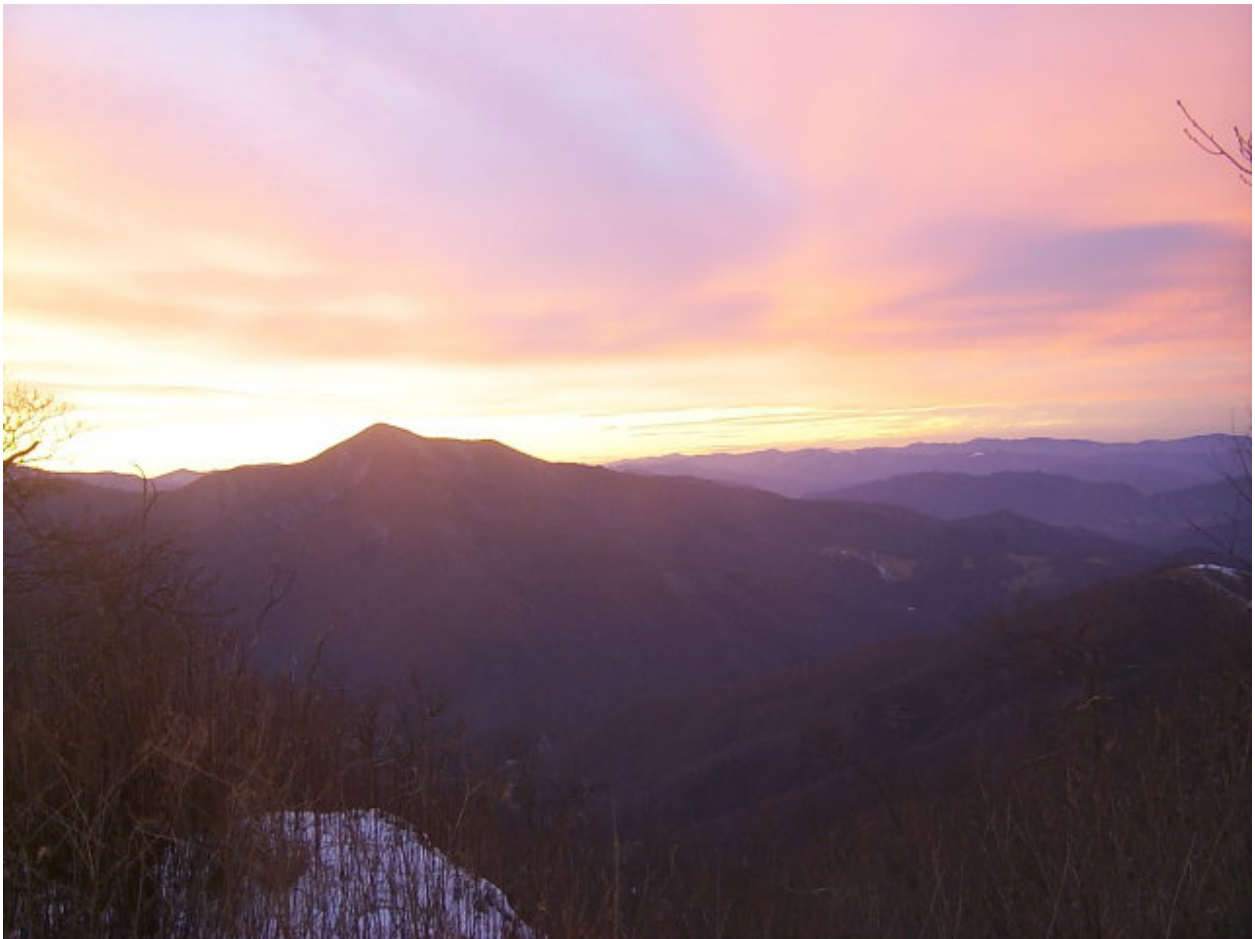


**Model-Based Assessment of the Effects of Acidic Deposition
on Sensitive Watershed Resources in the National Forests of
North Carolina, Tennessee, and South Carolina**

T.J. Sullivan, B.J. Cosby, K.U. Snyder, A.T. Herlihy, B. Jackson



October, 2007

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FINAL REPORT

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October, 2007

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Table of Contents

List of Figures	v
List of Tables	vii
Executive Summary	ix
I. Background	1
II. Objectives	5
III. Approach	6
A. Site Selection and Data Compilation	6
B. Model Application	12
1. Specification of Stream and Soil Data	13
2. Specification of Deposition and Meteorology Data	16
a. Wet Deposition Data (Reference Year and Calibration Values)	18
b. Dry and Occult Deposition Data and Historical Deposition Sequences	18
3. Protocol for MAGIC Calibration and Simulation at Individual Sites	19
4. Future Scenario Projections	21
a. Utilizing the VISTAS Results	21
b. Spatial Data	23
c. Future Scenarios	24
5. Critical Loads Analysis	26
6. Sensitivity of Model Output to Major Uncertainties	27
a. Uncertainty Due to Specification of Soils Data	28
b. Uncertainty Due to Specification of High Elevation Occult Deposition	29
c. Uncertainty Due to Specification of Stream Water Data for Calibration	29
d. Combined Model Calibration and Simulation Uncertainty	30
7. Development of Landscape Variables for Regionalization of Modeling Results	31
IV. Results and Discussion	32
A. Model Calibration	32
1. Calibration Data	37
2. Calibration Results	37
B. Hindcast Simulation Results	44
C. Future Simulation Results in Response to Emissions Control Scenarios	44
D. Model Estimates of Critical Loads	60
E. Characteristics of Modeled Sites	77
F. Regression Modeling to Estimate Critical Load at Locations That Were Not Modeled	77
G. Regionalization of Modeling Results	84
1. Statistical Frame	84
2. Watersheds Modeled for this Study	88
H. Episodic Variability in Stream Chemistry	93
1. Changes in ANC During Storm Events	93
2. Mechanisms of Episodic Acidification	96
3. Effects of Episodes on Biota	101
4. Summary of Episodic Effects	103
I. Model Uncertainty	104
1. Sensitivity analysis of simulation results and critical loads estimates	104
a. Sensitivity of Model Outputs to Specification of Stream Water Data	105

b.	Sensitivity of Model Outputs to Specification of Soils Data	110
c.	Sensitivity of Model Outputs to Specification of Occult Deposition.....	114
2.	Uncertainty in Model Simulations and Critical Loads Estimates	117
a.	Uncertainty in Simulated Variables	121
b.	Uncertainty in Critical Loads Estimates.....	123
J.	Effects of Streamwater Acidification on Aquatic Biota in the Southeastern U.S. ...	124
1.	Effects on Brook Trout.....	125
2.	Sublethal Effects on Fish.....	127
3.	Effects on Fish Species Richness	130
4.	Acidification Effects on Aquatic Invertebrates	139
5.	Species – ANC Relationships for Aquatic Invertebrates	142
V.	References Cited	148
VI.	Appendices	
Appendix A.	Estimated Total Deposition of Major Ions at Each Modeling Site for the Future Years 2009 and 2018	
Appendix B.	Hindcast Simulation Results for Selected Years in the Past	
Appendix C.	Time Traces of Simulated Values for Major Ions in Each of the 66 Study Watersheds	
Appendix D.	Deposition Unit Conversion Factors	
Appendix E.	Map Showing Locations of Upper and Lower Node National Stream Survey Sample Sites Within the Study Region	

List of Figures

1.	Glide path estimated by Visibility Improvement State and Tribal Association of the Southeast (VISTAS) to attain natural background visibility for Great Smoky Mountains National Park and Joyce Kilmer-Slickrock Wilderness by 2064 on the days classified as having the worst visibility conditions.....	23
2.	Average rate of change in visibility for three Class I areas utilizing the Environmental Protection Agency’s Models-3 Community Multiscale Air Quality (CMAQ) Modeling System.....	25
3.	Location of sites selected for modeling, coded by tier. Tier I sites had both soil and stream chemistry data for calibration	33
4.	Calibration results for the MAGIC model.....	46
5.	MAGIC estimates of historical change in CALK for the 66 modeled sites for multiple points in time	50
6.	CMAQ model wet deposition for the year 2002 at each modeling site location compared with three-year average (1998-2000) interpolated measurements from NADP/NTN.....	52
7.	Estimates of wet deposition under the three scenarios of emissions control	53
8.	Estimates of dry deposition under the three scenarios of emissions control.....	54
9.	Estimates of total (wet + dry) deposition under the three scenarios of emissions control.....	55
10.	Cumulative frequency distributions and histograms of simulated changes in a) stream ANC, b) soil percent base saturation, c) stream sum of base cations, and d) stream sulfate concentration.....	61
11.	Comparison of the extent to which various ANC criteria (0, 20, 50, 100 $\mu\text{eq/L}$) are simulated to be achievable in the year 2040 and 2100.....	71
12.	Site-by-site comparison of differences in simulated critical load of S (kg S/ha/yr) for the year 2040, depending on whether the target stream ANC value was selected to be 20 or 50 $\mu\text{eq/L}$	72
13.	Critical loads of S for the 66 modeled sites to achieve 4 different streamwater ANC levels (0, 20, 50 and 100 $\mu\text{eq/L}$), evaluated in 3 different years (2020, 2040 and 2100).....	74
15.	Distribution of modeling sites across key parameter values	78
16.	Predicted critical load (kg S/ha/yr), using five-variable empirical models based on landscape variables, versus MAGIC model estimates of critical loads.....	82
17.	Predicted critical load (kg S/ha/yr), using five-variable empirical models based on both landscape variables and water chemistry, versus MAGIC model simulated critical loads for the ANC critical level of 0 and 20 $\mu\text{eq/L}$	83
18.	Map of SAMI study area showing location of Class I areas and physiographic provinces	86
19.	Estimated number of stream segments by 7.5” (1:24,000 scale) topographic map Strahler order in the portion of the Southern Blue Ridge Province located in NC, TN, and SC, based on National Stream Survey Data	87
20.	Cumulative distribution function of sample site elevation for the population of NSS streams located in the Southern Blue Ridge Province in NC, TN, and SC.....	88
21.	Cumulative distribution functions for upper and lower node NSS streams in the portion of the Southern Blue Ridge Province located in NC, TN, and SC	89

22.	Map showing locations of streams surveyed by the USDA Forest Service for acid-base chemistry	91
23.	Locations of streams recently surveyed by the USDA Forest Service, showing stream ANC class in relation to the acid-sensitive geologic/elevational zone described by Sullivan et al. (2002b)	92
24.	Minimum streamwater ANC sampled at each site during each year versus median spring ANC for all samples collected at that site during that spring season	94
25.	Episodic stream chemistry data for Upper Creek in Pisgah National Forest	97
26.	Relationship between ANC and runoff for streamwater samples collected at intensively-studied sites in Shenandoah National Park.....	100
27.	Decrease in ANC and pH and increase in dissolved aluminum in response to a sharp increase in streamflow in three watersheds within Shenandoah National Park during a hydrological episode in 1995	102
28.	Sensitivity of model simulations to specification of stream water data	107
29.	Sensitivity of estimated critical loads to specification of stream water data.....	108
30.	Sensitivity of model simulations to specification of soils data	111
31.	Sensitivity of estimated critical loads to specification of soils data.....	113
32.	Sensitivity of model simulations to specification of occult deposition data	116
33.	Sensitivity of estimated critical loads to specification of occult deposition data.....	118
34.	Length-adjusted condition factor (K), a measure of body size in blacknose dace (<i>Rhinichthys atratulus</i>), compared with mean stream pH among 11 populations in Shenandoah National Park	129
35.	Number of fish species among 13 streams in SHEN	135
36.	Average number of families of aquatic insects in a sample for each of 14 streams in SHEN versus the mean or minimum ANC of each stream	145
37.	Average total number of individuals of aquatic insects in a sample for each of 14 streams in SHEN versus the mean or minimum ANC of each stream.....	147
38.	Average EPT index in a sample for each of 14 streams in SHEN versus the mean or minimum ANC of each stream.....	148

List of Tables

1.	Stream sites deleted from consideration as candidates for modeling.....	9
2.	Location of streams selected for modeling.....	10
3.	Location of streams selected for modeling to characterize a wilderness	12
4.	Watershed location from which soil data were borrowed for Tier II and Tier III modeling sites.....	15
5.	Columns from Environmental Protection Agency’s Models-3 Community Multiscale Air Quality (CMAQ) results summed to obtain the total sulfur, nitrate-nitrogen, and ammonia-nitrogen dry and wet deposition.....	24
6.	Slope intercept value (a) for the linear equation ($y = ax + b$) to describe the relationship between the year (x) and the estimated visibility metric called deciview	25
7.	Streamwater chemistry data used in model calibration.....	34
8.	Summary statistics for the 66 modeled sites for selected key variables.....	36
9.	Model input soil data.....	38
10.	Rainfall amount, precipitation concentrations of all ions, and dry deposition factors for SO ₄ , NO ₃ and NH ₄ in the simulation Reference Year 2005	40
11.	Total deposition (wet plus dry plus cloud) of ions at each modeled site for the reference year 2005	42
12.	Distribution of simulated and observed water chemistry and soil parameter values for the 66 modeled sites	45
13.	Simulated 1860 concentrations of various ions in streamwater for the modeled national forest streams.....	49
14.	Modeled past stream ANC (µeq/L) at three different points in time, in relation to four commonly used target ANC levels.....	51
15.	Simulated stream sulfate concentration (µeq/L) in 66 modeled streams in the past, present, and future under three scenarios of future emissions controls.....	56
16.	Simulated change in stream sulfate concentration (µeq/L) in the future (compared with values in 2005) under three scenarios of future emissions controls.....	58
17.	Simulated stream CALK (µeq/L) in 66 modeled streams in the past, present, and future under three scenarios of future emissions controls.....	65
18.	Simulated change in stream CALK (µeq/L) in the future (compared with values in 2005) under three scenarios of future emissions controls	67
19.	MAGIC critical loads results for the 66 study watersheds.....	69
20.	Achievability of ANC (µeq/L) endpoints in a variety of future years for 66 modeled streams.....	76
21.	List of variables used in empirical models to estimate critical load from landscape characteristics	79
22.	Selected multiple regression models to predict critical load for the year 2040 from either watershed variables only, or from a combination of stream chemistry and watershed variables	80
23.	Distribution of sites by measured current ANC value within modeled critical load categories.....	81
24.	Population statistics for the NSS upper node stream population in the Southern Blue Ridge	87
25.	Population estimates of stream segment condition in the portion of the Southern Blue Ridge Province that occurs within NC, TN, and SC from NSS data	90

26.	Prevalence of acidic, low-ANC, and low-pH streams on national forest lands included in recent surveys of stream chemistry of 256 stream reaches located in western North Carolina, eastern Tennessee, and South Carolina.....	92
27.	Sensitivity analysis results for stream water inputs, expressed as average percent change in simulated values resulting from recalibration of the model using alternate stream water data.....	109
28.	Sensitivity analysis results for stream water inputs, expressed as average percent change in estimated critical load values resulting from recalibration of the model using alternate stream water data	110
29.	Sensitivity analysis scenario results for soil inputs.....	114
30.	Sensitivity analysis critical loads results for soil inputs.....	114
31.	Sensitivity analysis scenario results for occult deposition inputs	119
32.	Sensitivity analysis critical loads results for occult deposition inputs	119
33.	Uncertainty widths for selected simulated variables and selected years.....	122
34.	Uncertainty widths for estimated critical loads.....	123
35.	Brook trout acidification response categories developed by Bulger et al. (2000).....	126
36.	Electrofishing stations in the St. Marys River watershed, Augusta County, Virginia ...	131
37.	Fish distribution in St. Marys River by sample year and sample station	132
38.	Critical pH thresholds for fish species which might be expected to occur within the study area.....	132
39.	Correlation of three physical stream parameters and fish species numbers on the Cherokee National Forest.....	136
40.	Median streamwater ANC and watershed area of streams used by Bulger et al. (1999) to evaluate the relationship between ANC and fish species richness.....	138
41.	Minimum, average and maximum ANC values in the 14 SHEN study streams during the period 1988 to 2001 for all quarterly samples.....	143

EXECUTIVE SUMMARY

The USDA Forest Service is concerned about the current and future health of terrestrial and aquatic resources within the Blue Ridge Province of the Southern Appalachian Mountains in western North Carolina, eastern Tennessee, and the upstate of South Carolina. Soils within some of these watersheds are inherently low in base cations. Adequate amounts of available calcium, magnesium, and potassium are essential to maintain healthy vegetation, and calcium is sometimes a factor limiting the development of healthy aquatic organisms. There is also a concern that the rate of base cation loss from the soil in the more acid-sensitive watersheds has been accelerated by the deposition of sulfur and nitrogen compounds from the atmosphere, and that some soil may be approaching a state of base cation depletion.

One way to measure the ability of a watershed to buffer acid inputs and the adequacy of the soil base cation supply is the acid-neutralizing capacity (ANC) of the drainage water. ANC equal to zero is defined as acidic. Streams having ANC values greater than 50 microequivalents per liter ($\mu\text{eq/L}$) are considered by many researchers in the southeastern United States to have adequate buffering capacity to offset the deposition of sulfur and nitrogen compounds, but some highly sensitive aquatic organisms may be adversely impacted at ANC values near or below 50 $\mu\text{eq/L}$. Low streamwater ANC (i.e., below 50 $\mu\text{eq/L}$) can signal the possibility of base-poor watershed soils. There are existing data on many hundreds of streams within the region, and these data provide the foundation for both aquatic and terrestrial resource evaluation.

An important tool in the evaluation of acidification damage to aquatic and terrestrial ecosystems is the critical load. This approach has been used extensively in Europe and Canada for characterizing the sensitivity of lake, stream, and forest soil resources to acidification damage. The critical load can be defined as the level of acidic deposition below which ecological damage would not be expected to occur, according to current scientific understanding. The critical load for protection of aquatic biota is generally based on maintaining surface water ANC at an acceptable level.

This study included calibration and application of the watershed model, MAGIC, to estimate the sensitivity of 66 watersheds in the Southern Blue Ridge province to changes in atmospheric sulfur deposition. The principal objectives of the research reported here were to:

1. Use MAGIC to estimate future trends for stream chemistry and soil base saturation under a range of future atmospheric deposition scenarios.
2. Use MAGIC to estimate changes in stream chemistry and soil base saturation since pre-industrial time.

3. Develop an approach to predict critical load or future stream ANC for a new stream, based on the known current stream water chemistry, geology, elevation, and forest cover of the catchment.
4. Estimate the critical loads of sulfur deposition needed to protect those streams that are not yet acidic, and to restore streams that are already acidic to specific chemical criteria values.
5. Evaluate model uncertainty and regional representativeness of the modeled streams.
6. Assess, based on available data, episodic variability in stream chemistry and biological dose-response relationships within the study area.

MAGIC model simulations predicted that stream ANC values were above 20 $\mu\text{eq/L}$ in all modeled watersheds in 1860, but below 50 $\mu\text{eq/L}$ in 38% of the watersheds and below 100 $\mu\text{eq/L}$ in 86% percent of the watersheds at that time. The minimum simulated ANC in 1860 among the modeled streams was 30 $\mu\text{eq/L}$. These hindcast water chemistry results are important, not only to assess the extent to which resources have been damaged by air pollution, but also to provide constraints regarding expectations for future chemical recovery. The hindcast simulation results suggested that the average of the modeled streams was acidified from $\text{ANC}=65 \mu\text{eq/L}$ in 1860 to $\text{ANC}=36 \mu\text{eq/L}$ in 2005.

In general, the model projected that soil and stream chemistry have changed substantially since pre-industrial times, but that future changes in response to emissions controls will be small. Simulation results suggested that modeled watersheds would not change to a large degree with respect to stream ANC or soil % base saturation, depending on the extent to which emissions are reduced in response to three emissions control scenarios (Base Case, Moderate, and Aggressive Additional Controls).

Site-to-site variability in critical loads was very high. Estimated critical loads for S deposition ranged from less than zero (ecological objective not attainable) to more than 1,000 kg S/ha/yr, depending on the selected site, ANC endpoint, and evaluation year. Thus, for some sites, one or more of the selected target ANC critical levels (0, 20, 50, 100 $\mu\text{eq/L}$) could not be achieved by the year 2100 (or alternative evaluation year) even if S deposition was reduced to zero and maintained at that level throughout the simulation. For other sites, the watershed soils contained sufficiently large buffering capacity that even very high sustained levels of atmospheric S deposition would not reduce stream ANC below common damage threshold criteria values.

With respect to selection of targets for land management, it appears that neither $\text{ANC}=0$ nor $\text{ANC}=100 \mu\text{eq/L}$ would be particularly useful. Almost all sites can maintain ANC above 0

$\mu\text{eq/L}$ without reducing S deposition below current values, and it has been well demonstrated that a variety of adverse ecological effects occur at such low ANC. A value of $\text{ANC} = 100 \mu\text{eq/L}$ is generally not attainable within 100 years, irrespective of the level of S deposition, and most of the study streams had ANC below $100 \mu\text{eq/L}$ prior to the onset of acidic deposition. Therefore, $\text{ANC} = 100 \mu\text{eq/L}$ is not an appropriate management target. ANC criteria values equal to 20 and $50 \mu\text{eq/L}$ are often achievable within a reasonable time frame and are associated with lower levels of ecological harm than $\text{ANC} = 0$.

Higher critical loads can be tolerated for some streams if one is willing to wait to 2100 to achieve the critical ANC target level, as compared with more stringent deposition reductions required to attain specified ANC values by 2020 or 2040. For other streams, higher critical loads can be tolerated for short-term protection versus more stringent deposition reductions required to protect ecosystems for a longer period of time. Higher critical loads are allowable if one wishes to prevent acidification to $\text{ANC} = 20 \mu\text{eq/L}$ (episodic acidification effects on brook trout likely) than if one wishes to be more restrictive and prevent acidification to ANC below $50 \mu\text{eq/L}$ (biological effects of acidification likely on biota other than brook trout).

The calculated critical loads of S deposition required to prevent streamwater acidification to ANC values below 0 and $20 \mu\text{eq/L}$ varied as a function of current ANC. These model data suggest that most modeled streams that had $\text{ANC} \leq 20 \mu\text{eq/L}$ would require low critical load values to maintain ANC above $20 \mu\text{eq/L}$ in the future. At low ANC values, critical loads were consistently near zero; at higher ANC values, critical loads were more variable. Some streams having ANC above $50 \mu\text{eq/L}$ exhibited low critical loads, whereas others had critical loads much higher than current deposition levels. Stronger relationships were observed when we plotted modeled critical load as a function of the ratio of streamwater ANC to streamwater SO_4^{2-} concentration. Watersheds having the lowest ANC and the highest SO_4^{2-} concentrations in streamwater had the lowest critical loads. This was especially true for calculations of the critical loads to achieve ANC values of 0 and $20 \mu\text{eq/L}$. In this analysis, the ANC reflects the current acid-base status, and the SO_4^{2-} concentration reflects the extent to which SO_4^{2-} is mobile within the watershed, as opposed to being retained on the soil. Lower critical loads occur where ANC is low and SO_4^{2-} is more mobile.

Values of annual average or spring season baseflow water chemistry are typically used to represent conditions at a given stream for purposes of characterization. However, streamwater chemistry undergoes substantial temporal variability, especially in association with hydrological

episodes. During such episodes, which are driven by rainstorms and/or snowmelt events, both discharge (streamflow volume per unit time) and water chemistry change, sometimes dramatically. This is important because streams may in some cases exhibit baseflow chemistry that is suitable for biota, but experience occasional episodic acidification with lethal consequences. Model projections of future streamwater chemistry response to acidic deposition are typically based on chronic chemistry. When interpreting model projections of chronic chemistry, it is important to also consider the likelihood of episodic excursions of water chemistry that are more acidic than is found during more typical baseflow periods.

We evaluated the extent to which the 66 study watersheds were representative of the broader region. For this analysis, we recalculated National Stream Survey (NSS) population statistics to generally match the Forest Service proclamation boundary that is the subject of this report. We defined the Southern Blue Ridge study area as the portion of the Omernik Level III ecoregion number 66 (Blue Ridge Mountains) that is located in the states of North Carolina, South Carolina, and Tennessee. Based on our analysis of the NSS data, there are 1,408 upper stream nodes and 1,404 lower stream nodes in the Southern Blue Ridge study area with an estimated stream length of 7,430 km.

Recent Forest Service surveys of 256 streams within the study region have found that acidic and low-ANC streams are much more prevalent than was represented by NSS for the Southern Blue Ridge Province. These recent Forest Service surveys were not statistically based, but do show widespread occurrence of streams having low ANC and pH. In general, these recently surveyed streams were located at higher elevation than the NSS streams. About 25% of the population of upper node NSS stream reaches in the Southern Blue Ridge Province within NC, TN, and SC were located at an elevation above 1,200 m. NSS watersheds were generally small, with 75% of the upper node sites having watersheds smaller than 11.1 km² (1,110 ha). Streams recently sampled by the Forest Service had smaller watersheds (median 143 ha), but were located at similar elevation, with 25% of the samples located at an elevation above 1,150 m. These recently surveyed stream sites commonly had low ANC and pH. Overall, 5% of the surveyed streams were chronically acidic, and 49% had ANC \leq 50 μ eq/L. Eleven percent had pH $<$ 6. Surveyed streams that were acidic (ANC \leq 0) or low in ANC ($<$ 50 μ eq/L) were located within the acid-sensitive areas within the Southern Appalachian Mountains that were mapped in conjunction with the Southern Appalachian Mountains Initiative (SAMI) Assessment on the basis of bedrock geology and elevation.

In order to aid in the process of extrapolating MAGIC model critical loads simulation results to watersheds within the study area that were not modeled, we developed a suite of multiple regression models to estimate critical loads from variables that are more widely available across the region than are the MAGIC model results. Separate multiple regression modeling efforts were conducted to estimate critical load from 1) landscape variables represented spatially in the GIS, 2) a combination of landscape variables and stream chemistry variables, and 3) streamwater chemistry only. Based on these relationships, critical loads can be estimated for the broader region.

I. BACKGROUND

The USDA Forest Service (FS) is concerned about the current and future health of terrestrial and aquatic resources within the Blue Ridge Province of the Southern Appalachian Mountains in western North Carolina, Eastern Tennessee, and the upstate of South Carolina. Soils within these watersheds have developed from the slow breakdown of parent rock material which can be inherently low in base cations. Adequate amounts of available calcium, magnesium, and potassium are essential to maintain healthy vegetation, and calcium is sometimes a factor limiting the maintenance of healthy aquatic organisms. These base cations are stored in vegetation (especially trees) and are cycled through the soils as vegetation decomposes. Base cations also become dissolved in the soil water and are transported into streams to be utilized by aquatic organisms.

Prior to the 1900s, there was minimal disturbance to the forests in uplands of the southern Blue Ridge Mountains with small areas cleared for agriculture and occasional wildland fires. Soil losses from erosion, and hence removal of base cations, are believed to have occurred in some locations when forests of western North Carolina were commercially harvested. In some cases, the soil losses are believed to have been substantial, for example if a devastating wildfire occurred in the logging slash. It has been estimated that seven feet of soil was lost following logging and two wildfires in the present day Shining Rock Wilderness (Vanderzanden et al., 1999).

There is also a concern that the rate of base cation loss has been accelerated by the deposition of sulfur and nitrogen compounds from the atmosphere, and that some soil may be approaching a state of base cation depletion. One way to measure the ability of a watershed to buffer acid inputs and the adequacy of the soil base cation supply is the acid-neutralizing capacity (ANC) of the drainage water. ANC can be calculated as the sum of the base cations minus the sum of the mineral acid anions (sulfate, nitrate, and chloride). The calculated ANC is also referred to by some people as the calculated alkalinity (CALK). Streams having ANC values of greater than 50 microequivalents per liter ($\mu\text{eq/L}$) are considered by many researchers in the southeastern United States to have adequate buffering capacity to offset the future deposition of sulfur and nitrogen compounds, but some highly sensitive aquatic organisms may be adversely impacted at ANC values near 50 $\mu\text{eq/L}$. ANC values less than 0 are considered acidic and many aquatic organisms would generally not occur under such conditions, including native brook trout. Marginal brook trout populations may be present in streams having ANC

between 0 and 20 $\mu\text{eq/L}$, but these streams are highly sensitive to periodic (episodic) acidification to ANC values below zero. Streams having ANC between 20 and 50 $\mu\text{eq/L}$ are potentially sensitive to chronic and episodic acidification and brook trout populations may or may not occur (Sullivan et al. 2002a).

The FS in western North Carolina (Pisgah and Nantahala National Forests), eastern Tennessee (Cherokee National Forest), and the upstate of South Carolina (Andrew Pickens Ranger District, Sumter National Forest) have been monitoring surface waters to determine if acid deposition could be adversely impacting the health of forested watersheds. Many streams in the national forests in this region show signs of acidification from atmospheric deposition, including streams in Class I areas that are administered by the Forest Service - Linville Gorge, Joyce Kilmer-Slickrock, and Shining Rock Wilderness Areas.

Ecosystem sensitivity to acidification and the potential effects of sulfur deposition on surface water quality have been well-studied in the southeastern United States, particularly within the National Acid Precipitation Assessment Program (NAPAP), the Fish in Sensitive Habitats (FISH) project, and the Southern Appalachian Mountains Initiative (SAMI). Major findings were summarized in a series of State of Science and Technology Reports (e.g., Sullivan 1990, Baker et al. 1990a,b), NAPAP Integrated Assessment (NAPAP 1991), the SAMI effects reports (Sullivan et al. 2002a,b), and the FISH report (Bulger et al. 1999). Although aquatic effects from nitrogen deposition have not been studied as thoroughly as those from sulfur deposition, concern has been expressed regarding the role of NO_3^- in acidification of surface waters, particularly during hydrologic episodes (e.g., Sullivan 1993, 2000; Sullivan et al. 1997; Wigington et al. 1993).

Soil and drainage water acidification developed in this region over a period of many decades in response to high levels of atmospheric sulfur and nitrogen deposition. However, sulfur deposition has been declining throughout the eastern United States since about the late 1970s and further decreases are expected in the future. More information is needed regarding the watershed responses that should be expected. There is also concern that base cations in the soil are being depleted in watersheds where stream acidification is occurring. This could adversely impact terrestrial site productivity. Therefore, the forest managers should be concerned with the following:

1. What will be the future trend in stream acid neutralizing capacity (ANC) as acid deposition continues to decrease in the eastern United States?

2. What was the historical stream chemistry and watershed soil base saturation for streams that have been surveyed?
3. Can the future stream ANC or extent of acid-sensitivity be estimated for a new stream based on available information on the geology, elevation, and forest cover of the catchment?
4. What level of sulfur deposition would be needed to protect those streams that are not yet acidic, and to restore streams that are already acidic or very low in ANC?

Computer models can be used to predict pollution effects on ecosystems and to perform simulations of future ecosystem response. The MAGIC (Model of Acidification of Groundwater in Catchments) model, a lumped-parameter, mechanistic model, has been widely used throughout North America and Europe to project stream water response. The Southern Appalachian Mountains Initiative (SAMI) used MAGIC to assess stream chemistry response to various emission reduction strategies throughout the southern Appalachian Mountains region (Sullivan et al. 2002a). MAGIC has also been used recently by the Shenandoah National Park (Sullivan et al. 2003) and Monongahela National Forest (Sullivan and Cosby 2004), to determine the sulfur deposition level at which unacceptable environmental damage would be expected to occur. ANC is the stream chemistry parameter most commonly used to identify various levels of environmental damage.

The need for emissions controls to protect resources has given rise to the concepts of critical levels of pollutants and critical loads of deposition. Critical levels and loads can be defined as "quantitative estimates of exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge." The basic concept of critical load is relatively simple, as the threshold concentration of pollutants at which harmful effects on sensitive receptors begin to occur. Implementation of the concept is, however, not at all simple or straight-forward. Practical thresholds for particular receptors (soils, fresh waters, forests) have not been agreed to easily. Different research groups have employed different definitions and different levels of complexity. Constraints on the availability of suitable, high-quality, regional data have been considerable.

Target load is somewhat different. It is based on both science, including in particular quantitative estimates of critical load, and also on policy. A target load is set on the basis of, in addition to model-based estimates of critical loads, such considerations as:

- desire to protect the ecosystem against chronic critical load exceedence;

- consideration of the temporal components of acidification/recovery processes, so that, for example, resources could be protected only for a specified period of time or allowed to recover within a designated window;
- seasonal and episodic variability, and probable associated biological responses; and
- model, data, and knowledge uncertainty and any desire to err on the side of resource protection.

A critical load is objectively determined, based on specific chemical criteria that are known or believed to be associated with adverse biological impacts. A target load is subjectively determined, but it is rooted in science and can incorporate allowances for uncertainty, ecosystem variability, temporal dynamics, and additional considerations.

Consideration of the efficacy of adopting one or more acid deposition critical loads for the protection of surface water quality from potential adverse effects of sulfur or nitrogen deposition is a multifaceted problem. It requires that sulfur and nitrogen be treated separately as potentially-acidifying agents, and that separate estimates be generated for all individual, well-defined watersheds, regions, or subregions of interest. Appropriate criteria must be selected as being indicative of damaged water quality, for example ANC or pH. Once a criterion has been selected, a critical value must be estimated, below which the criterion should not be permitted to fall. ANC criteria have been set at 0, 20, or 50 $\mu\text{eq/L}$ in various European and North American applications. Selection of critical values for ANC or pH is confounded by the existence of streams that are acidic or very low in pH or ANC due entirely to natural factors, irrespective of acidic deposition. In particular, low concentrations of base cations in solution, due to low weathering rates and/or minimal contact between drainage waters and mineral soils, geological sources of S, and high concentrations of organic acids, contribute to naturally low pH and ANC in some surface waters.

Previous work conducted for the FS has utilized the MAGIC model to estimate the maximum S deposition that can be tolerated to achieve a specific ANC for three specific years in the future (2010, 2040, and 2100) for the Joyce Kilmer-Slickrock and Shining Rock Wilderness Areas (Sullivan and Cosby 2002). From the array of critical loads presented, the FS Line Officers could select a target load based on the desired condition (ANC) to be reached by a specified time in the future. Different ANC thresholds can be selected depending on the extent of protection desired.

Sulfur is the primary determinant of precipitation acidity and SO_4^{2-} is the dominant acid anion associated with acidic streams throughout most of the southern Appalachian Mountains region (Sullivan et al., 2002a). Although a substantial proportion of atmospherically deposited S is retained in watershed soils, SO_4^{2-} concentrations in many mountain streams have increased as a consequence of acidic deposition. Nitrate concentrations in streamwater are also high in some high-elevation streams.

Key questions now facing scientists and policy makers concern the prognosis for future change in streamwater chemistry and the extent to which S, and in some cases also N, emissions and deposition will need to be reduced to allow ecosystem recovery and prevent further damage (c.f., Jenkins et al. 1998). Future streamwater and soil chemistry can be projected with a process-based watershed model, given various emissions control scenarios. Future emissions can be estimated on the basis of existing or expected regulations, or in response to more aggressive emissions control options. Critical loads can be calculated for streams, assuming chemical/biological dose-response relationships (Bull, 1992). Because different species respond at varying chemical indicator levels, multiple critical loads can be calculated or applied to a given stream. Federal land managers are now beginning to use model-based critical loads calculations for setting resource protection and restoration goals on federal lands (Porter et al. 2005). Process-based watershed acid-base chemistry models such as MAGIC can be used in an iterative fashion to calculate critical loads, based on the chemical indicator ANC. Streamwater ANC integrates geologic, edaphic, and hydrologic response functions. This kind of analysis requires specification of the critical ANC values, below which ecosystem damage would be likely to occur, and the time period in the future at which the critical load evaluation is to be made.

II. OBJECTIVES

The principal objectives of the study reported here were to:

1. Use MAGIC to estimate future trends for stream chemistry and soil base saturation under a range of future atmospheric deposition scenarios.
2. Use MAGIC to estimate changes in stream chemistry and soil base saturation since pre-industrial time.
3. Develop an approach to predict critical load or future stream ANC for a new stream, based on the known current stream water chemistry, geology, elevation, and forest cover of the catchment.

4. Estimate the critical loads of sulfur deposition needed to protect those streams that are not yet acidic, and to restore streams that are already acidic to specific chemical criteria values.
5. Evaluate model uncertainty and regional representativeness of the modeled streams.
6. Assess, based on available data, episodic variability in stream chemistry and biological dose-response relationships within the study area.

III. APPROACH

A. Site Selection and Data Compilation

Data are available from multiple sources with which to conduct this assessment. Snyder et al. (2004) compiled the available water chemistry data from western North Carolina and determined that there were 173 water samples collected by the FS or Environmental Protection Agency that could be used in a modeling study. A recent regional assessment (Sullivan et al. 2002a) of the southern Appalachians estimated 3 percent of the streams to be acidic, but Snyder et al. (2004) reported about 11 percent of the streams sampled on NF lands in western North Carolina are acidic and 40 percent are potentially acid-sensitive. Further water sampling has been conducted by the FS since the Snyder et al. (2004) report in western North Carolina and there are additional samples from eastern Tennessee (81 sites) and the upstate of South Carolina (13 sites), which were included in this analysis. Snyder et al. (2004) did note more samples should be collected from the most acid-sensitive (siliceous) lithology class. Additional water chemistry samples have now been taken from the siliceous lithology class on the Cherokee NF (about 45 sites), and of 8 of the 10 sites sampled in western North Carolina (near Linville Gorge Wilderness) in 2005 had titrated ANC values less than 0.

Snyder et al. (2004) also reported that adequate soil samples were available for watershed modeling from only 20 sites with FS ownership in western North Carolina and an additional 8 sites were available from other ownerships. In 2004 and 2005, the FS collected additional soil samples in 45 watersheds, following the procedures developed by Webb et al. (2004). Soil chemistry data are also available for other areas from the U.S. EPA's Direct Delayed Response Project and site-specific studies. These data were compiled for the SAMI Assessment (Sullivan et al. 2002).

MAGIC was used by SAMI for their regional assessment and all of their watersheds had water chemistry data that were used to calibrate the model. However, not all of the areas had soil chemistry data, and for some locations the soils data to perform the MAGIC analysis were

borrowed from another watershed thought to have similar characteristics. Sites having both stream and soil data were designated Tier I. Sites were designated Tier II if they had stream data, but no soil data, and for which there existed soil data for a nearby watershed on the same geology. Tier III sites had stream data and no soils data and soils data were borrowed from a site considered most similar with respect to location, geology (which could be different than that at the water chemistry site), stream ANC, elevation, and stream SO_4^{2-} and NO_3^- concentrations. Some watersheds lacking soils data were modeled for this study using a similar procedure to that used in the SAMI study. We used the same classification scheme in this project to identify sites being modeled with soil data collected within the catchment or “borrowed” from other sites.

Sites were selected for modeling in this project from several data sets. Initially the following sites were selected:

- All SAMI Tier I modeling sites located in NC, TN, or SC;
- All Forest Service watersheds sampled for soil chemistry in 2004 or 2005; and
- Sites in Shining Rock or Joyce Kilmer Wilderness Areas modeled by Sullivan and Cosby (2002).

Potential modeling sites were pre-screened to remove from consideration streams that had high concentrations of Cl^- ($> 70 \mu\text{eq/L}$) that could potentially have been caused by road salt application, and those that had high concentrations of NO_3^- ($> 30 \mu\text{eq/L}$) that could potentially have been caused by agricultural or silvicultural fertilization within the watershed. The potential for such anthropogenic disturbances, other than air pollution, was determined by stream chemistry and the location within the watersheds of roads, wilderness areas, and agricultural or forestry operations.

Samples were also pre-screened to remove sites for which the observed percent soil base saturation (% BS) was $> 60\%$. Such high values of % BS probably represent a sampling or analysis error, or reflect a local (and unrepresentative) heterogeneity in the soil matrix at the sampling site.

Based on these pre-screening criteria, 37 total sites having soil data available within the catchment were identified for modeling as a Tier I site in this project and 8 sites were removed during the pre-screening process. We examined the characteristics of these 37 sites to determine if they exhibited a good representation of different streamwater ANC, NO_3^- , and SO_4^{2-} concentrations; location; bedrock geology; soil type; elevation; and general vegetation type. Based on these analyses, gaps were identified in the variable distributions for these 37 Tier I

sites. To fill in these gaps in the landscape coverage, it was necessary to select additional sites for modeling. Recognizing that these additional sites would not have both streamwater and soils data available, the site selection was made based on availability of streamwater samples.

The USDA Forest Service has, in recent years, collected 308 stream chemistry samples at 256 streams locations in Pisgah and Nantahala National Forests in western North Carolina, Cherokee National Forest in eastern Tennessee, and the Andrew Pickens Ranger District, Sumter National Forest in South Carolina, all within the Southern Blue Ridge Province. This area includes three Class I areas administered by the Forest Service: Joyce Kilmer-Slickrock, Linville Gorge and Shining Rock Wilderness. Additional streams were selected for modeling from among these streams recently surveyed by the Forest Service for streamwater chemistry, but not sampled for soil chemistry. For each stream in this group of sites added to the selection process, soil data for model calibration were borrowed from a nearby watershed using the methods employed by SAMI, making these additional sites Tier II or Tier III modeling sites.

The additional sites were selected for modeling as follows. First, the available stream chemistry data were screened for possible watershed impacts other than from air pollution. Streams having high NO_3^- concentration ($> 30 \mu\text{eq/L}$) were deleted from consideration if there was evidence of possible agricultural contributions of N within the watershed. Streams having high Cl^- concentration ($> 70 \mu\text{eq/L}$) were deleted from consideration if there were roads within the watershed that might receive road salt application during winter. Sites were selected from this screened database in order to maximize the distribution of modeling sites across the gradients of streamwater ANC, SO_4^{2-} , and nitrate concentration, and also elevation, geology, and geographic location. Sites deleted from consideration as candidate modeling sites are listed in Table 1, along with a description of the reason for deletion.

After the site selection and calibration processes were complete, a total of 66 streams were modeled. These included 37 out of 39 streams within the Southern Blue Ridge for which we had both streamwater chemistry and watershed soil chemistry data (Tier I), plus 29 streams for which soil data were borrowed in order to calibrate the model (Tier II and Tier III). The final list of streams selected for modeling is given in Table 2, while Table 3 lists the 19 streams in wildernesses that are also found on the final list of streams. Some of the selected streams were modeled multiple times as part of the sensitivity analyses described in Section III.B.6.

Table 1. Stream sites deleted from consideration as candidates for modeling

Stream Name	Reason for removal
Baird Creek	Possible road salt
Cranberry Creek	Possible road salt -- State Hwy 194
Elk River	Agricultural influence; Possible road salt -- local hwy
Green Mountain Branch	Possible road salt. Adjacent to US 421 which traverses one side of the catchment
Horse Bottom Creek	Agricultural influence; Possible road salt -- local road
Middle Branch	Possible road salt. State Hwy TN143 goes through the top of the headwaters and below the sample site. Sample was taken 400 feet above the road and was 3000 feet to the Hwy at the top of the catchment
Middle Fork New River	Agricultural influence; Possible road salt -- local hwy
North Toe River	Agricultural influence
Paint Creek	Model calibration failure
Phillips Branch	Agricultural influence; Possible road salt
Puncheon Fork	Possible road salt. State Road 1502
Right Prong South Toe River	Model calibration failure
South Fork Citico Creek	Model calibration failure
UT Beaverdam Creek	Agricultural influence; Possible road salt-- a hwy adjoins side of the catchment and crosses the stream at the top of the catchment - - about 3000 feet above the sample site
UT Chattooga River	Model calibration failure
UT Clark Creek	Possible road salt -- local road
UT Forge Creek	Agricultural influence
UT Laurel Fork	Agricultural influence; Possible road salt -- local road
Watauga River	Agricultural influence; Possible road salt. Sample is 1000 feet downstream from nearest road. It is possible this is road salt since large portion of catchment has road paralleling stream(s)
Wilson Creek	Possible road salt. Along secondary state road (sample taken upstream from road)
Woodson Branch	Possible road salt. State Hwy 25/70 parallels the steam and is within 260 feet of sample

Table 2. Location of streams selected for modeling.

Stream ID	Tier	Stream Name	Forest	Longitude	Latitude	Elev. (m)
0839517353661	1	Adam Camp Branch	Nantahala	-83.951760	35.366180	834
0831227358653	3	Bear Branch	Cherokee	-83.122760	35.865360	602
0840591352378	1	Bearpen Branch	Nantahala	-84.059130	35.237830	1052
0831036359106	1	Beetree Branch	Cherokee	-83.103620	35.910650	593
0840805353091	2	Big Cove Branch	Cherokee	-84.080550	35.309110	1042
0835047350176	1	Big Laurel Brook	Nantahala	-83.504700	35.017650	1198
0840893352584	2	Big Oak Cove Creek	Cherokee	-84.089310	35.258490	909
0823767362291	3	Briar Creek	Cherokee	-82.376746	36.229149	755
0829119353006	2	Bubbling Spring Branch	Pisgah	-82.911930	35.300660	1591
0829160353045	1	Bubbling Spring West Tributary	Pisgah	-82.916050	35.304550	1609
0829321353099	1	Buckeye Cove Creek	Pisgah	-82.932150	35.309910	1674
0831509350320	3	Cane Creek Tributary	Nantahala	-83.150930	35.032030	901
0828955353809	3	Cathey Creek	Pisgah	-82.895510	35.380920	1025
0822102358016	1	Colberts Creek	Pisgah	-82.210240	35.801680	870
0828920352772	1	Courthouse Creek	Pisgah	-82.892030	35.277230	1061
0828670353323	1	Dark Prong	Pisgah	-82.867040	35.332340	1622
0828299352854	3	Davidson River	Pisgah	-82.829990	35.285440	806
0828261353375	1	East Fork Pigeon	Pisgah	-82.826180	35.337540	1211
0829079353270	1	Flat Laurel Creek	Pisgah	-82.907990	35.327060	1403
0831285350075	1	Glade Creek	Nantahala	-83.128530	35.007570	812
0830067351628	2	Greenland Creek	Nantahala	-83.006760	35.162820	1118
0830849349820	1	Indian Camp	Sumter	-83.084950	34.982020	735
0840675353310	1	Indian Branch	Cherokee	-84.067560	35.331040	839
0839574352637	2	Indian Spring Branch	Nantahala	-83.957460	35.263710	964
0835085350173	1	Kilby Creek	Nantahala	-83.508550	35.017350	1193
0840886352889	1	Kirkland Cove	Cherokee	-84.088630	35.288920	670
0822534357112	1	Left Prong South Toe River	Pisgah	-82.253430	35.711250	1245
0819654364692	3	Lindy Camp Branch	Cherokee	-81.965490	36.469240	919
JK5	1	Little Santetlah Cr. (NuCM site)	Nantahala	-83.929911	35.358406	693
0826852360155	3	Little Prong Hickey Fork	Pisgah	-82.685250	36.015500	830
0828351352745	3	Long Branch	Pisgah	-82.835100	35.274580	905
0822144357431	1	Lost Cove	Pisgah	-82.214400	35.743120	938
0822446357370	1	Lower Creek	Pisgah	-82.244640	35.737050	1076
0841287353269	3	McNabb Creek	Cherokee	-84.128750	35.326960	556
0822122357913	1	Middle Creek	Pisgah	-82.212240	35.791320	866
0829184352865	1	Mill Station Creek	Pisgah	-82.918500	35.286560	1453
0819269357923	3	Paddy Creek	Pisgah	-81.926967	35.792317	469
0823639357265	1	Peach Orchard Creek	Pisgah	-82.363950	35.726550	1307
0831049349229	3	Pigpen Branch	Sumter	-83.104950	34.922970	644

Table 2. Continued

Stream ID	Tier	Stream Name	Forest	Longitude	Latitude	Elev. (m)
0829510358422	3	Rattlesnake Branch	Cherokee	-82.951000	35.842240	978
0829630353646	1	Right Hand Prong	Pisgah	-82.963040	35.364620	1209
0840477353169	1	Roaring Branch	Cherokee	-84.047710	35.316910	1172
0840745352655	1	Rough Ridge Creek	Cherokee	-84.074590	35.265550	906
0818710358245	3	Russell Creek	Pisgah	-81.871000	35.824583	411
0831139350208	1	Scotsman Creek	Nantahala	-83.113980	35.020820	799
0830946350205	1	South Fork Fowler Creek	Nantahala	-83.094600	35.020540	820
0841126353049	1	Spivey Creek	Cherokee	-84.112660	35.304900	586
0826466361015	3	Squibb Creek	Cherokee	-82.646690	36.101560	614
0819433358564	3	Stillhouse Branch	Pisgah	-81.943383	35.856467	566
0840110352961	1	Unnamed creek	Nantahala	-84.011090	35.296110	1398
0840155353326	1	Unnamed creek	Nantahala	-84.015540	35.332620	1170
0840420353309	1	Unnamed Creek	Cherokee	-84.042080	35.330920	1160
0822486357318	1	Upper Creek	Cherokee	-82.248660	35.731830	1050
0828915353164	3	UT Flat Laurel Creek	Pisgah	-82.891590	35.316470	1719
0840993353234	2	UT Laurel Branch	Cherokee	-84.099930	35.323440	615
0819003359391	1	UT Linville River (NuCM site)	Pisgah	-81.900300	35.939933	1080
0840899353546	3	UT McNabb Creek	Cherokee	-84.089930	35.354680	1023
0819597358404	3	UT North Fork of Catawba	Pisgah	-81.959783	35.840482	524
0827442360239	3	UT Paint Creek	Cherokee	-82.744210	36.023930	844
0830204351641	1	UT Panthertown Creek (Boggy Creek)	Nantahala	-83.020450	35.164150	1119
0818710358242	3	UT Russell Creek	Pisgah	-81.871033	35.824283	412
0818853358259	3	White Creek	Pisgah	-81.885333	35.825967	493
0839262352702	1	Wildcat Branch	Nantahala	-83.926210	35.270260	823
0830458349846	1	Wilson Creek	Sumter	-83.045870	34.984670	744
0829674352788	3	Wolf Creek	Nantahala	-82.967440	35.278800	1105
0819415358260	3	Yellow Fork	Pisgah	-81.941517	35.826083	739

Table 3. Location of streams selected for modeling to characterize a wilderness.

Stream ID	Tier	Stream Name	CAA Designation	Wilderness	Elev. (m)
0839517353661	1	Adam Camp Branch	I	Joyce Kilmer - Slickrock	834
0835047350176	1	Big Laurel Brook	II	Southern Nantahala	1198
0829119353006	2	Bubbling Spring Branch	II	Middle Prong	1591
0829160353045	1	Bubbling Spring West Tributary	II	Middle Prong	1609
0829321353099	1	Buckeye Cove Creek	II	Middle Prong	1674
0828955353809	3	Cathey Creek	I	Shining Rock	1025
0828261353375	1	East Fork Pigeon	I	Shining Rock	1211
0829079353270	1	Flat Laurel Creek	II	Middle Prong	1403
0831285350075	1	Glade Creek	II	Ellicott Rock	812
0835085350173	1	Kilby Creek	II	Southern Nantahala	1193
JK5	1	Little Santetlah Cr. (NuCM site)	I	Joyce Kilmer-Slickrock	693
0818710358245	3	Russell Creek	I	Linville Gorge	411
0831139350208	1	Scotsman Creek	II	Ellicott Rock	799
0830946350205	1	South Fork Fowler Creek	II	Ellicott Rock	820
0826466361015	3	Squibb Creek	II	Sampson Mountain	614
0830849349820	1	Indian Camp	II	Ellicott Rock	735
0819003359391	1	UT Linville River (NUCM site)	I	Linville Gorge	1080
0818710358242	3	UT Russell Creek	I	Linville Gorge	412
0818853358259	3	White Creek	I	Linville Gorge	493

B. Model Application

MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on soil and surface water chemistry (Cosby et al., 1985a,b). The model simulates soil solution chemistry and surface water chemistry to predict the monthly and annual average concentrations of the major ions in these waters. MAGIC consists of 1) a section in which the concentrations of major ions are assumed to be governed by simultaneous reactions involving SO_4^{2-} adsorption, cation exchange, dissolution-precipitation-speciation of Al and dissolution-speciation of inorganic C; and 2) a mass balance section 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 loss to runoff. At the heart of MAGIC is the size of the pool of exchangeable base cations in the soil. As the fluxes to and from this pool change over time owing to changes in atmospheric deposition, the chemical equilibria between soil and soil solution shift to give changes in surface water chemistry. The degree and

rate of change of surface water acidity thus 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 SO_4^{2-} and F^- . Effects of 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. First-order rates are used for biological retention (uptake) of NO_3^- and NH_4^+ in the soils and streams. Weathering and the uptake rate of N are assumed to be constant. A set of mass balance equations for base 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 complete details of the model see Cosby et al. 1985 a, b; 1989). MAGIC has been used to reconstruct the history of acidification and to simulate the future trends on a regional basis and in a large number of individual catchments in both North America and Europe (e.g., Lepisto et al., 1988; Whitehead et al., 1988; Cosby et al., 1989, 1990, 1996; Hornberger et al., 1989; Jenkins et al., 1990a-c; Wright et al., 1990, 1994; Norton et al., 1992; Sullivan and Cosby, 1998).

The input data required in this project for aquatic and soils resource modeling with the MAGIC model (stream water, catchment, soils, and deposition data) were assembled and maintained in data bases for each site modeled (electronic spreadsheets and text-based MAGIC parameter files). Model outputs for each site were archived as text-based time-series files of simulated variable values. The outputs were also concatenated across all sites and maintained in electronic spreadsheets.

