, J.L. Stoddard, and K.C. Weathers. 2001. Acidic deposition in the northeastern United States: sources and inputs, ecosystem effects, and management strategies. *BioScience* 51(3):180-198.
- Driscoll, C.T., K.M. Driscoll, M.J. Mitchell, and D.J. Raynal. 2003a. Effects of acidic deposition on forest and aquatic ecosystems in New York State. *Environ. Pollut.* 123:327-336.
- Driscoll, C.T., K.M. Driscoll, K.M. Roy, and M.J. Mitchell. 2003b. Chemical response of lakes in the Adirondack region of New York to declines in acidic deposition. *Environ. Sci. Technol.* 37:2036-2042.
- Driscoll, C.T., K.M. Driscoll, K.M. Roy, and J. Dukett. 2007. Changes in the chemistry of lakes in the Adirondack region of New York following declines in acidic deposition. *Water Air Soil Pollut.* 22(6):1181-1188.
- Ferrier, C.A., R.F. Wright, B.J. Cosby, and A. Jenkins. 1995. Application of the MAGIC model to the Norway spruce stand at Solling, Germany. *Ecol. Model.* 83:77-84.
- Gbondo-Tugbawa, S.S. and C.T. Driscoll. 2003. Factors controlling long-term changes in soil pools of exchangeable basic cations and stream acid neutralizing capacity in a northern hardwood forest ecosystem. *Biogeochemistry* 63:161-185.
- Grimm, J.W. and J.A. Lynch. 1997. Enhanced Wet Deposition Estimates Using Modeled Precipitation Inputs. Final Report to U.S. Forest Service under Cooperative Agreement #23-721. Environmental Resources Research Institute, The Pennsylvania State University, University Park, PA.
- Hornberger, G.M., B.J. Cosby, and R.F. Wright. 1989. Historical reconstructions and future forecasts of regional surface water acidification in southernmost Norway. *Water Resour. Res.* 25:2009-2018.

- Jenkins, A., B.J. Cosby, R.C. Ferrier, T.A.B. Walker, and J.D. Miller. 1990a. Modelling stream acidification in afforested catchments: an assessment of the relative effects of acid deposition and afforestation. *J. Hydrol.* 120:163–181.
- Jenkins, A., P.G. Whitehead, B.J. Cosby, R.C. Ferrier, and D.J. Waters. 1990b. Modelling long term acidification: a comparison with diatom reconstructions and the implications for reversibility. *Phil. Trans. R. Soc. Lond.* 327:435-440.
- Jenkins, A., P.G. Whitehead, T.J. Musgrove, and B.J. Cosby. 1990c. A regional model of acidification in Wales. *J. Hydrol.* 116:403-416.
- Kretser, W., J. Gallagher, and J. Nicolette. 1989. Adirondack Lakes Study, 1984-1987: An Evaluation of Fish Communities and Water Chemistry. Adirondack Lakes Survey Corp, Ray Brook, NY.
- Larsen, D.P., K.W. Thornton, N.S. Urquhart, and S.G. Paulsen. 1994. The role of sample surveys for monitoring the condition of the nation's lakes. *Environ. Monitor. Assess.* 32:101-134.
- Lawrence, G.B., M.B. David, and W.C. Shortle. 1995. A new mechanism for calcium loss in forest-floor soils. *Nature* 378:162-165.
- Lawrence, G.B., K.M. Roy, B.P. Baldigo, H.A. Simonin, S.B. Capone, J.W. Sutherland, S.A. Nierzwicki-Bauer, and C.W. Boylen. 2008. Chronic and episodic acidification of Adirondack streams from acid rain in 2003-2005. *J. Environ. Qual.* 37:2264-2274.
- Lepistö, A., P.G. Whitehead, C. Neal, and B.J. Cosby. 1988. Modelling the effects of acid deposition: Estimation of long term water quality responses in forested catchments in Finland. *Nord. Hydrol.* 19:99-120.
- Likens, G.E., C.T. Driscoll, and D.C. Buso. 1996. Long-term effects of acid rain: response and recovery of a forest ecosystem. *Science* 272(5259):244-246.
- Linthurst, R.A., D.H. Landers, J.M. Eilers, D.F. Brakke, W.S. Overton, E.P. Meier, and R.E. Crowe. 1986. Characteristics of Lakes in the Eastern United States. Volume I. Population Descriptions and Physico-chemical Relationships. EPA/600/4-86/007a. U.S. Environmental Protection Agency, Washington, D.C.
- McDonnell, T.C., B.J. Cosby, and T.J. Sullivan. 2012. Regionalization of soil base cation weathering for evaluating stream water acidification in the Appalachian Mountains, USA. *Environ. Pollut.* 162:338-344.
- McNulty, S.G., E.C. Cohen, J.A.M. Myers, T.J. Sullivan, and H. Li. 2007. Estimates of critical acid loads and exceedances for forest soils across the conterminous United States. *Environ. Pollut.* 149:281-292.
- Moldan, F., R.F. Wright, R.C. Ferrier, B.I. Andersson, and H. Hultberg. 1998. Simulating the Gårdsjön covered catchment experiment with the MAGIC model. In H. Hultberg and R.A. Skeffington (Eds.). *Experimental Reversal of Acid Rain Effects. The Gårdsjön Roof Project*. Wiley and Sons, Chichester, UK. pp. 351-362.