1. Specification of Stream and Soil Data

For the 66 streams selected for modeling, at least one complete stream water chemical sample was available for each site during the period 1999 through 2005. No stream had samples taken in all years, and no single year was sampled in all streams. If multiple samples were available within a year (4 streams), the values were averaged to give a single annual mean for that year at the site. Of the 66 streams, 59 had samples taken in only one year. For these 59 sites,

the single sample available was used to calibrate MAGIC, regardless of the year it was taken. There were 7 streams for which stream water samples were available for multiple years at the site. All 7 of these streams had samples available in 2000, and that year was used for calibration of MAGIC at those sites. The uncertainty arising from using 2000 rather than another year to calibrate these sites was examined in a sensitivity analysis (see section below).

Soil data were assigned to modeling sites using protocols developed for the SAMI aquatic assessment (Sullivan et al. 2002a). For some sites (designated Tier I, n= 37), soils chemistry data were available from within the watershed to be modeled. In cases where data from multiple soil sampling sites were available for an individual watershed, the data were aggregated on an area-weighted basis to reflect the distribution of mapped soil types within the watershed. For a second group of sites (Tier II, n=6), soils data within the catchment were missing but were available from a nearby watershed underlain by similar geology. For a third group of sites (Tier III, n=23) soils data were neither available from within the watershed nor from nearby watersheds on similar geology.

Soil data for the Tier II and Tier III sites were obtained using a surrogate approach, whereby a watershed that lacked one or more input parameters was paired with a watershed for which all input data were available. This pairing was accomplished by comparing watershed similarity on the basis of streamwater characteristics (ANC, sulfate, and base cation concentrations), physical characterization (location, elevation), and bedrock geology data. The missing data were then “borrowed” from the data-rich paired watershed judged to be most similar (Table 4).

A site was designated as Tier II if the borrowed soil data were obtained from a site located on the same geologic sensitivity class within a distance of 5 km. If the location from which soils data were borrowed was further than 5 km away, or if the borrowed data were from a different geologic sensitivity class, the modeled site was designated as Tier III. Tier II and III sites were selected for modeling in order to ensure a distribution of sites across gradients of acid-base chemistry and elevation. A major emphasis was on selection of many acidic and low-ANC (< 50 $\mu\text{eq/L}$) streams, which exhibited varying SO_4^{2-} and NO_3^- concentrations, and which occurred at varying elevation. The uncertainty associated with this surrogate soil data assignment procedure was examined in a sensitivity analysis (see section below).

Table 4. Watershed location from which soil data were borrowed for Tier II and Tier III modeling sites

Stream	Tier	Location from which Soil Data Were Borrowed
Big Cove Branch	Tier II	Indian Branch
Big Oak Cove Creek	Tier II	Rough Ridge Creek
Bubbling Spring Branch	Tier II	Bubbling Spring West Tributary
Greenland Creek	Tier II	UT Panthertown Creek (Boggy Creek)
Indian Spring Branch	Tier II	Wildcat Branch
UT Laurel Branch	Tier II	Spivey Creek
Bear Branch	Tier III	Beetree Branch
Briar Creek	Tier III	Middle Creek
Cane Creek Tributary	Tier III	Buckeye Cove Creek
Cathey Creek	Tier III	Shining Rock
Davidson River	Tier III	Shining Rock
Lindy Camp Branch	Tier III	Rough Ridge Creek
Little Prong Hickey Fork	Tier III	Joyce Kilmer (NuCM site)
Long Branch	Tier III	Wilson Creek
McNabb Creek	Tier III	Beetree Branch
Paddy Creek	Tier III	Bubbling Spring West Tributary
Pigpen Branch	Tier III	UT East Fork
Rattlesnake Branch	Tier III	Shining Rock
Russell Creek	Tier III	Linville Gorge (NuCM site)
Squibb Creek	Tier III	Spivey Creek
Stillhouse Branch	Tier III	Bubbling Spring West Tributary
UT Flat Laurel Creek	Tier III	Buckeye Cove Creek
UT McNabb Creek	Tier III	Unnamed Creek
UT North Fork of Catawba	Tier III	Bubbling Spring West Tributary
UT Paint Creek	Tier III	Colberts Creek
UT Russell Creek	Tier III	Linville Gorge (NuCM site)
White Creek	Tier III	Glade Creek
Wolf Creek	Tier III	Glade Creek
Yellow Fork	Tier III	Bubbling Spring West Tributary

2. Specification of Deposition and Meteorology Data

MAGIC requires, as atmospheric inputs for each site, estimates of the total annual deposition (eq/ha/yr) of eight ions, and the annual precipitation volume (m/yr). The eight ions are: Ca, Mg, Na, K, NH₄, SO₄, Cl, and NO₃. Total deposition of an ion at a particular site for any year can be represented as combined wet, dry, and occult (cloud and fog) deposition:

$$\text{TotDep} = \text{WetDep} + \text{DryDep} + \text{OccDep}.$$

Inputs to the model are specified as wet deposition (the annual flux in meq/m²/yr) and a dry and occult deposition factor (DDF, unitless) used to multiply the wet deposition in order to get total deposition:

$$\text{TotDep} = \text{WetDep} * \text{DDF},$$

where

$$\text{DDF} = 1 + \text{DryDep} / \text{WetDep} + \text{OccDep} / \text{WetDep}.$$

Thus, given an annual wet deposition flux (WetDep), the ratio of dry deposition to wet deposition (DryDep / WetDep), and the ratio of occult deposition to wet deposition (OccDep / WetDep) for a given year at a site, the total deposition for that site and year is uniquely determined.

In order to calibrate MAGIC, time-series of total deposition are needed for the calibration year and the 140 years preceding the calibration for the historical reconstructions that are part of the calibration protocol. For future simulations using MAGIC, time-series of total deposition are needed to drive the model for the different future scenarios under consideration. The procedure for providing time-series of total deposition inputs to MAGIC is as follows.

The absolute values of wet deposition and DDF for each ion are provided for a Reference Year at each site. For this project, the MAGIC Reference Year was 2005 at all sites. Given the Reference Year deposition values, the deposition data for the historical and calibration periods and for future deposition scenarios can be calculated using the Reference Year absolute values and scaled time-series of wet deposition and DDF that give the values for a given year as a fraction of the Reference Year value. For instance, to calculate the total deposition of a particular ion in some historical or future year j:

$$\text{TotDep}(j) = [\text{WetDep}(0) * \text{WetDepScale}(j)] * [\text{DDF}(0) * \text{DDF Scale}(j)],$$

where $WetDep(0)$ is the Reference Year wet deposition ($meq/m^2/yr$) of the ion, $WetDepScale(j)$ is the scaled value of wet deposition in year j (expressed as a fraction of the wet deposition in the Reference Year), $DDF(0)$ is the dry and occult deposition factor for the ion for the Reference Year, and $DDFScale(j)$ is the scaled value of the dry and occult deposition factor in year j (expressed as a fraction of the DDF in the Reference Year).

The absolute value of wet deposition used for the Reference Year is time and space specific - varying geographically within the region, varying locally with elevation, and varying from year to year. It is desirable to have the estimates of wet deposition take into account the geographic location and elevation of the site as well as the year for which calibration data are available. Therefore, estimates of wet deposition used for the Reference Year should be derived from a procedure (model) that has a high spatial resolution and considers elevation effects. As described below, the absolute wet deposition values used for the Reference Year in this project were derived from observed data based on the National Atmospheric Deposition Network (NADP).

The absolute value of the DDF used for the Reference Year specifies the ratio between the absolute amounts of wet and total deposition. This ratio is less variable in space and time than is the estimate of wet deposition. That is, if in a given year the wet deposition goes up, then the total deposition usually goes up also (and conversely); and if the elevation or aspect of a given site results in lower wet deposition, the total deposition will often be lower also (and conversely). Estimates of the absolute values of DDF may, therefore, be derived from a procedure (model) that has a relatively low spatial resolution and/or temporally smoothes the data. Estimates of the absolute values of the DDF for the Reference Year at each site in this project were derived from the Advanced Statistical Trajectory Regional Air Pollution (ASTRAP) model (Shannon 1998) as described below.

The long-term scaled sequences used to specify time-series of deposition inputs for MAGIC simulations do usually not require detailed spatial or temporal resolution. Scaled sequences of wet deposition or DDF (normalized to the same reference year) at neighboring sites will be similar, even if the absolute wet deposition or DDF at the sites are different due to local aspect, elevation, etc. Therefore, if the scaled long-term patterns of any of these do not vary much from place to place, estimates of the scaled sequences (as for estimates of absolute DDF values) may be derived from a model that has a relatively low spatial resolution. Output from the

ASTRAP model was used to construct scaled sequences of both wet deposition and DDF for this project as described in the next sections.

a. *Wet Deposition Data (Reference Year and Calibration Values)*

The absolute values of wet deposition used for defining the Reference Year and for the MAGIC calibrations must be highly site-specific. We used estimated wet deposition data for each site derived from the spatial interpolation model of Grimm and Lynch (1997), referred to here as the Grimm model. The Grimm model is based on observed wet deposition at NADP monitoring stations, and provides a spatially interpolated value of wet deposition of each of the eight ions needed for MAGIC. The model also makes a correction for changes in precipitation volume (and thus wet deposition) based on the elevation at a given site. This correction arises from a model of orographic effects on precipitation volumes derived from regional climatological data.

The latitude, longitude, and elevation of the 66 MAGIC modeling sites were provided as inputs to the Grimm model. The model outputs were quarterly and annual wet deposition and precipitation estimates for each modeling site. The annual data were used for definition of the Reference Year and for MAGIC calibration and simulation. The NADP data (and thus the estimates provided by Grimm's model) cover the period 1983 to 2005. This period includes the MAGIC Reference Year and the calibration years for all of the modeling sites in this project.

b. *Dry and Occult Deposition Data and Historical Deposition Sequences*

Absolute values of DDF and the scaled sequences of wet deposition and DDF are derived for this project from simulations using the ASTRAP model as described in Appendix N of the SAMI aquatics report (Sullivan et al. 2002a). The ASTRAP model was used to provide estimates of historical wet, dry, and occult deposition of sulfur and oxidized nitrogen at 33 sites in and around the SAMI region (Shannon 1998). The ASTRAP sites included 21 existing NADP deposition monitoring stations, 7 sites in Class I areas, and 5 sites that were neither NADP nor Class I. For each of the sites, ASTRAP produced wet, dry, and occult deposition estimates of sulfur and oxidized nitrogen every ten years starting in 1900 and ending in 1990. The model outputs are smoothed estimates of deposition roughly equivalent to a ten-year moving average centered on each of the output years. The outputs of ASTRAP were used to estimate the absolute DDF for each site (using the DryDep/WetDep and OccDep/WetDep ratios from the ASTRAP

output), and to set up the scaled sequences of historical wet deposition and historical DDF for the calibration of each site modeled in this project.

The wet, dry, and occult deposition estimates provided by ASTRAP for each year (for both sulfur and oxidized nitrogen) at each ASTRAP site were used to calculate the MAGIC DDF for each year and each site. This provided time series of DDF for sulfur and oxidized nitrogen for each ASTRAP site extending from 1900 to 1990. The value of DDF for 1990 was used as the absolute value of DDF for the Reference Year (i.e., no change was assumed for DDF from 1990 to 2005). The resulting time series of DDF values from 1900 to 2005 for each ASTRAP site were normalized to the 2005 values to provide historical scaled sequences of DDF at each ASTRAP site.

The time series of wet deposition estimates for each ASTRAP site were used to construct historical scaled sequences of wet deposition. The absolute wet deposition outputs of ASTRAP were normalized to their 1990 values and converted to scaled sequences of wet deposition from 1900 to 1990 for each ASTRAP site. It was then necessary to couple these historical scaled wet deposition sequences from 1990 to the MAGIC Reference Year 2005. This coupling was accomplished using scaled observed changes in wet deposition from 1900 to 2005 derived from the Grimm model.

At very high elevations, the inputs of ions from cloud water can be very large. In the SAMI project (see Sullivan et al, 2002a,b), high elevation sites in the Great Smoky Mountain National Park (GSMNP) were determined to have DDF values (reflecting dry and occult but particularly cloud water inputs) that were approximately twice as large as those specified by the ASTRAP model. Accordingly, the SAMI project used the larger GSMNP DDF values for any site over 1500 meters in elevation. In this project, there were no sites in GSMNP, but there were five sites (Bubbling Spring Branch, Bubbling Spring West Tributary, Buckeye Cove Creek, Dark Prong, and UT Flat Laurel Creek (Table 2) at elevations over 1500 meters. These sites were assigned the higher DDF values used for the GSMNP high elevation sites in SAMI. The potential bias and uncertainty associated with the use of this high elevation DDF “correction” was examined in a sensitivity analysis (see section below).

3. Protocol for MAGIC Calibration and Simulation at Individual Sites

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 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 (streamwater 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 adjusting the optimized parameters, the simulated-minus-observed values of the criterion variables usually converge to zero (within some specified tolerance). The model is then considered calibrated.

There are eight parameters to be optimized in this procedure (the weathering and the selectivity coefficient of each of the four base cations), and there are eight observations that are used to drive the estimate (current soil exchangeable pool size and current output flux of each of the four base cations). If new assumptions or new values for any of the fixed variables or inputs to the model are adopted, the model must be re-calibrated by re-adjusting the optimized parameters until the simulated-minus-observed values of the criterion variables again fall within the specified tolerance.

Estimates of the fixed parameters, the deposition inputs, and the target variable values to which the model is calibrated all contain uncertainties. A “fuzzy optimization” procedure was utilized in this project to provide explicit estimates of the effects of these uncertainties. The procedure consists of multiple calibrations at each site using random values of the fixed parameters drawn from a *range* of fixed parameter values (representing uncertainty in knowledge of these parameters), and random values of Reference Year deposition drawn from a *range* of total deposition estimates (representing uncertainty in these inputs). The final convergence (completion) of the calibration is determined when the simulated values of the criterion variables are within a specified “acceptable window” around the nominal observed value. This “acceptable window” represents uncertainty in the target variable values being used to calibrate the site.

Each of the multiple calibrations at a site begins with (1) a random selection of values of fixed parameters and deposition, and (2) a random selection of the starting values of the adjustable parameters. The adjustable parameters are then optimized using an algorithm seeking to minimize errors between simulated and observed criterion variable. Calibration success is judged when all criterion values simultaneously are within their specified “acceptable windows”,

(which may occur before the absolute possible minimum error is achieved). This procedure is repeated ten times for each site.

For this project, the “acceptable windows” for base cation concentrations in streams were taken as +/- 2 µeq/L around the observed values. “Acceptable windows” for soil exchangeable base cations were taken as +/- 0.2% around the observed values. Fixed parameter uncertainty in soil depth, bulk density, cation exchange capacity, stream discharge, and stream area were assumed to be +/- 10% of the estimated values. Uncertainty in total deposition was +/- 10% for all ions.

The final calibrated model at the site is represented by the ensemble of parameter values of all of the successful calibrations at the site. When performing simulations at a site, all of the calibrated parameter sets in the ensemble are run for a given historical or future scenario. The result is multiple simulated values of each variable in each year, all of which are acceptable in the sense of the calibration constraints applied in the fuzzy optimization procedure. The median of all the simulated values within a year is the “most likely” response for the site in that year. For this project, whenever single values for a site are presented or used in an analysis, these values are the median values derived from running all of the ensemble parameter sets for the site.

An estimate of the uncertainty (or reliability) of a simulated response to a given scenario, can also be derived from the multiple simulated values within a year resulting from the ensemble simulations. For any year in a given scenario, the largest and smallest values of a simulated variable define the upper and lower confidence bounds for that site's response for the scenario under consideration. Thus for all variables and all years of the scenario, a band of simulated values can be produced from the ensemble simulations at a site that encompasses the likely response (and provides an estimate of the simulation uncertainty) for any point in the scenario. For this project, whenever uncertainty estimates are presented, the estimate is based on this range of simulated values in any year arising from the simulations using the ensemble parameter sets.

4. Future Scenario Projections

a. Utilizing the VISTAS Results

We used atmospheric modeling results developed by the Visibility Improvement State and Tribal Association of the Southeast (VISTAS) to specify emissions control scenarios and associated levels of future acidic deposition at modeling site locations. VISTAS recently performed a technical analysis for the state, local, and tribal air quality agencies for 10

southeastern states. VISTAS is evaluating pollution control strategies that currently exist or could be implemented to achieve reasonable progress to attain natural (not impacted by human activities) visibility conditions on the days having poorest visibility at the federally mandated Class I areas within the southeastern states. The year 2018 is the first year for which the affected states will evaluate whether air pollution emission reductions will provide reasonable progress to attain the visibility goal in 2064 (Figure 1). There are three Class I areas in eastern Tennessee and western North Carolina that VISTAS included in their analysis and are relevant to this study using MAGIC. The Class I areas are: Great Smoky Mountains National Park (which also represents Joyce Kilmer-Slickrock Wilderness), Linville Gorge Wilderness, and Shining Rock Wilderness.

VISTAS utilized the Environmental Protection Agency's Models-3 Community Multiscale Air Quality (CMAQ) Modeling System for their analysis to examine how fine particulate and ozone concentrations are predicted to change between 2002 and 2009, and how visibility (as measured by deciviews) is predicted to change between 2002 and 2018 (Figure 1). The CMAQ atmospheric modeling results also provide the best available estimates of how dry and wet sulfur and nitrogen deposition will change between 2002 and 2018 in the southeastern United States in response to changes in emissions. For example, the Clean Air Interstate Rule and North Carolina's Clean Smoke Stacks Act will significantly reduce sulfur dioxide emissions, which will result in improved visibility conditions and reduction in sulfur deposition at downwind locations. Also, it should be noted that the 2002 VISTAS modeling results represent "typical" annual emissions based upon the years 2000 through 2004.

The 2002, 2009 and 2018 CMAQ modeling results were obtained from the VISTAS contractor (Dr. Dennis McNally, Alpine Geophysics LLC) in Microsoft Excel[®] files on November 24, 2006. These files were then imported into Microsoft Access[®] to calculate the wet and dry nitrate-nitrogen (NO₃-N), ammonia-nitrogen (NH₄-N), and sulfur (S) deposition. Table 5 lists the modeled values from CMAQ outputs (CMAQ Definitions) that were utilized to make the estimates. CMAQ results, expressed as short tons of deposition for each grid cell (144 square kilometers), were multiplied by 0.0629862 to convert deposition estimates into units of kilograms of S or N per hectare.

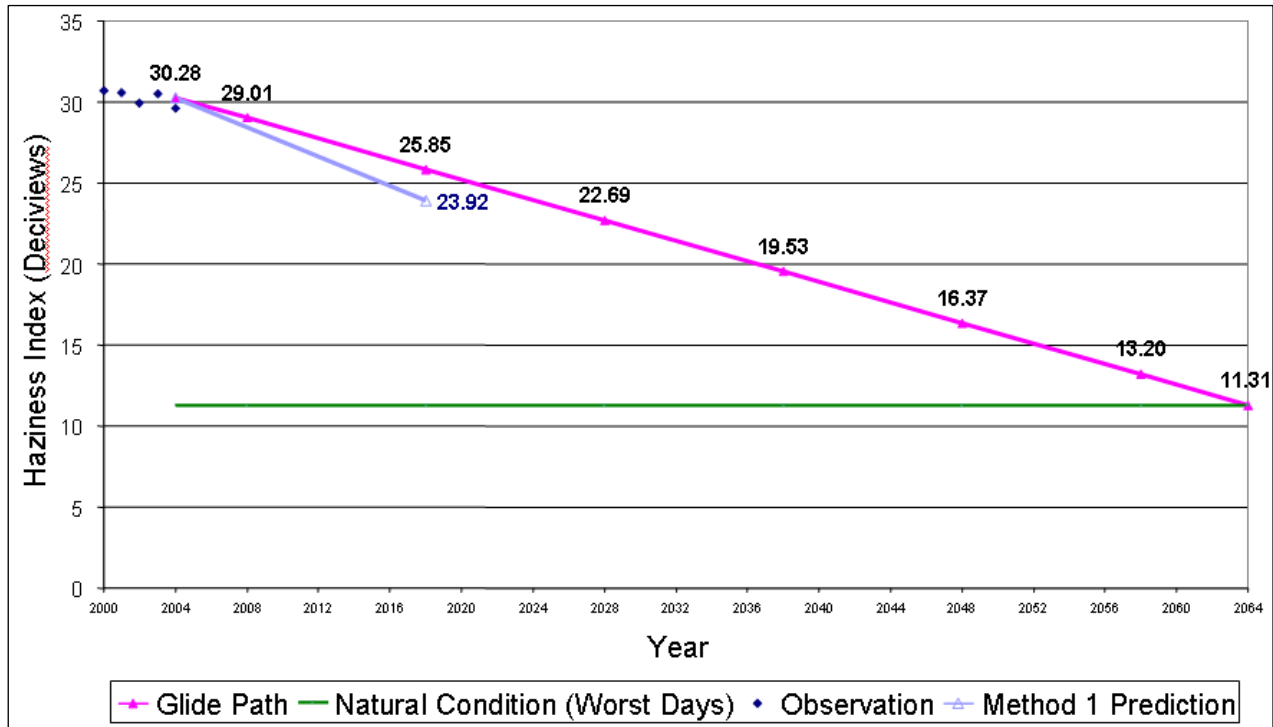


Figure 1. Glide path (pink line) estimated by Visibility Improvement State and Tribal Association of the Southeast (VISTAS) to attain natural background visibility (green line) for Great Smoky Mountains National Park and Joyce Kilmer-Slickrock Wilderness by 2064 on the days classified as having the worst visibility conditions. The blue line (Method 1 Prediction) indicates that the air pollution control programs currently in place will achieve greater improvement in visibility conditions in 2018 than the amount of improvement needed to maintain the glide path. (Graphic obtained from VISTAS.)

b. Spatial Data

Once the conversions of the tabular data were completed, results were imported into ArcMap[®] utilizing the northing and easting Lambert conformal conic projection (origin longitude: -97, origin latitude: 40, matching parallels of latitude: 33N and 45N, and false easting and northing were set to 0) coordinates provided for each cell in the CMAQ domain. Each of the deposition values represents the center of the grid, and the grid size was 12 km by 12 km. Eighteen separate spatially referenced raster maps were produced for the CMAQ results. The raster data were then used to estimate the average dry and wet sulfur and nitrogen deposition for each of the 66 catchments included in the MAGIC simulations.

Table 5. Columns from Environmental Protection Agency’s Models-3 Community Multiscale Air Quality (CMAQ) results summed to obtain the total sulfur, nitrate-nitrogen, and ammonia-nitrogen dry and wet deposition.

Desired Result	Variables Included	CMAQ Definition
Sulfur	SULF	Sulfuric Acid (H ₂ SO ₄)
	SO ₂	Sulfur Dioxide
	ASO4I	Aitken mode aerosol sulfate
	ASO4J	Accumulation mode aerosol sulfate
	ASO4K	Coarse mode aerosol sulfate
Nitrate - Nitrogen	HNO ₃	Nitric Acid
	NO ₃	Nitrogen Trioxide
	ANO3I	Aitken mode aerosol nitrate
	ANO3J	Accumulation mode aerosol nitrate
Ammonia - Nitrogen	NH ₃	Ammonia
	ANH4I	Aitken mode aerosol ammonium
	ANH4J	Accumulation mode aerosol ammonium

c. Future Scenarios

The CMAQ results produced by VISTAS provided the basis for estimating the sulfur, nitrate-nitrogen, and ammonia-nitrogen deposition for the years 2002, 2009, and 2018. To examine longer-term responses for the 66 catchments (as represented by stream ANC) in response to future changes in sulfur and nitrogen deposition, VISTAS visibility results were used to calculate the slope of visibility improvement between 2018 and 2064 for each of the three Class I areas (Table 6). Each of the three Class I areas do have different y-intercept and slope values, but the differences are not considered to be important. Therefore, we calculated the average visibility for the three Class I areas in 2018 (y-intercept) and 2064 to estimate the slope describing the change in visibility conditions over time (Table 6 and Figure 2).

MAGIC simulations were performed for three different future deposition scenarios:

1. No further sulfur or nitrogen reductions beyond those predict to occur by CMAQ for 2018. For this Base Case scenario, deposition levels were assumed to remain constant after 2018.

Table 6. Slope intercept value (a) for the linear equation ($y = ax + b$) to describe the relationship between the year (x) and the estimated visibility metric called deciview (y). The y-intercept values (b) were equal to the deciview values in 2018.

Class I Area	Extent of Visibility Improvement*	Year		Slope
		2018	2064	
Great Smoky Mountains	Visibility Goal Achieved	23.92	11.31	-0.27417
	Visibility Goal + 25%	23.92	14.14	-0.21272
Linville Gorge	Visibility Goal Achieved	22.03	11.28	-0.2336
	Visibility Goal + 25%	22.03	14.10	-0.1723
Shining Rock	Visibility Goal Achieved	22.21	11.64	-0.2298
	Visibility Goal + 25%	22.21	14.55	-0.1665
Average	Visibility Goal Achieved	22.72	11.41	-0.2459
	Visibility Goal + 25%	22.72	14.26	-0.1839

* Endpoints were specified as achievement of 2064 visibility goal and achievement of visibility improvement equal to zero human-caused visibility degradation (in deciviews) times 1.25. The latter endpoint assumes that the visibility goal will be missed by 25%.

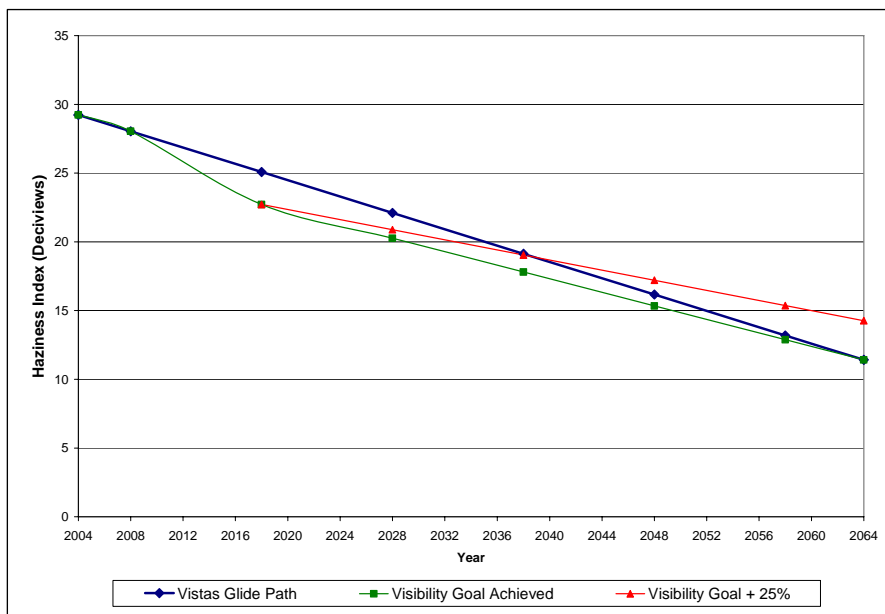


Figure 2. Average rate of change in visibility for three Class I areas utilizing the Environmental Protection Agency’s Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. The green line predicts the rate of visibility change between 2018 and 2064 if the Regional Haze goals are attained; while red line show the rate of visibility change if the Regional Haze goal is missed by 25 percent.

2. Assuming that percent changes in sulfur and nitrogen deposition will mimic visibility improvements (Figure 2) then the 2018 CMAQ results were reduced by 50 percent (Figure 2) to estimate the 2064 wet and dry deposition of sulfur and nitrogen compounds. After 2064, the deposition remained constant until 2100.
3. There is a possibility the Regional Haze visibility goals may not be achieved in 2064 either due to the cost of controls and/or international transport of pollutants. The third scenario addresses the possibility that the 2064 goal of natural visibility will be missed by 25 percent. For this scenario the 2018 CMAQ results were reduced by 37 percent (Figure 2) to estimate the 2064 wet and dry deposition of sulfur and nitrogen compounds. After 2064, the deposition remained constant until 2100.

Thus, under the aggressive additional emissions control scenario, both wet and dry deposition of S, NO_x, and NH₄ were reduced by 50% in a linear fashion from 2018 to 2064. This level of deposition reduction corresponded approximately with CMAQ estimates of the amount of ambient air pollution reduction that would need to occur in order to meet the national visibility goal.

Under the moderate additional emissions control scenario, both wet and dry deposition of S, NO_x, and NH₄ were also reduced in a linear fashion from 2018 to 2064. In this scenario, the deposition was reduced by 37% as compared with 2018 values.

5. Critical Loads Analysis

Critical loads modeling was performed independent of the scenario modeling discussed in the previous section. The critical loads analysis for each of the modeled streams determined threshold levels of sustained atmospheric deposition of S below which adverse effects to particular sensitive aquatic receptors would not be expected to occur, and evaluated interactions between the critical ANC endpoint value specified and the time period over which the critical load was examined. Critical loads for S deposition were calculated using the MAGIC model for each of the streams that were successfully calibrated in this project.

The MAGIC model was used in an iterative fashion to calculate the S deposition values that would cause the chemistry of each of the modeled streams to either increase or decrease streamwater ANC (depending on the current value) to reach the specified critical levels. For these analyses, the critical ANC levels were set at 0, 20, 50, and 100 µeq/L, the first two of which are believed to approximately correspond with chronic and episodic damage to relatively acid-tolerant brook trout populations (Bulger et al. 2000). Other more acid-sensitive species of aquatic biota may be impacted at higher ANC values. In order to conduct this critical loads

analysis for S deposition, it was necessary to specify the corresponding levels of N deposition. Nitrogen deposition accounts, however, for only a minor component of the overall acidification response of most streams in the forests under study. For this analysis, future N deposition was therefore held constant at recent levels.

It was also necessary to specify the times in the future at which the critical ANC values would be reached. We selected the years 2020, 2050, and 2100. It must be recognized that streamwater chemistry will continue to change in the future for many decades subsequent to stabilization of deposition levels. This is mainly because soils will continue to change in the degree to which they adsorb incoming S and because some watersheds will have become depleted of base cations. The former process contributes to a delayed acidification response. The latter process can cause streamwater base cation concentrations and ANC to decrease over time while SO_4^{2-} and NO_3^- concentrations maintain relatively constant levels. For these reasons, the sustained deposition loading that will cause the ANC of a given stream to decrease below a particular threshold value depends on the future year for which the evaluation is made.

In using MAGIC to estimate critical loads for the ANC levels and years discussed above, it was necessary to specify the temporal pattern of deposition changes as the critical load was approached. It was assumed that deposition from 2005 (the reference year for the MAGIC simulations) until 2009 would follow the VISTAS trajectory (see section 4a above). Changes in deposition leading to the critical load were begun in 2009 and completed by 2018, assuming a linear decrease (or increase) to the critical load value from 2009 to 2018. Deposition was then held constant at the critical loads value from 2018 until the end of the simulation, which was determined by the year selected for evaluation of the target ANC.

6. Sensitivity of Model Output to Major Uncertainties

The MAGIC model, like any process-based model of acid-base chemistry, is a simplification of an array of physical, chemical, and biological processes. Such simplification invariably results in uncertainty with respect to model structure and performance. Unfortunately, models of ecosystem behavior can never truly be validated because environmental systems are not closed and some processes might assume importance only under particular circumstances. Furthermore, with any model, it is possible to get the right answer for the wrong reason (c.f., Oreskes et al. 1994 for a discussion of model validation). Nevertheless, the MAGIC model has been extensively tested against independent measurements of chemical acidification and

recovery. These tests have included many comparisons between model projections of ANC or pH and the results of whole-watershed manipulation experiments and comparisons between model hindcasts of pH and diatom-inferred pH. In general, the MAGIC model has shown good agreement with these independent measurements or estimates of chemical change. It is beyond the scope of this project to provide additional tests of the model validity and accuracy. MAGIC was chosen for this project because of its well-documented history of tests of the model structure (see the review of Sullivan [2000] for additional information).

There are, however, numerous uncertainties associated with conducting an assessment of this type that can and should be examined, some of which are quantifiable, some not. Four major uncertainties that can be addressed here and that relate directly to the aims of this project are concerned with: 1) the assignment of soils data to specific watersheds; 2) assumptions regarding high-elevation occult deposition; 3) inter-annual variability in water chemistry data used for calibration; and 4) overall model simulation uncertainty. These errors and uncertainties are probably not additive, and might be expected to some extent to cancel each other out. Although it is not possible to rigorously quantify the overall uncertainty in the assessment results, analyses were conducted to evaluate and put into perspective these four major sources of uncertainties.

a. Uncertainty Due to Specification of Soils Data

Uncertainty was introduced into the modeling effort as a consequence of not having soils data available for all of the MAGIC modeling sites. For Tier II sites, soils data were borrowed from a nearby Tier I site located on the same geology. For Tier III sites, soils data were borrowed from the Tier I site judged to be most comparable with respect to streamwater ANC (an integrator of watershed soils conditions), geologic sensitivity class, location, elevation and streamwater sulfate concentration (an integrator of sulfur adsorption on soils).

The uncertainty associated with needing to borrow soils data for Tier II and Tier III sites can be examined by calibrating selected Tier I watersheds twice, once using the appropriate site-specific soils data, and a second time using borrowed soils data from an alternate site, using either Tier II or Tier III protocols. Both sets of calibrations for a site can be used for running historical or future simulations and for calculating critical loads. The simulated results for a site can then be compared to determine the magnitude of the differences in future simulated values (or estimated critical loads) resulting from the way soils data were supplied. If this sensitivity analysis is performed for a number of sites, it would be possible to determine if the differences in

soils data produced a consistent bias in results (all or most sites showing the same direction of change in simulated values). Performing the sensitivity analysis on a number of sites would also give a more robust estimate of the magnitude of the change produced, whether or not the change was biased.

Seven Tier I sites, selected to represent a range of acid-sensitivity, were calibrated a second time for this sensitivity analysis. For this second set of calibrations at each of the sites, watershed-specific soils data were not used. Rather, in each case soils data were borrowed from a nearby watershed, using the soils data borrowing procedures followed for Tier II and Tier III sites. Each “sensitivity calibration” for each site was then used for simulation of all future scenarios and for all critical loads calculations. Results were compared with those obtained on the basis of the original site calibrations for these watersheds.

b. Uncertainty Due to Specification of High Elevation Occult Deposition

The assumption of increased occult deposition at high elevations may introduce uncertainty in the modeling results. At very high elevations, the inputs of ions from cloud water can be very large. In this project there were five sites at elevations over 1500 meters for which the DDF values were increased to account for assumed increases in cloud water deposition.

The uncertainty associated with lack of observed data for high elevation occult deposition was examined by re-calibrating a number of modeled sites. The five high elevation sites (1,591 to 1,719 m) originally calibrated using the increased high elevation DDF values were re-calibrated using the lower ASTRAP DDF values. The next five highest sites (1245 to 1453 m) originally calibrated using the lower ASTRAP DDF values were re-calibrated using the increased high elevation DDF values. Both sets of calibrations for the ten sites were then used for running historical or future simulations and for calculating critical loads. The simulated results for the 10 sites were compared to determine the magnitude of the differences in future simulated values (or estimated critical loads) and to derive estimates of the bias and/or variance introduced in the assessment results because of the treatment (or lack thereof) of high elevation cloud water deposition.

c. Uncertainty Due to Specification of Stream Water Data for Calibration

There were 7 streams for which stream water samples were available for multiple years at the site. All 7 of these streams had samples available in 2000, and that year was used for

calibration of MAGIC at those sites. The uncertainty arising from using 2000 rather than another year to calibrate the model at these sites was examined by re-calibrating each of the seven sites using stream water data from another of the sampled years at the site. Three of the 7 sites had one more year of data available and that year was the adjacent year 1999. Four of the seven sites had one more year of data available in 2004. Two of the 7 sites each had a second year of data available in 2002.

This allowed nine re-calibrations using different observed stream water chemistry to examine the effects of inter-annual variability in stream water chemistry on calibration of MAGIC (five sites with one more year for re-calibration and two sites with two more years for re-calibration). Both sets of calibrations for the five sites and all three sets of calibrations for the two sites were then used for running historical or future simulations and for calculating critical loads. As for the other sensitivity analyses, the simulated results were compared to determine the magnitude of the differences in future simulated values (or estimated critical loads) and to derive estimates of the bias and/or variance introduced in the assessment results because of the selection of the calibration year.

In some cases where multiple years of streamwater chemistry are available, the MAGIC calibration procedure can be adapted: a) to use some average of the multiple years (to smooth through the inter-annual variability); or b) to match all years of available chemistry using a multiple year optimization routine. These approaches were not utilized in this project because the overwhelming majority of sites to be modeled (59 out of 66) only had observed stream water chemistry for a single year. It was decided that the seven sites with multiple years should be “officially” calibrated with only one year’s data for consistency with the majority of sites. The extra years were then included in this sensitivity analysis.

d. Combined Model Calibration and Simulation Uncertainty

The sensitivity analyses described above were designed to address specific assumptions or decisions that had to be made in order to assemble the data for the 66 modeled sites in a form that could be used for calibration of the model. In all cases, the above analyses address the questions of what the effect would have been if alternate available choices had been taken. These analyses were undertaken for a subset of sites for which the alternate choices were available at the same sites. As such, the analyses above are informative, but they provide no direct information about the uncertainty in calibration or simulation arising from the choices that were

incorporated into the final modeling protocol for *all* sites. That is, having made the choices about soils assignments, high elevation deposition, and stream samples for calibration (and provided an estimate of their inherent uncertainties), the need arises for a procedure for estimating uncertainty at each and all of the individual sites using the final selected calibration and simulation protocol.

These simulation uncertainty estimates were derived from the multiple calibrations at each site provided by the “fuzzy optimization” procedure employed in this project. For each of the 66 sites, 10 distinct calibrations were performed with the target values, parameter values, and deposition inputs for each calibration reflecting the uncertainty inherent in the observed data for the individual site. The effects of the uncertainty in the assumptions made in calibrating the model (and the inherent uncertainties in the data available) can be assessed by using all successful calibrations for a site when simulating the response to different scenarios of future deposition. The model then produces an ensemble of simulated values for each site. The median of all simulated values in a year is considered the most likely response of the site. The simulated values in the ensemble can also be used to estimate the magnitude of the uncertainty in the projection. Specifically, the difference in any year between the maximum and minimum simulated values from the ensemble of calibrated parameter sets can be used to define an “uncertainty” (or “confidence”) width for the simulation at any point in time. All ten of the successful model calibrations will lie within this range of values. These uncertainty widths can be produced for any variable and any year to monitor model performance.

7. Development of Landscape Variables for Regionalization of Modeling Results

Landscape variables including elevation, watershed area, ecoregion, lithology, forest type and geological sensitivity class were compiled for inclusion in multiple linear regression models to identify associations between characteristics of the landscape, modeled critical load, and stream chemistry. Watershed boundaries were provided by the Forest Service. EPA ecoregion types, as defined by Omernik, were identified, along with lithologic types from the USGS Draft Geologic Types of the Southern USA. USFS forest types were delineated from 1.1 km grid size satellite imagery (Haynes et al. 1995). Forest classes were grouped for some analyses into two categories: coniferous and hardwood forest.

Using the GIS, the percent area of each landscape variable was calculated for each watershed. Classes of landscape variables that were not represented in at least 10 modeled

watersheds were excluded from consideration for multiple regression modeling. The resulting set of landscape variables included ecoregion, lithology, geological sensitivity class, and forest type. Additionally, catchment area (ha) and elevation (m) at the sampling site were included in the analysis. Observed ANC and critical load to achieve the endpoint ANC of 20 $\mu\text{eq/L}$ in 2040 were selected as response variables.

The Statistix® software program was used to perform multiple linear regressions of the landscape variables. The Best Subsets Regression method was used to identify the models that showed the strongest associations between the landscape variables and CL.

IV. RESULTS AND DISCUSSION

A total of 66 stream sites in NC, TN, and SC were selected for modeling from among the 256 stream sites sampled by the Forest Service in recent years. Each of the 66 streams was successfully calibrated. The distribution of modeled sites by tier and geological sensitivity class is shown in Figure 3.

Site selection was not statistically-based, and we do not assume that the selected streams are representative of the overall population of streams on Forest Service land within the region. Rather, sites were generally selected for stream sampling by the Forest Service because they were suspected of being at least moderately acid-sensitive, based on geology, elevation, and the results of previous studies. Streamwater chemistry data for the sites selected for modeling are given in Table 7. For most sites, there was only one stream chemistry measurement. For sites having more than one sample, these data reflect an average of available sample results.

Summary statistics for selected key variables are provided in Table 8. In general, study watersheds were small (median 143 ha), located at high elevation (median 908 m), with low ANC (median 19 $\mu\text{eq/L}$) and exchangeable soil base cations (median % base saturation 10%). Median SO_4^{2-} (31 $\mu\text{eq/L}$) and NO_3^- (2 $\mu\text{eq/L}$) concentrations were low.

A. Model Calibration

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 is

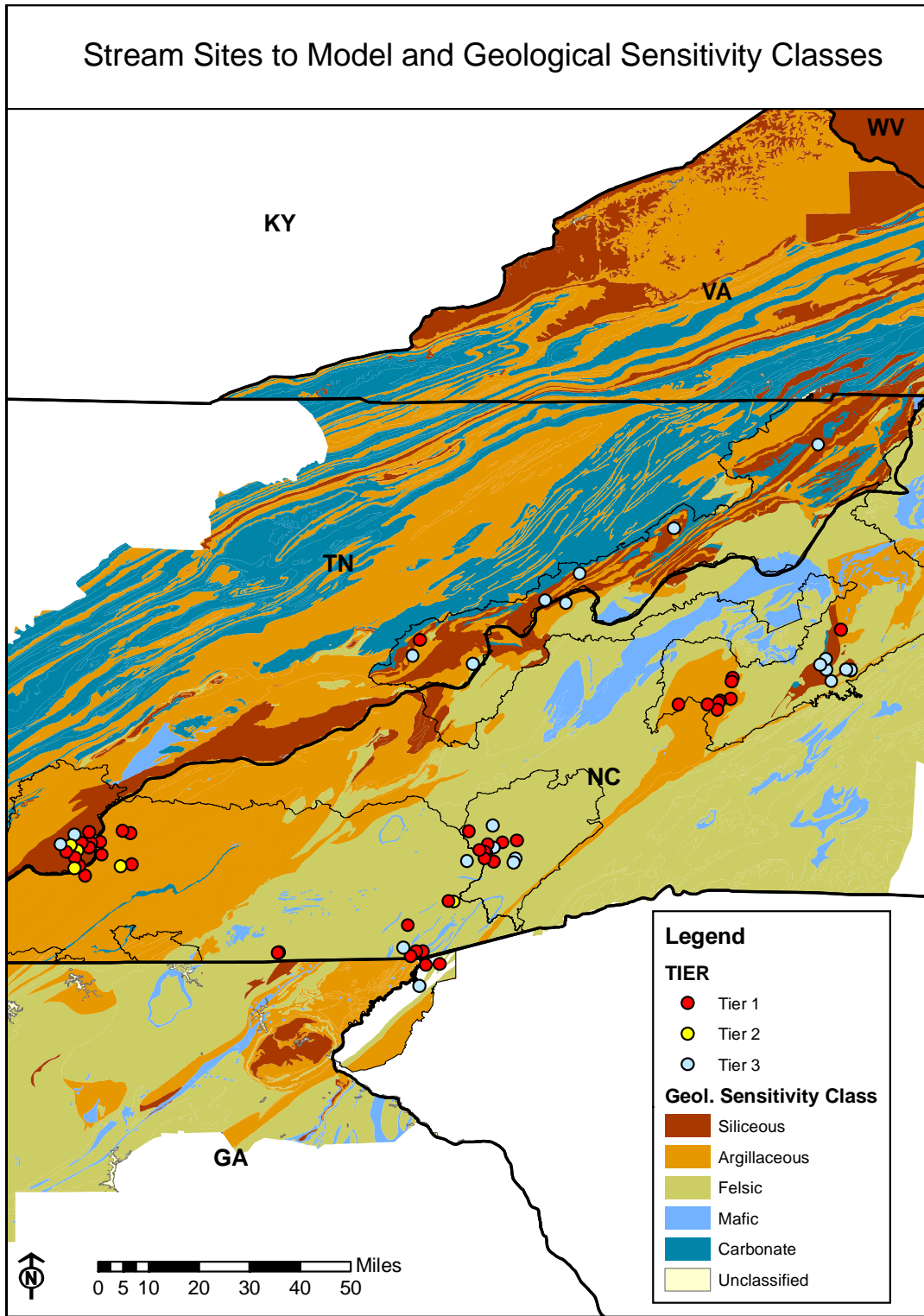


Figure 3. Location of sites selected for modeling, coded by tier. Tier I sites had both soil and stream chemistry data for calibration. Tier III sites had stream chemistry data for calibration, but soil data were borrowed from a nearby watershed that was judged to be similar with regard to acid-base chemistry.

Table 7. Streamwater chemistry data used in model calibration.

Site Name ³	Site ID	Calibration Year ¹	Observed Surface Water Concentrations ²												
			Qs	Ca	Mg	Na	K	NH4	SO4	Cl	NO3	SBC	SAA	CALK	pH
			m/yr	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	units
Adam Camp Branch	237	2003	1.08	16.2	14.6	27.9	9.9	0.0	17.4	11.7	0.0	68.6	29.1	39.4	6.55
Bear Branch	195	2003	0.58	45.1	41.7	30.4	16.2	0.0	71.2	10.7	0.0	133.3	82.0	51.3	6.49
Bearpen Branch	253	2004	0.98	18.2	15.0	28.4	8.1	0.5	30.4	13.8	3.8	69.7	48.0	22.2	6.16
Beetree Branch	190	2003	0.56	48.2	55.4	31.9	46.6	2.7	123.6	13.7	0.1	182.0	137.4	47.2	6.22
Big Cove Branch	259	2004	0.75	45.5	23.9	44.1	14.2	0.0	20.6	16.6	23.1	127.6	60.3	67.3	6.80
Big Laurel Brook	215	2003	1.41	11.3	12.1	22.8	6.8	0.0	10.6	13.1	0.5	53.0	24.1	28.9	6.32
Big Oak Cove Creek	261	2004	0.95	27.4	19.3	27.9	6.6	0.0	38.1	12.6	11.1	81.1	61.9	19.3	6.28
Briar Creek	72	2005	0.66	32.8	44.2	21.9	13.8	0.0	55.3	10.2	1.2	112.7	66.8	45.9	6.57
Bubbling Spring Branch	129	2003	1.15	20.3	11.9	21.1	6.3	0.0	44.2	9.8	1.0	59.6	55.0	4.6	5.23
Bubbling Spring West Tributary	131	2003	1.19	13.4	8.9	19.3	4.5	0.0	38.7	8.3	1.2	46.2	48.2	-2.1	4.89
Buckeye Cove Creek	133	2003	1.24	23.2	11.4	21.9	6.1	0.0	30.5	9.4	7.6	62.6	47.6	15.0	5.82
Cane Creek Tributary	204	2003	1.08	12.6	16.7	36.2	9.9	0.0	36.7	23.1	2.4	75.5	62.2	13.3	5.11
Cathey Creek	122	2003	0.84	46.3	31.5	38.5	15.0	0.0	36.0	13.7	10.5	131.3	60.3	71.0	6.85
Colberts Creek	53	2003	1.05	19.4	18.5	27.7	8.7	0.0	29.9	12.0	5.3	74.3	47.2	27.2	6.16
Courthouse Creek	121	2003	1.09	22.4	17.9	35.3	11.0	0.0	26.9	17.5	3.0	86.5	47.4	39.1	6.50
Dark Prong	114	2003	1.37	14.8	8.9	24.2	6.3	0.0	27.9	6.9	0.0	54.3	34.8	19.5	6.28
Davidson River	102	2000	1.51	34.5	23.7	38.1	10.2	5.1	27.3	11.2	3.1	106.6	41.7	70.0	6.45
East Fork Pigeon	101	2003	1.46	20.2	13.0	25.9	8.3	0.0	21.0	9.7	2.2	67.4	32.9	34.5	6.55
Flat Laurel Creek	127	2003	1.08	19.0	10.8	22.8	6.5	0.0	30.0	8.1	2.3	59.1	40.5	18.7	5.82
Glade Creek	198	2003	1.10	25.9	23.6	42.0	9.9	0.0	10.7	16.5	0.4	101.4	27.5	73.9	6.27
Greenland Creek	160	2003	1.15	13.8	10.3	33.6	6.8	0.0	28.6	14.3	1.8	64.5	44.6	19.9	5.48
Indian Branch	255	2004	0.70	53.4	23.0	38.9	13.2	0.0	34.0	23.7	22.9	128.4	80.6	47.8	6.69
Indian Camp	185	2004	0.76	38.9	22.7	51.5	10.4	1.7	12.8	36.9	0.7	123.5	50.4	74.9	6.47
Indian Spring Branch	239	2004	1.53	23.5	16.9	32.3	8.2	0.0	23.7	12.8	2.0	80.8	38.5	42.3	6.62
Kilby Creek	217	2003	1.30	14.4	13.7	25.9	8.4	0.0	10.4	12.9	0.7	62.4	23.9	38.5	6.54
Kirkland Cove	260	2004	0.84	33.0	15.8	33.1	9.9	0.0	31.0	13.9	11.3	91.8	56.1	35.7	6.58
Left Prong South Toe River	61	2000	1.11	26.5	18.0	33.8	6.5	5.7	17.7	13.8	14.6	84.8	46.1	44.4	6.23
Lindy Camp Branch	35	1999	0.79	31.6	27.9	19.8	16.6	0.0	48.0	11.0	0.6	95.8	59.7	36.1	5.89
Little Prong Hickey Fork	91	2000	0.86	44.5	51.3	41.9	10.4	8.1	21.4	6.2	6.3	148.1	33.9	122.4	6.67
Little Santetlah Cr. (NuCM site)	281	2000	1.32	30.9	16.8	25.7	8.6	0.4	32.0	12.5	6.7	82.1	51.2	31.3	6.23
Long Branch	103	2000	1.72	30.3	18.2	57.1	8.4	3.9	11.8	10.8	0.0	114.0	22.6	95.3	6.54
Lost Cove	56	2003	1.29	31.7	16.8	31.7	7.2	0.0	35.3	12.0	3.2	87.4	50.5	36.9	6.52
Lower Creek	59	2003	0.73	21.2	16.4	32.8	18.4	0.0	31.9	26.9	6.4	88.7	65.3	23.4	6.17
McNabb Creek	270	2004	0.53	123.3	61.0	69.7	14.8	0.0	207.4	57.2	0.4	268.7	265.1	3.7	6.00
Middle Creek	55	2003	1.05	19.1	17.1	28.2	8.2	0.0	29.4	12.3	4.1	72.5	45.8	26.7	6.12
Mill Station Creek	132	2004	0.71	35.6	19.9	49.5	11.7	0.0	59.5	44.0	4.0	116.6	107.5	9.1	5.87
Paddy Creek	26	2005	0.60	8.8	17.9	21.7	19.4	0.0	48.0	19.1	0.0	67.7	67.1	0.7	5.29

Table 7. Continued.

Site Name	Site ID	Calibration Year ¹	Observed Surface Water Concentrations ²												
			Qs	Ca	Mg	Na	K	NH ₄	SO ₄	Cl	NO ₃	SBC	SAA	CALK	pH
			m/yr	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L
Peach Orchard Creek	70	2000	0.63	43.2	18.8	35.2	6.7	6.4	50.3	11.0	20.1	104.0	81.4	29.0	5.86
Pigpen Branch	191	2000	0.91	30.5	30.2	53.2	13.9	0.0	9.6	20.7	0.0	127.8	30.3	97.5	6.84
Rattlesnake Branch	143	1999	0.52	28.3	20.1	60.0	12.9	0.0	39.3	14.3	0.5	121.2	54.1	67.1	6.36
Right Hand Prong	147	2003	1.02	32.1	23.3	31.9	9.9	0.0	29.4	12.0	12.7	97.2	54.1	43.1	6.59
Roaring Branch	251	2000	0.85	51.5	17.0	33.3	8.5	0.0	35.0	12.9	21.0	110.3	68.9	41.4	6.23
Rough Ridge Creek	257	2000	0.87	26.6	17.0	39.8	8.1	0.0	26.9	13.8	1.8	91.6	42.6	49.0	6.41
Russell Creek	15	2005	0.64	18.1	21.4	20.6	24.2	0.0	77.3	20.3	0.0	84.3	97.6	-13.4	4.81
Scotsman Creek	193	2003	1.00	17.4	17.7	38.6	9.0	0.0	16.5	17.7	1.5	82.6	35.7	46.9	6.59
South Fork Fowler Creek	188	2003	1.02	15.5	18.2	34.5	7.6	0.0	9.8	16.9	0.1	75.8	26.8	49.0	6.46
Spivey Creek	266	2004	0.71	42.7	27.0	46.9	17.5	1.9	44.1	16.7	5.0	134.1	65.8	70.2	6.73
Squibb Creek	88	1999	0.52	54.2	70.3	51.6	24.3	0.0	77.3	14.7	17.8	200.5	109.7	90.7	6.57
Stillhouse Branch	28	2005	0.67	6.1	16.0	17.1	12.9	0.0	33.5	16.2	0.0	52.0	49.7	2.2	5.27
Unnamed creek A	245	2004	1.22	22.5	12.3	26.2	8.0	0.0	30.1	11.5	11.7	69.1	53.3	15.8	6.17
Unnamed creek B	246	2004	0.63	57.1	21.6	44.7	7.4	0.0	45.4	48.1	16.9	130.8	110.5	20.3	6.26
Unnamed creek C	249	2004	0.70	42.3	19.8	36.7	11.4	0.0	27.9	25.3	18.3	110.2	71.5	38.7	6.49
Upper Creek	60	2003	0.74	20.5	16.6	27.8	16.9	0.0	33.3	25.1	6.1	81.8	64.6	17.2	6.06
UT Flat Laurel Creek	120	2000	0.75	21.8	13.2	34.5	6.3	4.3	36.0	17.7	3.2	75.8	56.9	23.2	5.68
UT Laurel Branch	265	2004	0.55	83.9	34.1	62.8	15.1	0.0	74.1	66.9	5.0	195.9	146.1	49.8	6.66
UT Linville River (NuCM site)	21	2004	0.71	14.6	10.8	19.3	10.1	0.0	52.7	17.8	1.2	54.8	71.6	-16.9	4.74
UT McNabb Creek	262	2004	0.76	15.8	10.9	28.8	9.5	0.0	20.3	15.4	0.5	65.1	36.2	28.8	6.32
UT North Fork of Catawba	33	2005	0.62	8.2	18.1	19.4	15.3	0.0	38.4	18.0	0.0	61.1	56.4	4.7	5.39
UT Paint Creek	94	1999	0.48	57.2	51.1	33.8	13.5	0.0	45.5	13.5	8.8	155.6	67.8	87.7	6.45
UT Panthertown Creek (Boggy Creek)	163	2000	1.10	24.3	9.4	40.5	5.6	2.9	31.3	12.7	1.2	79.7	45.2	37.3	5.70
UT Russell Creek	14	2005	0.67	11.5	14.7	15.7	18.1	0.0	58.7	20.7	0.0	60.0	79.4	-19.4	4.88
White Creek	16	2005	0.73	14.2	18.1	28.0	19.3	0.0	48.4	20.5	0.0	79.6	68.9	10.7	5.57
Wildcat Branch	232	2004	1.27	17.9	17.0	28.6	9.1	0.0	16.2	12.7	1.3	72.7	30.2	42.4	6.59
Wilson Creek	169	2000	0.96	32.4	22.5	59.1	10.4	0.0	10.5	39.3	0.7	124.4	50.5	73.9	6.32
Wolf Creek	150	2004	1.02	24.6	14.8	34.2	7.6	0.0	17.3	14.0	1.6	81.2	32.9	48.3	6.65
Yellow Fork	27	2005	0.61	9.3	19.2	16.1	14.7	0.0	48.5	16.5	0.0	59.3	65.0	-5.7	4.96

¹ Stream data used for calibration of MAGIC - If more than one sample was available for the calibration year, all samples were averaged for the year.

² Observed streamwater chemistry for the calibration year at each site. The streamwater concentrations were used as targets for calibration. The calibration year was not the same for all sites. Runoff (Qs) was calculated from the rainfall amount and a calculated yield at each site (the average yield across all sites was 55%). SBC is sum of base cations, SAA is sum of strong acid anions, CALK is calculated ANC.

³ See Table 2 for forest locations

Table 8. Summary statistics for the 66 modeled sites for selected key variables.

Parameter	Unit	Min	25th	Median	75th	Max	Mean
Watershed Area	ha	9.31	72.97	142.56	358.69	1,357.97	269.24
Stream ANC	µeq/L	-20.60	6.80	19.40	38.65	83.70	24.18
Elevation	m	411.00	740.25	907.50	1,149.75	1,719.00	955.59
Stream Nitrate	µeq/L	0.00	0.52	2.14	5.93	24.87	4.90
Stream pH	standard	4.74	5.83	6.30	6.55	6.85	6.14
Stream Sulfate	µeq/L	9.79	23.79	31.13	45.13	207.44	37.78
Soil BS	%	2.40	6.00	9.89	12.09	18.04	9.68
Soil Exch. Ca ²⁺	%	0.35	1.83	3.01	4.32	9.12	3.58
Soil Exch. Ca ²⁺ + Mg ²⁺	%	1.06	3.35	5.99	8.28	13.14	6.23
CEC	meq/kg	23.37	30.49	32.47	38.83	105.23	41.33

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 (streamwater 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 MAGIC model was calibrated 10 times at each of the 66 sites as part of a fuzzy optimization procedure (see discussion above) designed to provide estimates of simulation uncertainty. The model results presented in this report are based on the median values of the simulated water and soil chemistry variables from the multiple calibrations at each site using the fuzzy optimization procedure. The use of median values for each watershed helps to assure that the simulated responses are neither over- nor underestimates, but approximate the most likely behavior of each watershed (given the assumptions inherent in the model and the data used to constrain and calibrate the model). The uncertainty analyses make use of the maximum and minimum simulated values from the multiple calibrations for each site to calculate uncertainty “widths” (or confidence intervals) around the median simulated values.

1. Calibration Data

The calibration procedure requires that streamwater chemistry, soil chemical and physical characteristics, and atmospheric deposition data be available for each watershed. The water chemistry data needed for calibration are the concentrations of the individual base cations (Ca^{2+} , Mg^{2+} , Na^+ , and K^+) and acid anions (Cl^- , SO_4^{2-} , NO_3^-), and the stream pH. The soil data used in the model include soil depth and bulk density, soil pH, soil cation-exchange capacity, and exchangeable bases on the soil (Ca^{2+} , Mg^{2+} , Na^+ , and K^+). The atmospheric deposition inputs to the model include all major ions and must be estimates of total deposition, not just wet deposition.

The streamwater samples used for calibration at each site were collected by the USFS and pre-screened for use in this project as described above. The stream concentrations used for targets in calibrating MAGIC at each site are given in Table 7.

Soils data for model calibration were derived as vertically averaged values of soil parameters determined from the two soil layers sampled at each Tier I site. Soil samples represent the top 10 cm of mineral soil (shallow soil data) and an integrated sample taken at depth 10 cm to either bedrock or 50 cm, whichever came first. The soils data for the deep and shallow layers at each sampling site were averaged based on layer thickness and bulk density to obtain single vertically aggregated values for each soil pit. The vertically aggregated data were then spatially averaged (if necessary) to provide a single value for each soil variable in each watershed. Details of the soils data used for calibration at each site are given in Table 9.

The procedures used for deriving the deposition inputs at each site were described above. The wet concentrations, rainfall amounts and dry deposition factors used for the Reference Year during calibration of each site are summarized in Table 10. Total deposition estimates for the Reference Year 2005 are provided in Table 11. Total deposition estimates for 2009 and 2018 are provided in Appendix A.

2. Calibration Results

The procedures for calibrating and applying MAGIC to an individual site involved a number of steps, used a number of programs, and produced several discrete outputs. The input data required by the model (streamwater, watershed, soils, and deposition data) were assembled and maintained in databases (electronic spreadsheets) for each landscape unit. When complete,

Table 9. Model input soil data.

Site Name	Site ID	Soil Data Source	Observed Soil Variables						Calibrated Soil Parameters					
			ExCa %	ExMg %	ExNa %	ExK %	BS %	Soil pH	CEC meq/kg	WCa meq/m ² /yr	WMg meq/m ² /yr	WNa meq/m ² /yr	Wk meq/m ² /yr	Emx meq/kg
Adam Camp Branch	237	Tier 1	3.46	5.25	0.01	5.00	13.71	4.28	31.9	5.1	9.9	22.8	8.1	57.6
Bear Branch	195	Tier 3	1.65	1.29	0.01	2.58	5.53	3.96	36.1	19.6	21.7	14.0	8.0	7.8
Bearpen Branch	253	Tier 1	2.03	2.51	0.01	2.60	7.15	4.26	33.4	4.7	8.1	20.1	5.3	26.3
Beetree Branch	190	Tier 1	1.65	1.29	0.01	2.58	5.53	3.96	36.1	19.5	27.8	13.3	23.8	13.9
Big Cove Branch	259	Tier 2	7.29	3.13	1.38	3.39	15.19	4.39	30.5	16.4	11.0	25.2	7.7	46.2
Big Laurel Brook	215	Tier 1	1.14	5.21	0.83	3.32	10.50	4.12	36.2	4.9	11.5	19.7	7.4	91.7
Big Oak Cove Creek	261	Tier 2	5.59	3.33	0.01	3.17	12.09	4.44	29.2	5.6	7.8	19.3	3.0	24.7
Briar Creek	72	Tier 3	1.50	2.85	0.75	3.45	8.55	4.50	30.5	14.1	23.8	9.7	7.3	9.0
Bubbling Spring Branch	129	Tier 2	1.56	1.73	1.27	1.43	6.00	3.98	38.8	12.7	8.3	15.3	5.2	41.3
Bubbling Spring West Tributary	131	Tier 1	1.56	1.73	1.27	1.43	6.00	3.98	38.8	6.4	5.7	15.2	3.6	41.7
Buckeye Cove Creek	133	Tier 1	3.24	2.89	1.05	2.03	9.20	3.79	44.3	16.8	8.8	18.7	5.7	54.0
Cane Creek Tributary	204	Tier 3	3.24	2.89	1.05	2.03	9.20	3.79	44.3	0.0	9.9	22.9	7.6	23.5
Cathey Creek	122	Tier 3	1.84	1.25	0.16	0.94	4.19	4.26	103.7	28.2	21.6	24.8	10.9	18.4
Colberts Creek	53	Tier 1	3.00	3.37	1.15	4.16	11.68	4.25	32.5	7.5	11.6	19.6	6.4	23.8
Courthouse Creek	121	Tier 1	9.12	3.90	0.90	4.12	18.04	4.59	27.4	5.4	10.2	26.6	8.2	35.7
Dark Prong	114	Tier 1	2.50	1.83	1.18	1.72	7.23	4.12	34.3	10.2	7.5	25.3	6.7	58.7
Davidson River	102	Tier 3	1.84	1.25	0.16	0.94	4.19	4.26	103.7	38.8	30.2	46.6	13.4	39.3
East Fork Pigeon	101	Tier 1	1.84	1.25	0.16	0.94	4.19	4.26	103.7	16.6	13.3	28.0	9.9	38.1
Flat Laurel Creek	127	Tier 1	2.55	2.55	0.01	2.01	7.12	3.90	39.4	11.5	7.2	18.8	5.4	19.9
Glade Creek	198	Tier 1	2.29	3.57	1.32	2.93	10.10	4.31	31.3	17.7	20.9	34.1	8.9	79.0
Greenland Creek	160	Tier 2	3.01	0.87	0.01	1.03	4.91	4.06	35.6	3.9	7.2	28.1	5.8	27.0
Indian Branch	255	Tier 1	7.29	3.13	1.38	3.39	15.19	4.39	30.5	14.4	7.7	17.8	5.8	31.1
Indian Camp	185	Tier 1	5.20	4.38	0.96	4.07	14.61	4.50	29.3	15.2	11.4	24.2	5.6	61.6
Indian Spring Branch	239	Tier 2	4.29	3.08	0.01	3.66	11.04	4.37	30.4	16.2	17.1	39.2	9.0	49.9
Kilby Creek	217	Tier 1	2.08	2.92	0.58	4.10	9.68	4.48	27.8	7.4	12.6	22.1	8.8	86.8
Kirkland Cove	260	Tier 1	6.45	3.47	0.90	3.84	14.65	4.32	33.9	9.9	6.1	20.4	5.5	32.7
Left Prong South Toe River	61	Tier 1	3.98	3.08	0.39	3.30	10.76	3.92	38.1	16.7	14.6	28.7	5.3	55.1
Lindy Camp Branch	35	Tier 3	5.59	3.33	0.01	3.17	12.09	4.44	29.2	9.0	13.3	10.2	9.7	14.2
Little Prong Hickey Fork	91	Tier 3	8.57	2.75	0.31	2.04	13.67	4.26	105.2	30.0	39.5	32.4	7.9	22.8
Little Santetlah Cr. (NuCM site)	281	Tier 1	8.57	2.75	0.31	2.04	13.67	4.26	105.2	14.2	11.4	23.6	7.6	44.1
Long Branch	103	Tier 3	3.95	2.38	0.01	3.49	9.83	4.26	31.0	39.6	26.5	87.0	12.5	97.9
Lost Cove	56	Tier 1	4.33	4.02	1.02	3.74	13.11	4.05	37.6	24.4	13.4	31.3	6.6	27.1
Lower Creek	59	Tier 1	3.25	2.22	0.66	4.16	10.29	4.72	23.4	0.7	5.3	13.2	9.4	26.3
McNabb Creek	270	Tier 3	1.65	1.29	0.01	2.58	5.53	3.96	36.1	48.4	27.1	25.0	5.2	18.0
Middle Creek	55	Tier 1	1.50	2.85	0.75	3.45	8.55	4.50	30.5	9.7	11.0	21.1	6.1	27.4
Mill Station Creek	132	Tier 1	3.52	4.13	0.87	5.32	13.84	4.58	27.8	0.0	1.4	17.7	3.0	15.4

Table 9. Continued.

Site Name	Site ID	Soil Data Source	Observed Soil Variables						Calibrated Soil Parameters					
			ExCa %	ExMg %	ExNa %	ExK %	BS %	Soil pH	CEC meq/kg	WCa meq/m ² /yr	WMg meq/m ² /yr	WNa meq/m ² /yr	Wk meq/m ² /yr	Emx meq/kg
Paddy Creek	26	Tier 3	1.56	1.73	1.27	1.43	6.00	3.98	38.8	0.0	6.5	7.8	9.6	12.2
Peach Orchard Creek	70	Tier 1	4.79	3.29	0.45	3.38	11.91	4.12	38.1	13.1	5.7	17.4	2.6	9.2
Pigpen Branch	191	Tier 3	5.20	4.38	0.96	4.07	14.61	4.50	29.3	17.1	22.5	37.0	10.7	65.6
Rattlesnake Branch	143	Tier 3	1.84	1.25	0.16	0.94	4.19	4.26	103.7	6.5	7.0	26.5	5.3	12.3
Right Hand Prong	147	Tier 1	7.65	4.06	0.01	5.28	16.99	4.50	30.0	13.0	13.1	24.4	6.5	24.3
Roaring Branch	251	Tier 1	3.09	2.01	1.18	3.67	9.96	4.42	31.0	28.4	8.2	20.9	4.5	30.3
Rough Ridge Creek	257	Tier 1	5.59	3.33	0.01	3.17	12.09	4.44	29.2	8.5	8.4	27.6	4.7	32.0
Russell Creek	15	Tier 3	0.35	0.70	0.10	1.25	2.40	4.26	68.1	4.2	10.6	6.1	13.9	8.3
Scotsman Creek	193	Tier 1	1.53	1.50	2.05	1.29	6.37	4.40	27.7	7.6	13.7	27.6	7.3	46.8
South Fork Fowler Creek	188	Tier 1	1.83	1.68	0.85	2.36	6.71	4.25	29.0	6.5	14.6	24.1	6.1	87.9
Spivey Creek	266	Tier 1	8.70	4.44	0.01	4.80	17.95	4.42	31.5	11.8	10.6	26.8	8.8	19.8
Squibb Creek	88	Tier 3	8.70	4.44	0.01	4.80	17.95	4.42	31.5	10.4	22.8	22.4	8.6	4.2
Stillhouse Branch	28	Tier 3	1.56	1.73	1.27	1.43	6.00	3.98	38.8	0.0	7.3	7.0	7.3	21.0
Unnamed creek A	245	Tier 1	2.37	2.30	0.01	1.73	6.41	4.11	36.4	11.4	7.7	23.5	6.8	40.1
Unnamed creek B	246	Tier 1	1.89	1.91	0.01	3.79	7.60	4.29	30.3	16.3	6.2	15.8	1.5	26.1
Unnamed creek C	249	Tier 1	4.27	2.56	1.07	3.55	11.45	4.38	31.8	12.2	7.2	17.1	5.1	37.3
Upper Creek	60	Tier 1	4.28	2.70	1.01	2.54	10.53	4.33	31.3	0.2	5.1	9.5	9.2	23.3
UT Flat Laurel Creek	120	Tier 3	3.24	2.89	1.05	2.03	9.20	3.79	44.3	6.2	5.5	17.4	3.2	41.1
UT Laurel Branch	265	Tier 2	8.70	4.44	0.01	4.80	17.95	4.42	31.5	10.1	6.0	20.6	3.6	16.1
UT Linville River (NUCM site)	21	Tier 1	0.35	0.70	0.10	1.25	2.40	4.26	68.1	2.8	4.4	6.8	5.6	14.1
UT McNabb Creek	262	Tier 3	4.27	2.56	1.07	3.55	11.45	4.38	31.8	0.5	4.0	15.1	5.2	50.2
UT North Fork of Catawba	33	Tier 3	1.56	1.73	1.27	1.43	6.00	3.98	38.8	0.0	7.7	7.3	8.0	17.8
UT Paint Creek	94	Tier 3	3.00	3.37	1.15	4.16	11.68	4.25	32.5	19.7	19.8	12.0	5.2	9.2
UT Panthertown Creek (Boggy Creek)	163	Tier 1	3.01	0.87	0.01	1.03	4.91	4.06	35.6	15.7	6.4	36.1	4.5	20.6
UT Russell Creek	14	Tier 3	0.35	0.70	0.10	1.25	2.40	4.26	68.1	2.5	7.2	5.8	10.6	11.3
White Creek	16	Tier 3	2.29	3.57	1.32	2.93	10.10	4.31	31.3	0.0	4.9	10.4	10.0	15.6
Wildcat Branch	232	Tier 1	4.29	3.08	0.01	3.66	11.04	4.37	30.4	7.8	15.2	26.8	8.9	58.1
Wilson Creek	169	Tier 1	3.95	2.38	0.01	3.49	9.83	4.26	31.0	13.2	14.8	36.7	7.0	92.3
Wolf Creek	150	Tier 3	2.29	3.57	1.32	2.93	10.10	4.31	31.3	14.9	10.1	25.9	5.8	44.8
Yellow Fork	27	Tier 3	1.56	1.73	1.27	1.43	6.00	3.98	38.8	0.5	7.5	5.2	7.3	12.2

¹ The soils data for the two layers at each site were vertically aggregated using a bulk density weighting scheme. Observed soil exchangeable base cations and pH were used as targets for calibration. Average soil depth (.97 m), soil bulk density (1300 kg/m³), and soil porosity (50%) were assumed to be the same at all sites. Median soil logK_{al} across all sites. Weathering of each base cation (W_{xx}), and the maximum SO₄ adsorption capacity (Emx) were calibrated for each site - the values here are the median values for the ensemble calibrations.

Table 10. Rainfall amount, precipitation concentrations of all ions, and dry deposition factors (DDF) for SO₄, NO₃ and NH₄ in the simulation Reference Year 2005.

Site Name	Site ID	Rainfall Amount m/yr	Wet Concentrations								Dry Deposition Factors		
			Ca	Mg	Na	K	NH ₄	SO ₄	Cl ⁻	NO ₃	SO ₄	NO ₃	NH ₄
			µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	DDF	DDF	DDF
Adam Camp Branch	237	1.79	3.7	1.1	2.8	0.5	8.7	21.7	3.5	12.7	1.95	2.14	2.14
Bear Branch	195	0.99	3.7	1.0	2.5	0.5	9.0	21.1	3.1	12.7	1.95	2.14	2.14
Bearpen Branch	253	1.71	3.7	1.1	3.0	0.5	10.1	23.8	3.7	13.4	1.84	2.12	2.12
Beetree Branch	190	1.04	3.7	1.0	2.5	0.5	8.8	20.6	3.1	12.4	1.95	2.14	2.14
Big Cove Branch	259	1.43	3.7	1.1	2.8	0.5	8.7	20.8	3.6	12.2	1.95	2.14	2.14
Big Laurel Brook	215	2.32	2.9	1.0	3.4	0.4	8.6	19.9	4.0	11.3	1.84	2.12	2.12
Big Oak Cove Creek	261	1.58	3.8	1.1	3.0	0.5	9.1	22.2	3.8	12.8	1.95	2.14	2.14
Briar Creek	72	1.06	3.4	1.0	2.7	0.4	11.0	27.2	3.2	13.4	1.89	2.18	2.18
Bubbling Spring Branch	129	1.70	3.0	1.0	3.1	0.4	9.3	21.6	3.7	12.1	3.71	4.35	4.35
Bubbling Spring West Tributary	131	1.60	3.0	1.0	3.1	0.4	9.4	21.7	3.7	12.2	3.71	4.35	4.35
Buckeye Cove Creek	133	1.86	2.8	0.9	2.9	0.4	8.6	20.0	3.4	11.2	3.71	4.35	4.35
Cane Creek Tributary	204	2.25	2.8	1.0	3.4	0.4	8.5	19.8	3.9	11.3	1.84	2.12	2.12
Cathey Creek	122	1.49	2.9	1.0	3.0	0.4	9.0	21.0	3.6	11.8	1.84	2.12	2.12
Colberts Creek	53	1.81	2.7	0.9	2.7	0.4	9.3	23.8	3.3	11.5	1.89	2.18	2.18
Courthouse Creek	121	2.11	2.9	1.0	3.1	0.4	9.2	21.6	3.7	12.0	1.84	2.12	2.12
Dark Prong	114	1.66	2.9	1.0	3.1	0.4	9.2	21.5	3.7	12.0	3.71	4.35	4.35
Davidson River	102	2.42	2.9	1.0	3.0	0.4	9.1	21.6	3.6	12.0	1.84	2.12	2.12
East Fork Pigeon	101	2.21	2.8	0.9	2.9	0.4	8.6	20.5	3.5	11.4	1.84	2.12	2.12
Flat Laurel Creek	127	1.39	3.2	1.1	3.3	0.5	9.8	22.8	3.9	12.8	1.84	2.12	2.12
Glade Creek	198	2.00	2.8	1.0	3.5	0.4	8.6	19.8	4.0	11.3	1.84	2.12	2.12
Greenland Creek	160	1.97	2.8	1.0	3.4	0.4	8.6	20.0	4.0	11.4	1.84	2.12	2.12
Indian Branch	255	1.51	3.9	1.1	2.8	0.5	9.2	22.4	3.6	13.0	1.95	2.14	2.14
Indian Camp	185	1.81	2.9	1.0	3.4	0.4	8.7	20.0	4.0	11.4	1.84	2.12	2.12
Indian Spring Branch	239	2.62	3.4	1.0	2.8	0.5	9.5	23.0	3.6	13.0	1.84	2.12	2.12
Kilby Creek	217	1.84	3.9	1.4	4.5	0.6	11.7	27.2	5.3	15.4	1.84	2.12	2.12
Kirkland Cove	260	1.47	3.8	1.1	2.9	0.5	9.1	21.6	3.7	12.6	1.95	2.14	2.14
Left Prong South Toe River	61	2.07	2.7	0.9	2.6	0.4	9.7	24.8	3.2	11.6	1.89	2.18	2.18
Lindy Camp Branch	35	1.27	4.0	1.1	2.8	0.5	11.8	29.2	3.5	15.0	1.89	2.18	2.18
Little Prong Hickey Fork	91	1.08	3.2	0.9	2.6	0.4	10.6	26.9	3.1	12.9	1.89	2.18	2.18
Little Santetlah Cr. (NuCM site)	281	2.28	3.5	1.0	2.7	0.5	8.3	20.2	3.4	11.9	1.95	2.14	2.14
Long Branch	103	2.71	2.8	1.0	3.0	0.4	8.6	20.5	3.6	11.4	1.84	2.12	2.12
Lost Cove	56	2.23	2.7	0.9	2.7	0.4	9.4	23.8	3.3	11.4	1.89	2.18	2.18
Lower Creek	59	1.67	2.6	0.9	2.7	0.4	9.6	24.5	3.3	11.6	1.89	2.18	2.18
McNabb Creek	270	1.40	4.0	1.1	2.9	0.5	9.4	22.9	3.7	13.2	1.95	2.14	2.14
Middle Creek	55	1.81	2.7	0.9	2.8	0.4	9.4	24.0	3.4	11.6	1.89	2.18	2.18
Mill Station Creek	132	1.76	3.1	1.1	3.3	0.5	10.0	23.3	4.0	12.9	1.84	2.12	2.12

Table 10. Continued.

Site Name	Site ID	Rainfall Amount m/yr	Wet Concentrations								Dry Deposition Factors		
			Ca	Mg	Na	K	NH ₄	SO ₄	Cl ¹	NO ₃	SO ₄	NO ₃	NH ₄
			µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	µeq/L	DDF	DDF	DDF
Paddy Creek	26	1.20	2.9	1.0	2.9	0.4	10.4	25.7	3.6	12.2	1.89	2.18	2.18
Peach Orchard Creek	70	1.06	2.7	0.9	2.6	0.4	10.0	25.2	3.2	11.7	1.89	2.18	2.18
Pigpen Branch	191	1.81	2.9	1.0	3.5	0.4	8.6	19.9	4.1	11.4	1.84	2.12	2.12
Rattlesnake Branch	143	0.99	3.3	1.0	2.5	0.4	10.2	25.7	3.1	13.0	1.89	2.18	2.18
Right Hand Prong	147	1.67	3.0	1.0	3.1	0.5	9.3	22.0	3.7	12.4	1.84	2.12	2.12
Roaring Branch	251	1.45	3.9	1.1	2.9	0.5	9.1	21.8	3.7	12.8	1.95	2.14	2.14
Rough Ridge Creek	257	1.53	3.7	1.1	2.9	0.5	8.9	21.2	3.7	12.4	1.95	2.14	2.14
Russell Creek	15	1.32	2.9	1.0	2.9	0.4	10.7	26.4	3.6	12.5	1.89	2.18	2.18
Scotsman Creek	193	1.90	2.8	1.0	3.4	0.4	8.5	19.5	3.9	11.1	1.84	2.12	2.12
South Fork Fowler Creek	188	1.82	3.0	1.1	3.6	0.5	9.1	20.9	4.2	11.9	1.84	2.12	2.12
Spivey Creek	266	1.34	3.8	1.1	2.9	0.5	9.0	21.7	3.7	12.7	1.95	2.14	2.14
Squibb Creek	88	0.99	3.1	0.9	2.6	0.4	10.0	25.0	3.1	12.3	1.89	2.18	2.18
Stillhouse Branch	28	1.28	2.9	1.0	2.8	0.4	10.5	26.0	3.5	12.2	1.89	2.18	2.18
Unnamed creek A	245	1.96	3.7	1.1	2.9	0.5	9.0	22.1	3.7	12.8	1.95	2.14	2.14
Unnamed creek B	246	1.62	3.8	1.1	2.8	0.5	9.0	22.1	3.6	12.8	1.95	2.14	2.14
Unnamed creek C	249	1.55	3.7	1.1	2.8	0.5	8.9	21.7	3.5	12.6	1.95	2.14	2.14
Upper Creek	60	1.68	2.7	0.9	2.7	0.4	9.7	24.9	3.3	11.7	1.89	2.18	2.18
UT Flat Laurel Creek	120	1.44	3.0	1.0	3.1	0.5	9.5	22.0	3.7	12.3	3.71	4.35	4.35
UT Laurel Branch	265	1.49	3.9	1.1	2.8	0.5	9.2	22.5	3.7	13.0	1.95	2.14	2.14
UT Linville River (NUCM site)	21	1.40	2.9	1.0	2.9	0.4	10.7	26.7	3.5	12.6	1.89	2.18	2.18
UT McNabb Creek	262	1.33	4.3	1.2	3.1	0.6	10.2	25.3	4.1	14.5	1.95	2.14	2.14
UT North Fork of Catawba	33	1.24	2.9	0.9	2.8	0.4	10.4	25.7	3.4	12.1	1.89	2.18	2.18
UT Paint Creek	94	0.90	3.1	0.9	2.5	0.4	10.2	25.6	3.1	12.5	1.89	2.18	2.18
UT Panthertown Creek (Boggy Creek)	163	1.77	2.9	1.1	3.5	0.4	8.8	20.4	4.0	11.6	1.84	2.12	2.12
UT Russell Creek	14	1.37	2.9	1.0	2.9	0.4	10.4	25.6	3.6	12.2	1.89	2.18	2.18
White Creek	16	1.52	2.9	1.0	2.9	0.4	10.2	25.1	3.6	12.0	1.89	2.18	2.18
Wildcat Branch	232	2.17	3.4	1.0	2.9	0.5	9.5	23.0	3.6	13.0	1.84	2.12	2.12
Wilson Creek	169	2.27	2.9	1.1	3.6	0.5	8.9	20.9	4.2	11.9	1.84	2.12	2.12
Wolf Creek	150	1.75	3.1	1.1	3.2	0.5	9.6	22.4	3.9	12.5	1.84	2.12	2.12
Yellow Fork	27	1.16	2.9	1.0	2.9	0.4	10.5	26.0	3.6	12.3	1.89	2.18	2.18

¹ Dry deposition of Cl (and SeaSalt cations) at each site was added to produce Cl steady state with observed stream Cl and calculated runoff.

Table 11. Total deposition (wet plus dry plus cloud) of ions at each modeled site for the reference year 2005.

Name	Site ID	Precip (m/yr)	Total Deposition at Each Site (meq/m ² /yr) ¹										
			Ca	Mg	Na	K	NH ₄	SO ₄	Cl	NO ₃	SBC	SAA	CALK
Adam Camp Branch	237	1.8	9.5	2.7	7.1	1.3	33.6	76.1	12.6	48.9	20.6	137.5	-83.3
Bear Branch	195	1.0	5.2	1.4	3.5	0.7	19.0	40.7	6.2	26.8	10.8	73.7	-43.9
Bearpen Branch	253	1.7	9.4	2.8	7.6	1.3	36.6	74.9	13.3	48.7	21.1	136.9	-79.2
Beetree Branch	190	1.0	5.8	1.6	4.0	0.8	19.4	41.6	7.5	27.4	12.2	76.4	-44.8
Big Cove Branch	259	1.4	8.6	2.5	6.6	1.2	26.9	58.5	12.3	37.7	18.8	108.4	-62.7
Big Laurel Brook	215	2.3	9.9	3.6	11.8	1.5	42.1	84.6	18.2	55.4	26.9	158.2	-89.2
Big Oak Cove Creek	261	1.6	8.7	2.6	6.9	1.2	30.9	68.6	12.0	43.6	19.3	124.2	-74.0
Briar Creek	72	1.1	5.1	1.5	4.0	0.6	25.1	54.1	6.6	30.7	11.2	91.5	-55.2
Bubbling Spring Branch	129	1.7	6.9	2.4	7.3	1.0	68.1	134.7	11.2	88.3	17.6	234.1	-148.5
Bubbling Spring West Tributary	131	1.6	6.3	2.2	6.6	1.0	64.8	127.9	9.9	84.1	16.1	221.9	-141.0
Buckeye Cove Creek	133	1.9	7.3	2.5	7.5	1.1	70.0	138.6	11.7	91.2	18.4	241.4	-153.1
Cane Creek Tributary	204	2.3	12.0	4.4	14.4	1.9	40.9	82.2	24.2	53.8	32.6	160.3	-86.8
Cathy Creek	122	1.5	6.9	2.3	6.9	1.0	28.7	58.2	11.5	37.8	17.1	107.5	-61.7
Colberts Creek	53	1.8	7.4	2.5	7.6	1.0	36.8	81.3	12.5	45.2	18.6	138.9	-83.6
Courthouse Creek	121	2.1	10.4	3.6	11.0	1.6	40.9	83.6	18.7	53.6	26.5	155.9	-88.5
Dark Prong	114	1.7	6.3	2.1	6.5	0.9	66.5	133.2	9.6	87.3	15.9	230.1	-147.7
Davidson River	102	2.4	10.2	3.5	10.6	1.5	46.4	96.0	16.8	61.3	25.7	174.2	-102.0
East Fork Pigeon	101	2.2	8.6	2.9	9.0	1.3	40.2	83.2	14.0	53.4	21.8	150.6	-88.6
Flat Laurel Creek	127	1.4	5.7	1.9	5.9	0.8	28.9	58.0	8.8	37.6	14.5	104.5	-61.1
Glade Creek	198	2.0	9.2	3.4	11.3	1.4	36.6	73.0	17.8	47.9	25.3	138.7	-76.8
Greenland Creek	160	2.0	8.6	3.2	10.4	1.3	35.8	72.2	16.1	47.2	23.4	135.5	-76.4
Indian Branch	255	1.5	10.8	3.1	7.9	1.5	29.7	66.1	16.1	41.9	23.2	124.1	-71.2
Indian Camp	185	1.8	12.5	4.6	15.0	1.9	33.8	67.1	27.2	44.0	34.0	138.3	-70.5
Indian Spring Branch	239	2.6	13.4	4.1	11.1	1.9	53.0	110.9	19.3	72.2	30.4	202.4	-119.0
Kilby Creek	217	1.8	9.6	3.5	11.2	1.5	45.2	90.7	16.5	59.0	25.7	166.2	-95.3
Kirkland Cove	260	1.5	8.3	2.4	6.4	1.1	28.4	61.9	11.5	39.6	18.3	113.0	-66.3
Left Prong South Toe River	61	2.1	9.0	2.9	8.8	1.3	43.6	96.9	15.1	52.1	22.0	164.1	-98.5
Lindy Camp Branch	35	1.3	7.1	2.0	5.0	0.9	32.9	70.4	8.7	41.7	15.0	120.7	-72.8
Little Prong Hickey Fork	91	1.1	4.3	1.3	3.5	0.6	24.8	54.4	5.3	30.0	9.6	89.7	-55.4
Little Santetlah Cr. (NuCM site)	281	2.3	11.9	3.5	9.2	1.7	40.7	90.5	16.4	58.6	26.2	165.5	-98.6
Long Branch	103	2.7	10.9	3.7	11.6	1.6	49.2	102.1	18.3	65.6	27.9	185.9	-108.9
Lost Cove	56	2.2	9.4	3.0	9.2	1.3	45.9	100.5	15.4	55.7	23.0	171.5	-102.7
Lower Creek	59	1.7	9.8	3.2	10.1	1.3	35.3	78.2	18.9	42.6	24.5	139.8	-80.0
McNabb Creek	270	1.4	16.8	4.8	12.3	2.2	28.4	63.6	29.3	40.1	36.0	132.9	-68.5
Middle Creek	55	1.8	7.5	2.5	7.8	1.1	37.1	82.0	12.8	45.6	18.9	140.4	-84.4
Mill Station Creek	132	1.8	14.5	5.0	15.2	2.2	37.2	75.3	29.7	48.2	36.9	153.2	-79.1
Paddy Creek	26	1.2	3.8	1.3	3.9	0.5	27.3	58.7	11.2	32.1	9.4	101.9	-65.2
Peach Orchard Creek	70	1.1	4.2	1.4	4.2	0.6	22.9	50.2	6.8	26.9	10.4	83.9	-50.6
Pigpen Branch	191	1.8	9.2	3.4	11.3	1.4	33.0	66.0	18.4	43.5	25.2	127.8	-69.6
Rattlesnake Branch	143	1.0	5.2	1.5	4.0	0.7	22.0	48.0	7.3	28.0	11.4	83.3	-49.9
Right Hand Prong	147	1.7	7.5	2.5	7.6	1.1	33.2	67.6	12.2	44.0	18.7	123.8	-71.9
Roaring Branch	251	1.5	8.2	2.4	6.2	1.1	28.4	62.1	11.0	39.9	17.9	112.9	-66.6
Rough Ridge Creek	257	1.5	8.6	2.5	6.7	1.2	29.2	63.2	11.9	40.6	18.9	115.7	-67.6
Russell Creek	15	1.3	7.1	2.4	7.1	1.0	30.7	65.9	12.7	35.8	17.6	114.4	-66.1
Scotsman Creek	193	1.9	8.9	3.3	10.7	1.4	34.1	67.8	17.3	44.5	24.3	129.7	-71.3
South Fork Fowler Creek	188	1.8	8.8	3.3	10.8	1.4	35.5	70.8	17.0	46.4	24.3	134.2	-74.5
Spivey Creek	266	1.3	8.2	2.4	6.3	1.1	25.9	56.9	11.6	36.3	17.9	104.8	-61.0
Squibb Creek	88	1.0	5.0	1.5	4.1	0.6	21.5	46.4	7.3	26.4	11.2	80.1	-47.5

Table 11. Continued.

Name	Site ID	Precip (m/yr)	Total Deposition at Each Site (meq/m ² /yr) ¹										
			Ca	Mg	Na	K	NH ₄	SO ₄	Cl	NO ₃	SBC	SAA	CALK
Stillhouse Branch	28	1.3	3.6	1.2	3.6	0.5	29.2	62.9	10.6	34.0	8.9	107.5	-69.4
Unnamed creek A	245	2.0	10.3	3.0	8.1	1.4	37.5	84.0	13.9	53.5	22.8	151.4	-91.1
Unnamed creek B	246	1.6	16.7	4.8	12.5	2.3	31.2	69.8	28.8	44.5	36.2	143.1	-75.6
Unnamed creek C	249	1.6	11.2	3.2	8.4	1.5	29.7	66.1	17.2	42.1	24.3	125.4	-71.4
Upper Creek	60	1.7	9.5	3.1	9.7	1.3	35.7	79.1	18.0	43.0	23.6	140.0	-80.8
UT Flat Laurel Creek	120	1.5	7.4	2.5	7.7	1.1	60.2	118.3	13.1	77.5	18.7	208.9	-130.0
UT Laurel Branch	265	1.5	19.4	5.5	14.2	2.6	29.2	65.1	34.7	41.2	41.7	141.0	-70.2
UT Linville River (NUCM site)	21	1.4	7.0	2.3	7.0	1.0	32.5	70.6	12.3	38.2	17.4	121.2	-71.3
UT McNabb Creek	262	1.3	8.6	2.4	6.3	1.2	29.1	65.9	11.5	41.5	18.4	118.9	-71.4
UT North Fork of Catawba	33	1.2	3.7	1.2	3.7	0.5	27.8	59.9	10.8	32.5	9.2	103.2	-66.2
UT Paint Creek	94	0.9	4.4	1.3	3.6	0.6	20.0	43.5	6.4	24.5	9.9	74.3	-44.5
UT Panthertown Creek (Boggy Cr)	163	1.8	7.6	2.8	9.2	1.2	33.5	67.1	14.0	43.9	20.8	125.0	-70.7
UT Russell Creek	14	1.4	4.6	1.5	4.6	0.7	31.1	66.6	13.4	36.5	11.4	116.4	-73.9
White Creek	16	1.5	7.9	2.7	7.8	1.2	32.9	70.3	14.3	38.7	19.6	123.3	-70.9
Wildcat Branch	232	2.2	10.9	3.4	9.2	1.6	43.6	91.1	15.9	59.4	25.0	166.4	-97.9
Wilson Creek	169	2.2	16.0	5.9	19.8	2.5	42.5	86.4	35.5	56.5	44.2	178.4	-91.8
Wolf Creek	150	1.8	8.2	2.8	8.7	1.2	35.6	72.1	14.0	46.5	21.0	132.7	-76.1
Yellow Fork	27	1.2	3.3	1.1	3.3	0.5	26.6	57.2	9.8	31.1	8.2	98.1	-63.4

¹ Units of meq/m²/yr of deposition of SO₄²⁻, NO₃⁻, and NH₄⁺ can be converted to units of kg/ha/yr of either S or N by multiplying by 0.16, 0.14, and 0.14, respectively

these data bases were accessed by a program (MAGIC-IN) that generated the initial parameter files (xxx.PR) and optimization (xxx.OPT) files for each site. The initial parameter files contain observed (or estimated) soils, deposition, Mg and watershed data for each site. The optimization files contain the observed soil and streamwater data that are the targets for the calibration at each site, and the ranges of uncertainty in each of the observed values.

The initial parameter and optimization files for each site were sequentially passed to the optimization program (MAGIC-OPT). This program produced three outputs as each site was calibrated. The first (xxx.OUT) is an ASCII file of results that was passed to statistical routines for analysis and summary of model goodness-of-fit for the site. The second (xxx.PR1 ... xxx.PR10) was the multiple calibrated parameter set used in the fuzzy calibration procedure to assess model uncertainty (see below). The third (xxx.PAR) was the average parameter set for each site (average of the multiple calibration parameter sets) which represents the most likely response of the site.

The multiple calibrated parameter set (xxx.PR1...xxx.pr10) for each site was used by the program MAGIC-RUN with estimates of historical or future deposition to produce two outputs: 1) reconstructions of historical change at the site; and 2) forecasts of most likely future responses

for the applied future deposition scenario. The multiple calibrated parameter sets were also used with the same estimates of future deposition and the program MAGIC-RUN to produce an analysis of the uncertainty in model projections for that scenario. The results of the uncertainty analysis are in the form of an electronic spreadsheet giving simulated ranges (upper and lower values) for all modeled variables for each year of each scenario at each site.

Model Goodness-of-fit for Calibration Data

The multiple calibration procedure for each site produced summary statistics (mean, standard deviation, maximum and minimum) for the observed values, the simulated values and the differences (simulated-observed values) of each of the stream variables and soil variables simulated for each of the sites. These data are summarized in Table 12. In addition, plots of simulated versus observed values for stream variables were constructed (Figure 4).

These analyses showed that the model calibration results were not biased and did not contain unacceptably large residual errors.

B. Hindcast Simulation Results

Stream hindcast chemistry simulated for selected years in the past (1860, 1900, 1964, 1975, 1989, 1995, and 2005) are shown in Table 13 and Appendix B. Simulation results for all major streamwater chemistry variables for the year 1860 are shown in Table 13. Comparable simulation results for the other years are given in the appendix. MAGIC model simulations predicted that stream ANC values were above 20 $\mu\text{eq/L}$ in all modeled watersheds in 1860, but below 50 $\mu\text{eq/L}$ in 38% of the watersheds and below 100 $\mu\text{eq/L}$ in 86% percent of the watersheds at that time (Figure 5, Table 14). The minimum simulated ANC in 1860 among the modeled streams was 30 $\mu\text{eq/L}$ (Figure 5). The hindcast simulation results suggested that the average of the modeled streams was acidified from ANC=65 $\mu\text{eq/L}$ in 1860 to ANC=36 $\mu\text{eq/L}$ in 2005. The lowest simulated ANC in 2005 was -19 $\mu\text{eq/L}$, which was 49 $\mu\text{eq/L}$ lower than the minimum simulated value under pre-industrial (1860) conditions.

C. Future Simulation Results in Response to Emissions Control Scenarios

Simulation results for the future were based on estimates of atmospheric deposition in four years: 2002, 2009, 2018, 2064. There is some uncertainty in the MAGIC simulations caused by uncertainty in the CMAQ calibration of current (2002) conditions as well as projections of

Table 12. Distribution of simulated and observed water chemistry and soil parameter values for the 66 modeled sites.

Water Chemistry

n = 66		Simulated Variable Values										
	Ca	Mg	Na	K	NH4	SO4	Cl	NO3	SBC	SAA	Calk	pH
Ave	29.5	21.6	33.8	11.6	0.7	38.0	17.4	4.9	96.5	60.2	36.9	6.13
Max	123.3	70.4	69.7	46.6	8.1	207.6	63.0	23.2	268.7	263.4	122.2	6.83
Min	7.1	8.8	15.7	4.5	0.0	9.6	6.2	0.0	46.0	22.7	-19.2	4.73
		Observed Variable Values										
	Ca	Mg	Na	K	NH4	SO4	Cl	NO3	SBC	SAA	Calk	pH
Ave	29.4	21.6	33.9	11.6	0.7	37.8	17.8	4.9	96.5	60.4	36.7	6.13
Max	123.3	70.3	69.7	46.6	8.2	207.4	66.9	23.1	268.7	265.1	122.4	6.84
Min	6.1	8.9	15.7	4.5	0.0	9.6	6.2	0.0	46.2	22.6	-19.4	4.74
		Simulated-Observed										
	Ca	Mg	Na	K	NH4	SO4	Cl	NO3	SBC	SAA	Calk	pH
Ave	0.1	0.0	0.0	0.0	0.0	0.2	-0.4	0.0	0.0	-0.2	0.2	0.00
Max	3.5	0.1	0.0	0.0	0.1	1.0	0.5	0.4	2.9	1.1	3.7	0.18
Min	-0.3	-0.1	-0.4	-0.3	-0.1	0.0	-3.9	-0.6	-0.8	-3.8	-1.2	-0.28

Soil Chemistry

n = 66		Simulated Variable Values				
	pH1	BS1	ECa1	EMg1	ENa1	EK1
Ave	4.25	9.7	3.6	2.6	0.6	2.8
Max	4.73	18.0	9.1	5.2	2.1	5.3
Min	3.79	2.4	0.3	0.7	0.0	0.9
		Observed Variable Values				
	pH1	BS1	ECa1	EMg1	ENa1	EK1
Ave	4.24	9.7	3.6	2.7	0.6	2.8
Max	4.72	18.0	9.1	5.3	2.1	5.3
Min	3.79	2.4	0.4	0.7	0.0	0.9
		Simulated-Observed				
	pH1	BS1	ECa1	EMg1	ENa1	EK1
Ave	0.01	0.0	0.0	0.0	0.0	0.0
Max	0.03	0.0	0.0	0.0	0.0	0.0
Min	0.00	0.0	0.0	0.0	0.0	0.0

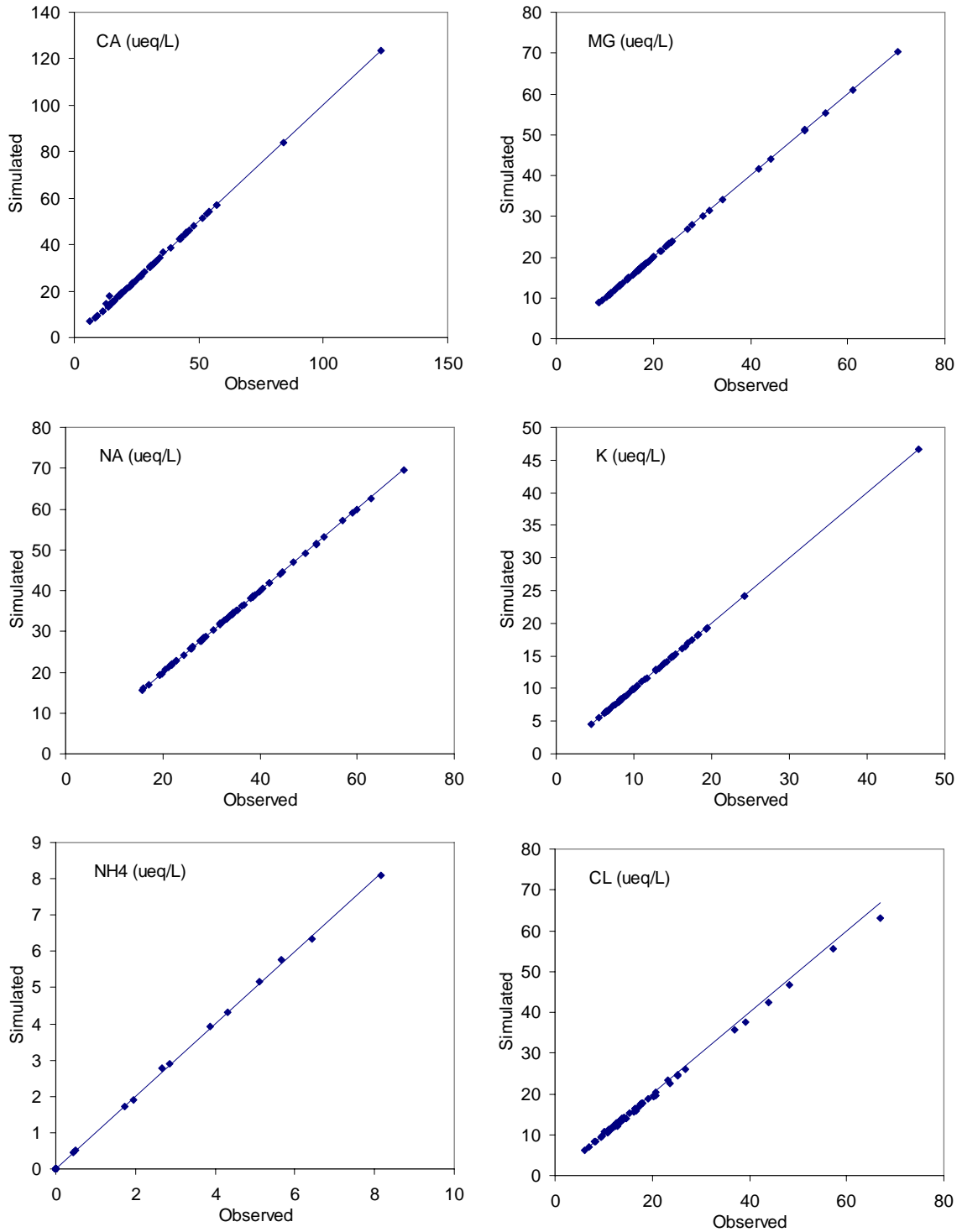


Figure 4. Calibration results for the MAGIC model. Predicted vs observed values of streamwater (first two pages) and soil (third page) variables for the 66 sites in the calibration year. (1:1 lines added)

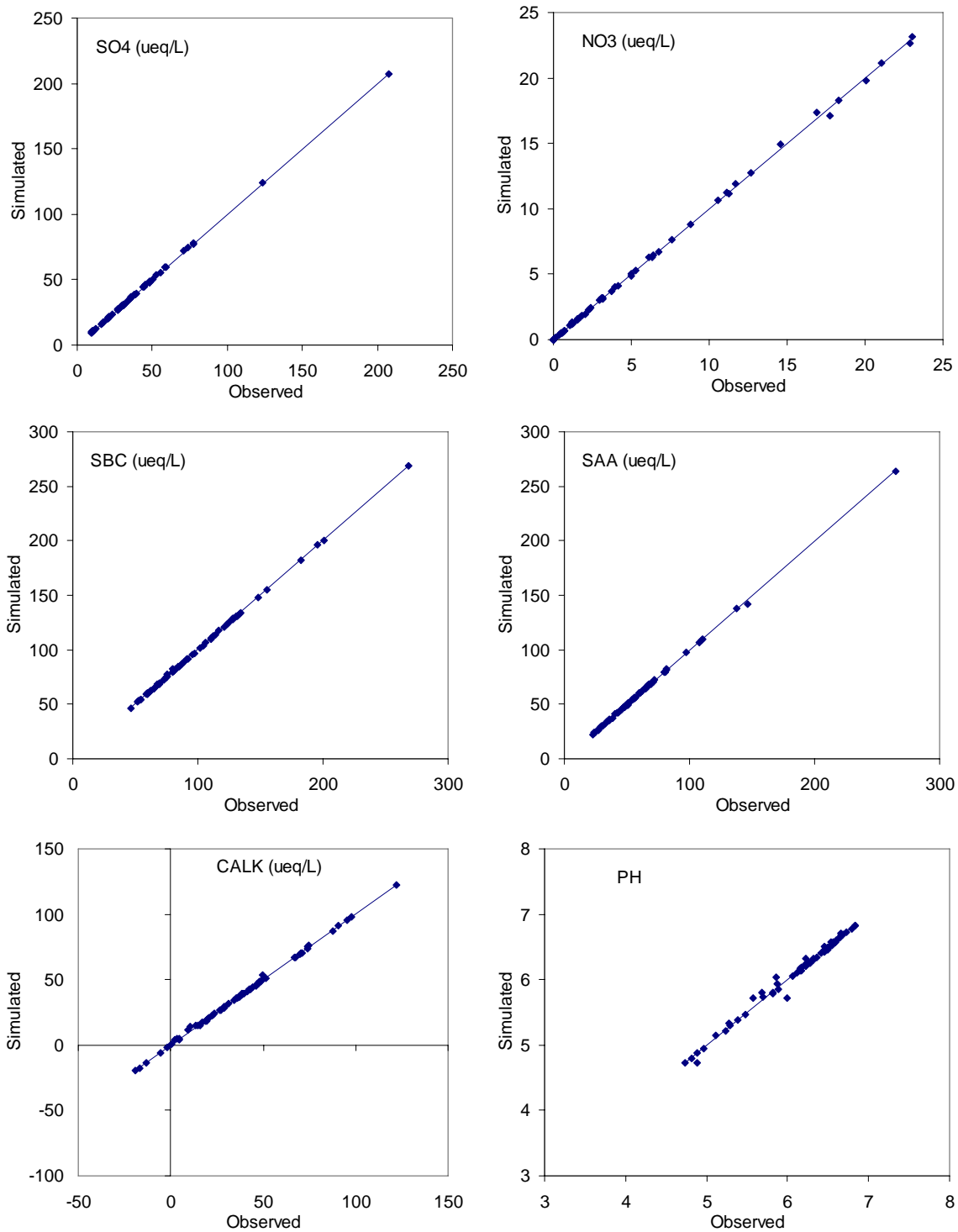


Figure 4. Continued.