- National Acid Precipitation Assessment Program (NAPAP). 1991. Integrated assessment report. National Acid Precipitation Assessment Program, Washington, DC.
- Nierzwicki-Bauer, S.A., C.W. Boylen, L.W. Eichler, J.P. Harrison, J.W. Sutherland, W. Shaw, R.A. Daniels, D.F. Charles, F.W. Acker, T.J. Sullivan, B. Momen, and P. Bukaveckas. 2010. Acidification in the Adirondacks: Defining the biota in trophic levels of 30 chemically diverse acid-impacted lakes. *Environ. Sci. Technol.* 44:5721-5727.
- Nizich, S.V., D. Misenheimer, A.P. T. Pierce, and P. Carson. 1996. National Air Pollutant Trends 1900-1995. EPA-454/R-96-007. U.S. Environmental Protection Agency, Office of Air Quality, Research Triangle Park, NC.
- Norton, S.A., R.F. Wright, J.S. Kahl, and J.P. Scofield. 1992. The MAGIC simulation of surface water acidification at, and first year results from, the Bear Brook Watershed Manipulation, Maine, USA. *Environ. Pollut.* 77:279-286.
- Paulsen, S.G., R.M. Hughes, and D.P. Larsen. 1998. Critical elements in describing and understanding our nation's aquatic resources. *J. Am. Water Resour. Assoc.* 34(5):995-1005.
- Posch, M., P.A.M. DeSmet, J.P. Hettelingh, and R.J. Downing. 2001. Calculation and mapping of critical thresholds in Europe. Status report 2001. Coordination Center for Effects, National Institute of Public Health and the Environment (RIVM), Bilthoven, The Netherlands.
- Rosenbrock, H.H. 1960. An automatic method for finding the greatest or least value of a function. *Computer Journal* 3:175-184.
- Scott, J.M., F. Davis, B. Csuti, R. Noss, B. Butterfield, C. Groves, H. Anderson, S. Caicco, F. D'Erchia, T.C. Edwards, Jr., J. Ulliman, and R.G. Wright. 1993. Gap analysis: A geographic approach to protection of biological diversity. Wildlife Monographs No. 123.
- Stoddard, J., J.S. Kahl, F.A. Deviney, D.R. DeWalle, C.T. Driscoll, A.T. Herlihy, J.H. Kellogg, P.S. Murdoch, J.R. Webb, and K.E. Webster. 2003. Response of surface water chemistry to the Clean Air Act Amendments of 1990. EPA 620/R-03/001. U.S. Environmental Protection Agency, Office of Research and Development, National Health and Environmental Effects Research Laboratory, Research Triangle Park, NC.
- Stoddard, J.L., C.T. Driscoll, J.S. Kahl, and J.H. Kellogg. 1998. Can site-specific trends be extrapolated to the regional level? A lake acidification example for the northeast. *Ecol. Appl.* 8(2):288-299.
- Sullivan, T.J. 1990. Historical Changes in Surface Water Acid-Base Chemistry in Response to Acidic Deposition. State of the Science, SOS/T 11. National Acid Precipitation Assessment Program, Washington, DC.
- Sullivan, T.J., D.L. Kugler, M.J. Small, C.B. Johnson, D.H. Landers, B.J. Rosenbaum, W.S. Overton, W.A. Krester, and J. Gallagher. 1990. Variation in Adirondack, New York, lakewater chemistry as a function of surface area. *Water Resour. Bull.* 26:167-176.