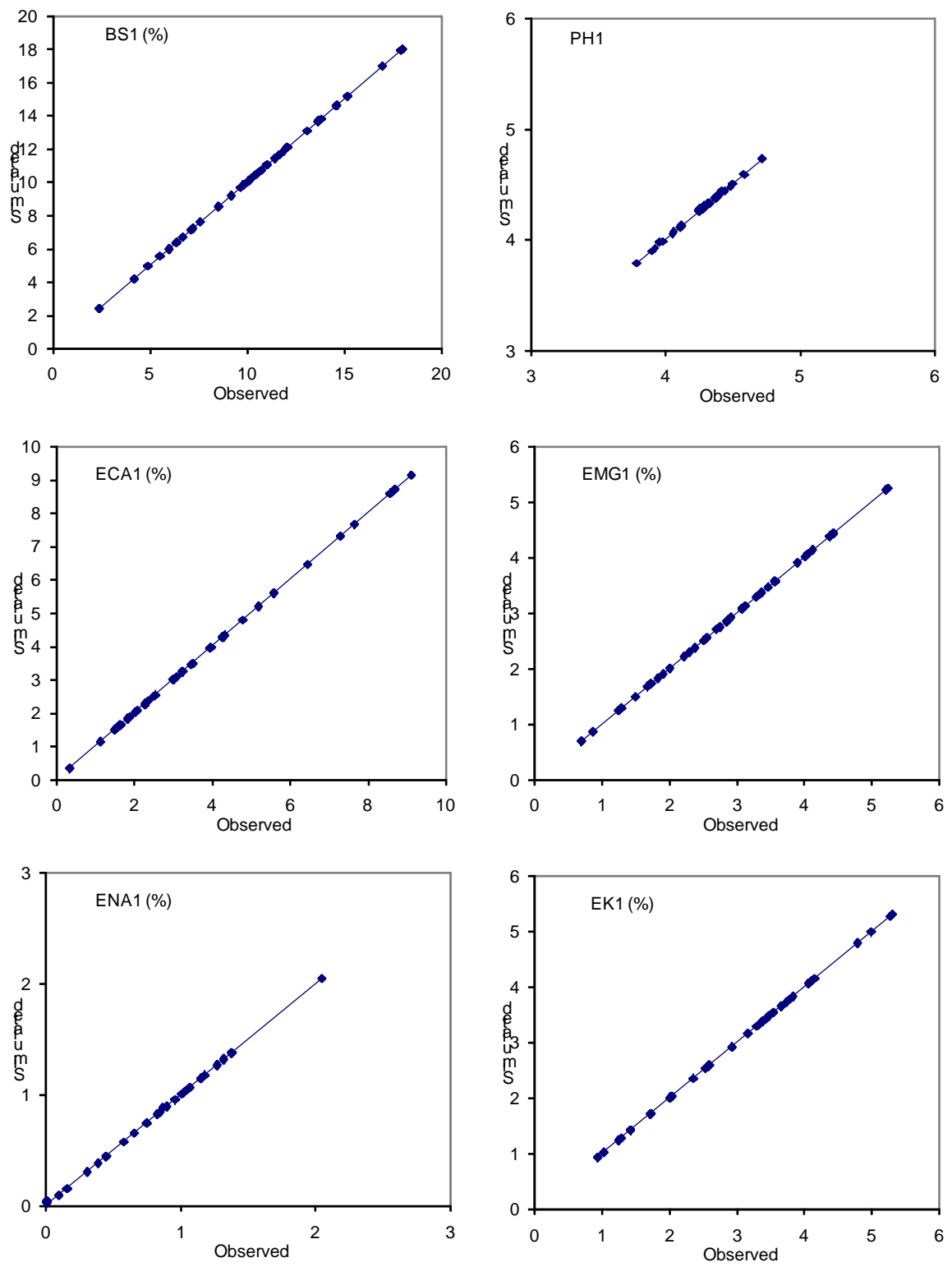


Figure 4. Continued.

Table 13. Simulated 1860 concentrations of various ions in streamwater for the modeled national forest streams.

Site Name	Site ID	1860 Simulated Stream Water Concentrations in µeq/L (except pH)											
		Ca	Mg	Na	K	NH ₄	SO ₄	CL	NO ₃	SBC	SAA	CALK	PH
Adam Camp Branch	237	13.5	11.7	27.9	8.8	0.0	0.0	11.5	0.0	61.8	11.5	50.3	6.7
Bear Branch	195	42.6	39.8	30.3	15.0	0.0	0.0	10.4	0.0	127.8	10.4	117.3	7.0
Bearpen Branch	253	14.4	11.1	28.4	6.8	0.0	0.0	13.6	0.0	60.6	13.6	47.1	6.6
Beetree Branch	190	45.9	53.7	31.9	45.0	0.0	45.9	14.1	0.0	176.4	59.5	117.7	6.9
Big Cove Branch	259	33.4	18.1	42.8	11.9	0.0	0.0	16.3	0.0	106.3	16.3	90.0	6.9
Big Laurel Brook	215	10.7	10.8	22.6	6.4	0.0	0.0	13.2	0.0	50.5	13.2	37.3	6.5
Big Oak Cove Creek	261	15.2	11.1	27.7	4.5	0.0	0.0	12.7	0.0	58.3	12.7	45.6	6.7
Briar Creek	72	29.7	38.4	21.1	12.0	0.0	0.0	10.8	0.0	101.2	10.8	90.6	6.9
Bubbling Spring Branch	129	17.2	9.3	19.9	5.5	0.0	0.0	9.7	0.0	51.8	9.7	41.8	6.4
Bubbling Spring West Tributary	131	10.6	6.6	18.0	3.8	0.0	0.0	8.2	0.0	39.0	8.2	30.6	6.0
Buckeye Cove Creek	133	19.6	9.1	21.2	5.5	0.0	0.0	9.4	0.0	55.3	9.4	45.8	6.5
Cane Creek Tributary	204	11.5	13.5	35.3	8.9	0.0	0.0	23.3	0.0	69.0	23.3	45.8	6.0
Cathey Creek	122	42.3	28.8	38.3	14.2	0.0	0.0	13.7	0.0	123.5	13.7	109.6	7.0
Colberts Creek	53	14.6	13.6	26.4	7.1	0.0	0.0	11.9	0.0	61.8	11.9	49.8	6.6
Courthouse Creek	121	14.3	12.5	34.4	9.0	0.0	0.0	17.1	0.0	70.1	17.1	52.9	6.7
Dark Prong	114	11.9	7.0	23.4	5.6	0.0	0.0	7.0	0.0	47.9	7.0	41.0	6.6
Davidson River	102	32.6	22.3	38.0	9.8	0.0	0.0	11.3	0.0	102.7	11.3	91.4	6.7
East Fork Pigeon	101	17.6	11.3	25.7	7.7	0.0	0.0	9.7	0.0	62.3	9.7	52.6	6.7
Flat Laurel Creek	127	15.9	8.5	22.8	5.8	0.0	0.0	8.4	0.0	53.0	8.4	44.6	6.4
Glade Creek	198	24.8	22.2	41.7	9.5	0.0	0.0	16.3	0.0	98.2	16.3	82.1	6.4
Greenland Creek	160	10.9	9.1	33.5	6.2	0.0	0.0	14.1	0.0	59.7	14.1	45.5	6.2
Indian Branch	255	36.0	15.6	36.8	10.4	0.0	0.0	22.7	0.0	99.0	22.7	75.4	6.9
Indian Camp	185	36.0	20.7	51.1	9.8	0.0	0.0	35.7	0.0	117.7	35.7	82.0	6.6
Indian Spring Branch	239	19.0	13.6	32.2	7.0	0.0	0.0	12.1	0.0	71.9	12.1	59.7	6.8
Kilby Creek	217	13.3	12.3	25.7	7.8	0.0	0.0	13.0	0.0	59.2	13.0	46.1	6.6
Kirkland Cove	260	21.7	10.1	32.0	7.8	0.0	0.0	13.6	0.0	71.5	13.6	58.0	6.8
Left Prong South Toe River	61	23.2	15.6	33.6	5.9	0.0	0.0	13.9	0.0	78.2	13.9	64.3	6.6
Lindy Camp Branch	35	20.4	19.2	19.6	13.4	0.0	0.0	11.2	0.0	72.4	11.2	61.5	6.4
Little Prong Hickey Fork	91	39.8	47.3	41.6	9.8	0.0	0.0	6.2	0.0	138.5	6.2	132.3	6.8
Little Santetlah Cr. (NuCM site)	281	20.0	11.3	25.1	7.0	0.0	0.0	12.3	0.0	63.3	12.3	50.9	6.6
Long Branch	103	29.3	17.6	57.1	8.1	0.0	0.0	10.8	0.0	112.0	10.8	101.1	6.6
Lost Cove	56	25.6	12.6	30.9	6.1	0.0	0.0	11.8	0.0	75.1	11.8	63.2	6.8
Lower Creek	59	14.3	11.4	31.8	14.8	0.0	0.0	26.1	0.0	72.3	26.1	46.7	6.6
McNabb Creek	270	122.0	59.7	69.7	14.0	0.0	77.1	55.6	0.0	265.3	131.8	133.5	7.1
Middle Creek	55	16.2	12.9	27.5	6.7	0.0	0.0	12.0	0.0	63.3	12.0	51.3	6.6
Mill Station Creek	132	20.8	9.1	46.7	7.4	0.0	0.0	42.5	0.0	84.0	42.5	41.7	6.6
Paddy Creek	26	6.4	13.1	19.4	17.1	0.0	0.0	18.8	0.0	56.1	18.8	37.2	6.5
Peach Orchard Creek	70	27.3	11.1	34.1	5.0	0.0	0.0	10.8	0.0	77.4	10.8	66.9	6.7
Pigpen Branch	191	28.6	28.4	52.9	13.3	0.0	0.0	20.4	0.0	123.3	20.4	103.0	6.9
Rattlesnake Branch	143	23.3	16.6	59.3	11.7	0.0	0.0	14.3	0.0	110.9	14.3	96.5	6.7
Right Hand Prong	147	20.3	15.6	31.8	7.5	0.0	0.0	12.0	0.0	75.0	12.0	62.8	6.8
Roaring Branch	251	43.1	12.5	31.9	6.6	0.0	0.0	12.9	0.0	94.1	12.9	81.1	6.7
Rough Ridge Creek	257	19.5	12.5	39.7	6.8	0.0	0.0	13.7	0.0	78.6	13.7	65.3	6.6
Russell Creek	15	17.4	20.1	20.5	23.2	0.0	0.0	19.4	0.0	81.2	19.4	61.8	6.8
Scotsman Creek	193	16.3	16.7	37.9	8.6	0.0	0.0	17.4	0.0	79.5	17.4	62.2	6.7
South Fork Fowler Creek	188	14.8	17.5	34.3	7.3	0.0	0.0	16.3	0.0	73.9	16.3	57.6	6.6
Spivey Creek	266	28.1	18.3	46.8	14.1	0.0	0.0	15.9	0.0	107.2	15.9	91.3	6.9

Table 13. Continued.

Site Name	Site ID	1860 Simulated Stream Water Concentrations in $\mu\text{eq/L}$ (except pH)											
		Ca	Mg	Na	K	NH ₄	SO ₄	CL	NO ₃	SBC	SAA	CALK	PH
Squibb Creek	88	29.7	47.2	51.3	17.9	0.0	0.0	13.9	0.0	146.1	13.9	132.3	6.9
Stillhouse Branch	28	5.3	12.6	15.6	11.5	0.0	0.0	15.5	0.0	44.9	15.5	29.6	6.3
Unnamed creek A	245	18.2	8.9	26.2	6.8	0.0	0.0	11.6	0.0	60.0	11.6	48.2	6.7
Unnamed creek B	246	52.8	17.6	44.7	6.1	0.0	0.0	46.6	0.0	121.2	46.6	74.5	6.9
Unnamed creek C	249	32.1	14.5	35.4	9.3	0.0	0.0	24.4	0.0	91.2	24.4	67.0	6.8
Upper Creek	60	13.3	11.1	26.0	14.1	0.0	0.0	24.8	0.0	64.5	24.8	39.8	6.5
UT Flat Laurel Creek	120	18.0	10.6	33.3	5.7	0.0	0.0	17.5	0.0	67.5	17.5	50.0	6.4
UT Laurel Branch	265	53.0	20.7	62.6	11.1	0.0	0.0	63.0	0.0	147.4	63.0	84.6	6.9
UT Linville River (NUCM site)	21	13.6	9.3	19.2	9.3	0.0	0.0	17.7	0.0	51.2	17.7	34.2	6.5
UT McNabb Creek	262	11.9	8.3	27.7	8.2	0.0	0.0	15.3	0.0	56.1	15.3	40.9	6.5
UT North Fork of Catawba	33	6.2	14.3	17.7	13.7	0.0	0.0	17.7	0.0	52.1	17.7	33.9	6.4
UT Paint Creek	94	49.9	43.5	32.2	11.9	0.0	0.0	13.3	0.0	137.5	13.3	124.5	6.8
UT Panthertown Creek (Boggy Cr)	163	21.0	8.3	40.5	5.1	0.0	0.0	12.8	0.0	74.7	12.8	61.7	6.3
UT Russell Creek	14	10.5	13.0	15.6	16.8	0.0	0.0	19.6	0.0	55.7	19.6	36.1	6.6
White Creek	16	10.9	10.1	24.6	15.0	0.0	0.0	19.7	0.0	60.5	19.7	41.1	6.4
Wildcat Branch	232	15.0	14.6	28.6	8.2	0.0	0.0	12.7	0.0	66.4	12.7	53.7	6.7
Wilson Creek	169	30.7	21.4	59.1	10.0	0.0	0.0	37.5	0.0	121.2	37.5	83.3	6.4
Wolf Creek	150	22.3	12.5	33.5	6.9	0.0	0.0	13.8	0.0	75.1	13.8	61.3	6.8
Yellow Fork	27	6.4	14.3	14.2	12.8	0.0	0.0	16.4	0.0	47.7	16.4	31.1	6.3

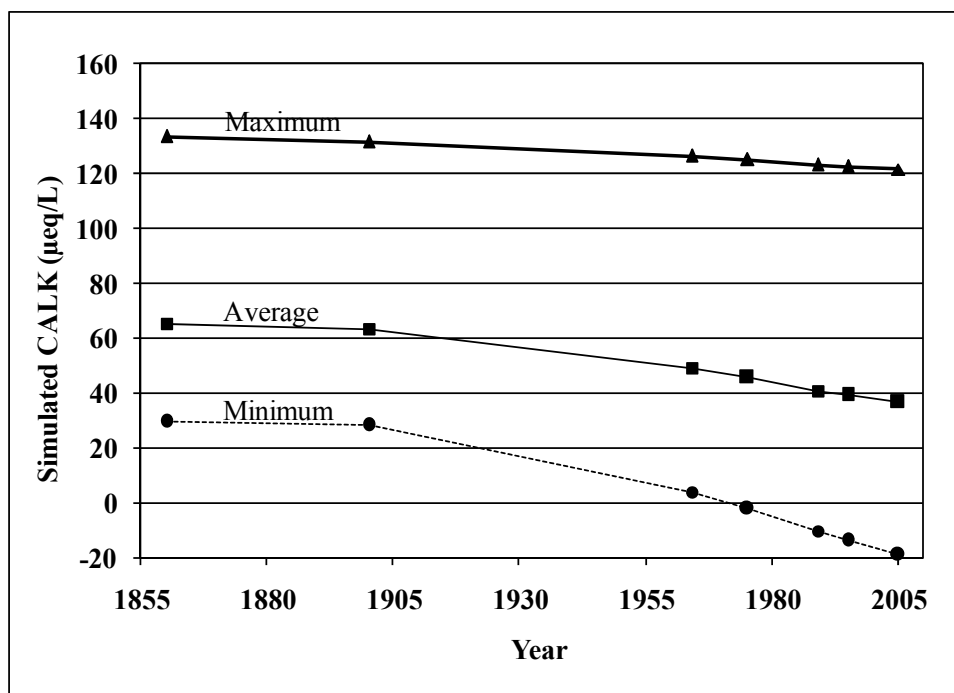


Figure 5. MAGIC estimates of historical change in CALK for the 66 modeled sites for multiple points in time. The average simulated value is represented by the solid line, and the maximum and minimum simulated values by the dashed lines.

Table 14. Modeled past stream ANC ($\mu\text{eq/L}$) at three different points in time, in relation to four commonly used target ANC levels.

	ANC \leq 0		ANC \leq 20		ANC \leq 50		ANC \leq 100	
	#	% ¹	#	%	#	%	#	%
1860	0	0	0	0	25	38	57	86
1975	2	3	9	14	43	65	64	97
2005	5	8	20	30	51	77	66	100

¹ percentages are expressed as the percent of the modeled streams (n=66) that were simulated to have ANC below the target level in the selected year.

future emissions and deposition. In Figure 6, we compare CMAQ simulations of wet deposition of $\text{NO}_3\text{-N}$, $\text{NH}_4\text{-N}$, and S with Grimm's interpolated values from the NADP/NTN monitoring network, averaged over a three-year period centered on 1999. CMAQ S results for 2002 (expressed as an average of emissions inventory data for the period 2000 to 2004) were generally higher than interpolated NADP/NTN values for 1998 to 2000. In addition, there was considerable scatter observed between modeled and interpolated measured values. Nevertheless, the CMAQ simulations represent the best available data with which to estimate future deposition to serve as the basis for MAGIC model projections of future stream chemistry.

Model projections of current and future deposition in the years 2002, 2009, 2018, and 2064 are shown for wet deposition (Figure 7), dry deposition (Figure 8), and total wet plus dry deposition (Figure 9) under the three emissions controls scenarios: Base Case, Moderate Additional Emissions Controls, and Aggressive Additional Emissions Controls. Stream SO_4^{2-} concentration projections for the future under the emissions controls scenarios are summarized in Table 15. Future changes in stream chemistry in response to changes in acidic deposition are driven mainly by changes in streamwater SO_4^{2-} concentration. For the reference year (2005), SO_4^{2-} concentrations varied, with half of the modeled streams having SO_4^{2-} concentration between 24 and 45 $\mu\text{eq/L}$ (Table 8). Two streams had SO_4^{2-} concentration higher than 80 $\mu\text{eq/L}$ (Table 15), presumably due in part to geological sources of S within the watershed. In response to the emissions controls scenarios, some of the modeled streams were projected to exhibit future decreases in SO_4^{2-} concentration, which were largest in more distant future years and under greater emissions reductions (Table 16).

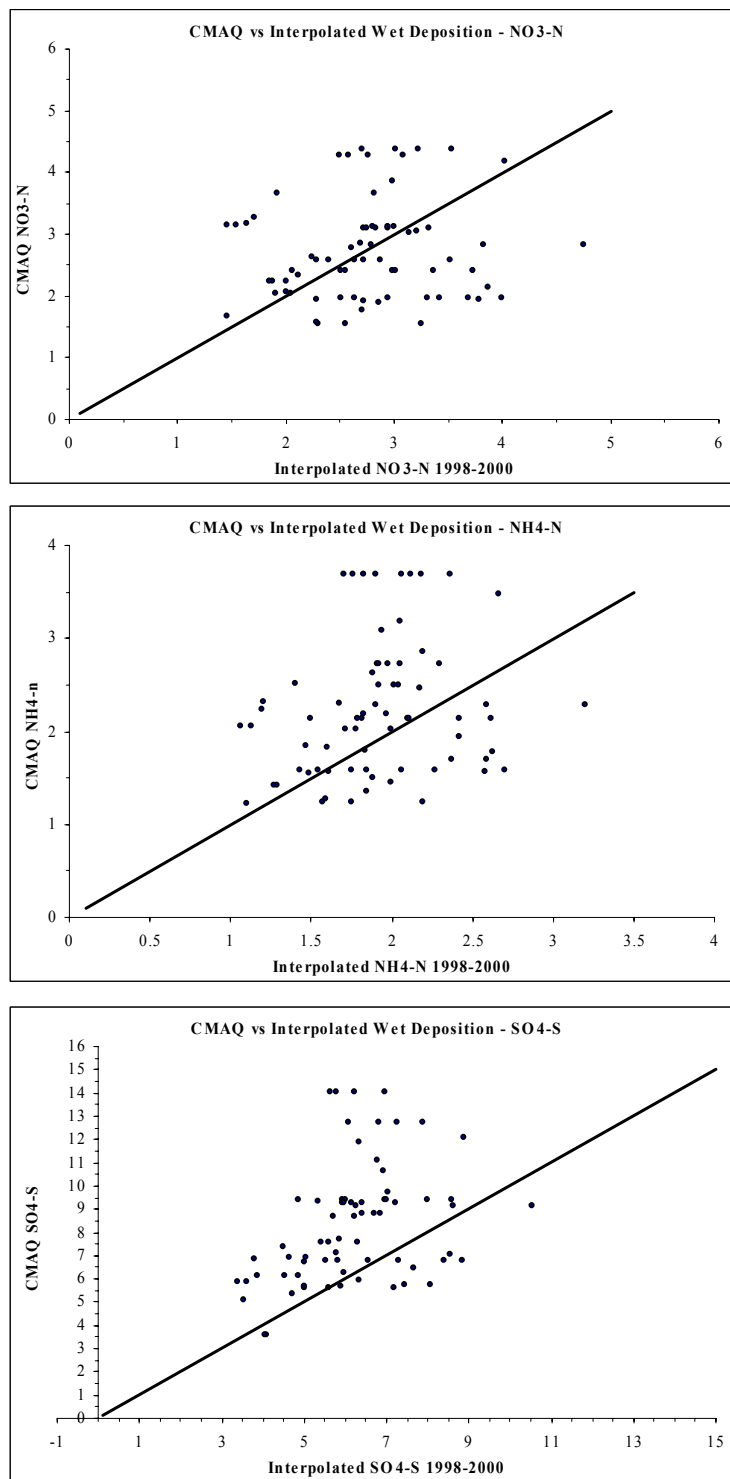


Figure 6. CMAQ model wet deposition for the year 2002 at each modeling site location compared with three-year average (1998-2000) interpolated measurements from NADP/NTN. Estimates are provided for S and for both oxidized and reduced N. Reference lines (1:1) are added.

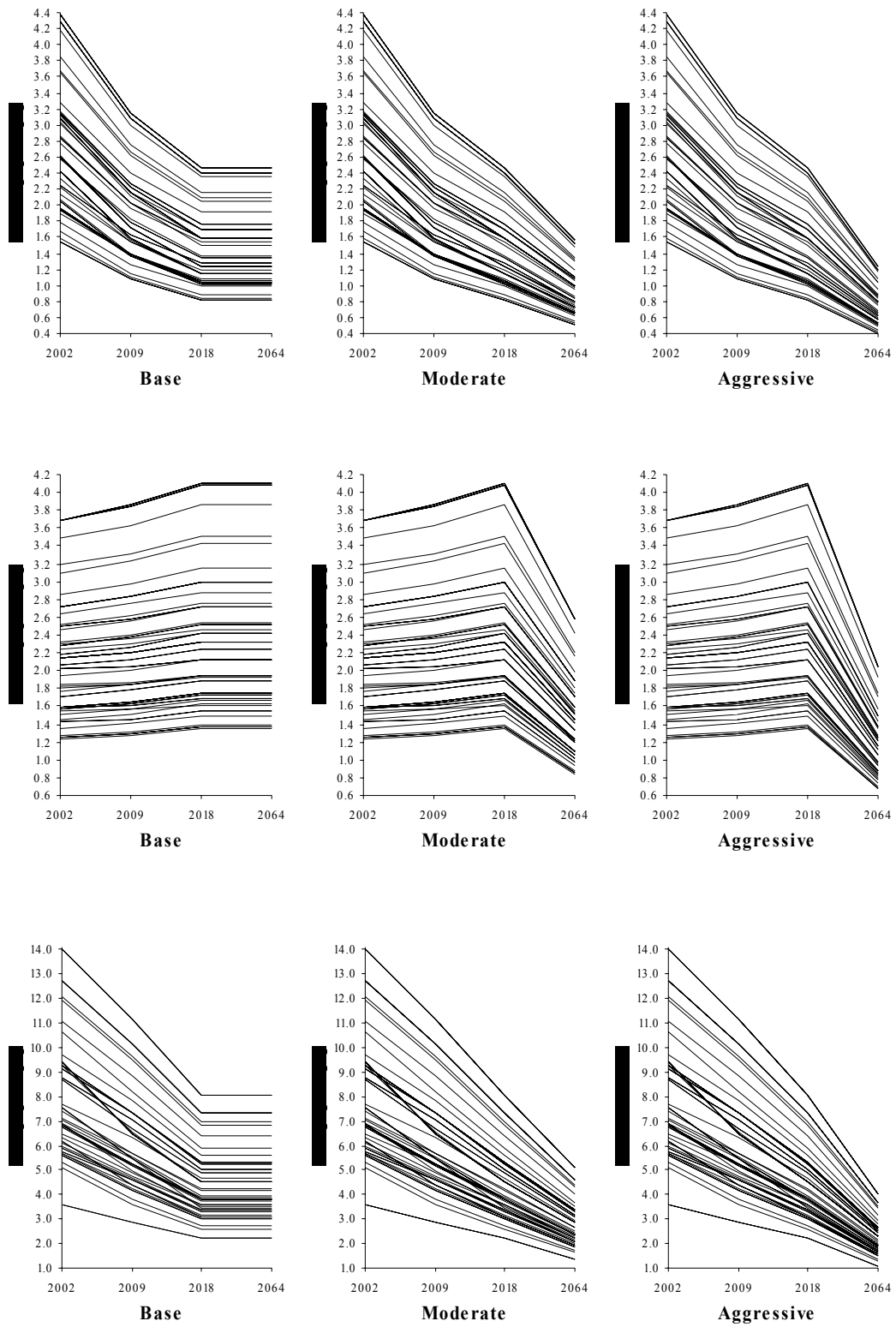


Figure 7. Estimates of wet deposition under the three scenarios of emissions control (n=66).

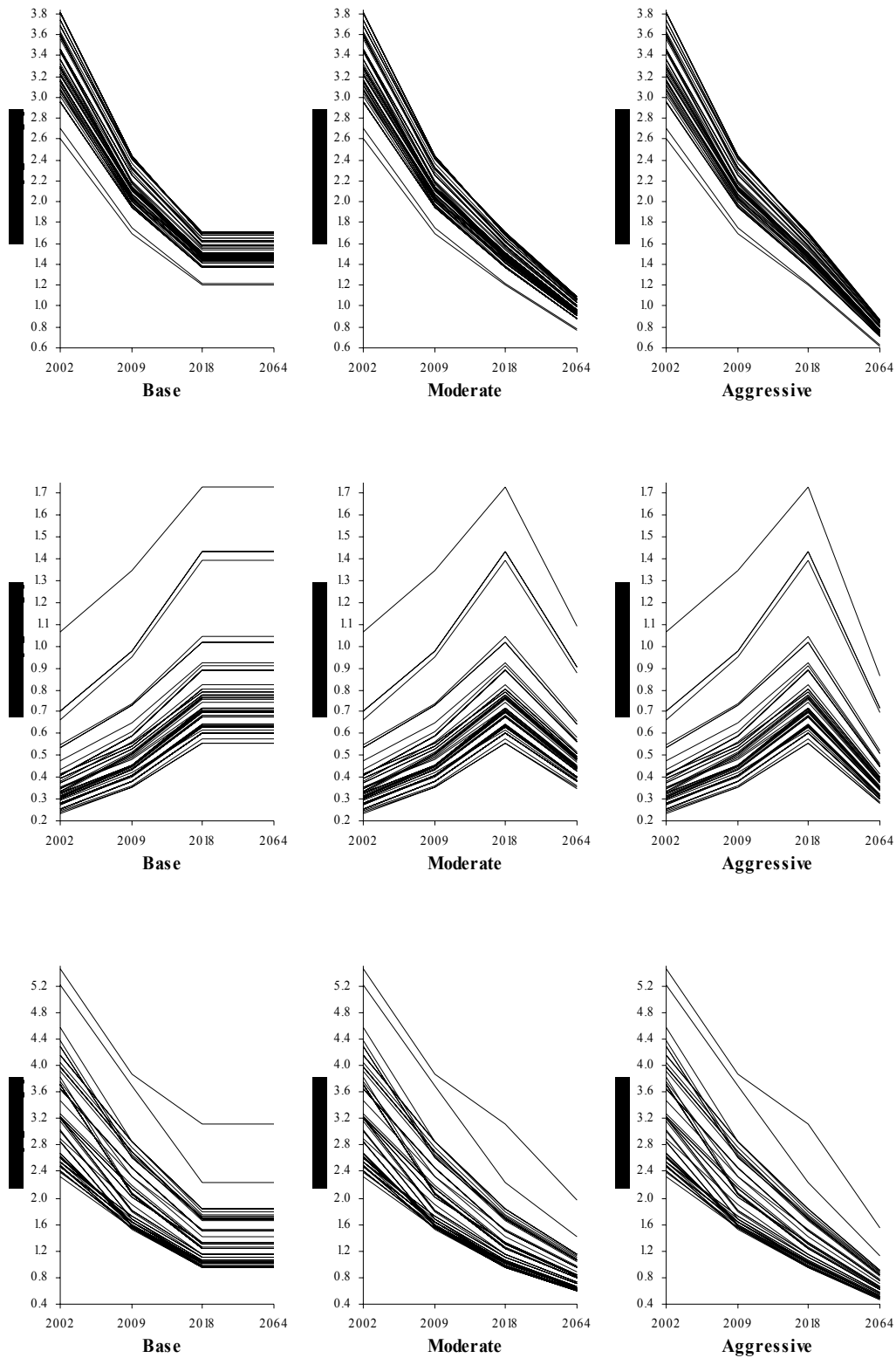


Figure 8. Estimates of dry deposition under the three scenarios of emissions control (n=66).

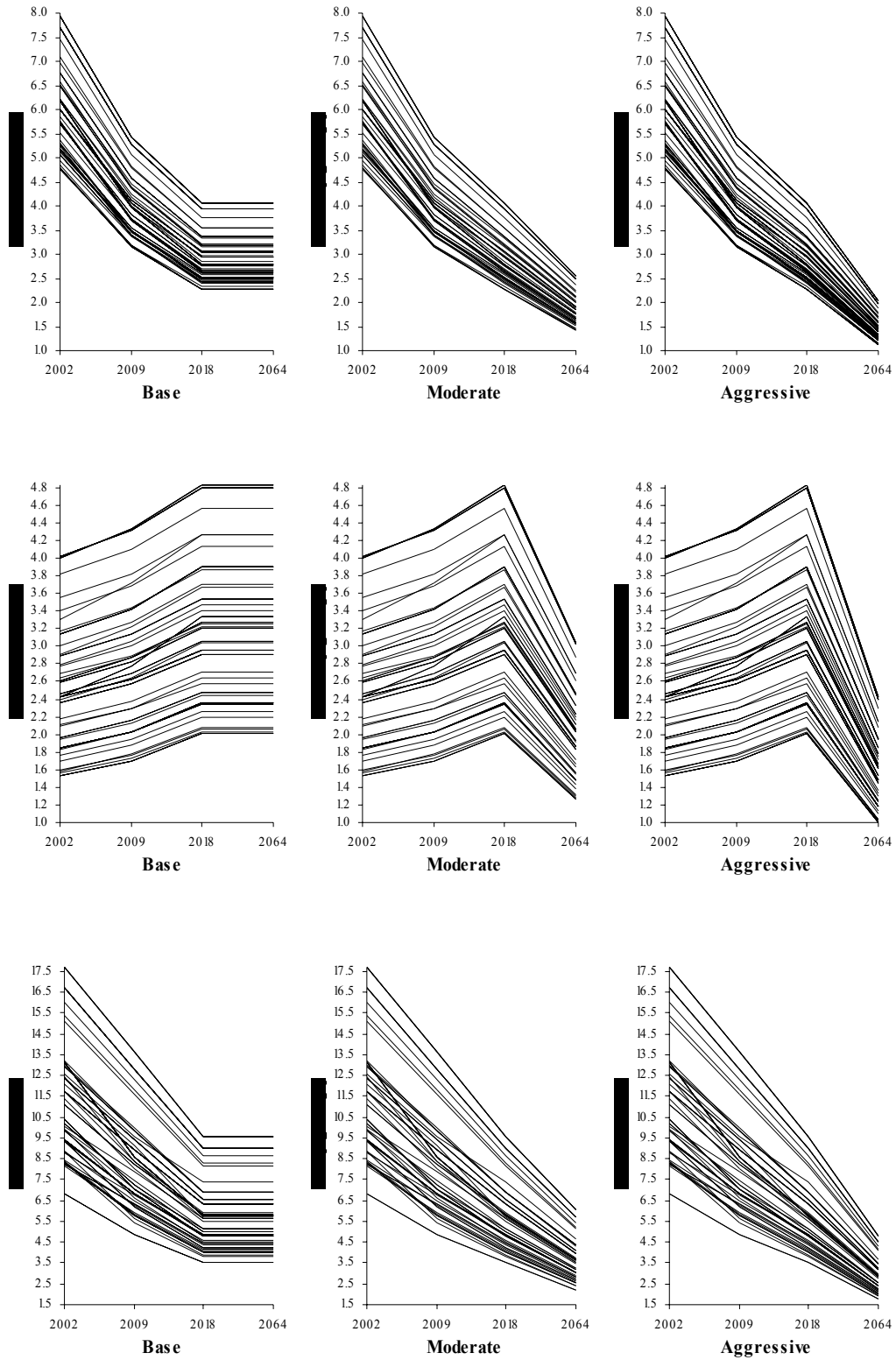


Figure 9. Estimates of total (wet + dry) deposition under the three scenarios of emissions control (n=66).

Table 15. Simulated stream sulfate concentration ($\mu\text{eq/L}$) in 66 modeled streams in the past, present, and future under three scenarios of future emissions controls.

Site Name	Site ID	1860	2005	Base Scenario			Moderate Scenario			Aggressive Scenario		
				2020	2040	2100	2020	2040	2100	2020	2040	2100
Adam Camp Branch	237	0.0	17.7	18.7	19.6	22.3	18.7	19.5	20.4	18.7	19.4	19.7
Bear Branch	195	0.0	71.9	66.1	57.9	45.7	66.1	56.8	37.9	66.1	56.4	35.4
Bearpen Branch	253	0.0	30.7	32.7	33.9	36.8	32.7	33.4	32.1	32.7	33.2	30.6
Beetree Branch	190	45.9	124.2	119.7	111.9	99.3	119.7	110.7	90.6	119.7	110.3	87.8
Big Cove Branch	259	0.0	20.8	22.0	23.1	26.0	22.0	22.8	23.7	22.0	22.8	23.0
Big Laurel Brook	215	0.0	10.8	11.7	12.5	15.0	11.7	12.4	13.6	11.7	12.4	13.1
Big Oak Cove Creek	261	0.0	38.4	39.6	39.9	40.4	39.6	39.4	35.7	39.6	39.2	34.3
Briar Creek	72	0.0	55.8	57.2	55.5	52.5	57.2	54.3	43.0	57.2	53.9	40.0
Bubbling Spring Branch	129	0.0	45.3	48.4	50.6	56.1	48.4	49.9	48.5	48.4	49.7	45.9
Bubbling Spring West Tributary	131	0.0	39.5	42.4	44.5	49.8	42.4	43.9	42.9	42.4	43.7	40.7
Buckeye Cove Creek	133	0.0	31.1	33.7	35.8	41.5	33.7	35.3	36.7	33.7	35.2	35.1
Cane Creek Tributary	204	0.0	37.4	39.4	40.1	41.9	39.3	39.5	36.2	39.3	39.3	34.3
Cathey Creek	122	0.0	37.1	38.5	38.6	39.0	38.5	37.9	33.4	38.5	37.7	31.7
Colberts Creek	53	0.0	30.6	33.0	34.4	37.8	33.0	33.8	32.2	33.0	33.6	30.6
Courthouse Creek	121	0.0	27.4	29.3	30.6	34.0	29.3	30.2	29.9	29.3	30.1	28.6
Dark Prong	114	0.0	28.4	30.7	32.5	37.4	30.7	32.1	33.2	30.7	32.0	31.8
Davidson River	102	0.0	28.8	30.4	31.1	32.8	30.4	30.6	28.7	30.4	30.4	27.4
East Fork Pigeon	101	0.0	21.4	22.7	23.7	26.0	22.7	23.3	23.0	22.7	23.2	22.0
Flat Laurel Creek	127	0.0	30.5	31.6	31.6	31.5	31.6	31.1	27.1	31.6	31.0	25.6
Glade Creek	198	0.0	10.9	11.9	12.7	15.3	11.9	12.6	13.9	11.9	12.6	13.5
Greenland Creek	160	0.0	29.1	30.6	31.2	32.7	30.6	30.8	28.6	30.6	30.6	27.3
Indian Branch	255	0.0	34.3	36.2	37.5	41.1	36.2	37.1	36.7	36.2	37.0	35.3
Indian Camp	185	0.0	12.9	14.0	15.2	18.4	14.0	15.0	16.7	14.0	14.9	16.2
Indian Spring Branch	239	0.0	23.9	25.7	27.1	30.3	25.7	26.7	26.4	25.7	26.6	25.3
Kilby Creek	217	0.0	10.6	11.5	12.4	15.1	11.5	12.3	13.7	11.5	12.2	13.2
Kirkland Cove	260	0.0	31.2	32.6	33.4	35.3	32.6	33.0	31.8	32.6	32.9	30.7
Left Prong South Toe River	61	0.0	18.7	20.5	22.1	26.6	20.5	21.8	23.6	20.5	21.7	22.6
Lindy Camp Branch	35	0.0	50.9	53.1	52.9	52.5	53.1	51.9	43.6	53.1	51.5	40.8
Little Prong Hickey Fork	91	0.0	22.5	24.3	25.5	28.6	24.3	25.1	24.9	24.3	25.0	23.6
Little Santetlah Cr. (NuCM site)	281	0.0	33.1	34.4	34.9	36.2	34.4	34.5	32.1	34.4	34.4	30.8
Long Branch	103	0.0	12.4	13.4	14.3	17.0	13.4	14.2	15.3	13.4	14.1	14.8
Lost Cove	56	0.0	36.0	38.3	39.2	41.1	38.3	38.5	34.9	38.3	38.3	32.9
Lower Creek	59	0.0	32.7	35.7	38.1	44.3	35.7	37.5	38.4	35.7	37.3	36.6
McNabb Creek	270	77.1	207.2	195.1	179.1	157.6	195.1	176.9	143.2	195.1	176.2	138.6
Middle Creek	55	0.0	30.1	32.3	33.7	36.9	32.3	33.1	32.0	32.3	33.0	30.4
Mill Station Creek	132	0.0	60.1	62.4	62.3	61.9	62.4	61.2	52.2	62.4	60.8	48.9

Table 15. Continued.

Site Name	Site ID	1860	2005	Base Scenario			Moderate Scenario			Aggressive Scenario		
				2020	2040	2100	2020	2040	2100	2020	2040	2100
Paddy Creek	26	0.0	48.2	51.2	52.2	54.0	51.1	51.1	44.7	51.1	50.8	41.9
Peach Orchard Creek	70	0.0	52.5	53.4	51.5	48.1	53.4	50.3	39.4	53.4	50.0	36.7
Pigpen Branch	191	0.0	10.1	10.9	11.8	14.2	10.9	11.6	12.9	10.9	11.6	12.5
Rattlesnake Branch	143	0.0	42.1	44.9	46.3	49.2	44.9	45.5	41.8	44.9	45.2	39.5
Right Hand Prong	147	0.0	29.9	31.5	32.3	34.0	31.5	31.8	29.8	31.5	31.7	28.4
Roaring Branch	251	0.0	36.3	37.5	38.0	39.1	37.5	37.6	34.6	37.5	37.4	33.3
Rough Ridge Creek	257	0.0	27.9	29.2	30.1	32.5	29.2	29.8	29.4	29.2	29.7	28.4
Russell Creek	15	0.0	78.3	76.8	70.4	61.6	76.8	68.4	48.5	76.8	67.7	44.4
Scotsman Creek	193	0.0	16.9	18.2	19.4	22.5	18.2	19.1	20.2	18.2	19.1	19.4
South Fork Fowler Creek	188	0.0	10.0	10.9	11.7	14.2	10.9	11.6	12.9	10.9	11.6	12.5
Spivey Creek	266	0.0	44.4	45.6	45.4	45.1	45.6	44.9	40.0	45.6	44.7	38.3
Squibb Creek	88	0.0	79.4	73.1	63.1	53.0	73.1	60.9	39.9	73.1	60.2	35.9
Stillhouse Branch	28	0.0	33.6	36.4	38.2	42.7	36.4	37.6	36.7	36.4	37.4	34.8
Unnamed creek A	245	0.0	30.4	31.7	32.4	34.3	31.7	32.0	30.8	31.7	31.9	29.6
Unnamed creek B	246	0.0	46.4	48.9	50.4	54.0	48.8	49.8	47.9	48.8	49.5	46.0
Unnamed creek C	249	0.0	28.2	29.8	31.1	34.6	29.8	30.8	31.6	29.8	30.7	30.6
Upper Creek	60	0.0	34.1	37.2	39.6	45.6	37.2	39.0	39.4	37.2	38.8	37.6
UT Flat Laurel Creek	120	0.0	38.0	41.4	44.3	52.7	41.3	43.7	46.8	41.3	43.5	44.9
UT Laurel Branch	265	0.0	74.7	76.1	74.8	71.9	76.1	73.7	62.3	76.1	73.3	59.3
UT Linville River (NUCM site)	21	0.0	53.9	56.7	56.8	57.2	56.7	55.7	48.2	56.7	55.4	45.3
UT McNabb Creek	262	0.0	20.5	21.8	22.9	26.2	21.8	22.7	23.9	21.8	22.6	23.2
UT North Fork of Catawba	33	0.0	38.5	41.7	43.8	48.3	41.7	43.0	40.8	41.7	42.7	38.5
UT Paint Creek	94	0.0	48.5	50.8	51.0	51.2	50.8	50.0	42.5	50.8	49.6	39.6
UT Panthertown Creek (Boggy Creek)	163	0.0	32.6	33.9	34.1	34.4	33.9	33.6	29.8	33.9	33.4	28.2
UT Russell Creek	14	0.0	59.6	61.6	60.1	57.7	61.5	58.8	47.5	61.5	58.4	44.3
White Creek	16	0.0	48.5	51.3	52.1	53.4	51.3	51.0	45.1	51.3	50.7	42.4
Wildcat Branch	232	0.0	16.4	17.8	19.0	22.4	17.8	18.8	20.0	17.8	18.7	19.3
Wilson Creek	169	0.0	11.0	12.0	13.0	15.9	12.0	12.8	14.4	12.0	12.8	14.0
Wolf Creek	150	0.0	17.5	18.9	20.0	23.3	18.9	19.8	21.0	18.9	19.7	20.2
Yellow Fork	27	0.0	48.6	51.5	52.3	53.3	51.5	51.2	44.1	51.5	50.8	41.2

Table 16. Simulated change in stream sulfate concentration ($\mu\text{eq/L}$) in the future (compared with values in 2005) under three scenarios of future emissions controls. Negative values imply that SO_4^{2-} concentration will decrease from 2005 values.

Site Name	Site ID	Base Scenario			Moderate Scenario			Aggressive Scenario		
		2020	2040	2100	2020	2040	2100	2020	2040	2100
Adam Camp Branch	237	1.1	2.0	4.6	1.1	1.8	2.7	1.1	1.7	2.1
Bear Branch	195	-5.8	-14.0	-26.2	-5.8	-15.1	-34.0	-5.9	-15.5	-36.6
Bearpen Branch	253	2.0	3.2	6.0	2.0	2.7	1.3	2.0	2.5	-0.1
Beetree Branch	190	-4.5	-12.3	-24.9	-4.5	-13.5	-33.6	-4.5	-13.8	-36.4
Big Cove Branch	259	1.2	2.2	5.1	1.2	2.0	2.9	1.2	1.9	2.1
Big Laurel Brook	215	0.9	1.8	4.3	0.9	1.6	2.8	0.9	1.6	2.4
Big Oak Cove Creek	261	1.2	1.5	2.0	1.2	1.0	-2.7	1.2	0.8	-4.1
Briar Creek	72	1.4	-0.3	-3.3	1.4	-1.5	-12.7	1.4	-1.9	-15.8
Bubbling Spring Branch	129	3.2	5.3	10.8	3.2	4.6	3.2	3.2	4.4	0.6
Bubbling Spring West Tributary	131	2.9	5.0	10.3	2.9	4.4	3.4	2.9	4.2	1.1
Buckeye Cove Creek	133	2.6	4.7	10.4	2.6	4.2	5.6	2.6	4.1	4.0
Cane Creek Tributary	204	2.0	2.8	4.6	2.0	2.2	-1.2	2.0	2.0	-3.1
Cathey Creek	122	1.4	1.4	1.9	1.4	0.8	-3.7	1.4	0.6	-5.4
Colberts Creek	53	2.4	3.8	7.2	2.4	3.2	1.6	2.4	3.0	0.0
Courthouse Creek	121	1.9	3.2	6.6	1.9	2.8	2.5	1.9	2.7	1.2
Dark Prong	114	2.3	4.1	9.0	2.3	3.7	4.7	2.3	3.5	3.4
Davidson River	102	1.7	2.3	4.0	1.7	1.8	-0.1	1.7	1.7	-1.4
East Fork Pigeon	101	1.3	2.2	4.5	1.3	1.9	1.5	1.3	1.8	0.6
Flat Laurel Creek	127	1.1	1.1	1.0	1.1	0.6	-3.5	1.1	0.4	-4.9
Glade Creek	198	0.9	1.8	4.4	0.9	1.7	3.0	0.9	1.6	2.5
Greenland Creek	160	1.5	2.1	3.6	1.5	1.7	-0.5	1.5	1.5	-1.9
Indian Branch	255	1.8	3.2	6.8	1.8	2.8	2.3	1.8	2.7	1.0
Indian Camp	185	1.1	2.2	5.4	1.1	2.1	3.8	1.1	2.0	3.2
Indian Spring Branch	239	1.8	3.2	6.4	1.8	2.8	2.5	1.8	2.7	1.4
Kilby Creek	217	0.9	1.8	4.5	0.9	1.7	3.1	0.9	1.7	2.6
Kirkland Cove	260	1.4	2.2	4.1	1.4	1.8	0.6	1.4	1.7	-0.5
Left Prong South Toe River	61	1.8	3.4	8.0	1.8	3.1	4.9	1.8	3.0	4.0
Lindy Camp Branch	35	2.2	2.0	1.6	2.2	1.0	-7.3	2.2	0.6	-10.1
Little Prong Hickey Fork	91	1.8	3.0	6.1	1.8	2.6	2.4	1.8	2.5	1.1
Little Santetlah Cr. (NuCM site)	281	1.3	1.9	3.2	1.3	1.5	-1.0	1.3	1.3	-2.2
Long Branch	103	1.0	1.9	4.6	1.0	1.8	2.9	1.0	1.7	2.4
Lost Cove	56	2.3	3.2	5.1	2.3	2.5	-1.1	2.3	2.3	-3.1
Lower Creek	59	3.0	5.4	11.6	3.0	4.8	5.7	3.0	4.6	3.9
McNabb Creek	270	-12.1	-28.1	-49.6	-12.1	-30.3	-64.0	-12.1	-31.0	-68.6

Table 16. Continued.

Site Name	Site ID	Base Scenario			Moderate Scenario			Aggressive Scenario		
		2020	2040	2100	2020	2040	2100	2020	2040	2100
Middle Creek	55	2.3	3.6	6.8	2.2	3.1	1.9	2.2	2.9	0.3
Mill Station Creek	132	2.3	2.1	1.8	2.3	1.1	-7.9	2.3	0.7	-11.2
Paddy Creek	26	3.0	4.0	5.9	3.0	3.0	-3.4	3.0	2.6	-6.2
Peach Orchard Creek	70	0.8	-1.1	-4.5	0.8	-2.2	-13.1	0.8	-2.6	-15.9
Pigpen Branch	191	0.9	1.7	4.2	0.9	1.6	2.9	0.9	1.5	2.4
Rattlesnake Branch	143	2.8	4.2	7.1	2.8	3.4	-0.3	2.8	3.2	-2.6
Right Hand Prong	147	1.6	2.4	4.1	1.6	1.9	-0.1	1.6	1.8	-1.5
Roaring Branch	251	1.3	1.8	2.8	1.3	1.3	-1.6	1.3	1.2	-3.0
Rough Ridge Creek	257	1.3	2.2	4.6	1.3	1.9	1.6	1.3	1.8	0.5
Russell Creek	15	-1.5	-7.9	-16.8	-1.6	-9.9	-29.8	-1.6	-10.6	-34.0
Scotsman Creek	193	1.3	2.5	5.6	1.3	2.3	3.3	1.3	2.2	2.6
South Fork Fowler Creek	188	0.9	1.7	4.2	0.9	1.6	2.9	0.9	1.6	2.5
Spivey Creek	266	1.2	1.0	0.7	1.2	0.4	-4.5	1.2	0.2	-6.1
Squibb Creek	88	-6.2	-16.2	-26.4	-6.3	-18.4	-39.4	-6.3	-19.2	-43.5
Stillhouse Branch	28	2.8	4.6	9.1	2.8	4.0	3.1	2.8	3.8	1.2
Unnamed creek A	245	1.3	2.0	3.9	1.3	1.6	0.4	1.3	1.5	-0.8
Unnamed creek B	246	2.5	4.1	7.6	2.5	3.4	1.5	2.5	3.2	-0.3
Unnamed creek C	249	1.6	2.9	6.4	1.6	2.6	3.3	1.6	2.5	2.4
Upper Creek	60	3.1	5.5	11.5	3.1	4.9	5.3	3.1	4.7	3.4
UT Flat Laurel Creek	120	3.4	6.3	14.7	3.3	5.7	8.8	3.3	5.5	6.9
UT Laurel Branch	265	1.4	0.0	-2.8	1.3	-1.0	-12.4	1.3	-1.4	-15.4
UT Linville River (NUCM site)	21	2.8	2.9	3.3	2.8	1.8	-5.8	2.8	1.5	-8.7
UT McNabb Creek	262	1.3	2.5	5.7	1.3	2.2	3.5	1.3	2.2	2.7
UT North Fork of Catawba	33	3.2	5.3	9.8	3.2	4.5	2.3	3.2	4.2	-0.1
UT Paint Creek	94	2.3	2.4	2.7	2.3	1.4	-6.1	2.3	1.1	-8.9
UT Panthertown Creek (Boggy C	163	1.3	1.5	1.8	1.3	1.0	-2.8	1.3	0.8	-4.4
UT Russell Creek	14	1.9	0.5	-2.0	1.9	-0.8	-12.1	1.9	-1.2	-15.4
White Creek	16	2.8	3.6	4.9	2.8	2.5	-3.4	2.8	2.2	-6.1
Wildcat Branch	232	1.4	2.6	6.0	1.4	2.4	3.6	1.4	2.3	2.9
Wilson Creek	169	1.0	2.0	4.8	1.0	1.8	3.4	1.0	1.8	2.9
Wolf Creek	150	1.4	2.6	5.8	1.4	2.3	3.5	1.4	2.2	2.7
Yellow Fork	27	2.9	3.7	4.7	2.9	2.5	-4.6	2.9	2.2	-7.5

In response to these simulated changes in stream SO_4^{2-} concentrations, stream base cation concentrations and ANC, and also soil percent base saturation were projected to change in the future (Figure 10). Simulated stream ANC values at each modeled site under each emissions scenario are summarized in Table 17 for the years 2020, 2040, and 2100. Simulated changes in ANC are presented in Table 18. In general, the model projected that soil and stream chemistry have changed substantially since pre-industrial times, but that future changes in response to emissions controls will be small. Model projections for individual sites are given in Appendix C.