- Sullivan, T.J., B.J. Cosby, S.A. Norton, D.F. Charles, R.F. Wright, and E. Gjessing. 1995. Multi-site testing and evaluation of a geochemical model of acid-base chemistry - confirmation of the MAGIC model using catchment manipulation experiments and historical diatom inferences. In: Ecosystem Manipulation Experiments: scientific approaches, experimental design and relevant results. In A. Jenkins, R.C. Ferrier and C. Kirby (Eds.). *Ecosystem Manipulation Experiments: Scientific Approaches, Experimental Design and Relevant Results*. Ecosystems Research Report 20. Commission of European Communities, Luxembourg. pp. 360-365.
- Sullivan, T.J., B.J. Cosby, C.T. Driscoll, D.F. Charles, and H.F. Hemond. 1996. Influence of organic acids on model projections of lake acidification. *Water Air Soil Pollut.* 91:271-282.
- Sullivan, T.J., D.F. Charles, J.A. Bernert, B. McMartin, K.B. Vaché, and J. Zehr. 1999. Relationship between landscape characteristics, history, and lakewater acidification in the Adirondack Mountains, New York. *Water Air Soil Pollut.* 112:407-427.
- Sullivan, T.J. 2000. Aquatic Effects of Acidic Deposition. Lewis Publ./CRC Press, Boca Raton, FL.
- Sullivan, T.J. and B.J. Cosby. 2002. Critical loads of sulfur deposition to protect streams within Joyce Kilmer and Shining Rock Wilderness Areas from future acidification. Report for the USDA Forest Service. April 2002.
- Sullivan, T.J., B.J. Cosby, J.R. Webb, K.U. Snyder, A.T. Herlihy, A.J. Bulger, E.H. Gilbert, and D. Moore. 2002. Assessment of the Effects of Acidic Deposition on Aquatic Resources in the Southern Appalachian Mountains. Report prepared for the Southern Appalachian Mountains Initiative (SAMI). E&S Environmental Chemistry, Inc., Corvallis, OR.
- Sullivan, T.J., B.J. Cosby, J.A. Laurence, R.L. Dennis, K. Savig, J.R. Webb, A.J. Bulger, M. Scruggs, C. Gordon, J. Ray, H. Lee, W.E. Hogsett, H. Wayne, D. Miller, and J.S. Kern. 2003. Assessment of Air Quality and Related Values in Shenandoah National Park. NPS/NERCHAL/NRTR-03/090. U.S. Department of the Interior, National Park Service, Northeast Region. [http://www.nps.gov/nero/science/FINAL/shen\\_air\\_quality/shen\\_airquality.html](http://www.nps.gov/nero/science/FINAL/shen_air_quality/shen_airquality.html).
- Sullivan, T.J., B.J. Cosby, A.T. Herlihy, J.R. Webb, A.J. Bulger, K.U. Snyder, P. Brewer, E.H. Gilbert, and D.L. Moore. 2004. Regional model projections of future effects of sulfur and nitrogen deposition on streams in the southern Appalachian Mountains. *Water Resour. Res.* 40: W02101. doi:10.1029/2003WR001998.
- Sullivan, T.J., C.T. Driscoll, B.J. Cosby, I.J. Fernandez, A.T. Herlihy, J. Zhai, R. Stemberger, K.U. Snyder, J.W. Sutherland, S.A. Nierzwicki-Bauer, C.W. Boylen, T.C. McDonnell, and N.A. Nowicki. 2006a. Assessment of the Extent to Which Intensively-Studied Lakes Are Representative of the Adirondack Mountain Region. Final Report 06-17. New York State Energy Research and Development Authority, Albany, NY.
- Sullivan, T.J., I.J. Fernandez, A.T. Herlihy, C.T. Driscoll, T.C. McDonnell, N.A. Nowicki, K.U. Snyder, and J.W. Sutherland. 2006b. Acid-base characteristics of soils in the Adirondack Mountains, New York. *Soil Sci. Soc. Am. J.* 70:141-152.

- Sullivan, T.J., B.J. Cosby, J.R. Webb, R.L. Dennis, A.J. Bulger, and F.A. Deviney Jr. 2008. Streamwater acid-base chemistry and critical loads of atmospheric sulfur deposition in Shenandoah National Park, Virginia. *Environ. Monitor. Assess.* 137:85-99.
- Sullivan, T.J., B.J. Cosby, C.T. Driscoll, T.C. McDonnell, and A.T. Herlihy. 2011. Target loads of atmospheric sulfur deposition protect terrestrial resources in the Adirondack Mountains, New York against biological impacts caused by soil acidification. *J. Environ. Stud. Sci.* 1(4):301-314.
- Sullivan, T.J., B.J. Cosby, C.T. Driscoll, T.C. McDonnell, A.T. Herlihy, and D.A. Burns. 2012. Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York. *Water Resour. Res.* 48 doi:10.1029/2011WR011171.
- Sullivan, T.J., G.B. Lawrence, S.W. Bailey, T.C. McDonnell, C.M. Beier, K.C. Weathers, G.T. McPherson, and D.A. Bishop. 2013. Effects of acidic deposition and soil acidification on sugar maple in the Adirondack Mountains, New York. *Environ. Sci. Technol.* 47:12687-12694. 10.1021/es401864w.
- Sverdrup, H. and P. Warfvinge. 1993. The effect of soil acidification on the growth of trees, grass and herbs as expressed by the (Ca+ Mg+ K)/Al ratio. *Rep. in Ecol. Eng.* 2:1993.
- Thornton, K., D. Marmorek, and P. Ryan. 1990. Methods for Projecting Future Changes in Surface Water Acid-Base Chemistry. National Acid Precipitation Assessment Program, Washington, DC.
- U.S. Environmental Protection Agency. 2008a. National Air Quality Status and Trends through 2007. EPA-454/R-08-006. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Air Quality Assessment Division, Research Triangle Park. NC.
- U.S. Environmental Protection Agency. 2008b. Integrated Science Assessment for Oxides of Nitrogen and Sulfur -- Ecological Criteria. EPA/600/R-08/082F. National Center for Environmental Assessment, Office of Research and Development, Research Triangle Park, NC.
- U.S. Environmental Protection Agency. 2009. Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur: Final. EPA-452/R-09-008a. Office of Air Quality Planning and Standards, Health and Environmental Impacts Division, Research Triangle Park, NC.
- Warby, R.A.F., C.E. Johnson, and C.T. Driscoll. 2009. Continuing acidification of organic soils across the northeastern USA: 1984 - 2001. *Soil Sci. Soc. Am. J.* 73(1):274-284.
- Whitehead, P.G., S. Bird, M. Hornung, J. Cosby, C. Neal, and P. Percios. 1988. Stream acidification trends in the Welsh uplands - a modelling study of the Llyn Brianne catchments. *J. Hydrol.* 101:191-212.
- Whittier, T.R., S.B. Paulsen, D.P. Larsen, S.A. Peterson, A.T. Herlihy, and P.R. Kaufmann. 2002. Indicators of ecological stress and their extent in the population of northeastern lakes: a regional-scale assessment. *BioScience* 52:235-247.

- Wright, R.F., E.T. Gjessing, N. Christophersen, E. Lotse, H.M. Seip, A. Semb, B. Sletaune, R. Storhaug, and K. Wedum. 1986. Project rain: changing acid deposition to whole catchments. The first year of treatment. *Water Air Soil Pollut.* 30:47-64.
- Wright, R.F. and B.J. Cosby. 1987. Use of a process-oriented model to predict acidification at manipulated catchments in Norway. *Atmos. Environ.* 21:727-730.
- Wright, R.F., B.J. Cosby, R.C. Ferrier, A. Jenkins, A.J. Bulger, and R. Harriman. 1994. Changes in the acidification of lochs in Galloway, southwestern Scotland, 1979-1988: the MAGIC model used to evaluate the role of afforestation, calculate critical loads, and predict fish status. *J. Hydrol.* 161:257-285.
- Wright, R.F., B.A. Emmett, and A. Jenkins. 1998. Acid deposition, land-use change and global change: MAGIC7 model applied to Risdalsheia, Norway (RAIN and CLIMEX projects) and Aber, UK (NITREX project). *Hydrol. Earth System Sci.* 2:385-397.