D. Model Estimates of Critical Loads

The levels of S deposition that were simulated to cause streamwater ANC to increase or decrease to four specified critical levels or ANC endpoints (0, 20, 50, and 100 $\mu\text{eq/L}$) are shown in Table 19 for each of the modeled streams. The first three of these critical levels have been utilized in critical loads studies elsewhere (c.f., Kämäri et al. 1992; Sullivan and Cosby 2002; Sullivan et al. 2005). Estimated critical loads for S deposition ranged from less than zero (ecological objective not attainable) to more than 1,000 S kg/ha/yr, depending on the selected site, ANC endpoint, and evaluation year. For the results shown in Table 19, critical load estimates that were less than zero (ANC criterion not attainable in the endpoint year regardless of deposition level) were set to zero. For most streams, ANC=0 was attainable in any year (2020, 2040, or 2100), and quite high levels of deposition could occur and the majority of streams would still have ANC above zero. In marked contrast, most of the modeled streams could not achieve ANC=100 $\mu\text{eq/L}$ in any of the future years, even if S deposition was reduced to zero and maintained at that level (Table 19). This feature of the model output is illustrated in Figure 11 using the years 2040 and 2100 as examples. Stream ANC above 0 was achievable in 2040 at 94% of the modeling sites, and for most of those sites (85% of total) the ANC criterion could be achieved with no reduction in S deposition from current levels. However, only 1.5% of the modeling sites could achieve ANC=100 $\mu\text{eq/L}$ in the year 2040. In order to achieve these ANC values in 2100, more of the sites would require reductions in deposition from current levels. With respect to selection of targets for land management, it appears that neither ANC=0 nor ANC=100 $\mu\text{eq/L}$ would be particularly useful. Almost all sites can maintain ANC above 0 $\mu\text{eq/L}$ without reducing S deposition, and it has been well demonstrated that a variety of adverse ecological effects occur at such low ANC. ANC = 100 $\mu\text{eq/L}$ is not attainable and most of the study streams had ANC below 100 $\mu\text{eq/L}$ prior to the onset of acidic deposition. Therefore, ANC

a)

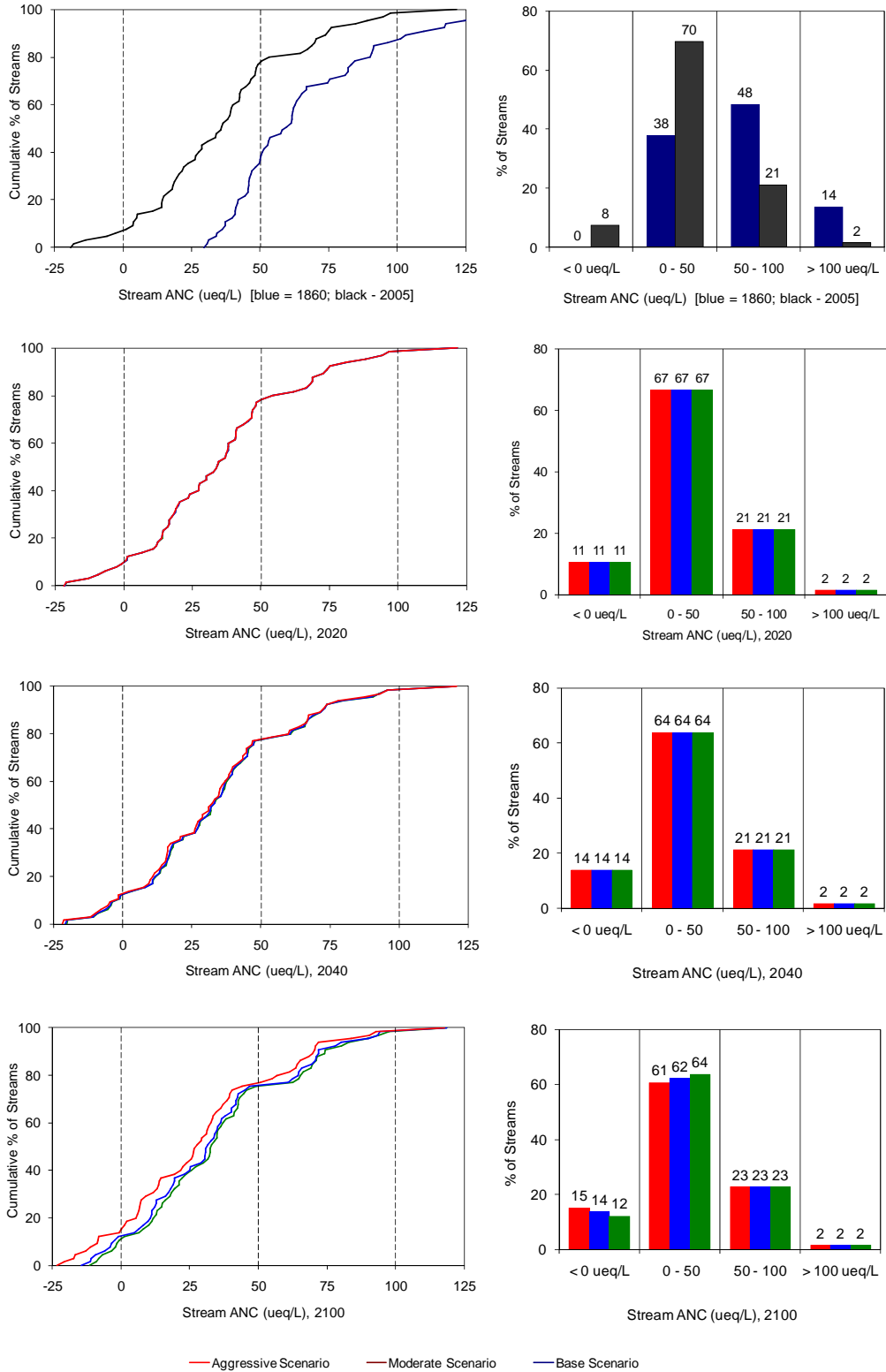


Figure 10. Cumulative frequency distributions and histograms of simulated changes in a) stream ANC, b) soil percent base saturation, c) stream sum of base cations, and d) stream sulfate concentration.

b)

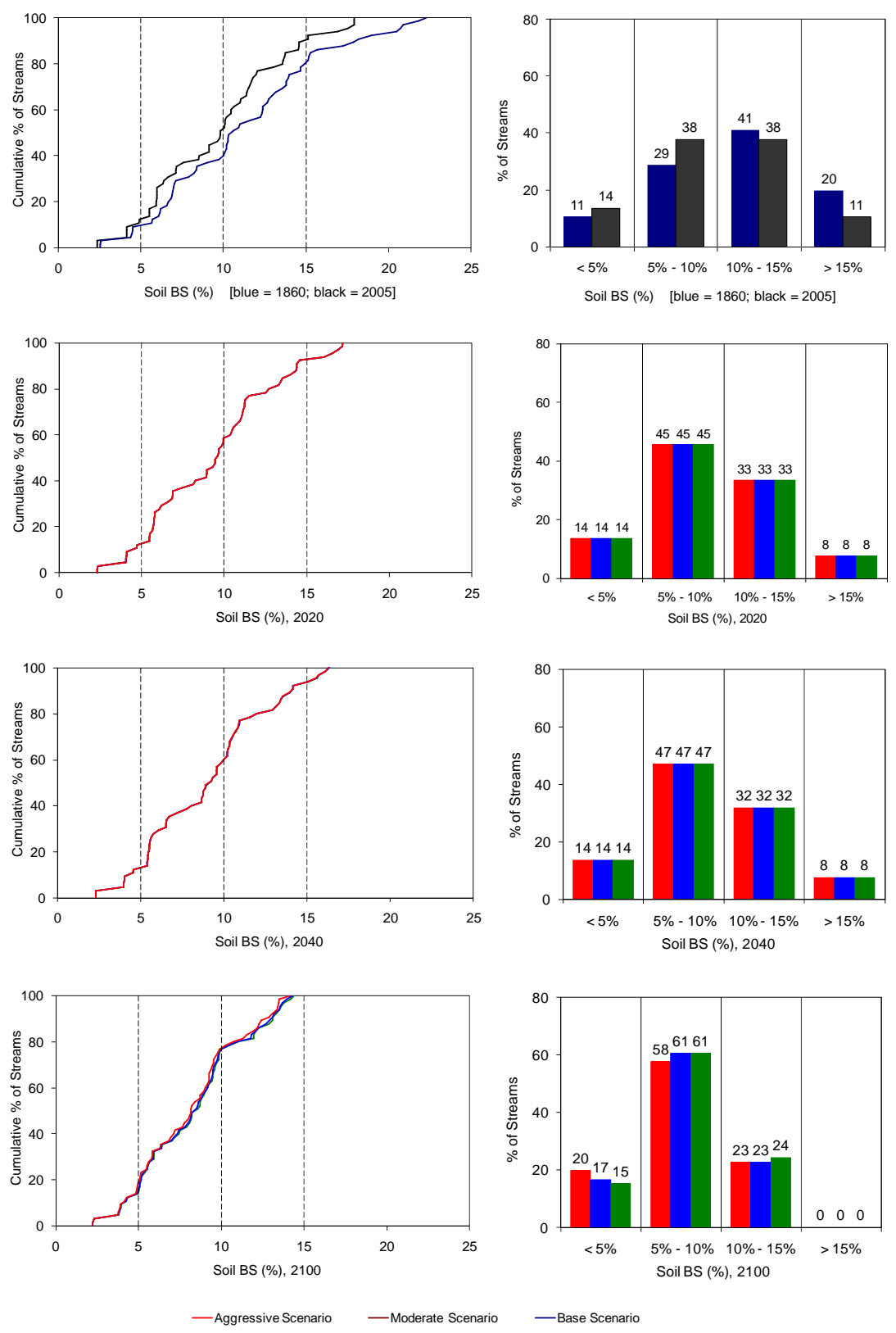


Figure 10. Continued.

c)

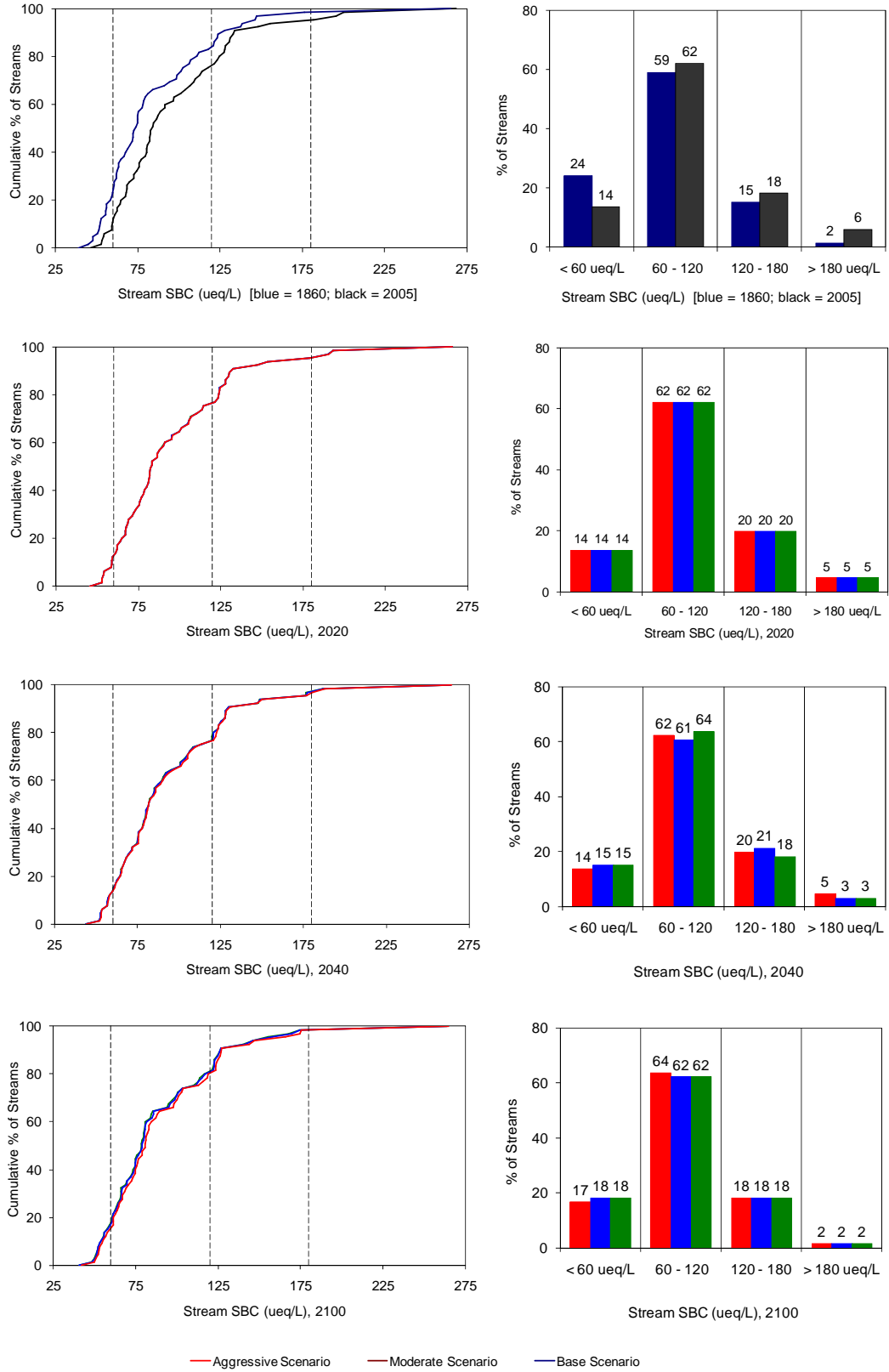


Figure 10. Continued.

d)

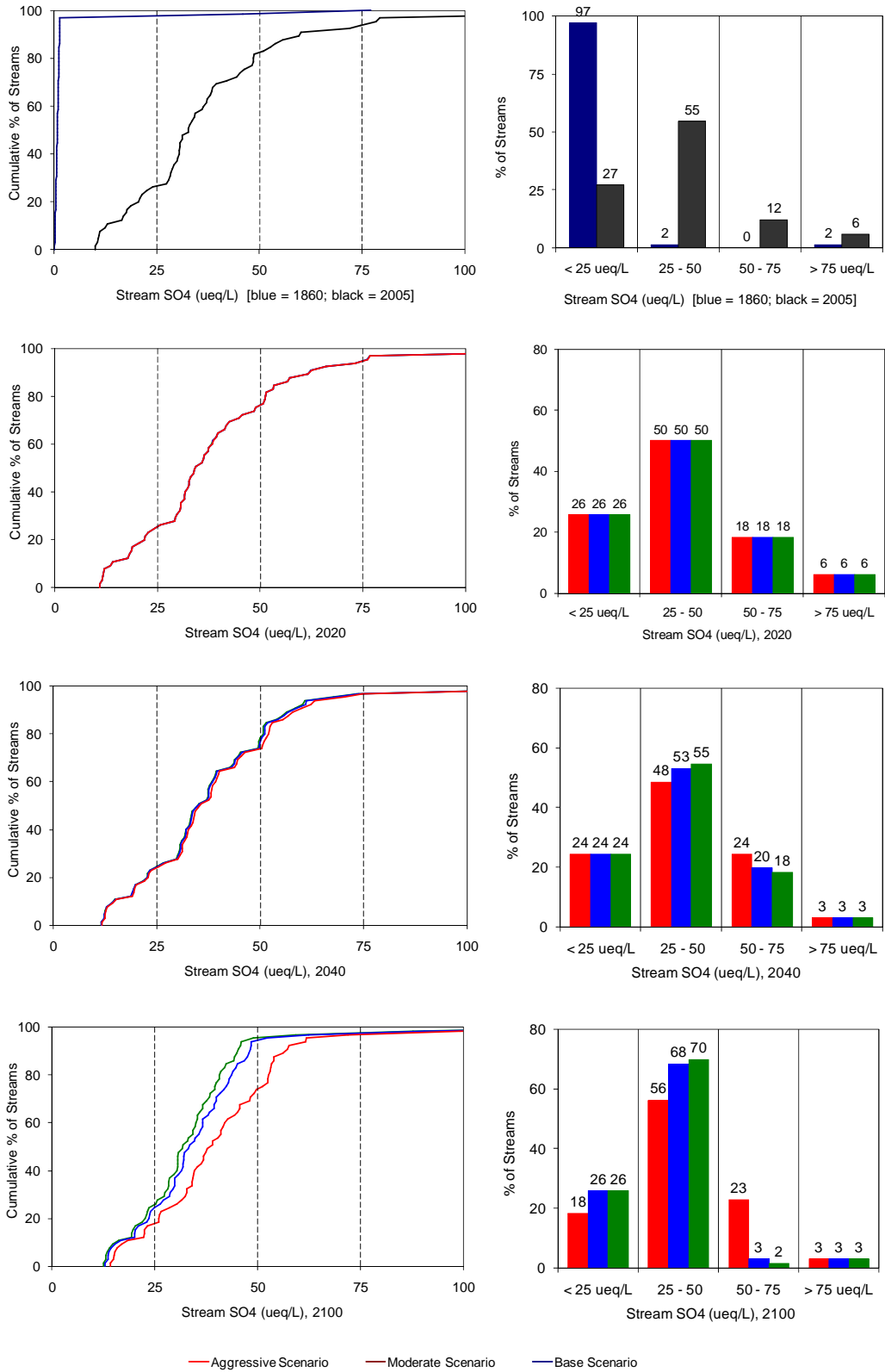


Figure 10. Continued.

Table 17. Simulated stream CALK (µeq/L) in 66 modeled streams in the past, present, and future under three scenarios of future emissions controls.

Site Name	Site ID	1860	2005	Base Scenario			Moderate Scenario			Aggressive Scenario		
				2020	2040	2100	2020	2040	2100	2020	2040	2100
Adam Camp Branch	237	50.3	39.3	38.2	37.0	33.6	38.2	37.1	34.8	38.2	37.1	35.2
Bear Branch	195	117.3	50.6	54.1	60.4	70.8	54.1	61.4	78.0	54.1	61.7	80.3
Bearpen Branch	253	47.1	22.0	19.7	17.4	11.8	19.7	18.0	16.3	19.7	18.3	17.8
Beetree Branch	190	117.7	46.1	48.3	53.9	65.2	48.3	54.4	71.9	48.3	54.5	74.0
Big Cove Branch	259	90.0	67.1	66.4	63.9	56.8	66.5	65.5	62.7	66.6	66.0	64.7
Big Laurel Brook	215	37.3	28.6	27.7	26.7	23.9	27.7	26.8	25.1	27.7	26.9	25.6
Big Oak Cove Creek	261	45.6	18.2	16.5	14.0	7.3	16.6	15.1	13.0	16.6	15.6	14.9
Briar Creek	72	90.6	44.8	41.2	39.7	38.6	41.2	40.7	46.7	41.2	41.1	49.3
Bubbling Spring Branch	129	41.8	3.5	-1.0	-4.7	-12.9	-0.9	-4.0	-5.8	-0.9	-3.7	-3.4
Bubbling Spring West Tributary	131	30.6	-3.0	-6.8	-10.0	-17.3	-6.8	-9.3	-10.9	-6.8	-9.1	-8.8
Buckeye Cove Creek	133	45.8	14.4	12.3	9.5	2.0	12.4	10.7	8.2	12.4	11.1	10.2
Cane Creek Tributary	204	45.8	14.1	11.8	10.0	6.0	11.8	10.6	11.2	11.8	10.9	12.9
Cathay Creek	122	109.6	69.9	68.7	67.3	64.3	68.8	69.0	71.8	68.9	69.6	74.3
Colberts Creek	53	49.8	26.4	24.0	21.0	14.0	24.0	21.8	19.4	24.0	22.1	21.2
Courthouse Creek	121	52.9	39.0	37.2	35.1	29.0	37.2	35.4	31.6	37.2	35.5	32.4
Dark Prong	114	41.0	18.7	15.9	13.2	6.4	15.9	13.5	9.9	15.9	13.6	11.1
Davidson River	102	91.4	68.8	68.0	66.6	63.7	68.0	66.4	65.9	68.0	66.3	66.6
East Fork Pigeon	101	52.6	33.9	32.6	31.2	27.7	32.6	31.6	30.6	32.6	31.8	31.6
Flat Laurel Creek	127	44.6	17.8	16.4	15.6	13.8	16.5	16.2	18.1	16.5	16.5	19.5
Glade Creek	198	82.1	73.8	72.7	71.5	68.3	72.7	71.6	69.4	72.7	71.7	69.8
Greenland Creek	160	45.5	19.5	17.6	16.2	13.2	17.7	16.8	17.2	17.7	16.9	18.5
Indian Branch	255	75.4	48.7	47.0	43.4	33.3	47.1	44.9	40.2	47.1	45.5	42.5
Indian Camp	185	82.0	75.9	75.1	74.0	70.7	75.1	74.0	71.2	75.1	74.0	71.4
Indian Spring Branch	239	59.7	42.6	40.5	38.1	32.0	40.6	38.5	35.4	40.6	38.6	36.3
Kilby Creek	217	46.1	38.0	37.0	35.9	32.7	37.0	36.0	33.8	37.1	36.1	34.2
Kirkland Cove	260	58.0	35.8	34.6	32.5	26.7	34.6	33.4	30.9	34.7	33.7	32.3
Left Prong South Toe River	61	64.3	43.0	43.5	41.6	36.3	43.6	42.4	40.1	43.6	42.7	41.4
Lindy Camp Branch	35	61.5	33.7	30.1	27.1	19.5	30.1	27.7	25.3	30.1	27.9	27.2
Little Prong Hickey Fork	91	132.3	121.7	121.7	120.7	117.8	121.7	120.4	118.5	121.7	120.3	118.7
Little Santetlah Cr. (NuCM site)	281	50.9	30.8	30.0	29.0	26.0	30.1	29.6	29.1	30.1	29.8	30.1
Long Branch	103	101.1	94.8	94.2	93.2	90.4	94.2	92.7	90.1	94.1	92.6	89.9
Lost Cove	56	63.2	36.4	33.7	31.1	25.5	33.7	31.8	30.7	33.7	32.0	32.4
Lower Creek	59	46.7	23.7	20.4	16.2	5.5	20.5	17.1	11.2	20.5	17.4	13.1
McNabb Creek	270	133.5	5.2	14.2	28.9	50.8	14.2	30.9	64.7	14.3	31.5	69.1

Table 17. Continued.

Site Name	Site ID	1860	2005	Base Scenario			Moderate Scenario			Aggressive Scenario		
				2020	2040	2100	2020	2040	2100	2020	2040	2100
Middle Creek	55	51.3	26.2	23.4	20.6	14.5	23.4	21.3	19.6	23.5	21.6	21.2
Mill Station Creek	132	41.7	10.8	6.6	2.3	-8.3	6.7	3.1	-1.0	6.7	3.4	1.3
Paddy Creek	26	37.2	1.1	-2.8	-5.5	-11.2	-2.8	-4.7	-3.9	-2.7	-4.5	-1.5
Peach Orchard Creek	70	66.9	27.5	27.2	26.0	22.6	27.3	27.4	30.5	27.3	27.9	33.1
Pigpen Branch	191	103.0	97.4	96.7	95.7	92.9	96.7	95.8	93.6	96.7	95.8	93.8
Rattlesnake Branch	143	96.5	64.6	61.8	59.8	55.2	61.8	60.5	60.9	61.8	60.7	62.7
Right Hand Prong	147	62.8	42.4	40.9	38.4	31.3	41.0	39.4	36.4	41.0	39.8	38.1
Roaring Branch	251	81.1	40.0	38.0	34.7	29.4	38.1	36.8	38.0	38.2	37.6	41.0
Rough Ridge Creek	257	65.3	48.4	46.7	44.8	39.6	46.7	45.1	41.9	46.7	45.2	42.7
Russell Creek	15	61.8	-13.7	-13.2	-7.9	0.1	-13.1	-6.2	12.4	-13.1	-5.6	16.3
Scotsman Creek	193	62.2	46.7	45.1	43.4	39.3	45.2	43.8	41.7	45.2	43.9	42.6
South Fork Fowler Creek	188	57.6	49.1	48.1	47.0	44.1	48.1	47.1	45.3	48.1	47.1	45.6
Spivey Creek	266	91.3	70.4	68.9	66.9	61.5	68.9	67.3	64.4	68.9	67.4	65.4
Squibb Creek	88	132.3	89.0	88.1	87.7	83.9	88.2	89.8	94.4	88.2	90.5	97.7
Stillhouse Branch	28	29.6	3.7	0.9	-1.6	-8.0	0.9	-1.2	-3.5	0.9	-1.1	-2.1
Unnamed creek A	245	48.2	14.8	13.8	11.6	7.0	13.9	13.1	12.9	13.9	13.7	15.0
Unnamed creek B	246	74.5	20.5	18.6	15.4	9.1	18.7	17.7	18.9	18.7	18.5	22.3
Unnamed creek C	249	67.0	39.4	37.9	34.8	26.5	38.1	36.3	32.8	38.1	36.9	35.0
Upper Creek	60	39.8	16.7	14.0	10.7	1.3	14.0	11.5	6.5	14.1	11.8	8.2
UT Flat Laurel Creek	120	50.0	21.5	19.0	15.8	6.8	19.0	15.9	10.5	19.0	15.9	11.7
UT Laurel Branch	265	84.6	53.2	50.2	46.8	37.3	50.2	47.5	42.8	50.2	47.7	44.5
UT Linville River (NUCM site)	21	34.2	-18.0	-21.0	-21.8	-23.2	-20.9	-20.7	-14.3	-20.9	-20.4	-11.5
UT McNabb Creek	262	40.9	28.6	27.3	25.8	21.8	27.3	26.0	23.2	27.3	26.0	23.7
UT North Fork of Catawba	33	33.9	4.9	1.4	-1.5	-8.5	1.4	-0.9	-2.4	1.4	-0.7	-0.6
UT Paint Creek	94	124.5	84.7	81.3	78.0	71.9	81.4	79.5	80.1	81.4	80.0	83.0
UT Panthertown Creek (Boggy Cr.)	163	61.7	35.7	34.3	33.2	31.1	34.3	33.2	34.1	34.3	33.2	35.1
UT Russell Creek	14	36.1	-19.2	-21.8	-21.4	-20.6	-21.8	-20.3	-11.1	-21.8	-19.9	-8.1
White Creek	16	41.1	14.2	10.7	7.5	-0.5	10.7	8.0	4.9	10.7	8.1	6.5
Wildcat Branch	232	53.7	42.2	40.9	39.3	34.7	40.9	39.5	36.6	40.9	39.6	37.2
Wilson Creek	169	83.3	74.9	74.0	72.9	69.7	74.1	73.1	70.9	74.1	73.2	71.2
Wolf Creek	150	61.3	48.0	46.5	44.8	40.4	46.6	45.1	42.4	46.6	45.2	43.1
Yellow Fork	27	31.1	-5.9	-9.6	-11.8	-16.6	-9.6	-11.0	-9.2	-9.6	-10.7	-6.9

Table 18. Simulated change in stream CALK ($\mu\text{eq/L}$) in the future (compared with values in 2005) under three scenarios of future emissions controls. Negative values imply that CALK concentration will decrease from 2005 values.

Site Name	Site ID	Base Scenario			Moderate Scenario			Aggressive Scenario		
		2020	2040	2100	2020	2040	2100	2020	2040	2100
Adam Camp Branch	237	-1.1	-2.3	-5.7	-1.1	-2.2	-4.4	-1.1	-2.1	-4.1
Bear Branch	195	3.4	9.8	20.2	3.5	10.7	27.4	3.4	11.0	29.6
Bearpen Branch	253	-2.3	-4.6	-10.2	-2.3	-4.0	-5.7	-2.3	-3.7	-4.2
Beetree Branch	190	2.3	7.8	19.1	2.2	8.3	25.8	2.2	8.5	28.0
Big Cove Branch	259	-0.7	-3.2	-10.3	-0.6	-1.6	-4.4	-0.5	-1.1	-2.4
Big Laurel Brook	215	-0.9	-2.0	-4.7	-0.9	-1.8	-3.5	-0.9	-1.8	-3.1
Big Oak Cove Creek	261	-1.6	-4.2	-10.9	-1.6	-3.0	-5.2	-1.5	-2.6	-3.3
Briar Creek	72	-3.6	-5.1	-6.1	-3.5	-4.0	2.0	-3.5	-3.7	4.5
Bubbling Spring Branch	129	-4.5	-8.2	-16.4	-4.5	-7.5	-9.4	-4.5	-7.2	-6.9
Bubbling Spring West Tributary	131	-3.9	-7.0	-14.4	-3.9	-6.4	-8.0	-3.9	-6.2	-5.9
Buckeye Cove Creek	133	-2.1	-4.9	-12.4	-2.0	-3.7	-6.2	-2.0	-3.3	-4.2
Cane Creek Tributary	204	-2.3	-4.1	-8.1	-2.3	-3.4	-2.9	-2.3	-3.2	-1.2
Cathey Creek	122	-1.2	-2.7	-5.6	-1.1	-1.0	1.9	-1.1	-0.4	4.4
Colberts Creek	53	-2.5	-5.4	-12.4	-2.4	-4.6	-7.0	-2.4	-4.3	-5.3
Courthouse Creek	121	-1.8	-3.9	-10.0	-1.8	-3.6	-7.4	-1.8	-3.5	-6.6
Dark Prong	114	-2.8	-5.6	-12.4	-2.8	-5.3	-8.8	-2.8	-5.1	-7.6
Davidson River	102	-0.8	-2.2	-5.2	-0.9	-2.4	-2.9	-0.9	-2.5	-2.3
East Fork Pigeon	101	-1.3	-2.8	-6.3	-1.3	-2.3	-3.3	-1.3	-2.2	-2.3
Flat Laurel Creek	127	-1.4	-2.2	-4.1	-1.3	-1.6	0.3	-1.3	-1.4	1.7
Glade Creek	198	-1.1	-2.3	-5.5	-1.1	-2.2	-4.4	-1.1	-2.2	-4.1
Greenland Creek	160	-1.8	-3.2	-6.3	-1.8	-2.7	-2.2	-1.8	-2.5	-0.9
Indian Branch	255	-1.7	-5.3	-15.4	-1.6	-3.8	-8.5	-1.6	-3.2	-6.2
Indian Camp	185	-0.7	-1.9	-5.2	-0.8	-1.9	-4.7	-0.8	-1.9	-4.5
Indian Spring Branch	239	-2.1	-4.6	-10.6	-2.1	-4.2	-7.3	-2.1	-4.0	-6.3
Kilby Creek	217	-1.0	-2.1	-5.3	-1.0	-2.0	-4.2	-1.0	-2.0	-3.8
Kirkland Cove	260	-1.2	-3.3	-9.1	-1.2	-2.4	-4.9	-1.1	-2.1	-3.5
Left Prong South Toe River	61	0.5	-1.4	-6.7	0.5	-0.6	-3.0	0.6	-0.3	-1.7
Lindy Camp Branch	35	-3.6	-6.6	-14.2	-3.6	-6.0	-8.3	-3.6	-5.8	-6.5
Little Prong Hickey Fork	91	0.0	-1.0	-3.9	0.0	-1.3	-3.2	0.0	-1.4	-3.0
Little Santetlah Cr. (NuCM site)	281	-0.8	-1.8	-4.8	-0.7	-1.2	-1.8	-0.7	-1.0	-0.7
Long Branch	103	-0.5	-1.6	-4.4	-0.6	-2.0	-4.7	-0.6	-2.2	-4.9
Lost Cove	56	-2.7	-5.3	-10.9	-2.7	-4.6	-5.7	-2.7	-4.4	-4.0
Lower Creek	59	-3.3	-7.4	-18.2	-3.2	-6.6	-12.5	-3.2	-6.3	-10.6
McNabb Creek	270	9.0	23.7	45.6	9.0	25.7	59.5	9.1	26.3	63.9

Table 18. Continued.

Site Name	Site ID	Base Scenario			Moderate Scenario			Aggressive Scenario		
		2020	2040	2100	2020	2040	2100	2020	2040	2100
Middle Creek	55	-2.8	-5.6	-11.7	-2.8	-4.9	-6.6	-2.8	-4.6	-5.0
Mill Station Creek	132	-4.2	-8.5	-19.1	-4.2	-7.8	-11.8	-4.1	-7.5	-9.6
Paddy Creek	26	-3.8	-6.5	-12.3	-3.8	-5.8	-4.9	-3.8	-5.6	-2.6
Peach Orchard Creek	70	-0.2	-1.5	-4.9	-0.2	-0.1	3.0	-0.1	0.5	5.6
Pigpen Branch	191	-0.8	-1.7	-4.5	-0.8	-1.6	-3.8	-0.8	-1.6	-3.6
Rattlesnake Branch	143	-2.8	-4.8	-9.4	-2.8	-4.1	-3.7	-2.8	-3.9	-1.9
Right Hand Prong	147	-1.5	-4.1	-11.2	-1.5	-3.0	-6.0	-1.4	-2.6	-4.3
Roaring Branch	251	-2.0	-5.3	-10.6	-1.8	-3.1	-2.0	-1.8	-2.4	1.0
Rough Ridge Creek	257	-1.7	-3.6	-8.8	-1.7	-3.3	-6.5	-1.7	-3.2	-5.7
Russell Creek	15	0.5	5.8	13.8	0.5	7.5	26.1	0.5	8.1	30.0
Scotsman Creek	193	-1.6	-3.3	-7.5	-1.6	-3.0	-5.0	-1.6	-2.8	-4.2
South Fork Fowler Creek	188	-1.0	-2.1	-4.9	-1.0	-2.0	-3.8	-1.0	-2.0	-3.5
Spivey Creek	266	-1.5	-3.4	-8.8	-1.5	-3.1	-5.9	-1.5	-3.0	-5.0
Squibb Creek	88	-0.9	-1.3	-5.1	-0.8	0.8	5.4	-0.8	1.5	8.7
Stillhouse Branch	28	-2.8	-5.3	-11.7	-2.8	-4.9	-7.3	-2.8	-4.8	-5.8
Unnamed creek A	245	-1.0	-3.1	-7.8	-0.9	-1.6	-1.8	-0.9	-1.1	0.2
Unnamed creek B	246	-2.0	-5.2	-11.4	-1.8	-2.8	-1.6	-1.8	-2.0	1.8
Unnamed creek C	249	-1.4	-4.6	-12.9	-1.3	-3.0	-6.6	-1.3	-2.4	-4.3
Upper Creek	60	-2.7	-6.0	-15.4	-2.6	-5.2	-10.2	-2.6	-4.9	-8.4
UT Flat Laurel Creek	120	-2.5	-5.6	-14.6	-2.5	-5.6	-10.9	-2.5	-5.6	-9.8
UT Laurel Branch	265	-3.0	-6.4	-16.0	-3.0	-5.7	-10.4	-3.0	-5.5	-8.7
UT Linville River (NUCM site)	21	-3.0	-3.9	-5.3	-3.0	-2.8	3.6	-3.0	-2.4	6.5
UT McNabb Creek	262	-1.3	-2.8	-6.9	-1.3	-2.6	-5.4	-1.3	-2.6	-4.9
UT North Fork of Catawba	33	-3.4	-6.4	-13.4	-3.4	-5.8	-7.3	-3.4	-5.5	-5.4
UT Paint Creek	94	-3.4	-6.7	-12.8	-3.3	-5.3	-4.6	-3.3	-4.8	-1.7
UT Panthertown Creek (Boggy C	163	-1.3	-2.5	-4.6	-1.3	-2.4	-1.5	-1.4	-2.5	-0.6
UT Russell Creek	14	-2.6	-2.3	-1.4	-2.6	-1.1	8.1	-2.6	-0.7	11.0
White Creek	16	-3.5	-6.7	-14.7	-3.5	-6.2	-9.3	-3.5	-6.1	-7.6
Wildcat Branch	232	-1.3	-2.9	-7.5	-1.3	-2.7	-5.6	-1.3	-2.6	-5.0
Wilson Creek	169	-0.9	-2.0	-5.2	-0.9	-1.8	-4.0	-0.9	-1.7	-3.7
Wolf Creek	150	-1.4	-3.2	-7.6	-1.4	-2.9	-5.6	-1.4	-2.8	-4.9
Yellow Fork	27	-3.7	-5.9	-10.7	-3.7	-5.1	-3.3	-3.7	-4.8	-0.9

Table 19. MAGIC critical loads results for the 66 study watersheds.

Site Name	Historical Stream ANC	2005 Stream ANC $\mu\text{eq/L}$	ANC in 2064 (Aggressive Scenario)	Ref Yr 2005 $\text{SO}_4\text{ TotDep Kg S/ha/yr}$	Critical Load of SO_4 kg S/ha/yr for Stream ANC = 0 $\mu\text{eq/L}$ in Year			Critical Load of SO_4 kg S/ha/yr for Stream ANC = 20 $\mu\text{eq/L}$ in Year			Critical Load of SO_4 kg S/ha/yr for Stream ANC = 50 $\mu\text{eq/L}$ in Year ¹			Critical Load of SO_4 kg S/ha/yr for Stream ANC = 100 $\mu\text{eq/L}$ in Year ¹		
					2020	2040	2100	2020	2040	2100	2020	2040	2100	2020	2040	2100
Adam Camp Branch	50	39	36.22	12.2	524.9	133.5	41.0	291.8	76.4	23.4	0.0	0.0	0.0			
Bear Branch	117	51	70.48	6.5	42.5	17.0	11.6	30.3	13.2	9.5	7.3	6.4	6.2	0.0	0.0	0.0
Bearpen Branch	47	22	17.77	12.0	111.4	30.4	11.7	5.2	2.7	2.7						
Beetree Branch	118	46	62.83	6.7	44.6	17.1	11.6	29.8	12.6	9.5	2.1	5.3	6.0	0.0	0.0	0.0
Big Cove Branch	90	67	66.38	9.4	574.4	143.1	40.1	466.0	115.9	31.2	229.2	54.0	11.8			
Big Laurel Brook	37	29	26.25	13.5	536.2	137.9	42.3	187.0	47.5	14.5						
Big Oak Cove Creek	46	18	15.53	11.0	108.3	27.5	9.4	0.0	0.0	0.0						
Briar Creek	91	45	43.93	8.7	52.8	18.7	11.0	34.2	13.1	8.4	0.0	0.1	3.1			
Bubbling Spring Branch	42	4	-4.58	21.6	6.1	5.3	5.9	0.0	0.0	0.0						
Bubbling Spring West Tributary	31	-3	-9.8	20.5	0.0	0.0	1.9	0.0	0.0	0.0						
Buckeye Cove Creek	46	14	10.99	22.2	133.6	35.6	13.9	0.0	0.0	0.0						
Cane Creek Tributary	46	14	11.4	13.1	62.7	19.7	10.2	0.0	0.0	1.1						
Cathy Creek	110	70	72.1	9.3	152.3	44.1	19.4	120.5	35.1	15.6	55.7	17.7	9.2	0.0	0.0	0.0
Colberts Creek	50	26	21.49	13.0	144.2	37.5	13.7	32.9	8.9	4.5	0.0	0.0	0.0			
Courthouse Creek	53	39	34.12	13.4	362.4	88.7	26.6	213.0	51.0	14.7	0.0	0.0	0.0			
Dark Prong	41	19	12.05	21.3	190.4	49.0	17.4	0.0	0.0	0.0						
Davidson River	91	69	65.58	15.4	344.0	94.5	38.5	273.7	75.5	30.9	131.0	37.5	16.9			
East Fork Pigeon	53	34	31.65	13.3	265.8	71.4	26.5	123.5	33.9	13.4	0.0	0.0	0.0			
Flat Laurel Creek	45	18	17.64	9.3	67.5	21.0	10.2	0.0	0.5	2.9						
Glade Creek	82	74	70.72	11.7	974.3	236.9	69.6	812.3	197.1	57.0	446.5	110.5	30.9			
Greenland Creek	45	19	17.37	11.5	90.4	26.5	12.3	0.0	0.9	3.1						
Indian Branch	75	49	44.95	10.6	294.4	71.5	19.5	188.1	45.1	12.5	0.0	0.0	0.0			
Indian Camp	82	76	72.72	10.7	910.0	227.0	65.3	778.8	195.1	55.2	460.6	118.0	31.4			
Indian Spring Branch	60	43	37.36	17.7	431.2	108.1	34.0	260.4	61.9	19.7	0.0	0.0	0.0			
Kilby Creek	46	38	35.23	14.5	767.7	188.4	54.5	421.4	103.1	29.1						
Kirkland Cove	58	36	33.47	9.9	250.7	62.9	19.2	124.5	30.4	9.5	0.0	0.0	0.0			
Left Prong South Toe River	64	43	42.41	15.5	443.2	113.7	36.5	277.9	71.6	23.3	0.0	0.0	0.0			
Lindy Camp Branch	62	34	27	11.3	84.9	25.1	10.9	36.5	11.8	6.3	0.0	0.0	0.0			
Little Prong Hickey Fork	132	122	119.11	8.7	377.8	103.2	40.2	353.9	96.1	36.8	305.6	82.3	30.6	148.0	39.9	14.6
Little Santetlah Cr. (NuCM site)	51	31	30.08	14.5	299.1	79.2	28.2	118.5	32.8	13.3	0.0	0.0	0.0			
Long Branch	101	95	90.89	16.3	1307.6	319.4	100.1	1150.5	281.2	87.0	831.2	204.6	61.5	0.0	0.0	0.0
Lost Cove	63	36	31.78	16.1	179.3	49.2	20.5	91.1	25.8	11.6	0.0	0.0	0.0			
Lower Creek	47	24	15.47	12.5	116.1	27.6	8.8	10.4	1.0	0.4						
McNabb Creek	134	5	49.96	10.2	16.5	12.4	11.5	1.7	8.1	9.5	0.0	0.7	6.1	0.0	0.0	0.0
Middle Creek	51	26	21.13	13.1	131.1	34.2	13.5	30.0	8.6	4.9	0.0	0.0	0.0			
Mill Station Creek	42	11	1.77	12.0	31.4	8.6	4.3	0.0	0.0	0.0						

Table 19. Continued.

Site Name	Historical Stream ANC	2005 Stream ANC µeq/L	ANC in 2064 (Aggressive Scenario)	Ref Yr 2005 SO ₄ TotDep Kg S/ha/yr	Critical Load of SO ₄ kg S/ha/yr for Stream ANC = 0 µeq/L in Year			Critical Load of SO ₄ kg S/ha/yr for Stream ANC = 20 µeq/L in Year			Critical Load of SO ₄ kg S/ha/yr for Stream ANC = 50 µeq/L in Year ¹			Critical Load of SO ₄ kg S/ha/yr for Stream ANC = 100 µeq/L in Year ¹		
					2020	2040	2100	2020	2040	2100	2020	2040	2100	2020	2040	2100
Paddy Creek	37	1	-3.93	9.4	0.0	2.4	3.2	0.0	0.0	0.0						
Peach Orchard Creek	67	27	30.4	8.0	49.4	16.3	8.6	18.6	7.8	5.0	0.0	0.0	0.0			
Pigpen Branch	103	97	94.93	10.6	1224.8	304.4	88.1	1113.0	276.9	79.1	866.0	216.9	60.4	0.0	0.0	0.0
Rattlesnake Branch	96	65	61.11	7.7	94.6	27.5	12.4	72.4	21.3	10.0	27.9	9.5	5.5			
Right Hand Prong	63	42	39.45	10.8	237.3	61.2	20.1	150.2	38.5	12.1	0.0	0.0	0.0			
Roaring Branch	81	40	39.66	9.9	177.8	43.9	15.9	101.6	25.2	9.6	0.0	0.0	0.0			
Rough Ridge Creek	65	48	44.02	10.1	314.2	79.4	25.0	217.8	54.9	17.1	0.0	0.0	0.0			
Russell Creek	62	-14	4.63	10.5	0.0	3.7	5.8	0.0	0.0	3.0	0.0	0.0	0.0			
Scotsman Creek	62	47	43.11	10.9	383.2	96.3	30.5	245.2	61.5	19.5	0.0	0.0	0.0			
South Fork Fowler Creek	58	49	46.4	11.3	732.7	182.0	54.5	489.7	122.5	36.4	0.0	0.0	0.0			
Spivey Creek	91	70	66.36	9.1	227.4	62.5	22.7	190.2	52.2	18.6	99.5	27.4	9.7			
Squibb Creek	132	89	94.52	7.4	52.6	22.6	13.1	46.7	19.7	11.3	34.6	13.8	8.3	0.0	0.0	1.3
Stillhouse Branch	30	4	-2	10.1	9.4	4.2	3.1	0.0	0.0	0.0						
Unnamed creek A	48	15	14.78	13.4	119.1	31.9	12.7	0.0	0.0	0.0						
Unnamed creek B	74	21	20.75	11.2	68.4	18.7	8.7	1.2	1.4	2.8	0.0	0.0	0.0			
Unnamed creek C	67	39	36.76	10.6	292.3	70.9	20.3	171.7	39.9	10.5	0.0	0.0	0.0			
Upper Creek	40	17	10.25	12.7	93.9	23.4	7.5	0.0	0.0	0.0						
UT Flat Laurel Creek	50	21	13.4	18.9	142.6	39.2	14.7	4.5	2.4	2.9	0.0	0.0	0.0			
UT Laurel Branch	85	53	46.11	10.4	136.0	38.2	14.0	98.9	27.1	9.9	6.5	2.2	1.4			
UT Linville River (NUCM site)	34	-18	-16.97	11.3	0.0	0.0	1.4	0.0	0.0	0.0						
UT McNabb Creek	41	29	24.93	10.5	322.0	82.5	24.7	116.2	28.8	8.2						
UT North Fork of Catawba	34	5	-1.2	9.6	9.1	4.4	3.3	0.0	0.0	0.0						
UT Paint Creek	125	85	80.93	7.0	88.7	27.3	12.8	75.7	23.3	10.8	49.5	15.4	7.4	0.0	0.0	0.0
UT Panthertown Creek (Boggy Cr)	62	36	33.26	10.7	121.2	35.9	16.7	63.9	20.2	10.3	0.0	0.0	0.0			
UT Russell Creek	36	-19	-15.14	10.6	0.0	0.0	2.1	0.0	0.0	0.0						
White Creek	41	14	6.8	11.3	44.3	133	6.4	0.0	0.0	0.0						
Wildcat Branch	54	42	38.49	14.6	558.9	141.0	42.1	331.9	85.0	25.1	0.0	0.0	0.0			
Wilson Creek	83	75	72.33	13.8	1117.8	278.7	81.7	924.3	230.9	66.8	505.5	133.5	38.3			
Wolf Creek	61	48	44.22	11.5	435.3	109.0	33.5	297.4	74.7	22.5	0.0	0.0	0.0			
Yellow Fork	31	-6	-9.65	9.2	0.0	0.0	1.9	0.0	0.0	0.0						

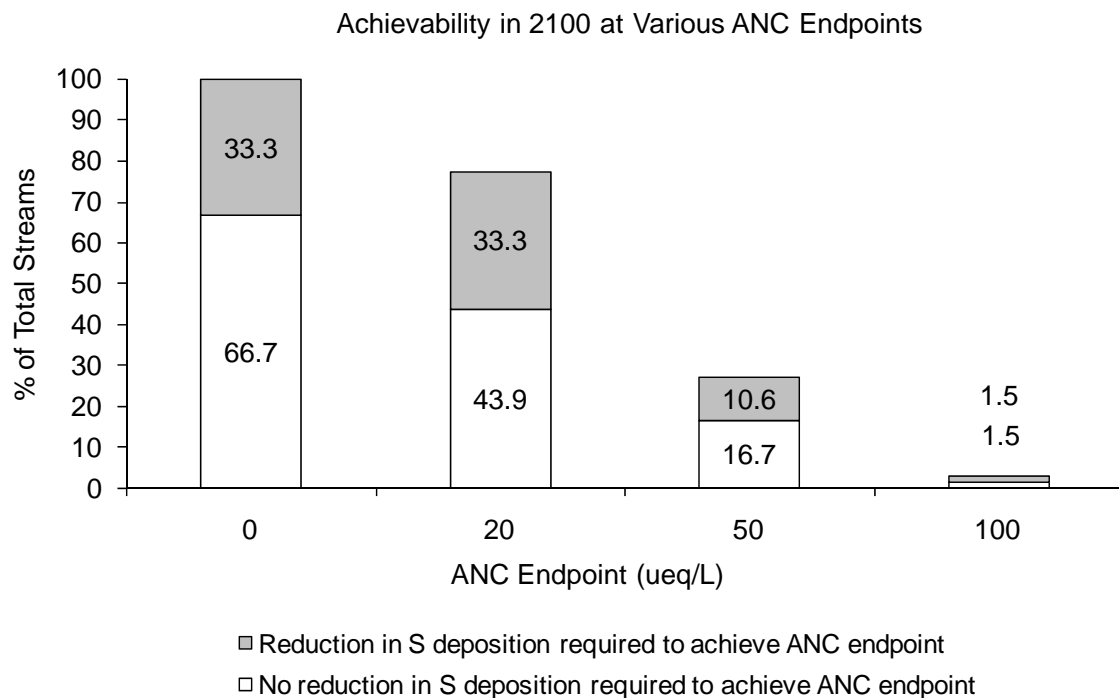
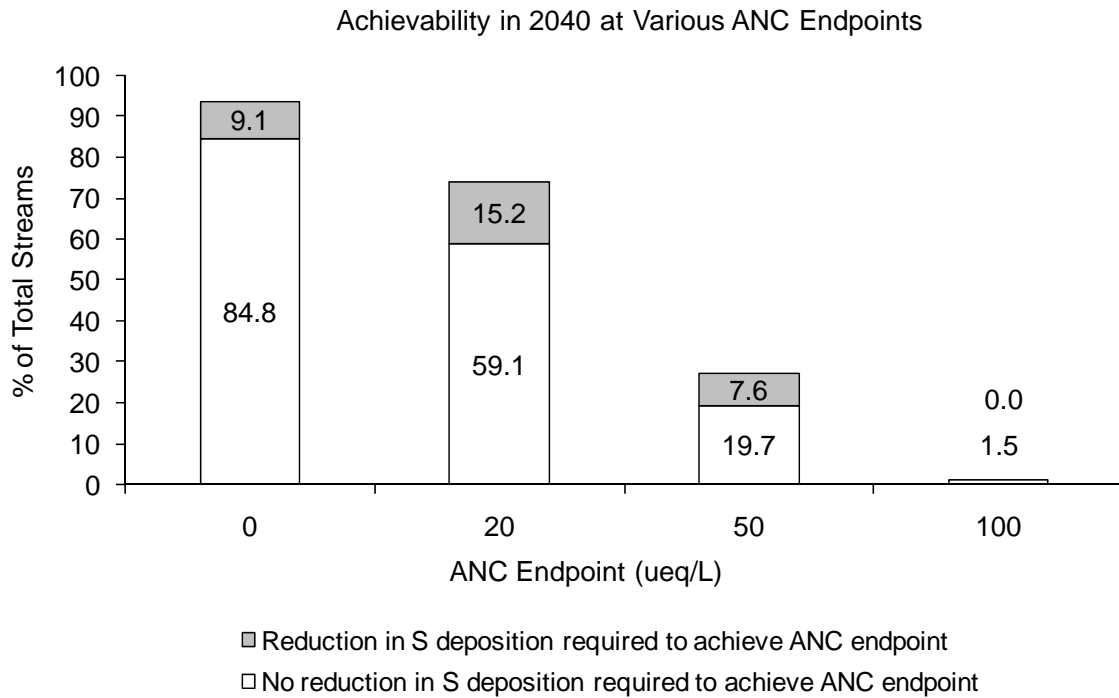


Figure 11. Comparison of the extent to which various ANC criteria (0, 20, 50, 100 $\mu\text{eq/L}$) are simulated to be achievable in the year 2040 (top panel) and 2100 (bottom panel). Results are presented as percent of modeled streams. Sites for which the endpoint criterion was achievable in the endpoint year are classified as either achievable without reduced S deposition compared with the reference year 2005, or achievable only with reduced S deposition.

= 100 $\mu\text{eq/L}$ is not a good management target. ANC criteria values equal to 20 and 50 $\mu\text{eq/L}$ are often achievable (Figure 11) and are associated with lower levels of ecological harm than ANC = 0. The critical load required to achieve ANC=20 or 50 $\mu\text{eq/L}$ varies greatly for those sites that have high critical loads. Figure 12 illustrates this pattern, using the year 2040 as an example. For sites having critical load closer to ambient deposition, however, the differences in critical load are much smaller, depending on whether the selected ANC criterion is 20 or 50 $\mu\text{eq/L}$ (Figure 12). The calculated S deposition critical load for modeled streams varied in relation to watershed sensitivity (as reflected in geologic, soils, and streamwater characteristics), the selected chemical criterion (critical ANC value), and the future year for which the evaluation was made.

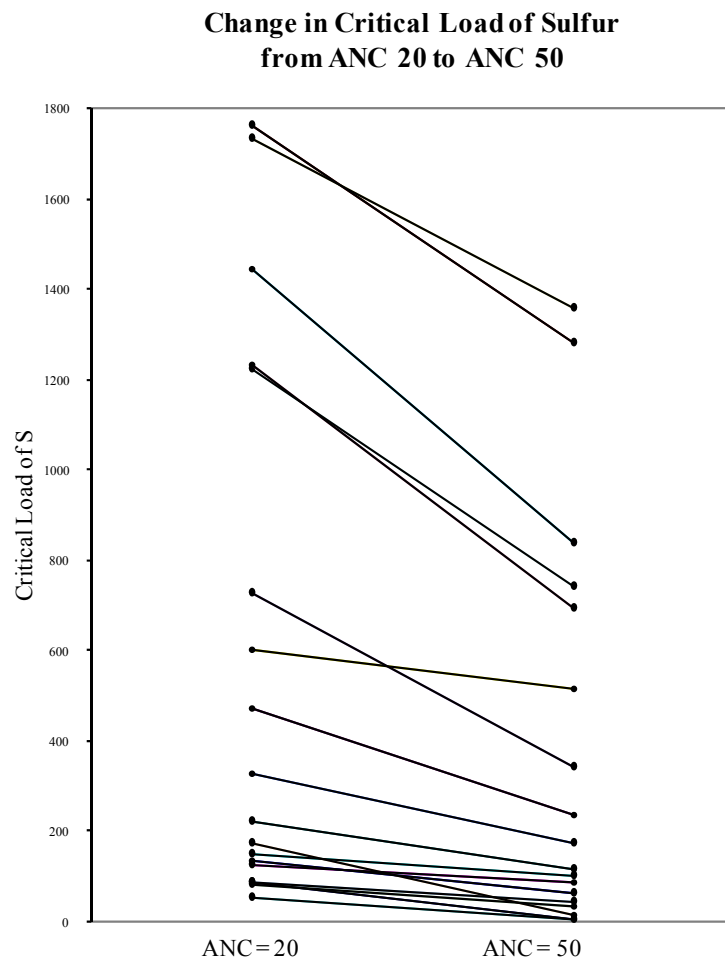


Figure 12. Site-by-site comparison of differences in simulated critical load of S (kg S/ha/yr) for the year 2040, depending on whether the target stream ANC value was selected to be 20 or 50 $\mu\text{eq/L}$ (n = 18).

The relationships between critical load, selection of ANC criterion value, and selection of evaluation year are important. Higher critical loads can be tolerated for some streams if one is willing to wait to 2100 to achieve the critical ANC target level, as compared with more stringent deposition reductions required to attain specified ANC values by 2020 or 2040. For other streams, higher critical loads can be tolerated for short-term protection versus more stringent deposition reductions required to protect ecosystems for a longer period of time. Higher critical loads are allowable if one wishes to prevent acidification to ANC = 20 $\mu\text{eq/L}$ (episodic acidification effects on brook trout likely) than if one wishes to be more restrictive and prevent acidification to ANC below 50 $\mu\text{eq/L}$ (biological effects of acidification likely on biota other than brook trout).