## 6 Glossary

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<b>Acid anion</b>	Negatively charged ion that does not react with hydrogen ion in the pH range of most natural waters.
<b>Acid-base chemistry</b>	The reaction of acids (proton donors) with bases (proton acceptors). In the context of this report, it means the reactions of natural and anthropogenic acids and bases, the result of which is described in terms of pH and acid neutralizing capacity of the system.
<b>Acid cation</b>	Hydrogen ion or metal ion that can hydrolyze water to produce hydrogen ions, e.g., ionic forms of aluminum, manganese, and iron.
<b>Acid neutralizing capacity)</b>	The equivalent capacity of a solution to neutralize strong acids. The components of ANC include weak bases (carbonate species, dissociated organic acids, alumino-hydroxides, borates, and silicates) and strong bases (primarily, OH <sup>-</sup> ). ANC can be measured in the laboratory by the Gran titration procedure or defined as the difference in the equivalent concentrations of the base cations and the mineral acid anions.
<b>Acidic deposition</b>	Transfer of acids and acidifying compounds from the atmosphere to terrestrial and aquatic environments via rain, snow, sleet, hail, cloud droplets, particles, and gas exchange.
<b>Acidic episode</b>	A hydrologic episode in a water body in which acidification of surface water to an acid neutralizing capacity less than or equal to 0 µeq/L occurs.
<b>Acidic lake or stream</b>	A lake or stream in which the acid neutralizing capacity is less than or equal to 0µeq/L.
<b>Acidification</b>	The decrease of acid neutralizing capacity in water or base saturation in soil caused by natural or anthropogenic processes.
<b>Acidified</b>	Pertaining to natural water that has experienced a decrease in acid neutralizing capacity or a soil that has experienced a reduction in base saturation.
<b>Acidophilic</b>	Describes organisms that thrive in an acidic environment.
<b>Aluminum, Total monomeric</b>	Operationally defined simple unpolymerized form of aluminum present in inorganic or organic complexes.
<b>Analyte</b>	A chemical species that is measured in a water sample.
<b>Anion</b>	A negatively charged ion.
<b>Anthropogenic</b>	Of, relating to, derived from, or caused by humans or related to human activities or actions.
<b>Base cation</b>	An alkali or alkaline earth metal cation (Ca <sup>2+</sup> , Mg <sup>2+</sup> , K <sup>+</sup> , Na <sup>+</sup> ); the first three of these are important plant nutrients.



<b>Base cation buffering</b>	The capacity of a watershed soil or a sediment to supply base cations ( $\text{Ca}^{2+}$ , $\text{Mg}^{2+}$ , $\text{K}^+$ , $\text{Na}^+$ ) to receiving surface waters in exchange for acid cations ( $\text{H}^+$ , $\text{Al}^{3+}$ ); may occur through cation exchange in soils or weathering of soil or bedrock minerals.
<b>Base cation supply</b>	The rate at which base cations can be supplied to buffer incoming acid cations; this rate is determined by the relative rate of mineral weathering, the availability of base cations on exchange sites, and the rate of mobile anion leaching.
<b>Base saturation</b>	The proportion of total soil cation exchange capacity that is occupied by exchangeable base cations, i.e., by $\text{Ca}^{2+}$ , $\text{Mg}^{2+}$ , $\text{K}^+$ , and $\text{Na}^+$ .
<b>Bias</b>	A systematic difference (error) between a measured (or predicted) value and its true value.
<b>Biological effects</b>	Changes in biological (organism-, population-, and community-level) structure and/or function in response to some causal agent; also referred to as biological response.
<b>Calibration</b>	Process of checking, adjusting, or standardizing operating characteristics of instruments or coefficients in a mathematical model with empirical data of known quality. The process of evaluating the scale readings of an instrument with a known standard in terms of the physical quantity to be measured.
<b>Catchment</b>	See watershed.
<b>Cation</b>	A positively charged ion.
<b>Cation exchange</b>	The interchange between a cation in solution and another cation on the surface of any surface-active material such as clay or organic matter.
<b>Cation exchange capacity</b>	The sum total of exchangeable cations that a soil can adsorb.
<b>Cation leaching</b>	Movement of cations out of soil, in conjunction with mobile anions in soil solution.
<b>Cation retention</b>	The physical, biological, and geochemical processes by which cations in watersheds are held, retained, or prevented from reaching receiving surface waters.
<b>Chronic acidification</b>	See long-term acidification.
<b>Circumneutral</b>	Close to neutrality with respect to pH (neutral pH= 7); in natural waters, pH6-8.
<b>Conductance</b>	See specific conductance.
<b>Confidence limits</b>	A statistical expression, based on a specified probability, that estimates the upper and/or lower value (limit) or the interval expected to contain the true population mean.
<b>Decomposition</b>	The microbially mediated reaction that converts solid or dissolved organic matter into its constituents (also called decay or mineralization).

<b>Denitrification</b>	Biologically mediated conversion of nitrate to gaseous forms of nitrogen (N <sub>2</sub> , NO, N <sub>2</sub> O); denitrification occurs during decomposition of organic matter.
<b>Dissolved inorganic carbon</b>	The sum of dissolved (measured after filtration) carbonic acid, bicarbonate, and carbonate in a water sample.
<b>Dissolved organic carbon</b>	Organic carbon that is dissolved or unfilterable in a water sample (0.45-µm pore size)
<b>Drainage basin</b>	See watershed.
<b>Drainage lake</b>	A lake that has a permanent surface water inlet and outlet.
<b>Dry deposition</b>	Transfer of substances from the atmosphere to terrestrial and aquatic environments via gravitational settling of large particles and turbulent transfer of trace gases and small particles.
<b>Dynamic model</b>	A mathematical model in which time is included as an independent variable.
<b>Empirical model</b>	Representation of a real system by a mathematical description based on experimental or observational data.
<b>Episodes</b>	A subset of hydrological phenomena known as events. Episodes, driven by rainfall or snowmelt, occur when acidification takes place during a hydrologic event. Changes in other chemical parameters, such as aluminum and calcium, are frequently associated with episodes.
<b>Episodic acidification</b>	The short-term decrease of acid neutralizing capacity from a lake or stream. This process has a time scale of hours to weeks and is usually associated with hydrological events.
<b>Equivalent</b>	Unit of ionic concentration, a mole of charge; the quantity of a substance that either gains or loses one mole of protons or electrons.
<b>Evapotranspiration</b>	The process by which water is returned to the air through direct evaporation or transpiration by vegetation.
<b>Forecast</b>	To estimate the probability of some future event or condition as a result of rational study and analysis of available data.
<b>Frame</b>	A structural representation of a population providing a sampling capability.
<b>Gran analysis</b>	A mathematical procedure used to determine the equivalence points of a titration curve for acid neutralizing capacity.
<b>Ground water</b>	Water in a saturated zone within soil or rock.
<b>Hindcast</b>	To estimate the probability of some past event or condition as a result of rational study and analysis of available data.

<b>Hydraulic residence time</b>	A measure of the average amount of time water is retained in a lake basin. It can be defined on the basis of inflow/lake volume, represented as "RT," or on the basis of outflow (outflow/lake volume) and represented as $T_w$ . The two definitions yield similar values for fast-flushing lakes, but diverge substantially for long-residence time seepage lakes.
<b>Hydrologic(al) event</b>	Pertaining to increased water flow or discharge resulting from rainfall or snowmelt.
<b>Hydrologic(al) flow paths</b>	Surface and subsurface routes by which water travels from where it is deposited by precipitation to where it drains from a watershed.
<b>Hydrology</b>	The study of the waters of the earth--their occurrence, circulation, and distribution; their chemical and physical properties; and their reaction with their environment, including their relationship to living things.
<b>Inorganic aluminum</b>	The sum of free aluminum ions ( $Al^{3+}$ ) and dissolved aluminum bound to inorganic ligands; operationally defined by labile monomeric aluminum.
<b>Labile monomeric aluminum</b>	Operationally defined as aluminum that can be retained on a cation exchange column and measured by one of the two extraction procedures used to measure monomeric aluminum. Labile monomeric aluminum is assumed to represent inorganic monomeric aluminum ( $Al_i$ ).
<b>Liming</b>	The addition of any base materials to neutralize surface water or soil acidity.
<b>Littoral zone</b>	The shallow, near-shore region of a body of water; often defined as the band from the shoreline to the outer edge of the occurrence of rooted vegetation.
<b>Long-term acidification</b>	The decrease of acid neutralizing capacity in a lake or stream over a period of hundreds to thousands of years, generally in response to gradual leaching of ionic constituents.
<b>Mineral acids</b>	Inorganic acids, e.g., $H_2SO_4$ , $HNO_3$ , $HCl$ , $H_2CO_3$ .
<b>Mineralization</b>	Process of converting organic nitrogen in the soil into ammonium, which is then available for biological uptake.
<b>Mineral weathering</b>	Dissolution of rocks and minerals by chemical and physical processes.
<b>Mitigation</b>	Generally described in the context of acidification as amelioration of adverse impacts caused by acidic deposition at the source (e.g., emissions reductions) or the receptor (e.g., lake liming).
<b>Mobile anions</b>	Anions that flow in solutions through watershed soils, wetlands, streams, or lakes without being adsorbed or retained through physical, biological, or geochemical processes.
<b>Model</b>	An abstraction or representation of a prototype or system.