The calculated critical loads of S deposition required to prevent streamwater acidification to ANC values below 0 and 20 $\mu\text{eq/L}$ varied as a function of ANC (Figure 13). These model data suggest that most modeled streams that had 1995 ANC $\leq 20 \mu\text{eq/L}$ would require low critical load values to maintain ANC above 20 $\mu\text{eq/L}$ in the future. At low ANC values, critical loads were consistently near zero; at higher ANC values, critical loads were more variable. Some streams having ANC above 50 $\mu\text{eq/L}$ exhibited low critical loads, whereas others had critical loads much higher than current deposition levels. Stronger relationships were observed when we plotted modeled critical load as a function of the ratio of streamwater ANC to streamwater SO_4^{2-} concentration (Figure 14). Watersheds having the lowest ANC and the highest SO_4^{2-} concentrations in streamwater had the lowest critical load. This was especially true for calculations of the critical loads to achieve ANC values of 0 and 20 $\mu\text{eq/L}$. In this case, the SO_4^{2-} concentration reflects the extent to which SO_4^{2-} is mobile within the watershed, as opposed to being retained on the soil. Lower critical loads occur where SO_4^{2-} is more mobile.

Achievability of ANC criteria in a variety of future years is summarized in Table 20. More sites could achieve ANC of 20 $\mu\text{eq/L}$ in the year 2100 (50 modeled sites) as compared with the year 2020 (42 modeled sites). Nevertheless, deposition would have to be reduced from ambient values at a third of the modeled sites to achieve ANC 20 $\mu\text{eq/L}$ in 2100, versus just 8% of the modeled sites to achieve ANC 20 $\mu\text{eq/L}$ in 2020. This occurs because the watershed can tolerate less deposition if the objective is to maintain the critical ANC over a longer period of time. Patterns were similar for the ANC criterion 50 $\mu\text{eq/L}$ (Table 20).

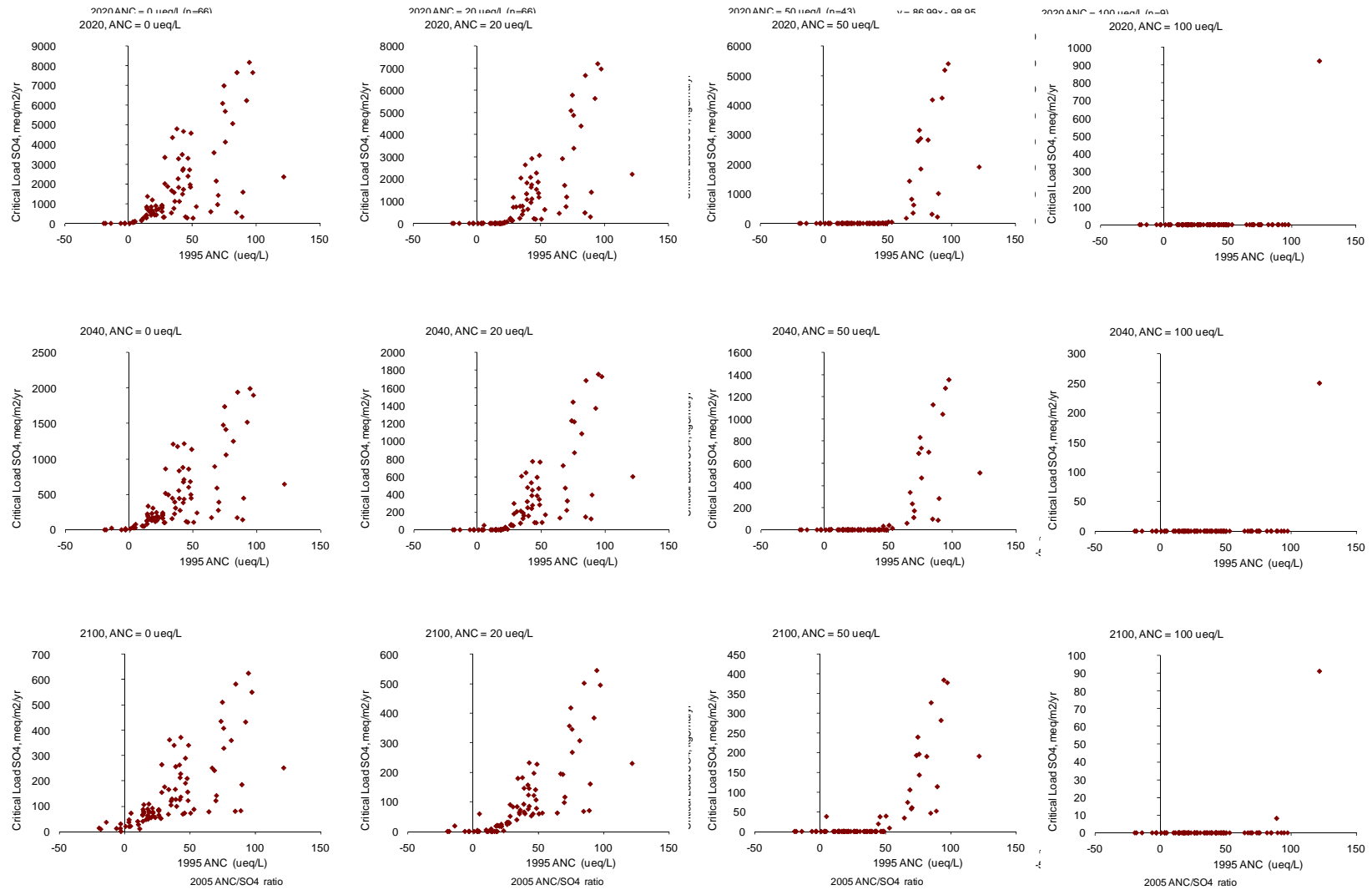


Figure 13. Critical loads of S for the 66 modeled sites to achieve 4 different streamwater ANC levels (0, 20, 50 and 100 $\mu\text{eq/L}$), evaluated in 3 different years (2020, 2040 and 2100). Critical load implementation was begun in 2009 and completed in 2018. Critical loads are depicted as median simulated values at each site, calculated from multiple (5 to 10) calibrations of MAGIC for each site.

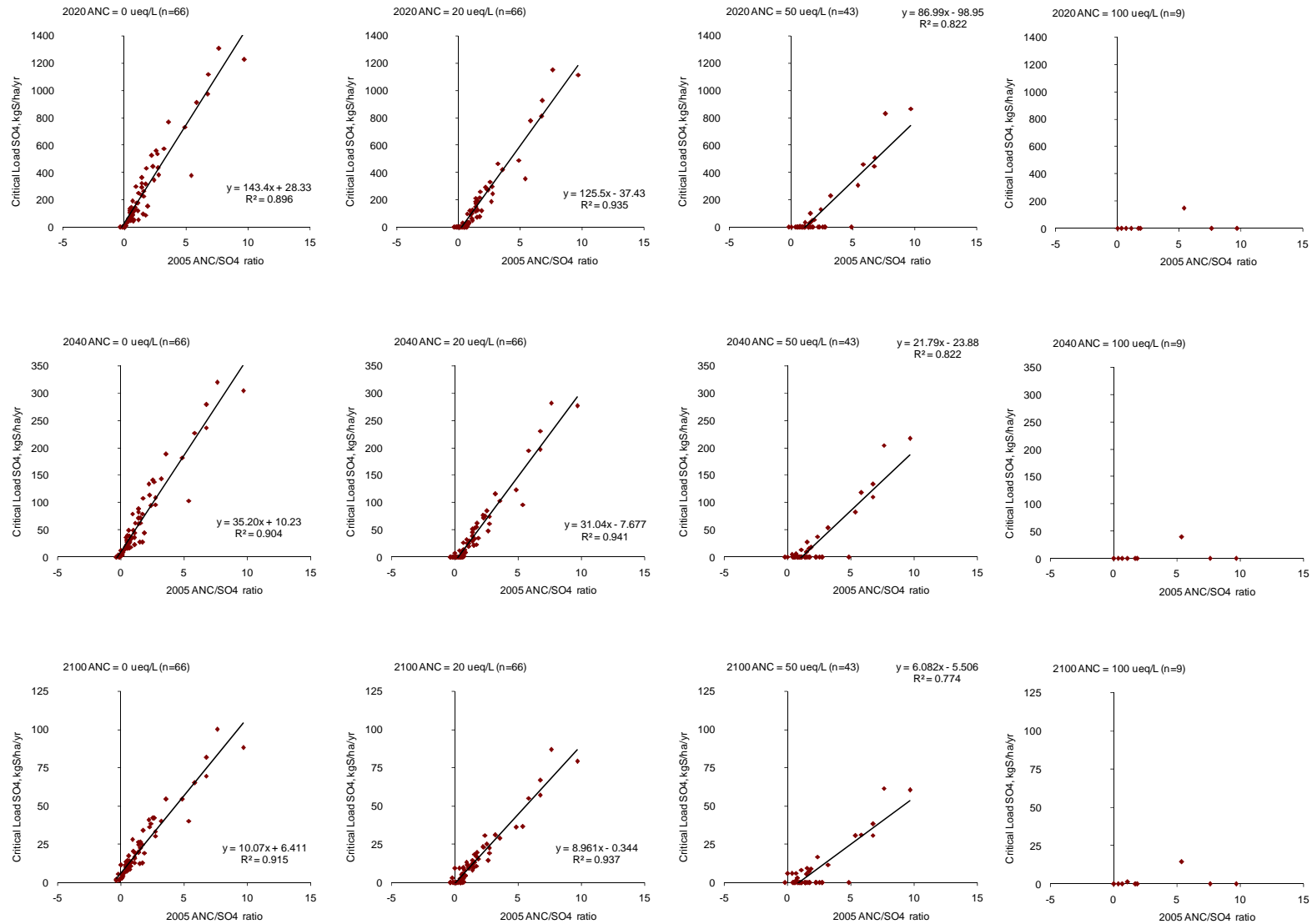


Figure 14. Critical loads of S for the 66 modeled sites to achieve four different streamwater ANC critical levels, evaluated in three different years, compared with the ratio of ANC to SO₄²⁻ concentration in streamwater in 2005.

Table 20. Achievability of ANC ($\mu\text{eq/L}$) endpoints in a variety of future years for 66 modeled streams.

	2020		2040		2100	
	#	%	#	%	#	%
Endpoint ANC = 0						
Total achievable streams	60	91	62	94	66	100
Streams requiring reduction in S deposition ¹	3	5	6	9	22	33
Endpoint ANC = 20						
Total achievable streams	42	64	47	71	51	77
Streams requiring reduction in S deposition	5	8	10	15	22	33
Endpoint ANC = 50						
Total achievable streams	14	21	18	27	18	27
Streams requiring reduction in S deposition	1	2	5	8	7	11
Endpoint ANC = 100						
Total achievable streams	1	2	1	2	2	3
Streams requiring reduction in S deposition	0	0	0	0	1	2

¹ Streams for which the critical ANC level was achievable by the indicated endpoint year, but only if S deposition is reduced below ambient (2005) values.

As illustrated in the analysis of critical loads of sulfur deposition presented here, there exists a range of important issues that should be considered in developing and implementing a critical loads approach. Key issues include the following:

- What is the environmental response indicator, and what does it tell us about the system?
- What is/are the selected critical endpoint criterion value(s) for the response indicator?
- What constitutes “recovery” in the context of this indicator?
- What is the time period of evaluation of the critical load?
- How representative of the broader region are the water bodies selected for modeling and/or how many waters are represented by the modeled sites?
- What are the major sources and levels of uncertainty?

Results of model simulations and critical loads calculations presented here will help to inform the development of the critical loads approach as a potential assessment and policy tool in the southeastern United States. This could aid the management of acid-sensitive resources in this region and elsewhere. Additional logical steps in the process could include selection of interim target loads of S deposition which would allow acid-sensitive soils and streams in the region to

begin the process of chemical recovery and move toward the long-term critical load values that would sustain sensitive aquatic and terrestrial life forms.

E. Characteristics of Modeled Sites

Distributions of the modeled sites across key parameter values are shown in Figure 15. Modeled sites were widely distributed across elevation (Figure 15A) and lithology (Figure 15B). Sites were relatively evenly distributed across the three most acid-sensitive geologic sensitivity classes (siliceous, argillaceous, and felsic). Only one modeled site was located on the less-sensitive mafic geology and none were located on insensitive carbonate geology (Figure 15C). Streamwater SO_4^{2-} concentrations were rather widely distributed, with some sites having SO_4^{2-} concentration below 15 $\mu\text{eq/L}$ and some above 65 $\mu\text{eq/L}$ (Figure 15D). Several streams had NO_3^- concentration higher than 15 $\mu\text{eq/L}$, but more than half had NO_3^- concentration less than 4 $\mu\text{eq/L}$ (Figure 15E). All modeled streams had calculated ANC below 150 $\mu\text{eq/L}$, and one-third had ANC below 20 $\mu\text{eq/L}$. Thus, many of these modeled streams are very acid-sensitive. Most (70%) had pH above 6.0, although some modeled sites (7%) had pH below 5.0 (Figure 15G).

Snyder et al. (2004) examined the relationship between stream ANC and landscape variables in western North Carolina and found the best predictors included the lithology, elevation, and whether the watershed had a conifer forest cover type. Also, streams with an ANC value of less than or equal to 20 $\mu\text{eq/L}$ had an average sulfate deposition of 25 kg/ha/yr or greater. Therefore, future stream ANC might be estimated for a new stream sample by knowing something about the geology, elevation, and forest cover of the catchment and the MAGIC results for an ANC category and/or landscape type.

F. Regression Modeling to Estimate Critical Load at Locations That Were Not Modeled

It is useful to put the results of this critical loads analysis into the perspective of the population of streams within the region. This cannot be done directly, however, because the modeled streams were not drawn from a statistical frame. This can be done indirectly by quantifying critical load within sensitivity classes or as a function of reference year chemistry and watershed characteristics.

In order to aid in the process of extrapolating MAGIC model critical loads simulation results to watersheds within the study area that were not modeled, we developed a suite of

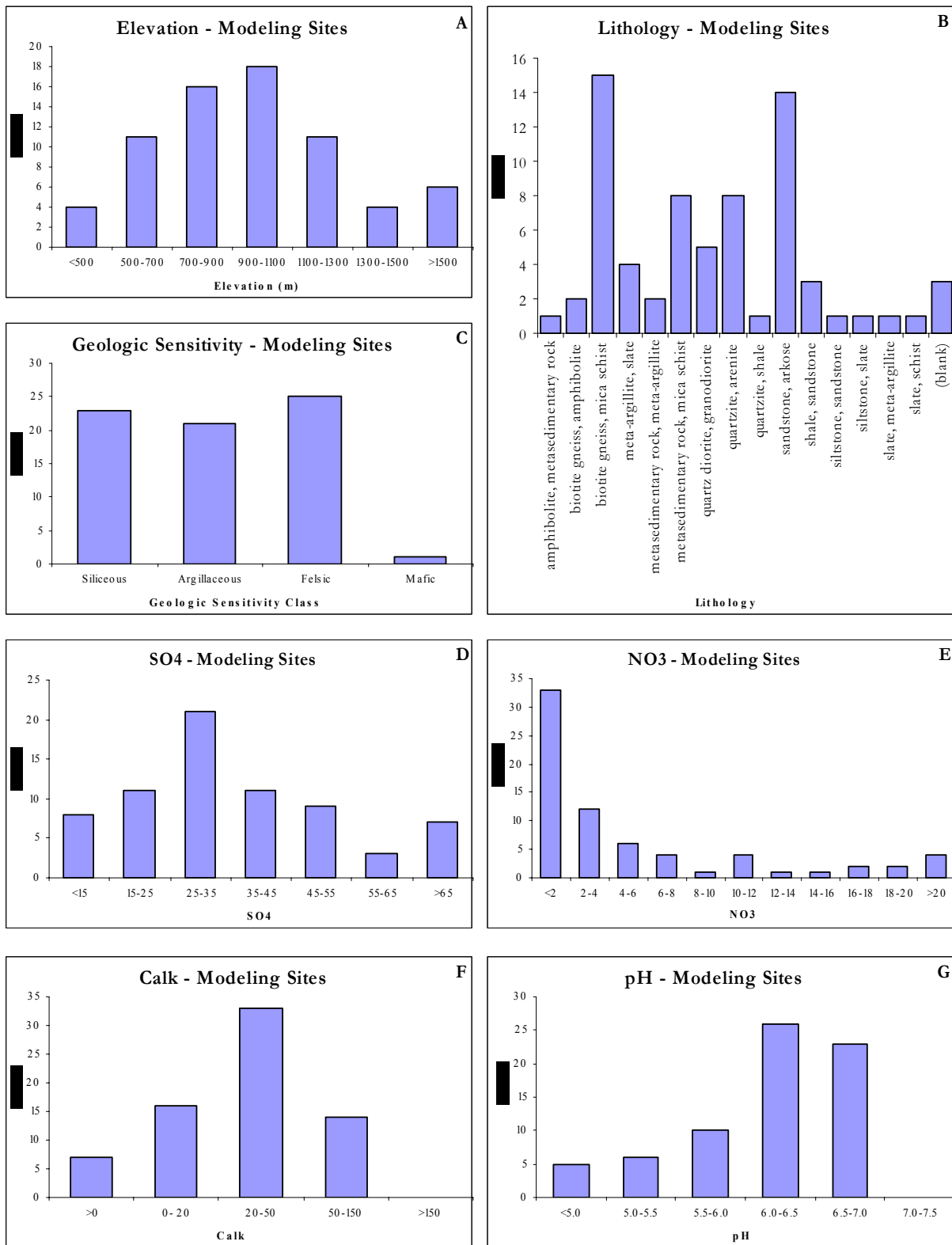


Figure 15. Distribution of modeling sites across key parameter values.

multiple regression models to estimate critical loads from variables that are more widely available across the region than are the MAGIC model results. Separate multiple regression modeling efforts were conducted to estimate critical load from 1) landscape variables represented spatially in the GIS, and 2) a combination of landscape variables and stream chemistry variables. Those that were significant predictors in one or more models are listed in Table 21. Results are summarized in Table 22. More robust predictions can be generated using water chemistry, in addition to landscape data. For watersheds lacking water chemistry data, landscape variables alone can be used, but the predictive relationships are weak.

In general, watersheds having low critical loads tend to have low streamwater ANC (Table 23). On this basis, much can be inferred about the critical load of the more acid-sensitive watersheds simply by knowing the ANC value. The best predictions of critical load were achieved using just water chemistry data, expressed as the ratio of stream ANC to stream SO_4^{2-}

Table 21. List of variables used in empirical models to estimate critical load from landscape characteristics.

Code	Candidate Predictor Variable	Unit
Watershed Variables		
A	Elevation	m
B	Watershed area	ha
C	High mountains ecoregion	%
D	Southern crystalline ridges and mountains ecoregion	%
E	Southern metasedimentary mountains ecoregion	%
F	Coniferous forest vegetation	%
G	Hardwood forest vegetation	%
H	Biotite gneiss lithology	%
I	Meta-argillite lithology	%
J	Meta-sedimentary lithology	%
K	Quartzite lithology	%
L	Sandstone lithology	%
M	Oak hickory forest	%
N	White-red-jack pine	%
Stream Chemistry Variables		
O	Stream ANC	$\mu\text{eq/L}$
P	Stream SO_4^{2-} concentration	$\mu\text{eq/L}$

Table 22. Selected multiple regression models to predict critical load for the year 2040 from either watershed variables only, or from a combination of stream chemistry and watershed variables.

Best Model	r²	Model Variables¹	Equation
To predict load needed to reach ANC = 20 µeq/L endpoint, landscape variables only			
Two variables	0.37	D, F	CL = 95.15 + 5.7668(D) + 3.9121(F)
Three variables	0.41	A, D, K	CL = 535.52 - 0.3582(A) + 5.0521(D) - 3.2330(K)
Four variables	0.47	D, E, G, K	CL = 330.22 + 6.2648(D) + 2.7050(E) - 3.7252(G) - 3.8293(K)
Five variables	0.50	A, B, D, G, K	CL = 862.18 - 0.4058(A) - 0.2253(B) + 4.5895(D) - 2.7527(G) - 4.3115(K)
To predict load needed to reach ANC = 0 µeq/L endpoint, landscape variables only			
Two variables	0.44	D, K	CL = 302.80 + 6.6196(D) - 2.8255(K)
Three variables	0.57	D, E, K	CL = 185.04 + 7.9817(D) + 2.5885(E) - 3.8179(K)
Four variables	0.59	D, E, K, M	CL = 355.25 + 7.9899(D) + 3.4423(E) - 4.8087(K) - 3.0525(M)
Five variables	0.61	B, D, E, K, M	CL = 405.26 - 0.2246(B) + 8.1086(D) + 3.2424(E) - 4.5866(K) - 2.8610(M)
To predict load needed to reach ANC = 20 µeq/L endpoint, water chemistry plus landscape variables			
Two variables	0.64	D, O	CL = -55.80 + 4.1705(D) + 9.1197(O)
Three variables	0.68	B, D, O	CL = 6.5892 - 0.2559(B) + 4.3496(D) + 9.2924(O)
Four variables	0.70	B, D, E, O	CL = -67.16 - 0.2449(B) + 5.1709(D) + 1.3372(E) + 9.1800(O)
Five variables	0.73	B, C, D, E, O	CL = -314.69 - 0.2789(B) + 3.2401(C) + 7.1231(D) + 3.6340(E) + 10.4186(O)
To predict load needed to reach ANC = 0 µeq/L endpoint, water chemistry plus landscape variables			
Two variables	0.62	D, O	CL = 59.09 + 5.3988(D) + 9.4971(O)
Three variables	0.66	B, D, O	CL = 134.71 - 0.3101(B) + 5.6157(D) + 9.5974(O)
Four variables	0.68	B, C, D, O	CL = 108.46 - 0.3216(B) + 0.7036(C) + 5.7334(D) + 9.8747(O)
Five variables	0.74	B, C, D, E, O	CL = -313.07 - 0.3489(B) + 4.8178(C) + 9.4063(D) + 4.8607(E) + 11.4149(O)

¹ See list of model variables in Table 21.

Table 23. Distribution of sites by measured current ANC value within modeled critical load categories.¹

Critical Load (kg S/ha/yr)	Number of Modeled Sites	Distribution of Sites by Current ANC ($\mu\text{eq/L}$) Class ² (Number of Sites)				
		< 0	0-20	20-50	50-100	> 100
Target Year 2040						
< 0	17	5	12			
0-2	4		2	2		
2-4	2			2		
4-8	1			1		
8-12	4		1	3		
>12	38			23	14	1
Target Year 2100						
< 0	15	4	11			
0-2	2	1	1			
2-4	6	1	2	3		
4-8	4		1	4		
8-12	14			8	5	
> 12	25			15	9	1

¹ Based on critical load to achieve stream ANC = 20 $\mu\text{eq/L}$ in the designated target year

² Current ANC based on MAGIC model simulations for 2005

concentration (Figure 14). For critical ANC values of 0, 20, and 50 $\mu\text{eq/L}$, the r^2 values ranged from 0.78 to 0.92, depending on the critical ANC value and endpoint year.

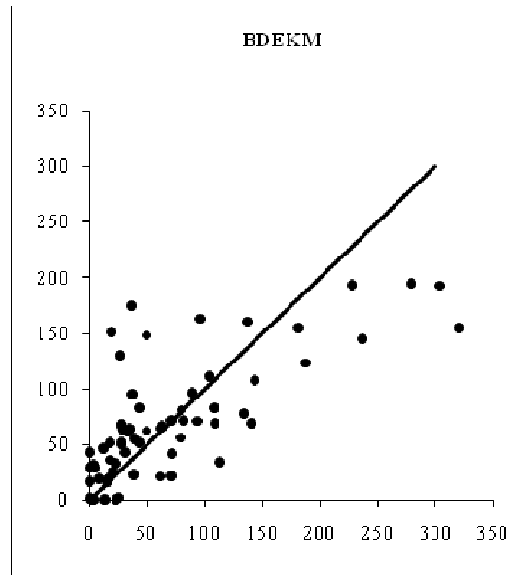
The significant water chemistry predictor variable was streamwater ANC. The landscape characteristics that were most commonly significant predictor variables in the regression models were:

- Southern Crystalline Ridges and Mountains ecoregion
- elevation
- quartzite lithology
- Southern Metasedimentary Mountains ecoregion
- oak-hickory forest
- hardwood forest

Five-variable predictive relationships explained about half or more of the variation in critical load. In Figure 16, predicted critical loads using these empirical relations are compared with MAGIC model simulated critical loads. Agreement is better for the models that used water chemistry (stream ANC) in addition to watershed variables in order to predict critical load (Figure 17).

ANC 0 Endpoint, Landscape Variables Only

A)



ANC 20 Endpoint, Landscape Variables Only

B)

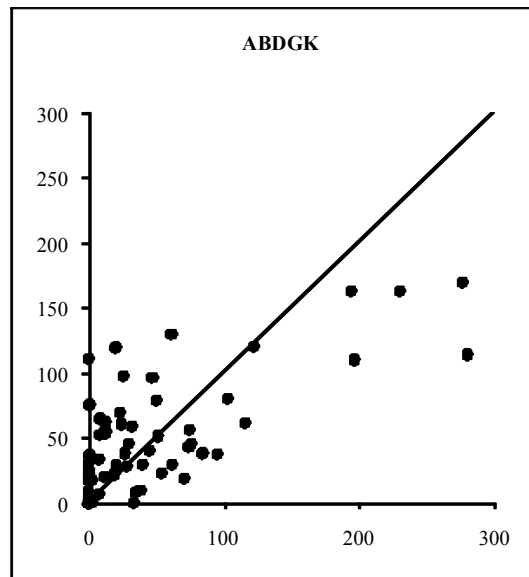
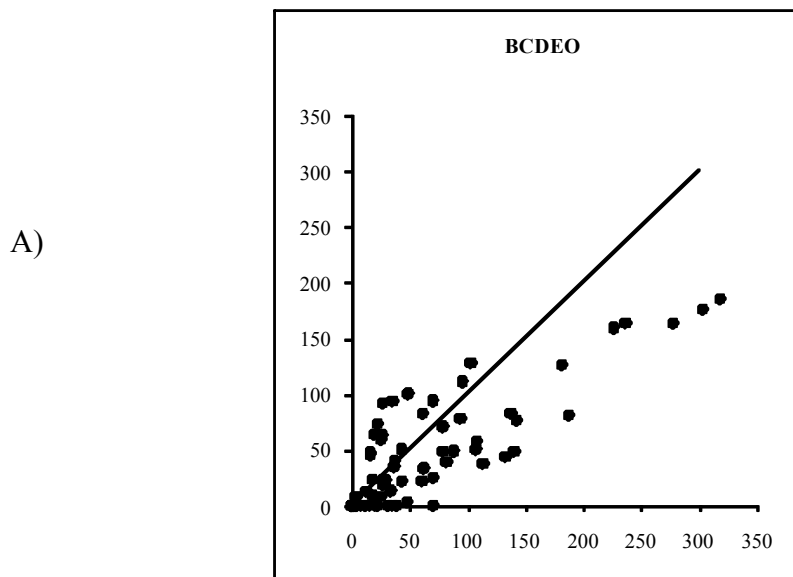


Figure 16. Predicted critical load (kg S/ha/yr), using five-variable empirical models based on landscape variables, versus MAGIC model estimates of critical loads. The critical loads estimates are shown for the ANC critical level of 0 in Panel A and 20 $\mu\text{eq/L}$ in Panel B. The r^2 values were 0.61 for panel A and 0.50 for panel B; $n=66$.

ANC 0 Endpoint, Landscape Variables Plus Water Chemistry



ANC 20 Endpoint, Landscape Variables Plus Water Chemistry

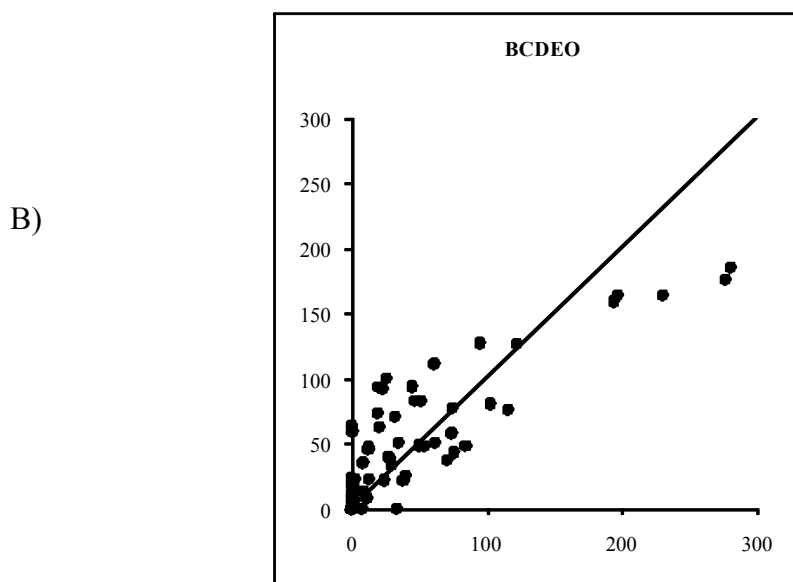


Figure 17. Predicted critical load (kg S/ha/yr), using five-variable empirical models based on both landscape variables and water chemistry, versus MAGIC model simulated critical loads for the ANC critical level of 0 in Panel A and 20 $\mu\text{eq/L}$ in Panel B. The r^2 values were 0.74 for panel A and 0.73 for panel B; $n=66$.

A linear regression was also developed to predict stream ANC from the landscape variables, using the Best Subsets Regression procedure. Elevation, the oak-hickory forest type, and the quartzite lithologic class showed statistically significant associations with stream ANC ($p < 0.001$, $r^2=0.57$). All three explanatory variables exhibited negative correlation with ANC. The resulting model was:

$$\text{ANC} = 98.6 - 0.0515(\text{elev}) - 0.2620(\text{oak-hickory}) - 0.5937(\text{quartzite})$$

Thus, a number of approaches can be used to estimate critical load or stream ANC in the absence of model simulations or measurements. For critical load estimation, the best predictor was stream ANC divided by stream SO_4^{2-} concentration. In situations where measurements of stream chemistry are not available, stream ANC or critical load can be estimated using only landscape variables, but these approaches only explain about 50% to 60% of the variability at best.

G. Regionalization of Modeling Results

1. Statistical Frame

The Forest Service land that is the subject of analyses for this report includes portions of three states. There has not been a statistically-based survey of streamwater chemistry confined to this area. However, the National Stream Survey (NSS), which was statistically based, covered the region of concern for this report. One of the subregions included in the NSS was the Southern Blue Ridge Province, and this province contains the watersheds modeled for this report.

The NSS was a randomized systematic sample of 500 stream reaches designed to estimate the characteristics of a target population of 64,300 stream reaches in acid-sensitive subregions of the eastern United States. The sampling unit in the NSS was stream reach, defined as a blue-line headwater segment or a segment between two confluences on U.S. Geological Survey 1:250,000-scale topographic maps. Each sampled reach was assigned a weight (inversely proportional to its selection probability) equivalent to the number of reaches it represented in the map population (see Kaufmann et al. [1991] for a detailed description of the statistical design). During site selection, stream reaches with watershed areas greater than 155 km² and reaches within mapped urban areas were excluded from the NSS target population. Streamwater chemical data were collected in the spring, between March 15 and May 15 of 1986 (1985 in the Southern Blue Ridge Pilot Survey subregion). Water samples were taken from both the

upstream end and downstream end of each randomly selected stream segment (as shown on the 1:250,000 scale maps) and they are designated as the "upper node" and the "lower node". Samples were not collected during, or for 24 hr after, precipitation events in order to minimize possible storm influences.

The boundary of the Southern Blue Ridge Province encompasses a large area of western North Carolina, along with smaller portions of eastern Tennessee, western South Carolina, northern Georgia, and southern Virginia (Figure 18). All of the acid-sensitive streams located on Forest Service lands that are included in this assessment occur within the boundaries of the Southern Blue Ridge Province. The National Stream Survey (NSS) target population in this region was 2,031 stream reaches, with an overall length of 9,036 km. The NSS survey design included sampling each statistically-selected stream at two locations, the upstream node and the downstream node (Kaufmann et al. 1991; Appendix E). The NSS was conducted during spring baseflow in order to represent chronic chemistry. Because the NSS used a probability design, it provides the best available picture of the regional status and extent of chronic acid-base stream chemistry in the Southern Blue Ridge (Baker et al. 1990a, Sullivan et al. 2002a). The target population did not include lower-order tributary reaches that are not represented as blue lines on 1:250,000-scale maps. However, more recent stream survey work by the Forest Service does include many of these smaller, and sometimes more highly acidic, streams.

Results of the NSS in the Southern Blue Ridge Province indicated that only 6% of upper node sampling sites that were sampled (4 of 67 sampled streams) had $ANC \leq 50 \mu\text{eq/L}$ and none were chronically acidic (Kaufmann et al. 1988, Elwood et al. 1991). All sampled streams had $\text{pH} > 6.0$.

NSS stream length interpolations suggested that 7.8% of the stream length in the Southern Blue Ridge Province had ANC in 1985-86 less than $50 \mu\text{eq/L}$. This equates to 706 km of stream. Results of the Eastern lakes Survey (ELS) in this province were similar. Only 3.9% of the surveyed lakes and reservoirs had $ANC < 50 \mu\text{eq/L}$, and none were chronically acidic (Landers et al. 1988).

The median ANC of the Southern Blue Ridge upper node stream population was $100 \mu\text{eq/L}$, and 25% of the stream population had $ANC < 75 \mu\text{eq/L}$. The median pH was 6.98, and 25% of the stream population had $\text{pH} < 6.8$ (Table 24). DOC concentrations were very low, with 75% of the stream population having DOC below $58 \mu\text{M/L}$.

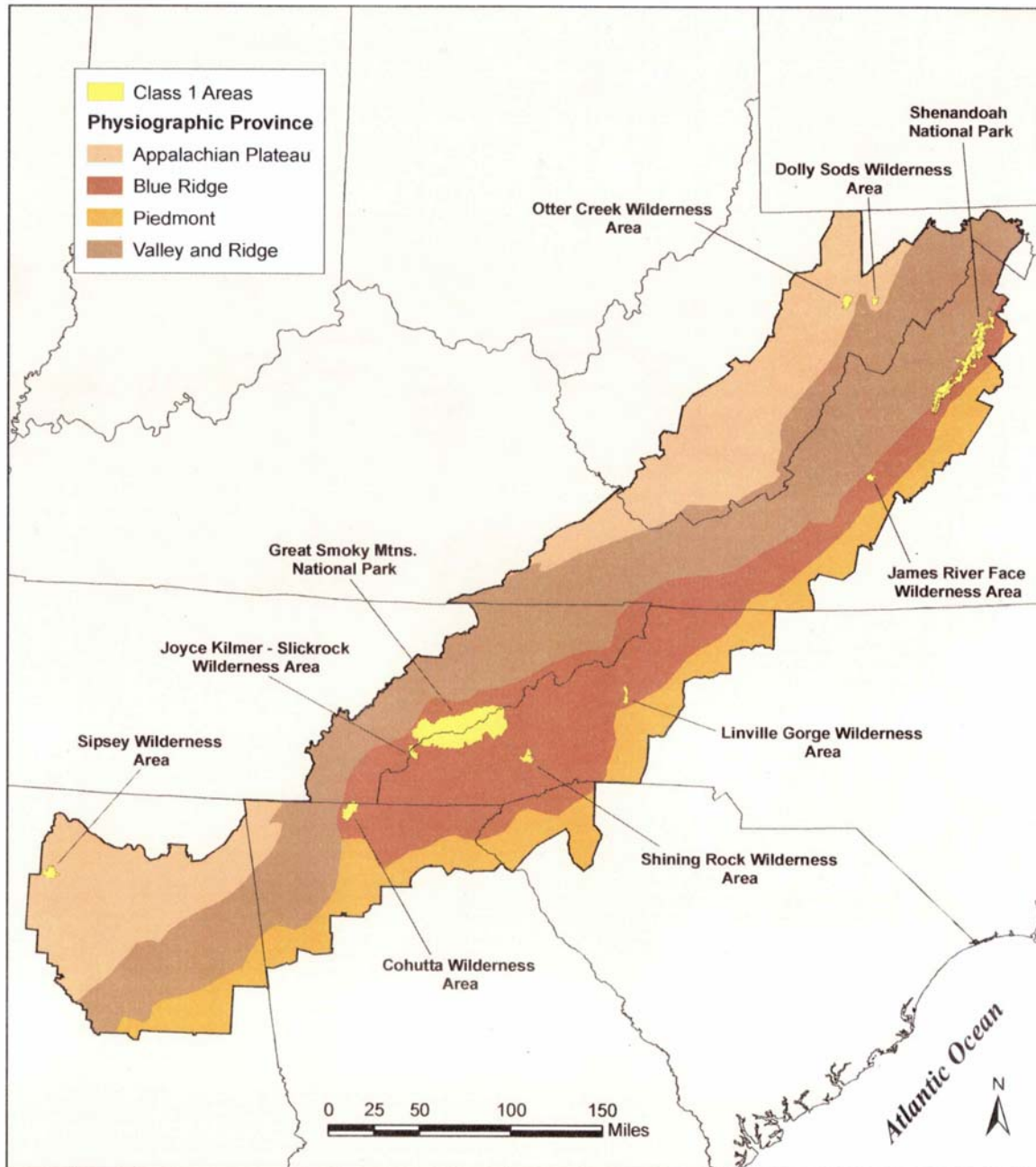


Figure 18. Map of SAMI study area showing location of Class I areas and physiographic provinces. The study reported here was confined to the Southern Blue Ridge province. (Source: Sullivan et al. 2002a).

Table 24. Population statistics for the NSS upper node stream population in the Southern Blue Ridge.

Statistic	Variable ¹						
	ANC	pH	SO ₄ ²⁻	NO ₃ ⁻	Cl ⁻	Ca ²⁺	DOC
Median	100	6.98	20.7	6.9	20.7	44.7	48.0
25 th percentile	75	6.80	13.1	1.7	15.8	32.0	42.6
75 percentile	141	7.06	33.1	15.8	23.7	81.5	58.2

¹ units are µeq/L for all except pH (standard units) and DOC (µmol/L)

For this analysis, we recalculated NSS population statistics for the portion of the Southern Blue Ridge Province that is the subject of this report. We defined the Southern Blue Ridge study area as the portion of the Omernik Level III ecoregion number 66 (Blue Ridge Mountains) that is located in the states of North Carolina, South Carolina, and Tennessee. Based on our analysis of the NSS data, there are 1,408 upper nodes and 1,404 lower nodes in the Southern Blue Ridge study area with an estimated stream length of 7,430 km.

NSS upper node stream sampling locations were mainly first through third order streams, with the largest number being first order streams if plotted on a 1:24,000 scale map. In contrast, the lower node stream sampling sites were largely third order streams located at lower elevation (Figures 19, 20). Population statistics for the upper and lower node sites are presented in Table 25. Median ANC values were 100 and 114 µeq/L, respectively. Streamwater SO₄²⁻

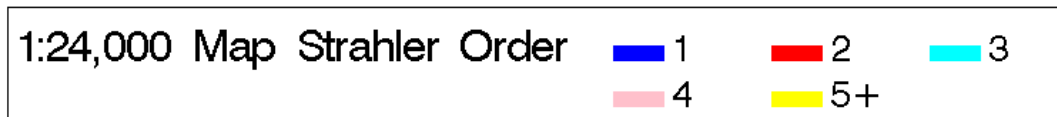
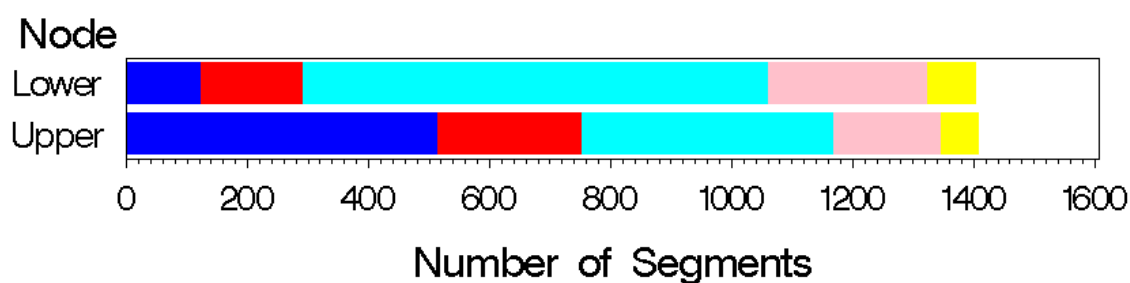


Figure 19. Estimated number of stream segments by 7.5'' (1:24,000 scale) topographic map Strahler order in the portion of the Southern Blue Ridge Province located in NC, TN, and SC, based on National Stream Survey Data.

Southern Blue Ridge Population Estimates – NSS Data

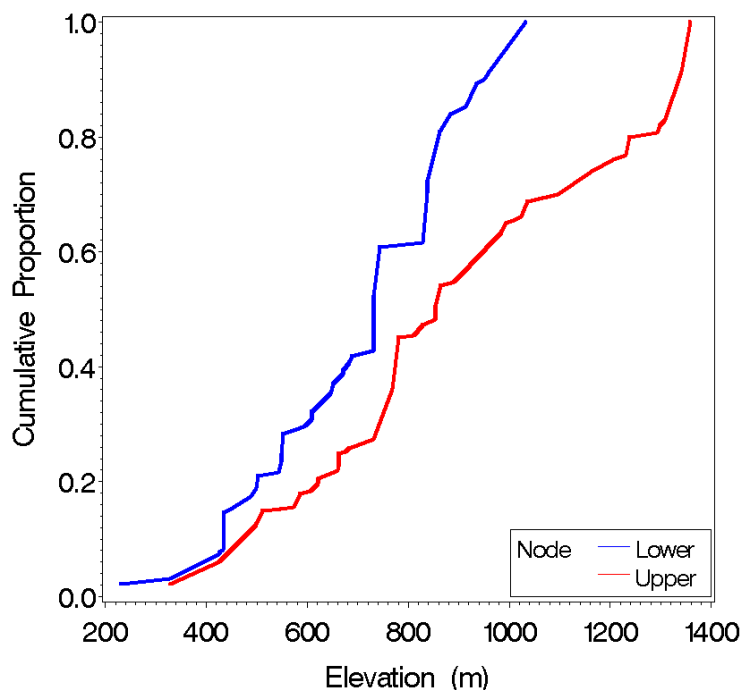


Figure 20. Cumulative distribution function of sample site elevation for the population of NSS streams located in the Southern Blue Ridge Province in NC, TN, and SC.

concentrations were generally fairly low. An estimated three-fourths of the upper node streams had SO_4^{2-} concentrations below $60 \mu\text{eq/L}$. Nitrate concentrations were about one third of the SO_4^{2-} concentrations. Lower node SO_4^{2-} and NO_3^- concentrations were generally similar to upper node concentrations, although ANC and pH tended to be lower at the upper nodes (Figure 21). Comparative statistics for the modeled streams are shown in Table 8 for comparison.

2. Watersheds Modeled for this Study

It is likely that many upper tributary stream reaches located upstream from the blue line streams (those on the USGS maps that constitute the frame population) have lower ANC than those in the NSS frame population. This expectation is based on the finding that ANC typically increases from upstream to downstream (Kaufmann et al. 1988, Elwood et al. 1991). It is therefore likely that the lowest ANC streams are upstream from the blue-line reaches or in the upper tributaries that flow into the upper blue-line reaches, and therefore outside of the NSS statistical frame. For example, a non-statistical sampling was conducted by Winger et al. (1987)

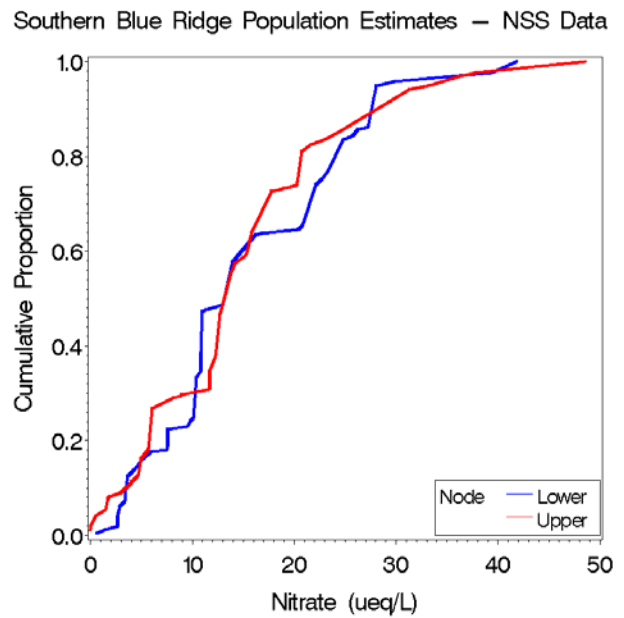
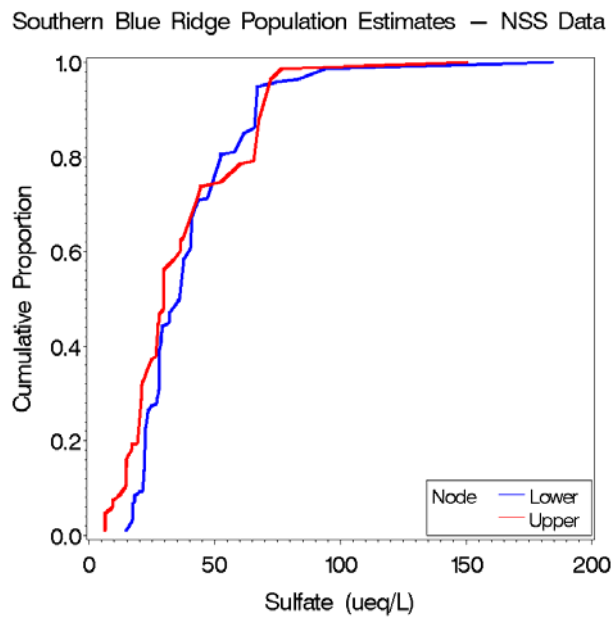
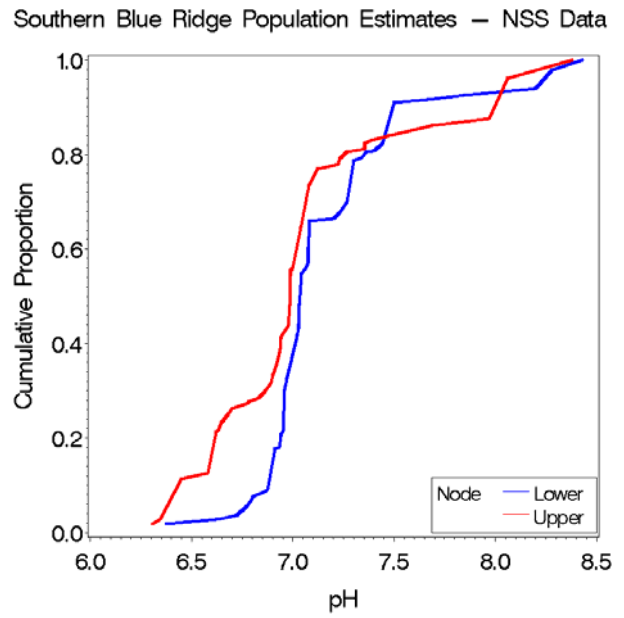
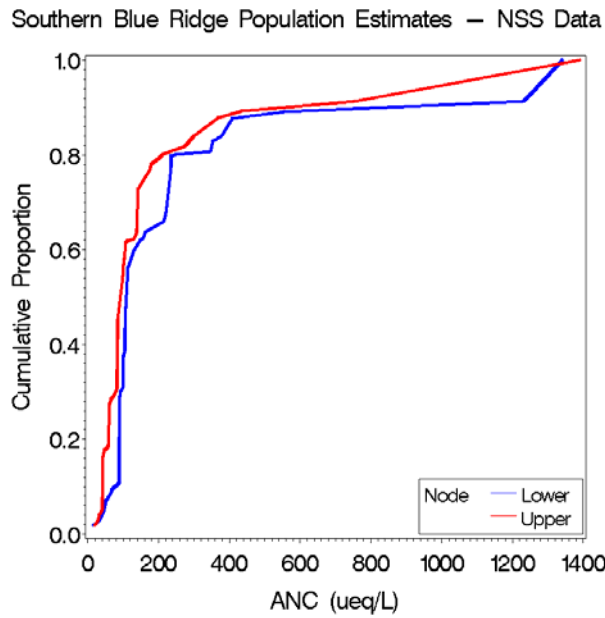


Figure 21. Cumulative distribution functions for upper and lower node NSS streams in the portion of the Southern Blue Ridge Province located in NC, TN, and SC.

Table 25. Population estimates of stream segment condition in the portion of the Southern Blue Ridge Province that occurs within NC, TN, and SC from NSS data¹.

Parameter	Min	25 th	Median	75 th	Max	Mean	SD
Upper Nodes							
Watershed Area (km ²)	0.1	0.3	1.4	11.1	85.0	12.3	21.1
ANC (µeq/L)	20.9	60.8	99.5	159.8	1,390.3	240.1	374.6
Elevation (m)	329.2	676.6	853.4	1,207.0	1,359.3	910.3	302.7
Nitrate (µeq/L)	0.0	6.0	13.8	20.7	48.6	15.3	10.3
pH	6.3	6.7	7.0	7.1	8.4	7.1	0.5
Sulfate (µeq/L)	6.2	20.8	29.8	60.4	150.3	37.8	24.7
Lower Nodes							
Watershed Area (km ²)	2.1	7.6	14.0	29.9	138.9	22.7	23.4
ANC (µeq/L)	16.5	90.2	114.1	235.9	1,341.2	278.6	376.1
Elevation (m)	231.6	551.7	731.5	862.5	1,033.2	712.1	197.1
Nitrate (µeq/L)	0.7	10.4	13.9	22.7	41.8	15.7	9.5
pH	6.4	7.0	7.0	7.3	8.4	7.2	0.4
Sulfate (µeq/L)	14.7	23.2	37.7	52.3	184.4	40.9	24.7

¹ Comparable statistics for the streams modeled for this report are given in Table 8.

of first- and third-order reaches of streams in eastern Tennessee, western North Carolina, and northern Georgia. All sampled streams were located within the Southern Blue Ridge Province. Samples were collected near baseflow during the period 1982-1984. They found that 3% of the sampled first-order streams, and none of the third-order streams, were acidic. These results could not, however, be directly extrapolated to the population of streams because the selection process was not random. An additional consideration was that the two acidic streams that were sampled by Winger et al. (1987) drained watersheds containing a pyritic phyllite of the Anakeesta formation, which is a potential source of geological SO_4^{2-} in streamwater. The concentration of SO_4^{2-} in streamwater draining these two watersheds was eight to nine times higher than that of the streams draining watersheds without an apparent bedrock source of SO_4^{2-} (Winger et al. 1987). Nitrate was also two to three times higher in the streams affected by the Anakeesta formation (Elwood et al. 1991).

Recent Forest Service surveys of 256 streams within the study region have found that acidic and low-ANC streams are much more prevalent than was represented by NSS for the Southern Blue Ridge Province. These surveys were not statistically based, but do show widespread occurrence of streams having low ANC and pH (see Figure 22). In general, these

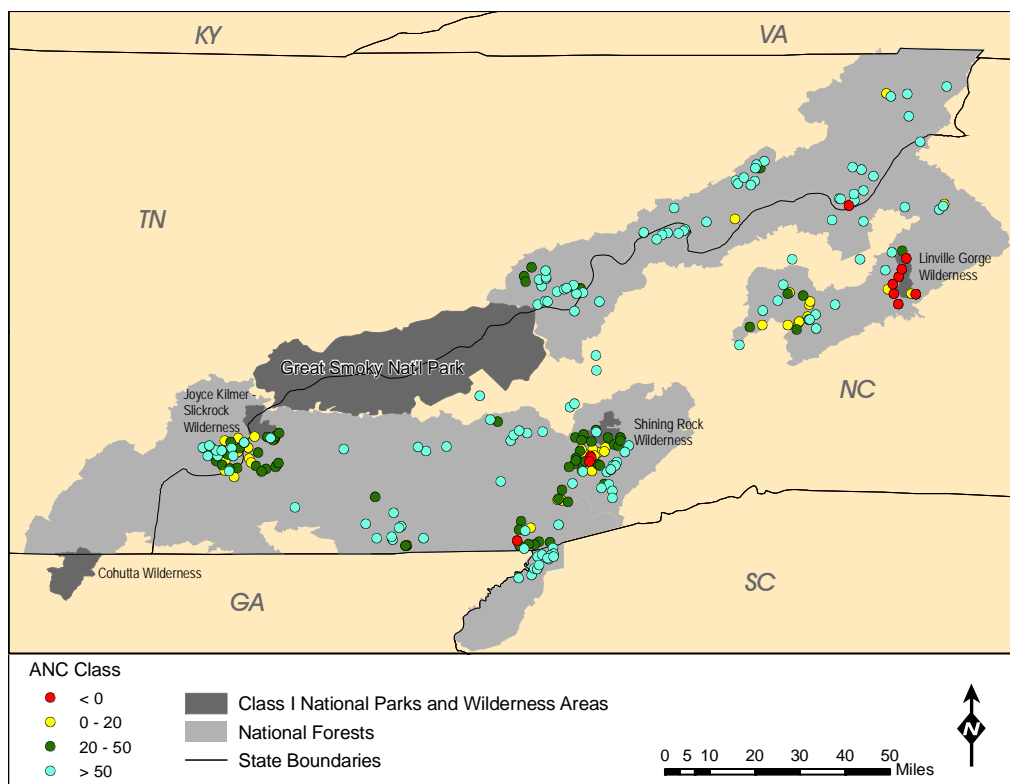


Figure 22. Map showing locations of streams surveyed by the USDA Forest Service for acid-base chemistry. Acidic and low-ANC streams are widely distributed on Forest Service lands, especially in Linville Gorge Wilderness.

recently surveyed streams were located at higher elevation than the NSS streams. About 25% of the population of upper node NSS stream reaches in the Southern Blue Ridge Province within NC, TN, and SC were located at an elevation above 1,200 m. NSS watersheds were generally small, with 75% of the upper node sites having watersheds smaller than 11.1 km² (Table 25).