<b>Monomeric aluminum</b>	Aluminum that occurs as a free ion ( $\text{Al}^{3+}$ ), simple inorganic complexes (e.g., $\text{Al}(\text{OH})_n^{3-n}$ , $\text{AlF}_n^{3-n}$ ), or simple organic complexes, but not in polymeric forms; operationally, extractable aluminum measured by the pyrocatechol violet method or the methyl-isobutyl ketone method (also referred to as the oxine method) is assumed to represent total monomeric aluminum. Monomeric aluminum can be divided into labile and non-labile components using cation exchange columns.
<b>Monte Carlo method</b>	Technique of stochastic sampling or selection of random numbers to generate synthetic data.
<b>Natural acids</b>	Acids produced within terrestrial or aquatic systems through natural, biological, and geochemical processes; i.e., not a result of acidic deposition or deposition of acid precursors.
<b>Nitrification</b>	Oxidation of ammonium to nitrite or nitrate by microorganisms.
<b>Nitrogen fixation</b>	Biological conversion of elemental nitrogen ( $\text{N}_2$ ) to organic N.
<b>Nitrogen saturation</b>	Condition whereby nitrogen inputs to an ecosystem exceed plant uptake requirements.
<b>Non-labile monomeric aluminum</b>	Operationally defined as aluminum that passes through a cation exchange column and is measured by one of the two extraction procedures used to measure monomeric aluminum; assumed to represent organic monomeric aluminum ( $\text{Al}_o$ ).
<b>Organic acids</b>	Heterogeneous group of acids generally possessing carboxyl ( $-\text{COOH}$ ) or phenolic ( $\text{C}-\text{OH}$ ) groups; includes fulvic and humic acids.
<b>Organic aluminum</b>	Aluminum bound to organic matter, operationally defined as that fraction of aluminum determined after sample is passed through a cation exchange column.
<b>Parameter</b>	(1) a characteristic factor that remains at a constant value during the analysis, or (2) a quantity that describes a statistical population attribute.
<b>pH</b>	The negative logarithm of the hydrogen ion activity. The pH scale is generally presented from 1 (most acidic) to 14 (most alkaline); a difference of one pH unit indicates a ten-fold change in hydrogen ion activity.
<b>Physiography</b>	The study of the genesis and evolution of land forms; a description of the elevation, slope, and aspect of a study area.
<b>Plankton</b>	Plant or animal species that spend part or all of their lives in open water.
<b>Pool</b>	In ecological systems, the supply of an element or compound, such as exchangeable or weatherable cations or adsorbed sulfate.

<b>Population</b>	For the purpose of this report, (1) the total number of lakes within a given geographical region or the total number of lakes with a given set of defined chemical, physical, or biological characteristics; or (2) an assemblage of organisms of the same species inhabiting a given ecosystem.
<b>Precision</b>	A measure of the capacity of a method to provide reproducible measurements of a particular analyte (often represented by variance).
<b>Probability sample</b>	A sample in which each unit has a known probability of being selected.
<b>Project</b>	To estimate future possibilities based on rational study and current conditions or trends.
<b>Quality assurance</b>	A system of activities for which the purpose is to provide assurance that a product (e.g., data base) meets a defined standard of quality with a stated level of confidence.
<b>Quality control</b>	Steps taken during sample collection and analysis to ensure that data quality meets the minimum standards established in a quality assurance plan.
<b>Regionalization</b>	Describing or estimating a characteristic of interest on a regional basis.
<b>Retention time</b>	The estimated mean time (usually expressed in years) that water resides in a lake prior to leaving the system. (See hydraulic residence time.)
<b>Scenario</b>	One possible deposition sequence following implementation of a control or mitigation strategy and the subsequent effects associated with this deposition sequence.
<b>Short-term acidification</b>	See episode.
<b>Simulation</b>	Description of a prototype or system response to different conditions or inputs using a model rather than actually observing the response to the conditions or inputs.
<b>Simulation model</b>	Mathematical model that is used with actual or synthetic input data, or both, to produce long-term time series or predictions.
<b>Species richness</b>	The number of species occurring in a given aquatic ecosystem, generally estimated by the number of species caught using a standard sampling regime.
<b>Specific conductance</b>	The conductivity between two plates with an area of 1 cm <sup>2</sup> across a distance of 1 cm at 25 °C; a measurement that reflects the ionic strength of a solution.
<b>Steady state</b>	The condition that occurs when the sources and sinks of a property (e.g., mass, volume, concentration) of a system are in balance (e.g., inputs equal outputs; production equals consumption).
<b>Stratified design</b>	A statistical design in which the population is divided into strata, and a sample selected from each stratum.

<b>Strong acid anion sum (SAA or C<sub>A</sub>)</b>	Refers to the equivalent sum of SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup> , Cl <sup>-</sup> , and F <sup>-</sup> . The term specifically excludes organic acid anions.
<b>Strong acids</b>	Acids with a high tendency to donate protons or to completely dissociate in natural waters, e.g., H <sub>2</sub> SO <sub>4</sub> , HNO <sub>3</sub> , HCl, and some organic acids. (See acid anions.)
<b>Strong bases</b>	Bases with a high tendency to accept protons or to completely dissociate in natural waters, e.g., NaOH.
<b>Subpopulation</b>	Any defined subset of the target population.
<b>Sulfate adsorption</b>	The process by which sulfate is chemically exchanged (e.g., for OH <sup>-</sup> ) or adsorbed onto positively charged sites on the soil matrix; under some conditions this process is reversible, and the sulfate may be desorbed.
<b>Sulfate reduction</b>	(1) the conversion of sulfate to sulfide during the decomposition of organic matter under anaerobic conditions (dissimilatory sulfate reduction) and (2) the formation of organic compounds containing reduced sulfur compounds (assimilatory sulfate reduction).
<b>Sulfate retention</b>	The physical, biological, and geochemical processes by which sulfate in watersheds is held, retained, or prevented from reaching receiving surface waters.
<b>Sum of base cations (SBC or C<sub>B</sub>)</b>	Refers to the equivalent sum of Ca <sup>2+</sup> , Mg <sup>2+</sup> , Na <sup>+</sup> , and K <sup>+</sup> . The term specifically excludes cationic Al <sup>n+</sup> and Mn <sup>2+</sup> .
<b>Surficial geology</b>	Characteristics of the earth's surface, especially consisting of unconsolidated residual, colluvial, alluvial, or glacial deposits lying on the bedrock.
<b>Target population</b>	A subset of a population explicitly defined by a given set of exclusion criteria to which inferences are to be drawn from the sample attributes.
<b>Turnover</b>	The interval of time in which the density stratification of a lake is disrupted by seasonal temperature variation, resulting in the entire water mass becoming mixed.
<b>Variable</b>	A quantity that may assume any one of a set of values during analysis.
<b>Watershed</b>	The geographic area from which surface water drains into a particular lake or point along a stream.
<b>Weak acids</b>	Acids with a low proton-donating tendency that tend to dissociate only partially in natural waters, e.g., H <sub>2</sub> CO <sub>3</sub> , H <sub>4</sub> SiO <sub>4</sub> , and most organic acids. (See acid anions.)
<b>Weak bases</b>	Bases with a low proton-accepting tendency that tend to dissociate only partially in natural waters, e.g., HCO <sub>3</sub> <sup>-</sup> , Al(OH) <sub>4</sub> <sup>-</sup> .
<b>Wet deposition</b>	Transfer of substances from the atmosphere to terrestrial and aquatic environments via precipitation, e.g., rain, snow, sleet, and hail.

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State of New York  
Andrew M. Cuomo, Governor

# Critical Loads of Sulfur and Nitrogen for Protection of Acid-Sensitive Aquatic and Terrestrial Resources in the Adirondack Mountains

Final Report  
March 2014

New York State Energy Research and Development Authority  
Richard L. Kauffman, Chair | John B. Rhodes, President and CEO