Streams recently sampled by the Forest Service had smaller watersheds. These recently surveyed stream sites commonly had low ANC and pH (Table 26). Overall, 5% of the surveyed streams were chronically acidic, and 49% had ANC ≤ 50 µeq/L (Table 26). Eleven percent had pH < 6. Surveyed streams that were acidic (ANC ≤ 0) or low in ANC (< 50 µeq/L) were located within the acid-sensitive areas within the Southern Appalachian Mountains that were mapped by Sullivan et al. (2002a) on the basis of bedrock geology and elevation (Figure 23).

Sulfate concentrations in NSS streams in the study region were low, with a median value of 30 µeq/L and 75th percentile of 60 µeq/L at the upper nodes (Table 25). Some surface waters in the region have much higher concentration of SO₄²⁻. For example, 4% of the lakes sampled by the ELS had lakewater SO₄²⁻ concentrations higher than 115 µeq/L. Elwood et al. (1991) interpreted this finding as suggestive of watershed sources of SO₄²⁻.

Table 26. Prevalence of acidic, low-ANC, and low-pH streams on national forest lands included in recent surveys of stream chemistry of 256 stream reaches located in western North Carolina, eastern Tennessee, and South Carolina.

ANC/pH Category	Percent of Stream Reaches Surveyed Having Average Value Below Criterion
ANC < 0	5
ANC 0-20	15
ANC 20-50	29
pH < 5	2
pH 5-5.5	3
pH 5.5-6.0	5

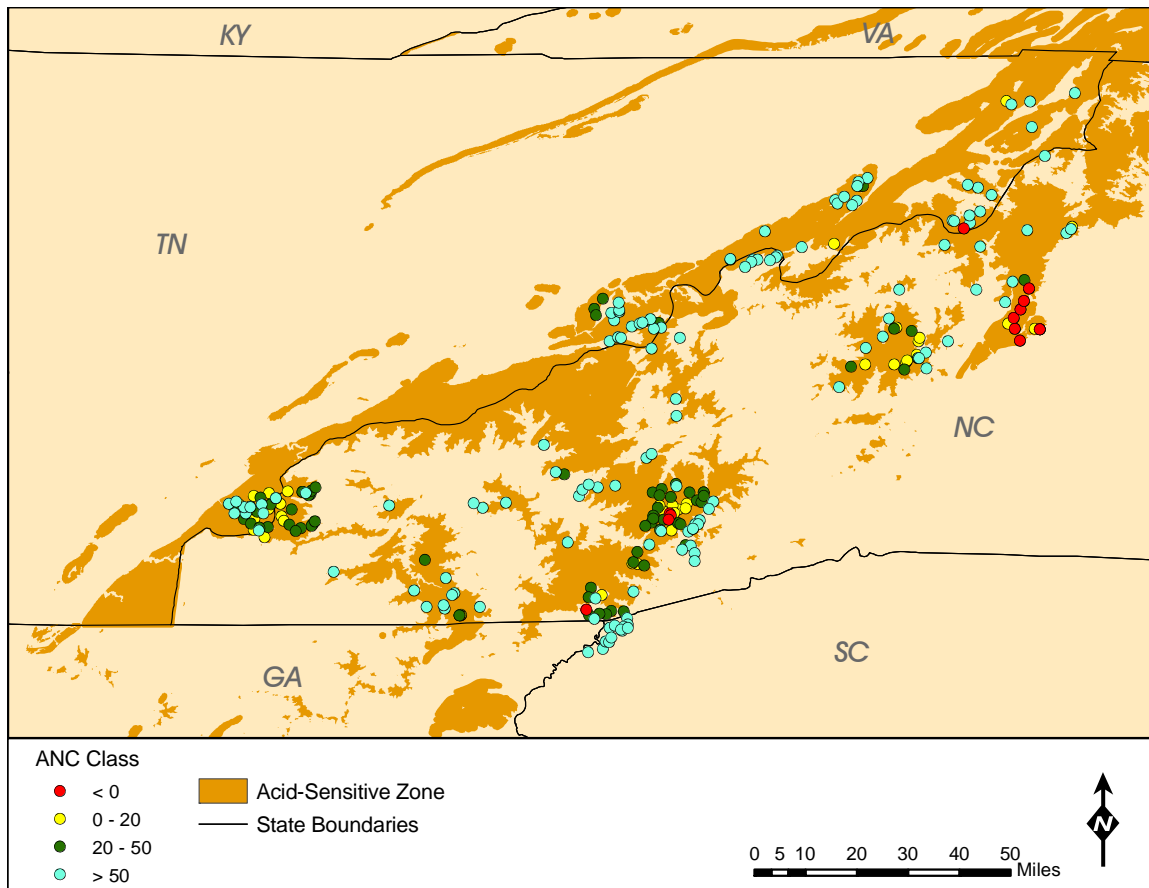


Figure 23. Locations of streams recently surveyed by the USDA Forest Service, showing stream ANC class (color coded circles) in relation to the acid-sensitive geologic/elevational zone described by Sullivan et al. (2002b).

H. Episodic Variability in Stream Chemistry

1. Changes in ANC During Storm Events

Values of annual average or spring season baseflow water chemistry are typically used to represent conditions at a given stream for purposes of characterization. However, streamwater chemistry undergoes substantial temporal variability, especially in association with hydrological episodes. During such episodes, which are driven by rainstorms and/or snowmelt events, both discharge (streamflow volume per unit time) and water chemistry change, sometimes dramatically. This is important because streams may in some cases exhibit chronic chemistry that is suitable for aquatic biota, but experience occasional episodic acidification with lethal consequences (c.f., Wigington et al. 1993). Model projections of future streamwater chemistry response to acidic deposition are typically based on chronic chemistry. When interpreting model projections of chronic chemistry, it is important to also consider the likelihood of episodic excursions of water chemistry that are more acidic than is found during more typical baseflow periods. In order to do this, it is important to know something about the magnitude of typical episodic acidification, expressed as loss of ANC, that occurs during rainstorms and/or snowmelt within the region of interest.

Data regarding episodic variability in streamwater ANC are not widely available within the study area for this report. However, some useful data are available for some acid-sensitive streams. Some of the best available data were collected in Shenandoah National Park, Virginia. Sullivan et al. (2003) presented data for six intensively-studied sites within the park for the period 1993 to 1999 (Figure 24). The minimum measured ANC each year at each site (which generally is recorded during a large hydrological episode) is plotted against the median spring ANC for that year at that site. Sites that exhibited median spring ANC below about 20 $\mu\text{eq/L}$ (Paine Run, White Oak Run, Deep Run) generally had minimum measured ANC about 10 $\mu\text{eq/L}$ lower than median spring ANC. In contrast, at the high-ANC Piney River site (median spring ANC > 150 $\mu\text{eq/L}$), the minimum measured ANC was generally more than about 40 $\mu\text{eq/L}$ lower than the respective median spring ANC. At sites having intermediate ANC values, with median spring ANC in the range of about 30 to 90 $\mu\text{eq/L}$, the minimum ANC measured each year was generally about 20 to 30 $\mu\text{eq/L}$ lower than the respective median spring ANC. Thus, there is a rather clear pattern of larger episodic ANC depressions in streams having higher median ANC and smaller episodic ANC depressions in streams having lower median ANC. This pattern has been reported previously for streams in Shenandoah National Park by Eshleman (1988) and Hyer

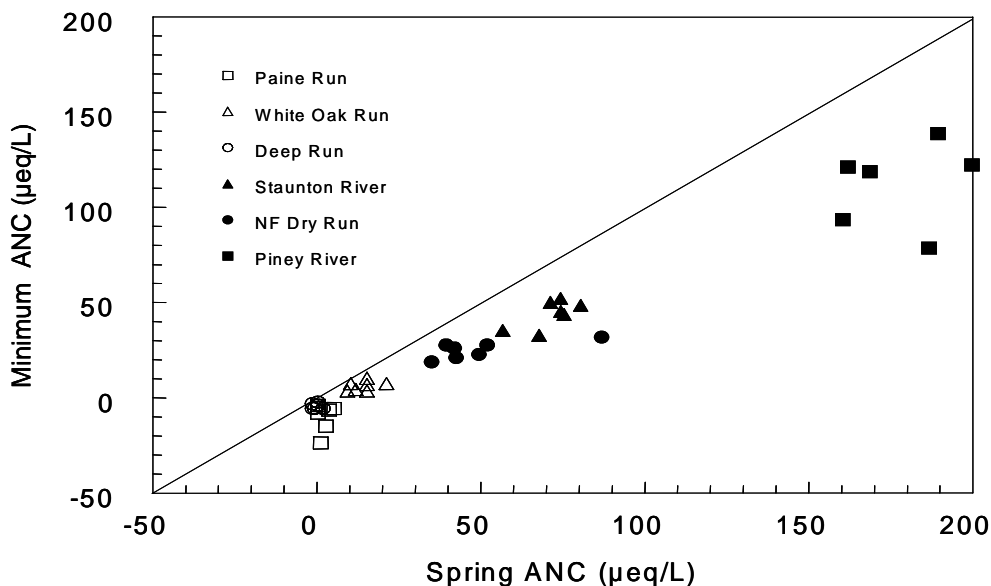


Figure 24. Minimum streamwater ANC sampled at each site during each year versus median spring ANC for all samples collected at that site during that spring season. Data are provided for all intensively-studied streams within Shenandoah National Park during the period 1993-1999. A 1:1 line is provided for reference. The vertical distance from each sample point upwards to the 1:1 line indicates the ANC difference between the median spring value and the lowest sample value for each site and year. (Source: Sullivan et al. 2003)

et al. (1995). The two sites that had median spring ANC between about 0 and 10 µeq/L consistently showed minimum measured values below 0. Streams having low (near zero) chronic ANC can be expected to experience relatively small episodic ANC depressions. However, those depressions often result in minimum ANC values that are associated with toxicity to aquatic biota.

In general, pre-episode ANC is a good predictor of minimum episodic ANC and also a reasonable predictor of episodic Δ ANC. Higher values of pre-episode ANC lead to larger Δ ANC values, but minimum ANC values of such streams are generally not especially low. Lowest minimum ANC values are reached in streams that have low pre-episode ANC, but the Δ ANC values for such streams are generally small.

Webb et al. (1994) developed an approach to calibration of an episodic acidification model for VTSSS long-term monitoring streams in western Virginia that was based on the regression method described by Eshleman (1988). Median, spring quarter ANC concentrations for the period 1988 to 1993 were used to represent chronic ANC, from which episodic ANC was predicted. Regression results were very similar for the four lowest ANC watershed classes, and

they were therefore combined to yield a single regression model to predict the minimum measured ANC from the chronic ANC. Extreme ANC values were about 20% lower than chronic values, based on the regression equation:

$$\text{ANC}_{\text{min}} = 0.79 \text{ANC}_{\text{chronic}} - 5.88 \quad (r^2=0.97; \text{se of slope}=0.02, p \leq 0.001)$$

Because the model was based on estimation of the minimum ANC measured in the quarterly sampling program, it is likely that the true minimum ANC values were actually somewhat lower than 20% below the measured chronic ANC. Nevertheless, regression approaches for estimation of the minimum episodic ANC of surface waters, such as was employed by Webb et al. (1994) for western Virginia, provide a basis for predicting future episodic acidification. It must be recognized, however, that future episodic behavior might vary from current behavior if chronic conditions change dramatically.

Webb (2003) reported variation in streamwater ANC as a function of runoff (in mm/hr) for three streams in Virginia situated on different types of bedrock. The stream located on siliceous bedrock (Paine Run) showed ANC generally varying from about $-5 \mu\text{eq/L}$ at runoff values near 1 mm/hr to over $10 \mu\text{eq/L}$ at runoff values near 0.005 mm/hr.

Based on an analysis of 105 stormflow/baseflow sample pairs for White Oak Run during the period 1980-1992, about 15% of the episodes exhibited episodic ΔANC more than about $20 \mu\text{eq/L}$ (Eshleman et al. 1995). The mean ΔANC was $-12.4 \mu\text{eq/L}$. Baseflow ANC in this stream varies from about 20 to $60 \mu\text{eq/L}$ (Sullivan et al. 2003).

Cook et al. (1994) found that ANC in acid-sensitive high-elevation streams in the Great Smoky Mountains decreased by as much as $15 \mu\text{eq/L}$ during storm events. Baseflow ANC values for the eight study streams ranged from $-31 \mu\text{eq/L}$ to $28 \mu\text{eq/L}$.

O'Brien et al. (1993), in a study of five mid-Atlantic stream watersheds, found that the streams on less reactive bedrock (lowest baseflow stream ANC) exhibited the smallest changes in ANC during storms, but were much more likely to lose all ANC during a given storm. The study streams having baseflow ANC near 20 to $40 \mu\text{eq/L}$ (Fishing Creek Tributary, MD and Reedy Creek, VA) showed episodic ANC depressions of about 15 to $25 \mu\text{eq/L}$. The stream that was chronically acidic (Mill Run, VA) showed very small ($< 10 \mu\text{eq/L}$) episodic ANC depression (O'Brien et al. 1993) during storms.

Elwood et al. (1985) sampled several sites in the Walker Camp Prong drainage in GSMNP during both baseflow and stormflow. As expected, ANC and pH were lower during stormflow, whereas inorganic monomeric Al was higher. Decreases in pH and ANC were generally greater at the downstream sites which had higher baseflow ANC. At the higher elevation, chronically-acidic stream sites, the observed episodic depression in ANC was generally less than about 10 $\mu\text{eq/L}$. At the lower elevation sites, which exhibited positive ANC, the episodic ANC depression was about 20 $\mu\text{eq/L}$. Episodic increases in Al_i were 2 to 3 $\mu\text{mol/L}$ at the three uppermost sites, but were negligible at the lowest elevation site, which had baseflow pH of 6.2 and stormflow pH of 5.5.

Significant decreases in pH and increases in Al during a major storm were reported for Raven Fork in North Carolina (Jones et al. 1983). Baseflow ANC was 20 $\mu\text{eq/L}$ and pH was 5.7. The stream pH declined to below 5.0 throughout the storm, with minimum pH of 4.3 (Elwood et al. 1991).

Episodic acidification during rainstorms of two small streamlets in the Noland Divide watershed was reported by Nodvin et al. (1995). They showed episodic pH depressions of one-half to one pH unit, to minimum values near pH 5.0. Corresponding episodic ANC depressions were about 5 to 30 $\mu\text{eq/L}$. Episodic stream chemistry data for Upper Creek (0822486357318) on the Pisgah National Forest are presented in Figure 25. Each point represents a sample and the Celo stream gage is down river from the sample site on the main stream. ANC decreased to below zero on two occasions during rainstorms, from baseline values near 10 $\mu\text{eq/L}$.

2. Mechanisms of Episodic Acidification

The routing of water as it flows through a watershed determines the degree of contact with acidifying or neutralizing materials and therefore influences (along with soils and bedrock characteristics) the amount of episodic acidification that occurs. In any given watershed, surface water ANC may vary in time depending upon the proportion of the flow that has contact with deep versus shallow soil horizons; the more subsurface contact, the higher the surface water ANC (Turner et al. 1990). This can be attributed in part to higher base saturation and greater SO_4^{2-} adsorption capacity in subsurface soils. It may also be related to the accumulation in the upper soil horizons of acidic material derived from atmospheric deposition and decay processes (Lynch and Corbett 1989, Turner et al. 1990). Storm flow and snowmelt are often associated with episodes of extreme surface water acidity due to an increase in the proportion of flow

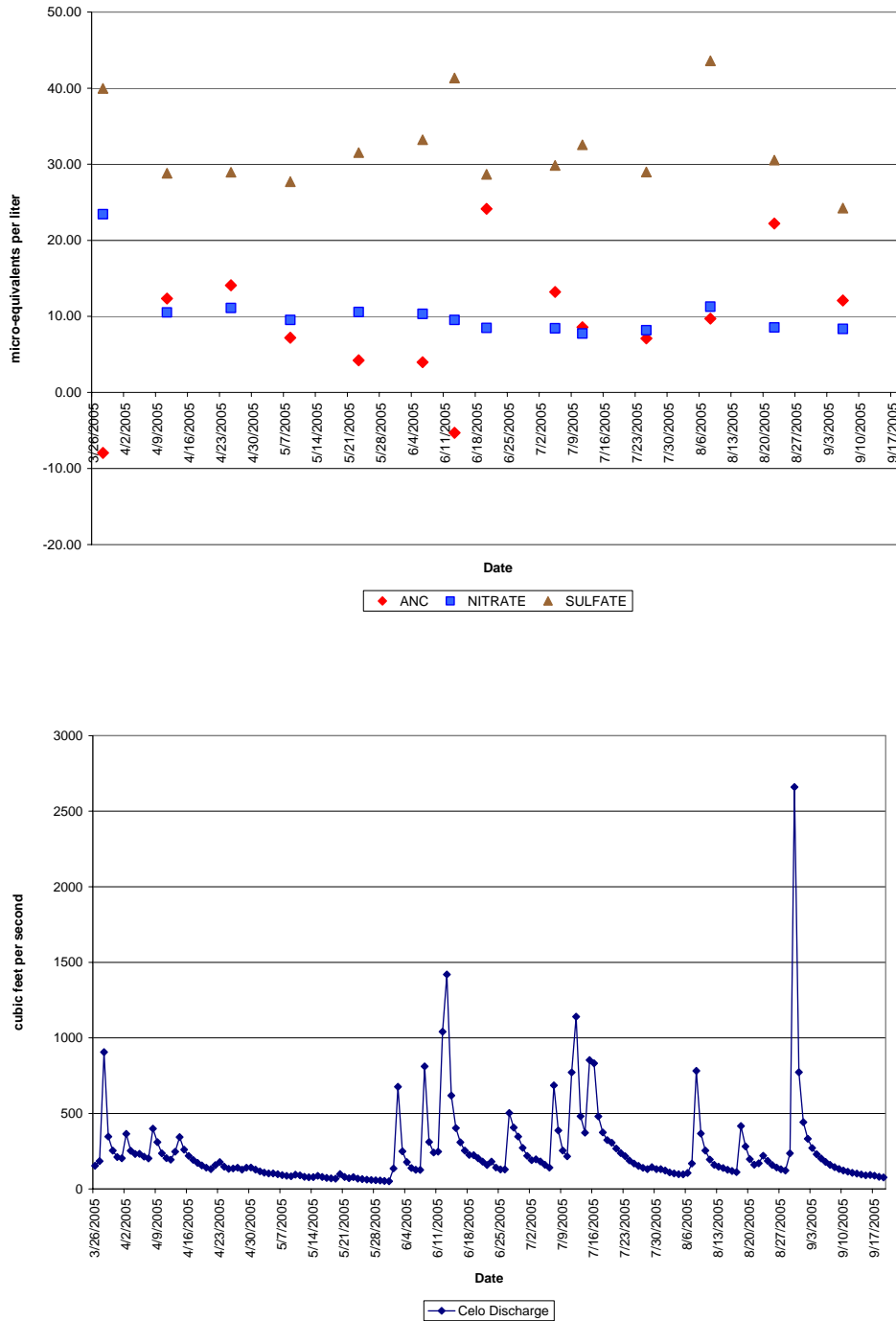


Figure 25. Episodic stream chemistry data for Upper Creek in Pisgah National Forest. The top panel shows the data for ANC, NO₃⁻, and SO₄²⁻; the bottom panel shows the stream discharge.

derived from water that has moved laterally through the surface soil without infiltration to deeper soil horizons (Wigington et al. 1990). Episodic acidification may be the limiting condition for aquatic organisms in southern Appalachian streams that are marginally suitable for aquatic life under baseflow conditions.

Miller-Marshall (1993) analyzed data from the University of Virginia's Surface Water Acidification Study (SWAS) for the period 1988-1991 for White Oak Run, North Fork Dry Run, Deep Run, and Madison Run in Shenandoah National Park, and also conducted a field experiment in 1992 at White Oak Run and North Fork Dry Run. Acid anion flushing was the predominant acidification mechanism during hydrological episodes. Base cation dilution frequently also played a large role, depending on the underlying bedrock geology and baseflow ANC. At the site exhibiting the lowest baseflow ANC (Deep Run), base cations increased during episodes. At the other sites, base cation concentrations were diluted during episodes, with the greatest dilution occurring in the streams that were highest in baseflow ANC (Miller-Marshall 1993).

Streams can lose ANC during a storm due to an increase in anionic solutes, such as SO_4^{2-} , NO_3^- , or organic acid anions. Alternatively, streams can lose ANC as a consequence of base cation dilution, as the increased volume of discharge dilutes the baseflow. Any or all of these ionic changes can be important in a given stream.

There are several different mechanisms of episodic acidification in operation in southeastern streams, depending at least in part on the bedrock geology of the stream. Eshleman and Hyer (2000) estimated the contribution of each major ion to observed episodic ANC depressions in Paine Run, Staunton River, and Piney River during a three-year period. During the study, 33 discrete storm events were sampled and water chemistry values were compared between antecedent baseflow and the point of minimum measured ANC (near peak discharge). The relative contribution of each ion to the ANC depressions was estimated using the method of Molot et al. (1989), which normalized the change in ion concentration by the overall change in ANC during the episode. At the low-ANC (≈ 0) Paine Run site on siliciclastic bedrock, increases in NO_3^- and SO_4^{2-} , and to a lesser extent organic acid anions, were the primary causes of episodic acidification. Base cations tended to compensate for most of the increases in acid anion concentration. ANC declined by 3 to 21 $\mu\text{eq/L}$ (median 7 $\mu\text{eq/L}$) during the episodes studied. At the intermediate-ANC (≈ 60 to 120 $\mu\text{eq/L}$) Staunton River site on granitic bedrock, increases in SO_4^{2-} and organic acid anions, and to a lesser extent NO_3^- , were the primary causes

of episodic acidification. Base cation increases compensated these changes to a large degree, and ANC declined by 2 to 68 $\mu\text{eq/L}$ during the episodes (median decrease in ANC was 21 $\mu\text{eq/L}$). At the high-ANC (\square 150 to 200 $\mu\text{eq/L}$) Piney River site on basaltic (69%) and granitic (31%) bedrock, base cation concentrations declined during episodes (in contrast with the other two sites where base cation concentrations increased). Sulfate and NO_3^- usually increased. The change in ANC during the episodes studied ranged from 9 to 163 $\mu\text{eq/L}$ (median 57 $\mu\text{eq/L}$; Eshleman and Hyer 2000). Changes in base cation concentrations during episodes contributed to the ANC of Paine Run, had little impact in Staunton River, and consumed ANC in Piney River (Hyer 1997).

The relative importance of the major processes that contribute to episodic acidification varies among the streams, in part as a function of bedrock geology and baseflow streamwater ANC. Sulfur-driven acidification was an important contributor to episodic loss of ANC at all three sites, probably because S adsorption by soils occurs to a lesser extent during high-flow periods. This is partly due to diminished contact between drainage water and potentially adsorbing soils surfaces. Dilution of base cation concentrations was most important at the high-ANC site.

Similar conclusions were reached by Miller-Marshall (1993). Acid anion flushing was the predominant acidification mechanism during episodic acidification. Base cation dilution also played a large role for most of the watersheds, but the extent of its importance depended largely on the underlying bedrock.

The importance of NO_3^- to episodic acidification in Shenandoah National Park is a relatively recent development, attributed to the effects of gypsy moth infestation in many watersheds within the park (Webb et al. 1995, Eshleman et al. 1999). Consumption of foliage by the moth larvae converted foliar N, which is normally tied up in long-term N cycling processes, into more labile N forms on the forest floor. Nitrate has more commonly been associated with both chronic and episodic acidification of higher elevation streams in the Great Smoky Mountains.

Eshleman et al. (1999) concluded that episodic acidification of streams in Shenandoah National Park is controlled by a complex set of natural, anthropogenic, and disturbance factors that together produce a transient response that varies dramatically from watershed to watershed. They further hypothesized that the results of recent studies in the park can be largely explained

by a biogeochemical response to forest disturbance by gypsy moth larvae, which temporarily overwhelmed the normal controls on N and base cation dynamics.

The most acidic stream conditions generally occur during high-flow periods, in conjunction with storm or snowmelt runoff. The general relationship between flow level and ANC is evident in Figure 26, which plots ANC measurements against flow for three intensively-studied streams representing different bedrock types. The response of all three streams is similar in that most of the lower ANC values occur in the upper range of flows levels. However, consistent with observations by Eshleman (1988), the minimum ANC values that occur in

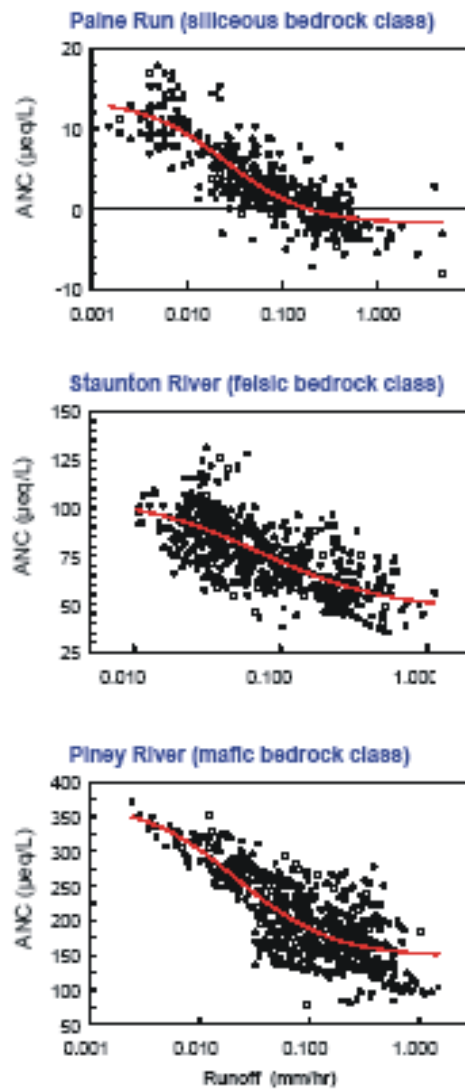


Figure 26. Relationship between ANC and runoff for streamwater samples collected at intensively-studied sites in Shenandoah National Park. The data represent samples collected during the 1992-1997 period. (Source: Sullivan et al. 2003)

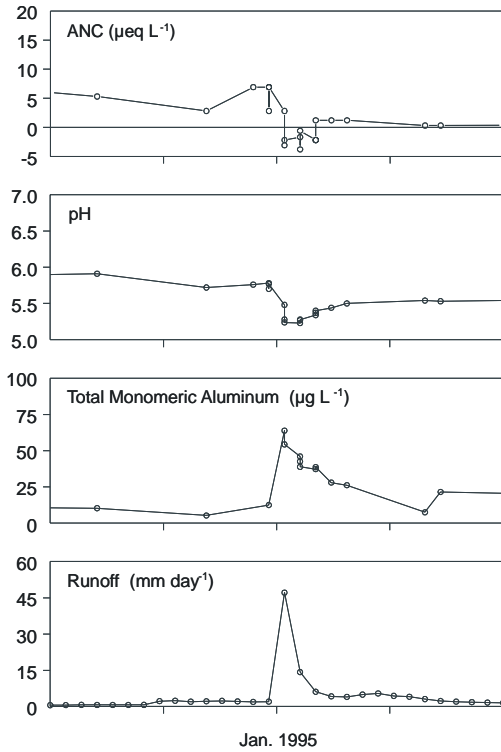
response to high flow are related to baseflow ANC values. Paine Run (siliciclastic bedrock) had a mean weekly ANC value of about 6 $\mu\text{eq/L}$ and often had high-flow ANC values that were less than 0 $\mu\text{eq/L}$. Staunton River (granitic bedrock) had a mean weekly ANC value of about 82 $\mu\text{eq/L}$ and had only a few high-flow ANC values less than 50 $\mu\text{eq/L}$. Piney River (basaltic bedrock) had a mean weekly ANC value of 217 $\mu\text{eq/L}$ and no values as low as 50 $\mu\text{eq/L}$.

3. Effects of Episodes on Biota

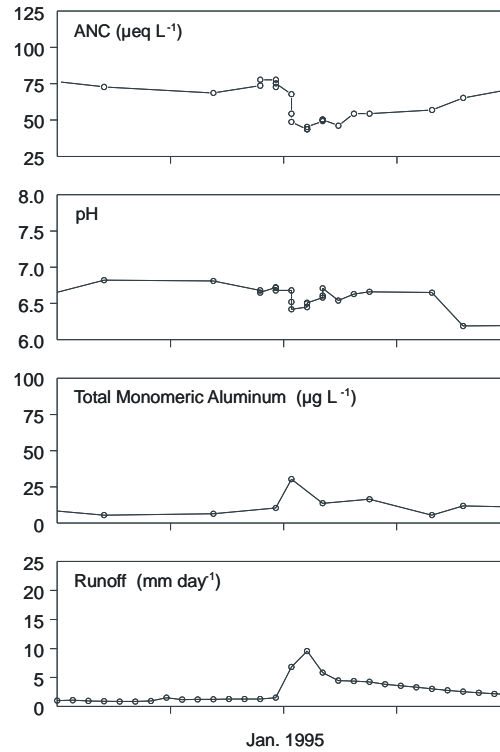
Previous studies have shown that mobilization of dissolved Al during episodic acidification is a primary cause of fish mortality in streams that have low ANC under baseflow conditions (Wigington et al. 1993). Streams with higher ANC during baseflow are less likely to become sufficiently acidic during episodes to bring much Al into solution. Figure 27 provides an example of changes in ANC, pH, and dissolved Al that occurred in Paine Run, Staunton River, and Piney River during a high-flow episode in January 1995. Under baseflow conditions, ANC at the Paine Run site was above 0 $\mu\text{eq/L}$, pH was above 5.5, and Al concentration was less than 25 $\mu\text{g/L}$. Discharge levels increased dramatically during the episode, resulting in depression of ANC to less than 0 $\mu\text{eq/L}$, pH values less than 5.5, and an increase in Al concentration to near 75 $\mu\text{g/L}$, above the threshold for adverse effects on some species of aquatic biota. That same episode also resulted in substantial declines in ANC in the granitic (Staunton River) and basaltic (Piney River) watersheds. However, ANC values at these two sites were relatively high prior to the episode (about 75 and 175 $\mu\text{eq/L}$, respectively) and did not decline to below about 50 $\mu\text{eq/L}$ during the episode at either site, and pH values remained above 6.0 and 6.5, respectively (Figure 27).

Results from the U.S. EPA's Episodic Response Project demonstrated that episodic acidification can have long-term adverse effects on fish populations. Streams with suitable chemistry during low-flow, but low pH and high Al levels during high flow, had substantially lower numbers and biomass of brook trout than were found in non-acidic streams (Wigington et al. 1996). Streams having acidic episodes showed significant mortality of fish. Some brook trout avoided exposure to stressful chemical conditions during episodes by moving downstream or into areas with higher pH and lower Al. This movement of brook trout only partially mitigated the adverse effects of episodic acidification, however, and was not sufficient to sustain fish biomass or species composition at levels that would be expected in the absence of acidic episodes. These findings suggested that stream assessments based solely on chemical

Paine Run (siliciclastic bedrock class)



Staunton River (granitic bedrock class)



Piney River (basaltic bedrock class)

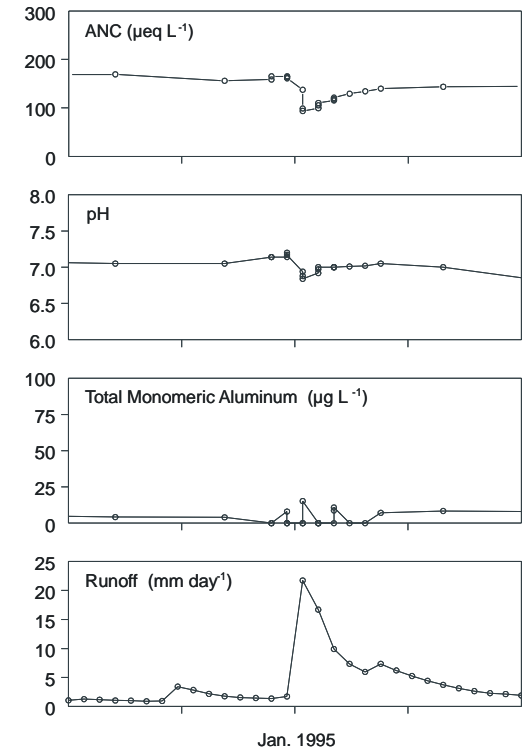


Figure 27. Decrease in ANC and pH and increase in dissolved aluminum in response to a sharp increase in streamflow in three watersheds within Shenandoah National Park during a hydrological episode in 1995. The watersheds were selected to be representative of the three geologic sensitivity classes within the park. Data are shown for the month of January, 1995. (Source: Sullivan et al. 2003)

measurements during low-flow conditions will not accurately predict the status of fish populations and communities in small mountain streams unless some adjustment is made for episodic processes (Baker et al. 1990b, Wigington et al. 1996, Sullivan 2000, Sullivan et al. 2003).

4. Summary of Episodic Effects

Thus, episodic acidification of streams can be attributed to a number of causes, including dilution of base cations and increased concentrations of sulfuric, nitric, and organic acids (Eshleman et al. 1995, Hyer et al. 1995). For streams having low pre-episodic ANC, episodic decreases in pH and ANC and increases in toxic Al concentrations can have adverse impacts on fish populations. Not all of the causes of episodic acidification are related to acidic deposition. Base-cation dilution and increase in organic acid anions during high-flow conditions are natural processes. The contribution of nitric acid, indicated by increased NO_3^- concentrations, can be related to disturbance, such as forest defoliation by the gypsy moth (Webb et al. 1995, Eshleman et al. 1998), or to acidic deposition. Significant contributions of sulfuric acid, indicated by increased SO_4^{2-} concentrations during episodes in some streams, is an effect of atmospheric deposition and the dynamics of S adsorption on soils (Eshleman and Hyer 2000).

Samples that have been collected from streams on Forest Service lands within the study area that is the focus of this report were collected during different seasons. It would be of interest to determine the extent of seasonal chemical variability that occurs in at least a subset of these streams. This is especially important because of the timing of the life stages of brook trout. Spawning occurs in Appalachian Mountain streams in the fall; hatching occurs in mid-winter. Thus, the most acid-sensitive stages of brook trout (eggs, sacfry, and swim-up fry) are present generally between about October and March.

At pH values below about 5.5 to 6.0, inorganic monomeric Al is mobilized from soils to surface waters, and can reach toxic levels in streamwater ($\sim 2 \mu\text{mol/L}$; which is equivalent to $54 \mu\text{g/L}$). The inorganic monomeric fraction of Al includes the toxic species; organically-complexed Al is generally considered to be nontoxic. Stream monitoring and stream survey programs in North Carolina and surrounding states have generally not measured the concentrations of dissolved Al fractions. Anecdotal information indicates that inorganic monomeric Al concentrations are probably fairly low in most streams in this region during baseflow, especially in comparison with concentrations commonly measured in surface waters in

the northeastern U.S. (Charles 1991). Notable exceptions include some highly acidic streams in GRSMNP (Rosemond et al. 1992), Linville Gorge and Shining Rock Wilderness. It is entirely possible that higher, and potentially toxic, levels of inorganic Al might be attained during rainfall episodes in low-ANC streams throughout the region. Such a response was documented by Webb (2003) in Paine Run in Shenandoah National Park (Figure 27). Episodic streamwater chemistry data are relatively rare in this region, and should be collected. Future studies to fill this data gap should include fractionation and analysis of Al.

Mobilization of dissolved Al from the soil and the streambed into dissolved form in the stream water during rainfall events is a major cause of episodic fish mortality in the northern Appalachian Mountains and northeastern U.S. (Wigington et al. 1993). The potential for biologically-meaningful episodic acidification related to acidic deposition is determined largely by the baseflow ANC of the stream and the extent to which SO_4^{2-} and/or NO_3^- concentrations increase with increasing discharge. Streams having low, but positive (less than about 20 $\mu\text{eq/L}$) ANC during baseflow are highly susceptible to episodic acidification to negative ANC values, with concomitant increases in inorganic Al into the toxic range (c.f., Figure 27). Streams in the southeastern U.S. having somewhat higher ANC (20-50 $\mu\text{eq/L}$) might be expected to typically experience episodic ANC depressions in the range of about 10 to 30 $\mu\text{eq/L}$, but are generally not expected to become acidic during episodes.

I. Model Uncertainty

1. Sensitivity analysis of simulation results and critical loads estimates.

In the following sensitivity analyses, the values of certain input data were altered for a number of sites and the model was then recalibrated for the sites. An analysis is presented here for selected simulated variables or critical loads derived from the “original calibrations” compared to the same variable and critical loads derived from the “sensitivity calibrations” using altered input data. Sensitivity analyses were performed for stream water data, soils data, and occult deposition data used to calibrate the model.

The primary comparisons for the sensitivity analyses for emissions control scenarios are presented as x-y plots showing model projection results from the “sensitivity calibrations” on the y-axis and values derived from the “original calibrations” on the x-axis. Four plots are presented for simulated variables (SO_4 , SBC, CALK, and % BS). Each plot contains simulated values of the variable in question from a number of sites (the sites for which the input data were changed)

for three future years (2020, 2040, and 2100) for three different future deposition scenarios (base, moderate, and aggressive). A 1:1 line is presented on each plot to provide a reference for the changes in variable values resulting from the changed input data.

The primary comparisons for the sensitivity analyses for critical loads are also presented as x-y plots showing results derived from the “sensitivity calibrations” on the y-axis and values of the “original calibrations” on the x-axis. Two plots are presented for each sensitivity analysis. Each plot contains critical loads estimates from a number of sites (the sites for which the input data were changed) for three target ANCs (0, 20 and 50 µeq/L) for three future years (2020, 2040, and 2100). A 1:1 line is presented on each plot to provide a reference for the changes in critical loads estimates resulting from the changed input data.

Measures of the *average change in variable values* (across the sites) for a given year and scenario, and the *average change in critical loads* (across sites) for a given target ANC and year are presented in tables for each sensitivity analysis. The average changes are calculated as the absolute values of the change in output between the values derived from the *Sensitivity* calibration and those derived from the *Original* calibration, expressed as a percentage of the absolute value derived from the *Original* Calibration:

$$\text{Ave Change (\%)} = 100 \times [(\text{Sensitivity} - \text{Original})] / \text{Original}.$$

Both simulated values of CALK and estimates of critical loads can have values approaching zero making percentage calculations problematic. Therefore, the average change due to the Sensitivity recalibration at each site is expressed as either 1) the Percentage Change as defined above; or 2) just the Absolute Change when the absolute values from the “original calibrations” are less than 5 µeq/L for CALK or less than 20 kg S/ha/yr for critical load estimates.

a. *Sensitivity of Model Outputs to Specification of Stream Water Data*

For the 66 sites modeled, 59 had stream water samples taken in only one year over the period 1995 to 2005. For these 59 sites, the single sample available was used to calibrate MAGIC, regardless of the year it was taken. There were 7 streams, however, for which stream water samples were available for multiple years at the site. All 7 of these streams had samples available in 2000, and that year was used for the original calibration of MAGIC at those sites. The stream water values in the other years at each site (all within +/- 5 years of 2000) differed,

however, from the values measured in 2000. This inter-annual variation is expected, but calibration of the model to a different suite of stream water values (all other inputs to the model being the same) can be expected to produce a different set of model outputs.

The sensitivities of simulated variable values and estimated critical loads to the specification of the stream water data used for calibration were examined by recalibrating the 7 sites using the streamwater data from the alternate years as target values for the fuzzy optimization procedure. Two of the streams had two years each of additional data. For these streams two sensitivity recalibrations were performed, giving 9 “site” recalibrations that could be compared to the original calibrations. All other inputs to the model remained the same. All 9 recalibrated “sites” were used for running the three future deposition scenarios and for calculating critical loads.

Changing the surface water data used for calibrating the model produced offsets in the simulated values of all streamwater variables (Figure 28). For instance, if the alternate chemistry data contained higher base cation and/or SO_4 values, the optimization procedure adjusted weathering and/or sulfate adsorption parameters to match the higher or lower values. In general, this adjusted offset in the calibration year persisted through all years of the various forecast scenarios as can be seen in Figure 28 for SBC, SO_4 , and CALK. On the other hand, the observed soil data were not changed in this sensitivity analysis, so the optimization procedure calibrated soil selectivity coefficients to simulate the same soil base saturation values. Without an offset in the calibration year, soil base saturation (% BS, Figure 28) in the forecast years was essentially unchanged.

Changing the stream water data used for calibrating the model had mixed effects on the estimates of critical loads derived from the model (Figure 29). Some critical loads estimates increased with the altered stream water input data and some decreased. In general, if the stream water ANC used to recalibrate the model at a site was higher than the ANC used in the original calibration, the critical load estimates increased for that site. This is expected because the “site” represented by the “sensitivity calibration” apparently has a higher buffer capacity (higher ANC) than the original “site”, all else being the same, and therefore can tolerate a higher critical load both at the present and into the future. The same reasoning applies to sites that were recalibrated with lower stream water ANC. Three of the nine “sites” recalibrated with altered stream water inputs had higher ANC in the calibration year; six “sites” had lower ANC values in the calibration year.

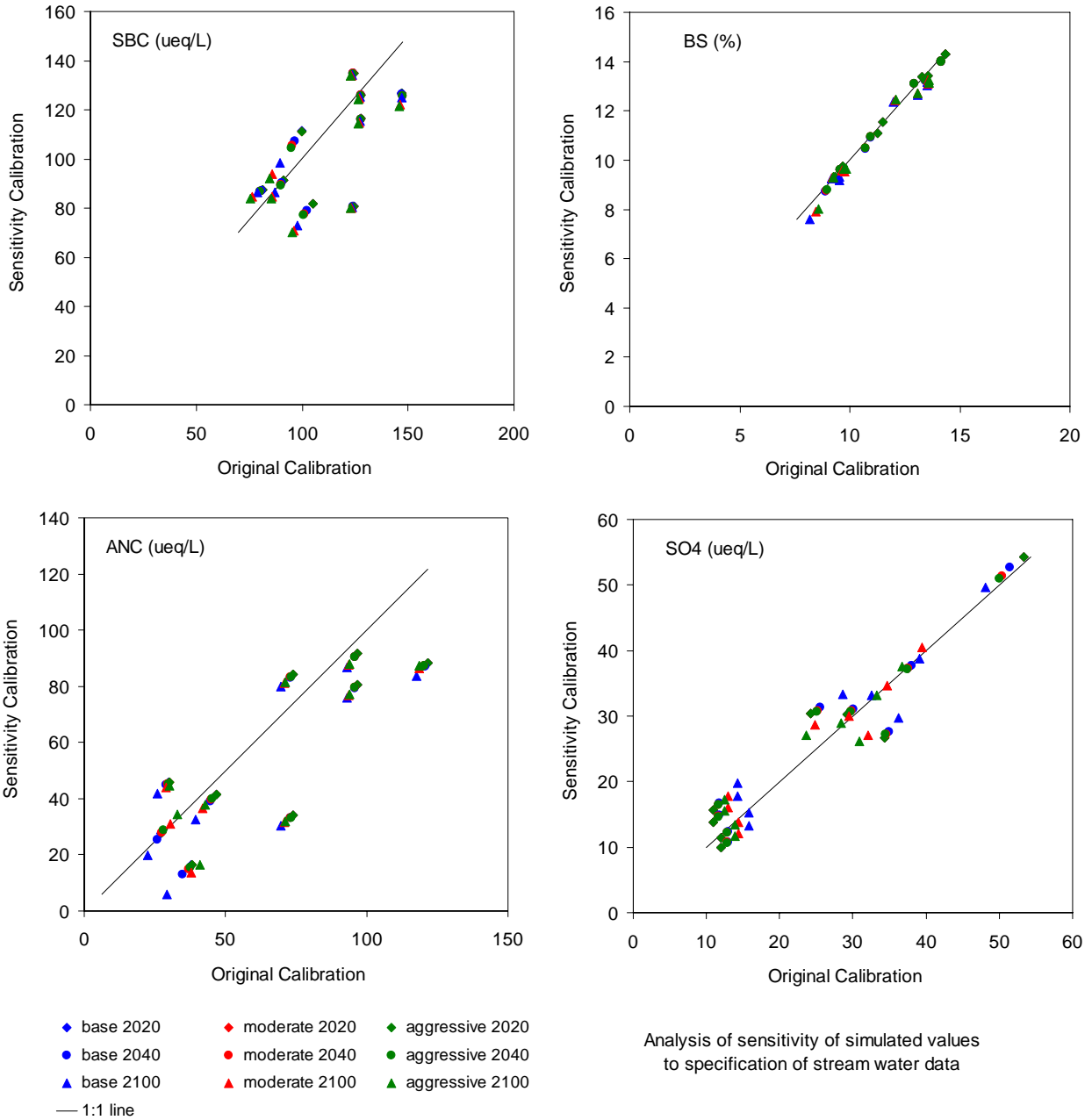


Figure 28. Sensitivity of model simulations to specification of stream water data. Seven sites were recalibrated using stream water data from alternate years to calibrate the model. Five sites had 1 alternate year of data; two sites had 2 alternate years. Alternate years were sampled within 5 years of the original calibration year. MAGIC model projections of ANC, SO₄, SBC and soil BS are presented for the years 2020, 2040, and 2100 for three future deposition scenarios. Values on the x-axis are based the original model calibrations of each site. Values on the y-axis are based on the sensitivity re-calibration of each site using stream data from the alternate year.

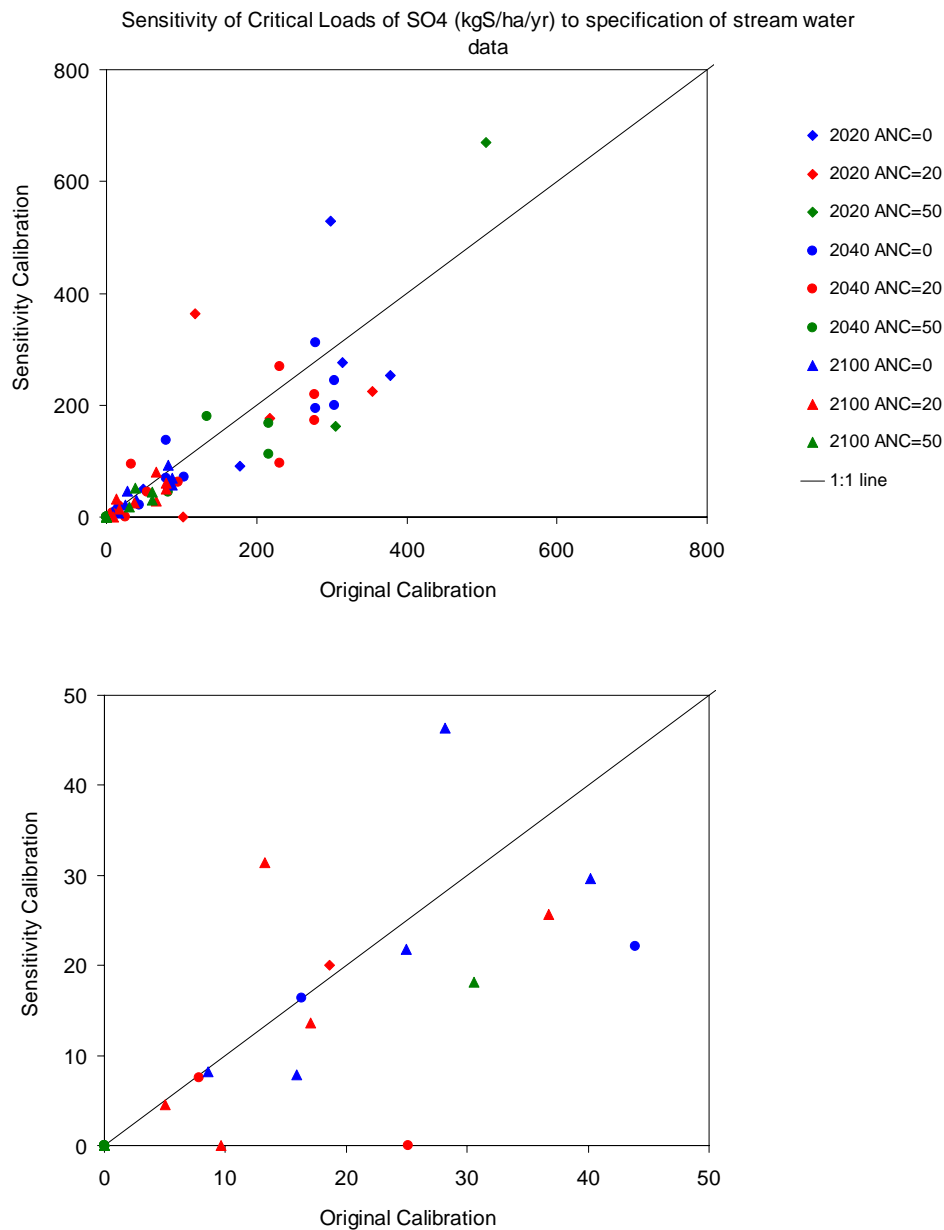


Figure 29. Sensitivity of estimated critical loads to specification of stream water data. Seven sites were recalibrated using stream water data from alternate years to calibrate the model. Five sites had 1 alternate year of data; two sites had 2 alternate years. Alternate years were sampled within 5 years of the original calibration year. MAGIC model estimates of critical loads of SO₄ deposition which produce three different ANC levels in three different years are presented. Values on the x-axis are based the original model calibrations of each site. Values on the y-axis are based on the sensitivity recalibration of each site using stream data from the alternate year. (The lower panel shows an expansion of the axes for the lower critical loads values).

The average changes in variable values (n=9) that resulted from changing the stream water data used for calibrating the model were greatest for SBC (~12%), SO₄ (~15%), and CALK (~27%) (Table 27). As explained in the discussion above, the changes in % BS (~1%) were relatively small compared to the changes in stream water variables. The average changes in all simulated variables appear to be of about the same size in all three years and for all three future scenarios.

Table 27. Sensitivity analysis results for stream water inputs, expressed as average percent change in simulated values (for 9 sites) resulting from recalibration of the model using alternate stream water data. Average changes are presented for selected variables in selected years for the three future deposition scenarios.

	Average Percent Change in Simulated Chemistry			
	Calk	SO ₄	SBC	BS
Base Scenario				
2020	27	16	12	1
2040	28	16	12	1
2100	33	14	13	3
Moderate Scenario				
2020	27	16	12	1
2040	27	15	12	1
2100	28	13	13	3
Aggressive Scenario				
2020	27	16	12	1
2040	27	15	12	1
2100	27	13	13	2

The average change in estimated critical load values (n=9) resulting from changing stream water inputs used for calibration was smallest (~30%) for a target ANC of 0 µeq/L (Table 28). Average changes in estimated critical loads using target ANC values of 20 or 50 µeq/L were approximately 50%. For target ANC values of 0 and 20 µeq/L there was an indication that average changes decreased as the target year increased (Table 28).

Table 28. Sensitivity analysis results for stream water inputs, expressed as average percent change in estimated critical load values (for 9 sites) resulting from recalibration of the model using alternate stream water data. Average changes are presented for three target ANC values in three target years.

Year	Average Percent Change in Simulated Critical Load		
	ANC=0 $\mu\text{eq/L}$	ANC = 20 $\mu\text{eq/L}$	ANC = 50 $\mu\text{eq/L}$
2020	30	62	50
2040	33	60	50
2100	29	34	50

b. Sensitivity of Model Outputs to Specification of Soils Data

If soils data were available within the catchment for a given site, the site was designated as a Tier I site. For Tier II sites, soils data were borrowed from a nearby Tier I site located on the same geology. For Tier III sites, soils data were borrowed from the Tier I site judged to be most comparable with respect to streamwater ANC (an integrator of watershed soils conditions), geologic sensitivity class, location, elevation and streamwater sulfate concentration (an integrator of sulfur adsorption on soils).

The sensitivities of simulated variable values and estimated critical loads to the necessity of having to “borrow” soils data for Tier II and Tier III sites were examined by calibrating 7 Tier I watersheds twice, the first time using the appropriate site-specific soils data, and the second time using borrowed soils data from an alternate site, using either Tier II or Tier III protocols. Both sets of calibrations for the 7 sites were used for running the three future deposition scenarios and for calculating critical loads.

Changing the soil data used for calibrating the model produced pronounced offsets in the simulated values of the soil variable % BS while having little effect on simulated values of stream water variables (SO_4 , SBC and CALK; Figure 30). If the alternate soil chemistry data contained higher observed base saturation in the calibration year, the optimization routine adjusted the selectivity coefficients to produce a higher simulated base saturation in the calibration year (and conversely). Simulated base saturation values in all future years of all scenarios reflected the offset in base saturation values in the calibration year (Figure 30). Because the stream water variables used to calibrate the sites were not changed in this analysis,

there was no appreciable offset produced in these variables and their values in future years were essentially unchanged (Figure 30).

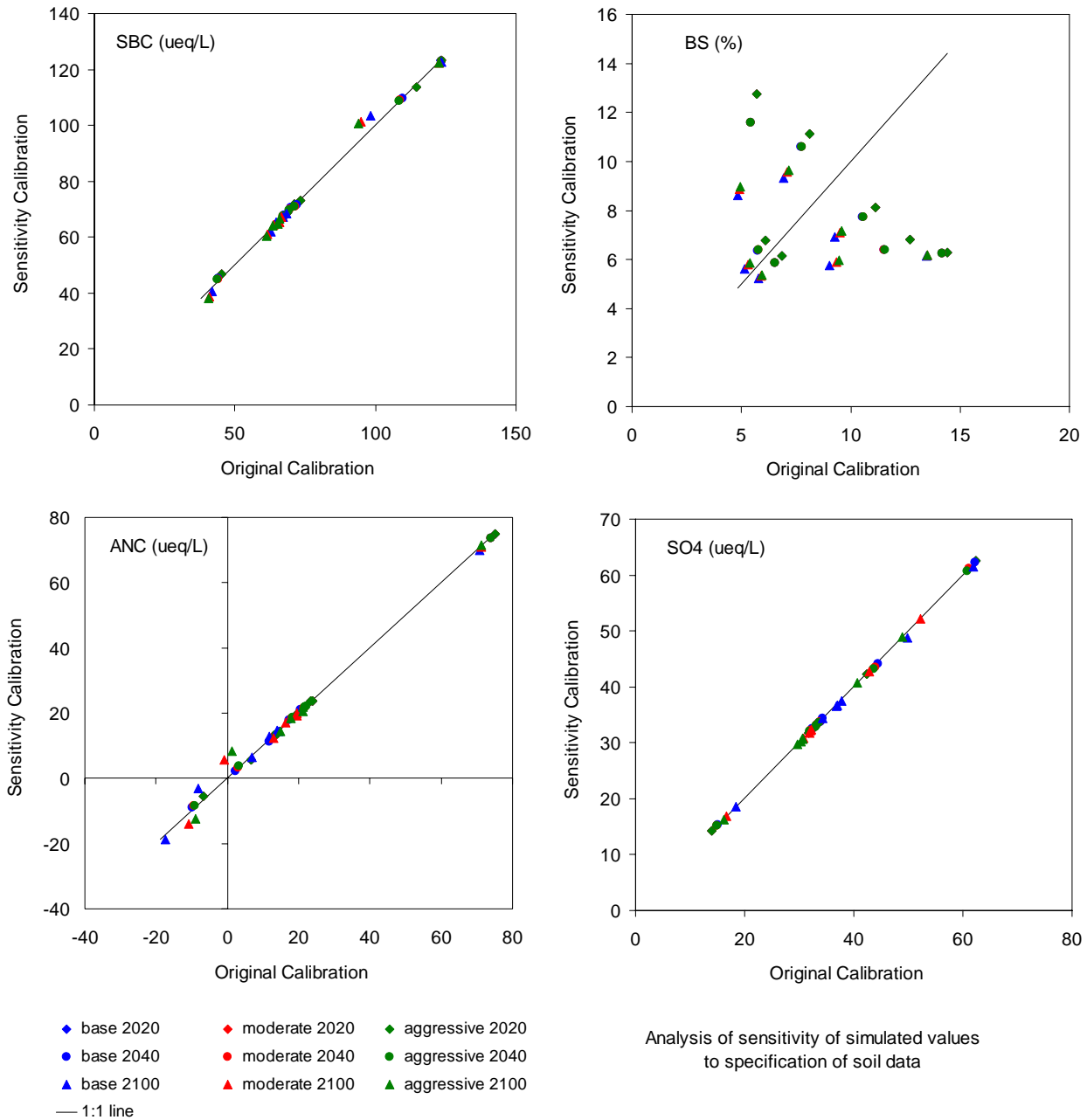


Figure 30. Sensitivity of model simulations to specification of soils data. Three Tier I sites were recalibrated using Tier II protocols, and four Tier I sites were recalibrated using Tier III protocols. MAGIC model projections of ANC, SO₄, SBC and soil BS are presented for the years 2020, 2040, and 2100 for three future deposition scenarios. Values on the x-axis are based the original model calibrations of each site using soils data collected within the watersheds (Tier I). Values on the y-axis are based on the

sensitivity re-calibration of each site using borrowed soils data from an alternate site using Tier II or Tier III protocols.

Changing the soil data used for calibrating the model had only a small effect on the estimates of critical loads derived from the model (Figure 31). All critical load estimates (with exception of the estimates the one site which had the highest critical load values) were essentially unchanged (see especially Figure 31 expanded axis). This is primarily because the critical load estimates derived for this project are all based on streamwater ANC, which in turn depend most critically on weathering rates at the sites. While soil base saturation can have an effect on the lag time associated with achievement of a critical load (delaying that achievement through the buffering action of exchangeable cations on the soil matrix), the ultimate value of the critical load depends most heavily on weathering. In the calibration of MAGIC, weathering rates are derived (in part) by calibrating to stream water SBC concentrations which were not changed in this sensitivity analysis.

The average changes in variable values (n=7) that resulted from changing the soil data used for calibrating the model were greatest for soil base saturation BS (~40%) (Table 29). Average changes in simulated stream water SO₄ (~1%) and SBC (~1%) were very small because the input data used to calibrate these variables was not changed in this sensitivity analysis. Even though individual stream water ions showed little sensitivity to changed soil input data, the average change in future simulated stream ANC (~6%) (Table 29) did show the effect of changing the soil buffer pools (% BS). The average changes in simulated stream water variables appear to be of about the same size in all three years and for all three future scenarios. The average change in simulated soil base saturation, however, appears to decrease as the length of the simulation increases (for all three scenarios).

The average change in estimated critical load values (n=7) resulting from changing soil inputs used for calibration was smallest (~15%) for a target ANC of 0 µeq/L (Table 30). The average change appeared to get larger as the target ANC increased to 20 µeq/L (~25%) and to 50 µeq/L (~33%). For target ANC values of 50 µeq/L there was an indication that average changes decreased as the target year increased (Table 30), but this pattern was not observed for the other two target ANC values.

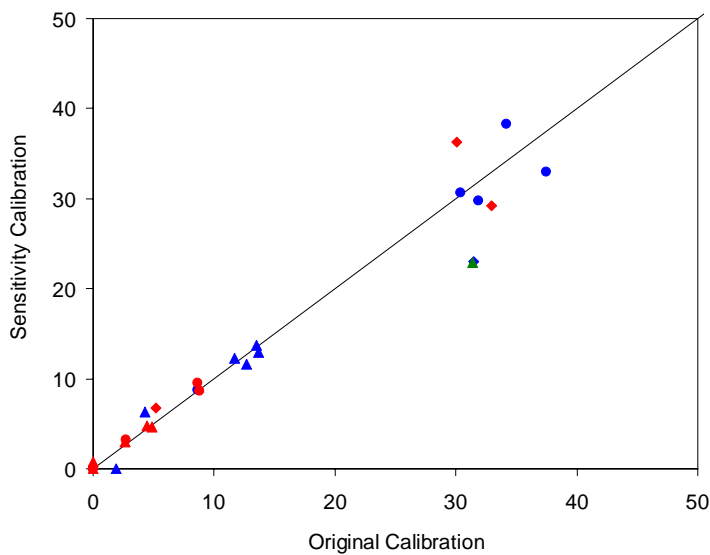
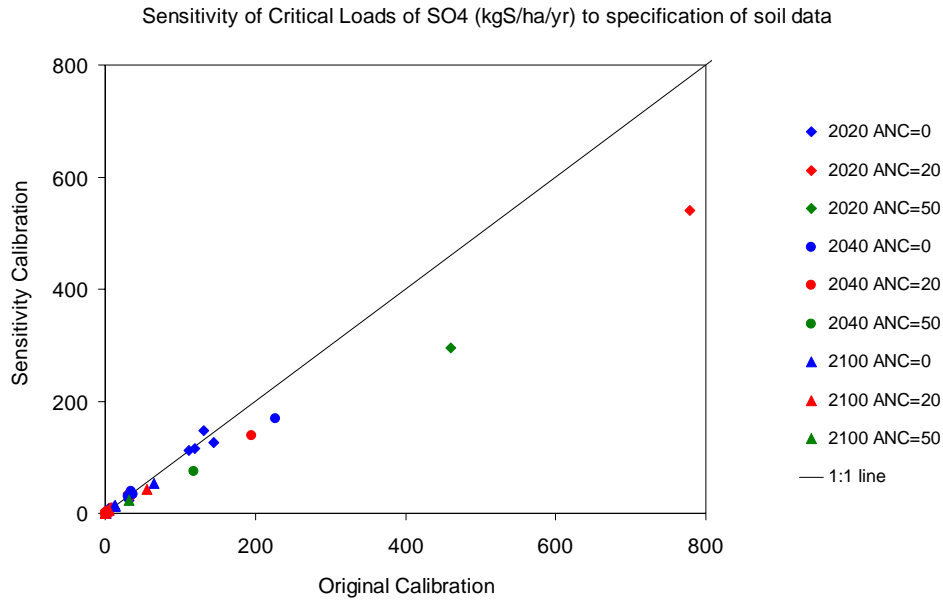


Figure 31. Sensitivity of estimated critical loads to specification of soils data. Three Tier I sites were recalibrated using Tier II protocols, and four Tier I sites were recalibrated using Tier III protocols. MAGIC model estimates of critical loads of SO₄ deposition which produce three different ANC levels in three different years are presented. Results presented on the x-axis are based the original model calibrations of each site using soils data collected within the watersheds (Tier I). Results presented on the y-axis are based on the sensitivity re-calibration of each site using borrowed soils data from an alternate site using Tier II or Tier III protocols. (The lower panel shows an expansion of the axes for the lower critical loads values).

Table 29. Sensitivity analysis scenario results for soil inputs. Average percent change in simulated values (for 7 sites) resulting from recalibration of the model using alternate soil data. Average changes are presented for selected variables in selected years for the three future deposition scenarios.

	Average Percent Change in Simulated Chemistry			
	Calk	SO ₄	SBC	BS
Base Scenario				
2020	6	0	1	45
2040	3	1	1	43
2100	13	1	2	35
Moderate Scenario				
2020	6	0	1	45
2040	3	1	1	43
2100	7	0	2	36
Aggressive Scenario				
2020	6	0	1	45
2040	3	1	1	43
2100	9	0	3	36

Table 30. Sensitivity analysis critical load results for soil inputs. Average percent change in estimated critical load values (for 7 sites) resulting from recalibration of the model using alternate soil data. Average changes are presented for three target ANC values in three target years.

	Average Percent Change in Critical Load		
	ANC=0 µeq/L	ANC = 20 µeq/L	ANC = 50 µeq/L
2020	14	21	36
2040	11	29	37
2100	19	23	27

c. Sensitivity of Model Outputs to Specification of Occult Deposition

At very high elevations, the inputs of ions from cloud water (one form of occult deposition) can be very large. In this project there were five sites at elevations over 1500 meters. These sites were assigned higher occult deposition values than the other sites in the original

calibration procedure to account for the likelihood of increased occult deposition from cloud water based on observations at one high elevation site in the Great Smoky Mountain National Park. All other sites were assigned lower occult deposition values based on the atmospheric deposition model ASTRAP.

The sensitivities of simulated variable values and estimated critical loads to the assumptions regarding high elevation occult deposition were examined by re-calibrating 10 of the modeled sites. The five highest elevation sites (1591 to 1719 m) originally calibrated using the increased high elevation occult deposition values were re-calibrated using the lower occult values assigned to all other sites. The next five highest sites (1245 to 1453 m) originally calibrated using the lower occult values were re-calibrated using the increased high elevation occult values. Both sets of calibrations for the ten sites were then used for running the three future deposition scenarios and for calculating critical loads.

Changing the occult deposition data used for calibrating the model produced offsets in the simulated values of SO_4 and CALK (Figure 32) but had a much smaller effect SBC and % BS. The effect on simulated stream water SO_4 values is less than straightforward, with competing effects occurring. For those sites recalibrated with higher occult SO_4 deposition, the optimization procedure calibrated a higher SO_4^{2-} adsorption capacity to maintain the same target SO_4 value in stream water in the calibration year (only deposition inputs were changed in this analysis). The converse is true for those sites recalibrated with lower occult SO_4 deposition.

When forecasts were run into the future, the stream water SO_4 simulated by sites with higher occult deposition and higher SO_4^{2-} adsorption capacity were affected by two factors: 1) the scaled future deposition of SO_4 was higher; and 2) the higher sulfate adsorption meant either greater adsorption or greater desorption of SO_4 from the adsorbed soil pool (depending on the calibration year stream water SO_4 concentration). The converse of these effects applies to streams with lower occult deposition used for recalibration. As a result, the future simulated values of SO_4 (Figure 32) show both increases and decreases.

The stream water SBC values and soil base saturation values (Figure 32) were essentially unchanged as a result of changing the occult SO_4 deposition. This is because the optimization routine was using the same stream water and soil targets for the base cation variables. The optimization routine calibrated different base cation weathering and selectivity coefficients to match these base cation targets even though occult SO_4 deposition (and adsorption) had changed. With stream water SO_4 altered and stream water SBC essentially unchanged, the simulated CALK

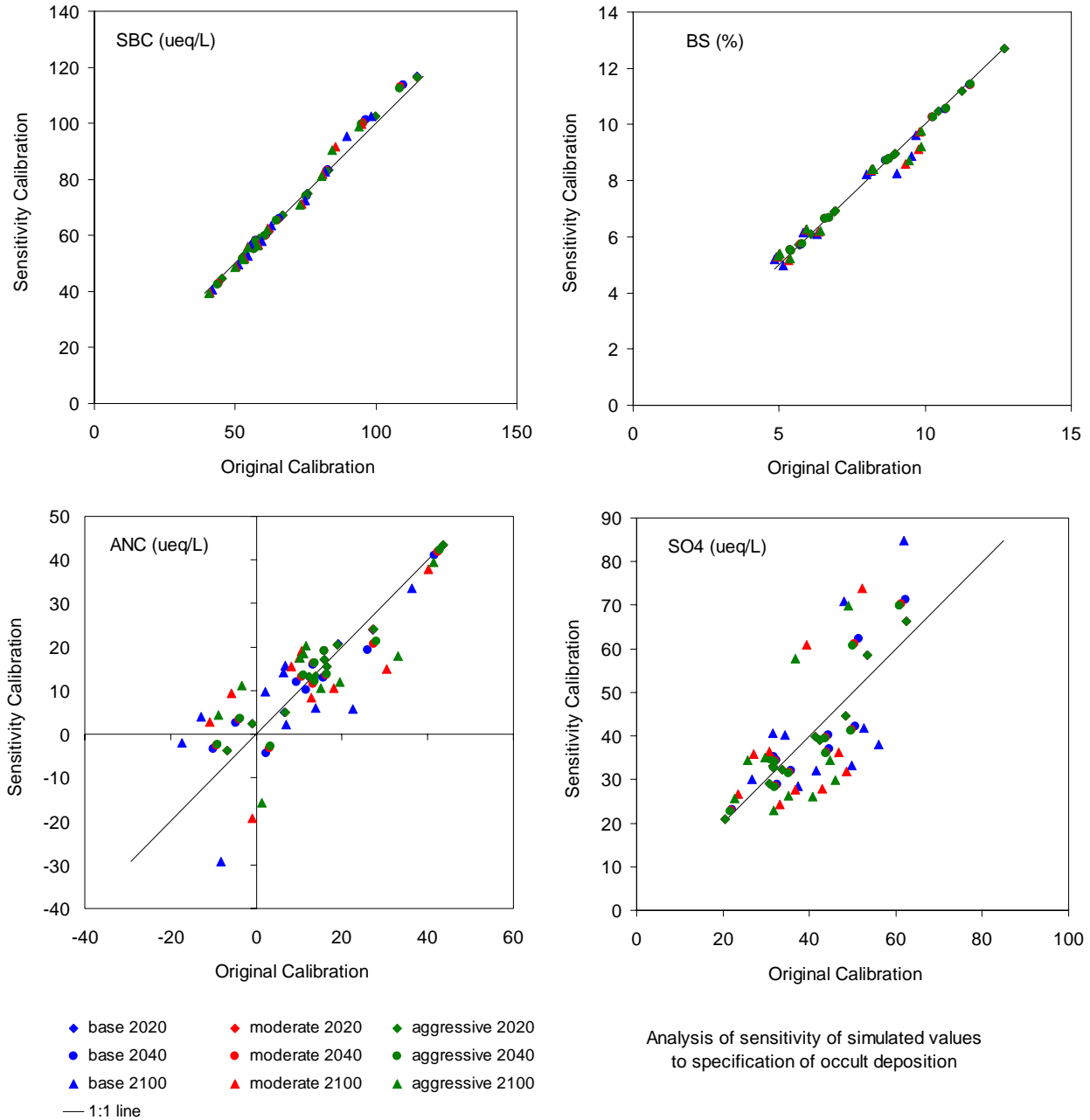


Figure 32. Sensitivity of model simulations to specification of occult deposition data. Five high elevation sites (1591m-1719m) were recalibrated using lower occult deposition, and five intermediate elevation sites (1245m-1453m) were recalibrated using higher occult deposition. MAGIC model projections of ANC, SO₄, SBC and soil BS are presented for the years 2020, 2040, and 2100 for three future deposition scenarios. Values on the x-axis are based the original model calibrations of each site using elevation-appropriate occult deposition. Values on the y-axis are based on the sensitivity recalibration of each site using either increased or decreased occult deposition.

values show the same magnitude (but opposite direction) of changes as those in stream water SO_4 (Figure 32).

Changing the occult deposition data used for calibrating the model had mixed effects on the estimates of critical loads derived from the model (Figure 33). Some critical loads estimates increased with the altered occult deposition input data and some decreased. In general, increased occult SO_4 deposition produced lower critical load estimates, and conversely. This is expected because the recalibration with higher occult deposition and the same stream water and soil base cation status resulted in lower stream water ANC. As discussed above, lower stream water ANC generally indicates a lower buffer capacity and thus a lower critical load for any future year.

The average changes in variable values ($n=10$) that resulted from changing the occult deposition data used for calibrating the model were greatest for stream water SO_4 and CALK, with patterns across both the future scenarios and future years (Table 31). In general, changing occult deposition produced average changes in stream water SO_4 that increased into the future, starting at 6% in 2020 and increasing about 5-fold (to ~30%) by 2100. A similar pattern was noted for average changes in ANC, starting at 13% in 2020 and increasing into the future. Unlike, SO_4 , however, the average changes in ANC for the year 2100 also showed an effect of the scenario, with the average changes in ANC being largest for the base scenario (the scenario with the highest SO_4 deposition in 2100). Average changes in the base cation variables SBC and % BS were relatively small (1% to 5%) (Table 31) compared to the changes in stream water variables, primarily because the target values used to calibrate these variables were not changed in this sensitivity analysis.

The average change in estimated critical load values ($n=10$) resulting from changing the occult deposition inputs was approximately the same (~60% to 65%) for all target years of target ANC's 0 and 20 $\mu\text{eq/L}$ (Table 32). Average changes in critical loads to produce 50 $\mu\text{eq/L}$ were not calculable for the 10 sites because both the original calibration and sensitivity recalibration for these sites produced critical load estimates of 0 $\mu\text{eq/L}$ for all of the ensemble parameter files.

2. Uncertainty in Model Simulations and Critical Loads Estimates

The sensitivity analyses in the preceding three sections were designed to address specific assumptions or decisions that had to be made in order to assemble the data for the 66 modeled sites in a form that could be used for calibration of the model. In all cases, the analyses addressed the questions of what the effect would have been if alternate available choices had been taken.

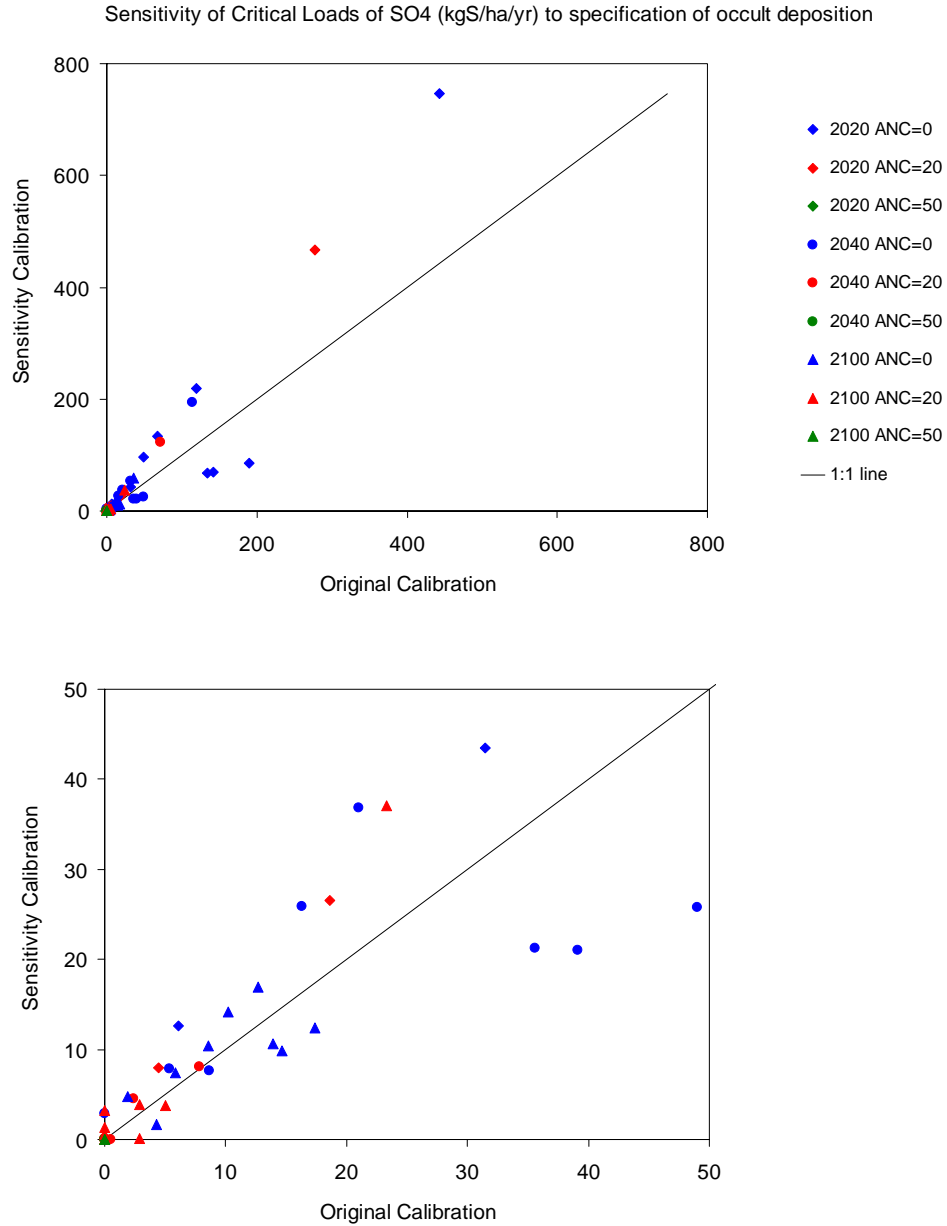


Figure 33. Sensitivity of estimated critical loads to specification of occult deposition data. Five high elevation sites (1591m-1719m) were recalibrated using lower occult deposition, and five intermediate elevation sites (1245m-1453m) were recalibrated using higher occult deposition. MAGIC model estimates of critical loads of SO₄ deposition which produce three different ANC levels in three different years are presented. Values on the x-axis are based the original model calibrations of each site using elevation-appropriate occult deposition. Values on the y-axis are based on the sensitivity recalibration of each site using either increased or decreased occult deposition. (The lower panel shows an expansion of the axes for the lower critical loads values).

Table 31. Sensitivity analysis scenario results for occult deposition inputs. Average percent change in simulated values (for 10 sites) resulting from recalibration of the model using either increased or decreased occult deposition data. Average changes are presented for selected variables in selected years for the three future deposition scenarios.

		Average Percent Change in Simulated Chemistry			
		Calk	SO ₄	SBC	BS
Base Scenario					
2020		13	6	1	0
2040		24	12	2	1
2100		104	28	3	5
Moderate Scenario					
2020		13	6	1	0
2040		24	13	2	1
2100		85	30	3	5
Aggressive Scenario					
2020		13	6	1	0
2040		24	13	2	1
2100		60	31	3	5

Table 32. Sensitivity analysis critical loads results for occult deposition inputs. Average percent change in estimated critical load values (for 10 sites) resulting from recalibration of the model using either increased or decreased occult deposition data. Average changes are presented for three target ANC values in three target years.

		Average Percent Change in Critical Load		
Year		ANC=0 µeq/L	ANC = 20 µeq/L	ANC = 50 µeq/L
2020		67	68	na
2040		58	73	na
2100		63	59	na

These analyses were undertaken for a subset of sites for which the alternate choices were available at the same sites. As such, the analyses above are informative, but they provide no direct information about the uncertainty in calibration or simulation arising from the choices that were incorporated into the final modeling protocol for *all* sites. The results in this section provide estimates of the overall model simulation uncertainty (and uncertainty in critical loads estimates) at all 66 sites using the final calibration, simulation, and critical loads analysis protocols.

The uncertainty estimates were derived from the multiple calibrations at each site provided by the “fuzzy optimization” procedure employed in this project. For each of the 66 sites, 10 distinct calibrations were performed with the target values, parameter values, and deposition inputs for each calibration reflecting the uncertainty inherent in the observed data for the individual site. The effects of the uncertainty in the assumptions made in calibrating the model (and the inherent uncertainties in the data available) can be assessed by using all successful calibrations for a site when simulating the response to different scenarios of future deposition, or using all successful calibrations at a site when calculating a critical loads estimate.

When implemented with the ensemble parameter sets attached to each site, the model produces an ensemble of simulated values for each year at each site, or an ensemble of critical loads estimates for each analysis at each site. The median of all simulated or calculated values at a site is considered the most likely response of the site. The projections from MAGIC reported throughout this document are the median values from the ensemble of calibrations for each of the 66 modeled sites. The simulated or calculated values in the ensemble can also be used to estimate the magnitude of the uncertainty in the projection. Specifically, the difference in any year between the maximum and minimum simulated values from the ensemble of calibrated parameter sets at a site can be used to define an “uncertainty” (or “confidence”) width for the simulation at any point in time for that individual site. Similarly, the difference between the maximum and minimum critical load calculated for a site (10 ensemble parameter sets used in the same critical loads analysis at the site) can be used to define an “uncertainty” (or “confidence”) width for that particular critical load estimate at the site.

The uncertainty widths may vary through time for a given variable and site. It is expected, for instance, that uncertainties near the calibration year (when the model is constrained by observed data) should generally be smaller than uncertainties in the distant past or far future. Uncertainty widths may differ among variables being simulated for a given year and site. The model is calibrated to observations of base cations in the stream and exchangeable base cations

in the soil, so it might be expected that the uncertainty in these variables should be relatively small. ANC, on the other hand, is by definition the sum of a number of simulated variables, and it might be expected that the uncertainty in ANC would be relatively larger, reflecting the aggregate uncertainty in all variables being simulated. Finally, uncertainty widths may vary from site to site for a given variable and year. The characteristics of individual sites (soil depth, runoff, deposition, etc.) vary and the effects of these variations may make the simulated values of some variables more or less sensitive to uncertainties in the inputs of other parameters or variables.

Given these three broad categories affecting the variability of uncertainty widths (site-to-site, year-to-year, and variable-to-variable), the presentation of a complete analysis of simulation uncertainty is problematic. A summary is given here for the following 4 variables: SO₄ concentration in stream water; SBC (sum of base cation concentrations) in stream water; charge balance ANC (CALK) in stream water; and soil base saturation (% BS). The uncertainty widths for these 4 variables are given for the reference year (2005; the year nearest to the calibration year for all sites), and for three years (2020, 2040, 2100) in each of three future scenarios (base, moderate, aggressive).

The uncertainty widths presented are the average of the individual uncertainty widths at each of the 66 sites for a given variable and year. The widths at any individual site may be lower or higher, but the average across all 66 sites gives a robust indication of the size of simulation uncertainty in general for this project. The uncertainty width for each variable in a given year at a given site is expressed as a percentage (+/-) of the median simulated variable value:

$$\text{uncertainty width (\%, +/-)} = 100 \times [(\text{max} - \text{min})/2] / \text{median},$$

where max, min, and median are statistics of the 10 simulated values in a given year at a given site resulting from the 10 ensemble parameter sets at the site. Uncertainty widths were defined similarly for the each of the critical loads estimates at a site.

a. *Uncertainty in Simulated Variables*

As anticipated, the uncertainty widths for the calibrated variables SBC and BS were relatively small (+/- 1% to 2%) for all years and all scenarios (Table 33). Uncertainty widths for SO₄ were larger than for the base cations, but still very reasonable (+/- 1% to 7%, Table 33).

Table 33. Uncertainty widths (% +/-, see text) for selected simulated variables and selected years. The uncertainty widths are the average widths for all 66 sites modeled.

	Uncertainty Width (%)			
	CALK	SO ₄	SBC	BS
Reference Year				
2005	9	1	1	1
Base Scenario				
2020	14	2	1	1
2040	15	4	1	1
2100	21	7	1	2
Moderate Scenario				
2020	14	2	1	1
2040	14	4	1	1
2100	19	7	1	2
Aggressive Scenario				
2020	14	2	1	1
2040	14	4	1	1
2100	18	7	1	2

Even though SO₄²⁻ adsorption was calibrated for the sites, a number of other processes affecting SO₄ were not calibrated (most importantly the total deposition inputs of SO₄), resulting the higher uncertainty estimates. The uncertainty widths for charge balance ANC (CALK) were +/- 9% in the reference year, +/- 14% in the near future and approximately +/- 20% in 2100 (Table 33). Because CALK is the sum of a number of variables, the relatively narrow uncertainty width in the reference year is very encouraging. It suggests that the overall calibration procedure was robust and that all variables in the model were well-constrained (at least given the levels of uncertainty assumed for parameters and inputs in the fuzzy optimization procedure). The slightly larger, but still reasonable, uncertainty widths in future years suggest that relatively small differences in simulated ANC between different deposition scenarios can be accepted as “real” with some confidence.

The uncertainty widths for CALK and SO₄ (Table 33), were smallest in the reference year, but become larger the further into the future the simulations were taken. There does not seem to be an effect on SO₄ uncertainty of the particular future scenario. The SO₄ uncertainty

widths in 2100 were the same for all three scenarios. On the other hand, the uncertainty in CALK does vary by scenario, with the uncertainty width being greatest in 2100 for the base scenario and least for the aggressive scenario.

b. Uncertainty in Critical Loads Estimates

Uncertainty widths for critical loads estimates were derived for 12 different critical loads analysis protocols: four different target ANC values in three different years (Table 34). Unlike the analysis of average uncertainty widths for simulated variables given above (where each average uncertainty width was based on 66 sites), the number of sites contributing to an average uncertainty width for a particular critical load analysis protocol was variable. Uncertainty widths could only be calculated for sites where a particular critical loads analysis was possible (target ANC less than historical ANC at the site), and where at least one of the ensemble estimates of the critical load at the site was greater than zero. For instance, there were only 9 modeled sites that had an historical ANC greater than 100 µeq/L. Therefore, only those 9 sites could be analyzed for a critical load producing a target ANC = 100 µeq/L. Of those 9 sites, only 1 site gave at least one of the ensemble critical loads estimates as greater than zero. Therefore, in Table 34, the uncertainty width estimate for Target ANC=100 in any of the three years is based on only one site (n=1). The largest number of sites used to calculate an average uncertainty width for a particular critical load protocol was for a target ANC = 0 µeq/L in year 2020, for which 60 sites (n=60) produced at least one critical load estimate greater than zero. The average

Table 34. Uncertainty widths (UW, % +/-, see text) for estimated critical loads. The uncertainty widths are the average widths for all sites for which a given critical load could be estimated. The number of sites averaged (n) for each uncertainty width (UW) are indicated. The different critical loads analysis are based on target stream ANC values (left column) in target years (top row).

	Uncertainty Width for Critical Loads (%)					
	Target Year 2020		Target Year 2040		Target Year 2100	
	UW%	n	UW%	n	UW%	n
Target ANC = 0 µeq/L	31	60	24	58	17	58
Target ANC = 20 µeq/L	32	44	23	43	18	41
Target ANC = 50 µeq/L	36	15	23	15	21	16
Target ANC = 100 µeq/L	25	1	23	1	18	1

uncertainty widths presented in Table 34 for target ANC of 0 and 20 $\mu\text{eq/L}$ are based on between 41 and 60 sites for all years, and are probably robust estimates of the uncertainty. Average uncertainty widths for ANC of 50 $\mu\text{eq/L}$ are based on 15-16 samples in a given year and are probably reliable. The result for target ANC of 100 $\mu\text{eq/L}$ ($n=1$) will not be discussed further.

The average uncertainty widths for critical load estimates were not excessively large, ranging from +/- 17% to +/- 36% depending on the analysis conditions. The estimated (but not observed) dry and occult deposition of SO_4 to these sites ranges from 80% to 270% of wet deposition. The uncertainty in estimated critical load (given the assumed dry and occult deposition with which the sites were calibrated) is well within this range, suggesting that one of the largest limitations in obtaining reliable estimates of future critical loads for a site is obtaining a reasonable estimate of current deposition.

The average uncertainty widths for critical load estimates are greatest (+/- 31% to 36%) for target year 2020, and least (+/- 17% to 21%) for target year 2100 (Table 34). For a given target year, there is no apparent variation in the uncertainty width across the target ANC values (Table 34). Contrary to the pattern in average uncertainty widths for simulated variables (which increased into the future), the uncertainty in critical loads estimates seems to be greatest for target years nearest to the present. Estimates of critical loads derived for farther into the future are apparently more reliable for a given site than estimates of loads necessary to produce changes on relatively short time scales.

J. Effects of Streamwater Acidification on Aquatic Biota in the Southeastern U.S.

The status of stream biology within the study area and the effects of acidic deposition on aquatic chemistry and biology have been evaluated in conjunction with a number of regional studies (e.g., Baker et al. 1991; Charles 1991; Herlihy et al. 1993, 1996; SAMAB 1996; Sullivan et al. 2002a). Stream chemistry and biological resources have been intensively studied at a few locations, including some streams in Shenandoah National Park (Bulger et al. 1999, Sullivan et al. 2003) and the St. Marys River (Webb et al. 1989) in Virginia and also in Great Smoky Mountains National Park (GSMNP) in Tennessee and North Carolina (Rosemond et al. 1992). These case studies illustrate the types of responses to loadings of acidic deposition that would be expected in sensitive stream reaches on national forest lands within the study area.

Changes in stream acid-base chemistry, including pH, ANC, inorganic Al, and Ca^{2+} , can affect in-stream biota. The organisms most likely to respond include fish species (such as trout,

dace, sculpins, and minnows) and aquatic insects (SAMAB 1996). Acidification effects on aquatic biota are most commonly evaluated using either ANC or pH as the chemical indicator criterion. pH is more closely tied to physiological response mechanisms. For modeling studies, however, ANC is generally preferred because stream acidification models do a better job projecting ANC than they do pH. ANC criteria have been used for evaluation of potential acidification effects on fish communities. The utility of these criteria lies in the association between ANC and the surface water constituents that directly contribute to or ameliorate acidity-related stress, in particular pH, Ca^{2+} , and Al.

In most stream survey areas, quantification of biological responses to streamwater acidification is not possible, given the scarcity or absence of dose-response data. Most available dose-response data for the southeastern United States have been generated from studies of streams in Virginia or in GRSMNP. In general, we expect dose-response functions throughout North Carolina, South Carolina, and Tennessee to be similar. Data with which to evaluate acidification relationships have been scarce from North Carolina and Tennessee until the recent stream survey work by the Forest Service. This is unfortunate in view of the richness of cold-water stream resources in this state. For example, of the 33,000 miles of potential wild trout streams in the Southern Appalachian Assessment area, 32% are in North Carolina (SAMAB 1996).

Aquatic impacts of acidic deposition have been most thoroughly studied for fish. Effects of low pH and ANC on several individual fish species, including many that are found in southern Appalachian Mountain streams, have been well documented (Baker et al. 1990b). In particular, a great deal of research has focused on brook trout.

1. Effects on Brook Trout

Fish communities of Appalachian Mountain streams may contain a variety of species, but are often dominated by trout, especially native brook trout. Brook trout is often selected as an appropriate indicator species for acidification effects on in-stream biota because it is the only trout species native to streams in the southern Appalachian Mountains and because residents place great recreational and aesthetic value on this species (SAMAB 1996). It is important to note, however, that brook trout is a relatively acid-tolerant species. Many other fish species, including non-native rainbow (*Oncorhynchus mykiss*) and brown (*Salmo trutta*) trout, as well as a variety of other native fish species, are more acid-sensitive than brook trout. In many

Appalachian Mountain streams that have been acidified by acidic deposition, brook trout is the last species to disappear; it is generally lost at pH near 5.0 (MacAvoy and Bulger 1995), which generally corresponds in these streams with ANC near zero.

Introduced rainbow and brown trout are found at lower elevations throughout the study region. Brook trout dominate at higher elevations, with up to several kilometers of coexistence with the non-native trout species (Larson and Moore 1985).

Effects on biota are generally evaluated either with respect to impacts on a particular species or as impacts on the diversity of fish or other potentially sensitive life form. Bulger et al. (2000) developed ANC thresholds for brook trout, which are presented in Table 35. These values were based on annual average streamwater chemistry, and therefore represent chronic exposure conditions. The likelihood of additional episodic stress is incorporated into the categories in the manner in which they are interpreted. For example, the episodically acidic response category, which has chronic ANC in the range of 0 to 20 $\mu\text{eq/L}$, represents streams which are expected to acidify to ANC near or below zero during rainfall episodes. In such streams, sublethal and/or lethal effects on brook trout are possible.

Table 35. Brook trout acidification response categories developed by Bulger et al. (2000).

Response Category	Chronic ANC Range ($\mu\text{eq/L}$)	Expected Response
Suitable	> 50	Reproducing brook trout expected if other habitat features are also suitable
Indeterminate	20 to 50	Brook trout response expected to be variable
Episodically acidic	0 to 20	Sub-lethal and/or lethal effects on brook trout are possible
Chronically acidic	< 0	Lethal effects on brook trout probable

Streams that have annual average ANC greater than about 50 $\mu\text{eq/L}$ are generally considered suitable for brook trout because they have a large enough buffering capacity that persistent acidification poses no threat and there is little likelihood of storm-induced acidic episodes lethal to brook trout. As a result, reproducing brook trout populations are expected if the habitat is otherwise suitable (Bulger et al. 2000). Streams having ANC between 50 and 150 $\mu\text{eq/L}$, however, may periodically experience episodic chemistry that affects species more sensitive than brook trout. Streams having annual average ANC from 20 to 50 $\mu\text{eq/L}$ may or may not experience episodic acidification during storms that can be lethal to juvenile brook trout,

as well as other fish. The occurrence of episodic acidity depends on a number of hydrologic, physical, and chemical characteristics that cannot be readily predicted (Bulger et al. 2000). Streams that are episodically acidic (annual average ANC from 0 to 20 $\mu\text{eq/L}$) are marginal for brook trout because acidic episodes are likely (Hyer et al. 1995), although the frequency and magnitude of episodes vary. Streams that are chronically acidic (annual average ANC less than 0 $\mu\text{eq/L}$) generally cannot support healthy brook trout populations (Bulger et al. 2000).

Survival of brook trout eggs and fry were evaluated in the Fish in Sensitive Habitats (FISH) Project in three Shenandoah National Park streams exhibiting different ANC. These early life stages are more sensitive to the adverse effects of acidity than are adult brook trout. MacAvoy and Bulger (1995) conducted four one to three-month long field bioassays in three streams of differing ANC in Shenandoah National Park. Paine Run experienced episodic chemistry as low as ANC -3 $\mu\text{eq/L}$ and pH 5.3. At Staunton River, the ANC remained above 45 $\mu\text{eq/L}$ and pH ranged from 6.2 to 6.8. Piney River maintained ANC above 120 $\mu\text{eq/L}$ and pH above 6.8. A total of 18,000 hatchery brook trout eyed eggs through fry were exposed to ambient water chemistry, including significant rain events. In three of the four bioassays, embryos/fry showed poorer survivorship in the low-ANC Paine Run compared to the high-ANC Piney River. In the fourth bioassay, poor survivorship occurred in all three streams due to drought conditions. Trout in the intermediate ANC Staunton River showed variable survivorship even though pH never fell below 6.0. The investigators attributed the observed differential survival to both chronic and episodic water chemistry (Bulger et al. 1999).

It is important to note, however, that acidity is not the only stress factor that influences the distribution of brook trout in southern Appalachian Mountain streams. Other habitat characteristics, including water temperature, can be important. In addition, it is likely that brook trout populations at many locations have been affected by encroachment by introduced rainbow trout (c.f., Larson and Moore 1985).

2. Sublethal Effects on Fish

Sensitive species, such as blacknose dace (*Rhinichthys atratulus*) are impacted in the pH range 5.5 to 6.0. Sublethal effects, such as reduction in the condition factor (an index to describe the relationship between fish weight and length), have been shown for blacknose dace near pH 6.0 (Dennis and Bulger 1995). This species is widely distributed in Appalachian Mountain streams and is nearly as tolerant of low pH and ANC as the brook trout.

Fish with higher condition factor are more robust than fish having low condition factor. Condition factor, expressed as fish weight/length³ multiplied by a scaling constant (Everhart and Youngs 1981), is interpreted as depletion of energy resources such as stored liver glycogen and body fat. Dennis and Bulger (1995) measured condition factor in 1,202 blacknose dace from three streams of different ANC values in Shenandoah National Park. K-values in the low-ANC stream were significantly lower (11%) than measurements for fish in the two higher-ANC streams. Whole-body sodium concentrations during summer baseflow, measured as an additional test of sublethal stress, were highest in the low-ANC stream, and lowest in the stream having highest ANC. Dennis and Bulger (1995) suggested that ion regulation in the lowest-ANC stream may be more metabolically costly because of chronic sublethal pH stress. The fish may maintain higher body Na⁺ concentrations in order to provide a buffer against episodic pH depressions in the more acidic stream.

The strongest relationship between condition factor and stream acid-base chemistry was found for the minimum pH recorded over the previous three years (corresponding with the approximate life span of blacknose dace). Observed differences in condition factor with decreasing pH were attributed to the likelihood that maintenance of internal chemistry in the more acidic streams would require energy that otherwise would be available for growth and weight gain (Dennis and Bulger 1995, Webb 2003).

As another component of the FISH project, condition factor was compared in populations of blacknose dace in Shenandoah National Park in 11 streams spanning a range of pH/ANC conditions (Bulger et al. 1999). Figure 34 shows the highly significant relationship between mean stream pH and condition factor in blacknose dace. Note that the four populations represented on the left side of the figure all have mean pH values within or below the range of critical pH values, at which negative populations effects are likely for the species (Baker and Christensen 1991). That poor condition is related to population survival is suggested by the extirpation in 1997 of the blacknose dace population from the stream (Meadow Run) with the lowest pH and ANC (J. Atkinson, pers. comm.2002; Figure 34).

The results of the condition factor comparisons among the 11 streams indicated that the mean length-adjusted condition factor of fish from the stream with the lowest ANC was about 20% lower than that of the fish in best condition. No previous studies had reported changes in condition factor of blacknose dace during acidification. Comparisons with the work of Schofield and Driscoll (1987) and Kretser et al. (1989) suggest that pH in the low-pH Shenandoah National

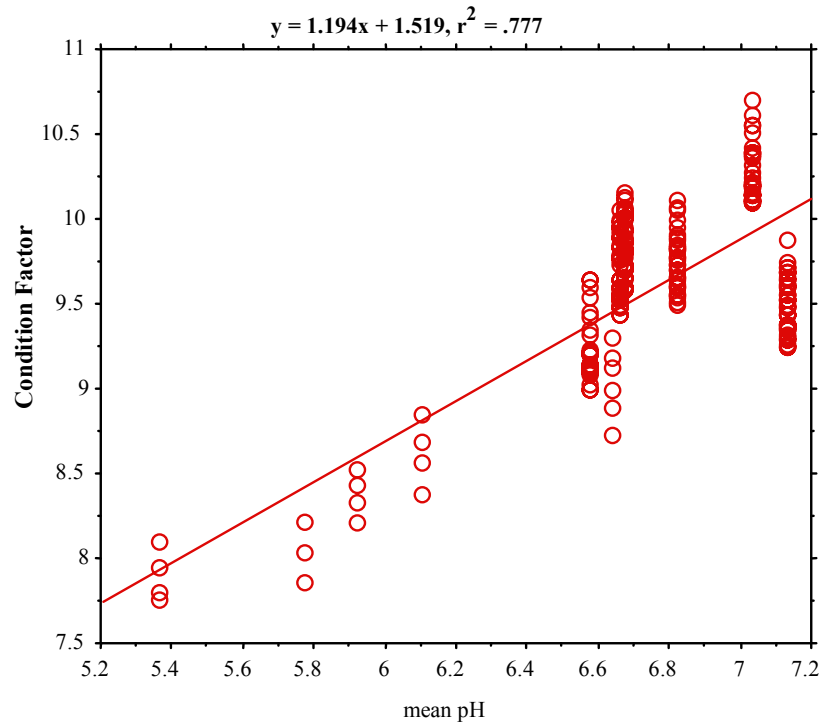


Figure 34. Length-adjusted condition factor (K), a measure of body size in blacknose dace (*Rhinichthys atratulus*) compared with mean stream pH among 11 populations (n=442) in Shenandoah National Park. Values of pH are means based on quarterly measurements, 1991-94; K was measured in 1994. The regression analysis showed a highly significant relationship ($p \leq 0.0001$) between mean stream pH and body size, such that fish from acidified streams were less robust than fish from circumneutral streams.

Park streams is also near or below the limit of occurrence for blacknose dace populations in the Adirondack region of New York (Sullivan et al. 2003).

Smaller blacknose dace body size could result from direct toxicity (e.g., elevated energy use to compensate for sublethal ionoregulatory stress) or from reduced access to food or lower food quality (Baker et al. 1990b). Primary productivity is low in headwater streams and lower still in softwater headwaters, which are more likely to be acidified. Production of invertebrates is likely to be low in such streams as well (Wallace et al. 1992). Thus, lower food availability cannot be ruled out as a potential contributor to lowered condition in Shenandoah National Park blacknose dace populations. Nevertheless, reduced growth rates have been attributed to acid stress in a number of other fish species, including Atlantic salmon, chinook salmon, lake trout, rainbow trout, brook trout, brown trout, and Arctic char. Furthermore, the blacknose dace population in poorest condition in Shenandoah National Park occurred in a stream with mean pH below the minimum recorded for blacknose dace populations in Vermont, New Hampshire,

Maine and New York (Baker et al. 1990b). The four blacknose dace populations in poorest condition in Shenandoah National Park occurred in streams at or below the critical pH for the species, where adverse effects due to acidification are likely to be detectable at the population level (Baker et al 1990a). Consequently, acid stress is probably at least partly responsible for the lower condition of blacknose dace populations in Shenandoah National Park, though lower food availability, either resulting from the nature of softwater streams or exacerbated by acidification, cannot be ruled out (Sullivan et al. 2003).

It is possible that smaller body size in blacknose dace is the result of energy transfer from somatic growth to physiological maintenance, secondary to chronic sublethal acidification stress. It is well known that chronic sublethal stress reduces growth in fish, as well as reproductive success (Wedemeyer et al. 1990). Chronic sublethal stress caused by pH levels below about 6.0 may have serious effects on wild trout populations. There is an energy cost in maintaining physiological homeostasis; the calories used to respond to stress are a part of the fish's total energy budget and are unavailable for other functions, such as growth (Schreck 1981, 1982).

The energy costs to fish for active iono-osmoregulation can be substantial (Farmer and Beamish 1969, Bulger 1986). The concentrations of serum electrolytes (such as sodium [Na^+] and chloride [Cl^-]) are many times higher (often 100-fold higher) in fish blood than in the freshwaters in which they live. The active uptake of these ions occurs at the gills. Because of the steep gradient in Na^+ and Cl^- concentrations between the blood and freshwater, there is constant diffusional loss of these ions, which must be replaced by energy-requiring active transport. Low pH increases the rate of passive loss of blood electrolytes (especially Na^+ and Cl^-); and Al elevates losses of Na^+ and Cl^- above the levels due to acid stress alone (Wood 1989).

3. Effects on Fish Species Richness

Effects of streamwater acidification on fish species richness have been studied in the St. Marys River and in Shenandoah National Park. At both locations, fish species richness has been found to be closely associated with stream acid-base chemistry. The St. Marys River studies have examined changes in one stream over time. The Shenandoah National Park studies have examined differences across streams at a given time (space-for-time substitution analysis).

Bugas et al. (1999) conducted electrofishing in the St. Marys River in 1976, and every two years from 1986 through 1998. Systemic streamwater acidification occurred during the study period. Sampling occurred at six sites between the wilderness area boundary at the

downstream end (Station A) and the headwaters (Station F) over a distance of about 8 km (Table 36). The number of fish species in the St. Marys River within the wilderness declined from 12 in 1976 to 4 in 1998. Three of the four species present in 1998 (brook trout, blacknose dace, fantail darter) are typically the only fish species present in streams having similar levels of acidity in Shenandoah National Park (Bulger et al. 1999, Webb 2003). Bugas et al. (1999) reported that successful brook trout reproduction in the St. Marys River occurred only one year out of four during the period 1995 through 1998. Eight fish species were recorded in one or more early years, but have not been observed in more recent years (Table 37). Several, including blacknose dace, rainbow trout, and torrent sucker, showed a pattern of being progressively restricted over time to lower reaches (Table 37), which generally have higher ANC.

Rosyside dace (*Clinostomus funduloides*) and torrent sucker (*Thoburnia rhothocea*) were last present in 1996; Johnny darter (*Etheostoma nigrum*) and brown trout were last present in 1994; rainbow trout and longnose dace (*Rhinichthys cataractae*) were last present in 1992; bluehead chub (*Nocomis leptocephalus*) and smallmouth bass (*Micropterus dolomieu*) were last present in 1990 and 1988, respectively; white sucker (*Catostomus commersoni*) and central stoneroller (*Campostoma anomalum*) were last present in 1986. Of the four remaining species, three (blacknose dace, fantail darter [*Etheostoma flabellare*]), and mottled sculpin [*Cottus bairdi*]) have declined in density and/or biomass; the fourth remaining species is brook trout, the region's most acid tolerant species; this population has fluctuated, and reproductive success has been sporadic. Blacknose dace, once abundant throughout the river, remain only at the lowest sampling station, which has the highest pH, and at such low numbers (five individuals in 1998) that they might be strays from downstream. For some of the species (smallmouth bass, white sucker, the three trout, and blacknose dace) the critical pH is known (see Table 38), and their decline and/or extirpation, given the pH of the river, is not surprising.

Table 36. Electrofishing stations in the St. Marys River watershed, Augusta County, Virginia.

Station	Elevation (m)	Stream km from Wilderness Boundary	Sample Length (m)	Sample Area (ha)
A	524	0.35	171	0.12
B	570	2.61	123	0.08
C	610	3.97	127	0.10
D	646	5.11	76	0.04
E	661	5.98	161	0.07
F	722	8.13	91	0.02

Table 37. Fish distribution in St. Marys River by sample year and sample station. Letter denotes uppermost station in the watershed where individual species were collected. ^a (Source: Bugas et al. 1999)

Fish Species	1976	1986	1988	1990	1992	1994	1996	1998
Brook Trout	F	F	F	F	F	F	F	F
Blacknose Dace	E	E	E	C	A	B	A	A
Fantail Darter	C	C	C	C	C	B	B	B
Mottled Sculpin	B	B	B	B	B	B	B	B
Rosyside Dace	B	B	B	B	A	B	A	
Torrent Sucker	C	B	B	B	B		A	
Rainbow Trout	E	E	C	C	C			
Longnose Dace	B	A			A			
Johnny Darter	A					A		
White Sucker	B	A						
Bluehead Chub	A			A				
Central Stoneroller		A						
Smallmouth Bass			B					
Brown Trout	C					A		
Total Species	12	10	8	8	8	7	6	4

^a Stream reaches are described in Table 36. Station A is the lowest (524 m) and Station F is the highest (722 m)

Table 38. Critical pH thresholds for fish species which might be expected to occur within the study area. (Source: Bulger et al. 1999)

Common Name	Latin Name	Family	Critical pH ^a Threshold
Blacknose Dace	<i>Rhinichthys atratulus</i>	Cyprinidae	5.6 to 6.2
Creek Chub	<i>Semotilus atromaculatus</i>	Cyprinidae	5.0 to 5.4
White Sucker	<i>Catostomus commersoni</i>	Catostomidae	4.7 to 5.2
Brook Trout	<i>Salvelinus fontinalis</i>	Salmonidae	4.7 to 5.2
Brown Trout	<i>Salmo trutta</i>	Salmonidae	4.8 to 5.4
Rainbow Trout	<i>Oncorhynchus mykiss</i>	Salmonidae	4.9 to 5.6
Rock Bass	<i>Ambloplites rupestris</i>	Centrarchidae	4.7 to 5.2
Smallmouth Bass	<i>Micropterus dolomieu</i>	Centrarchidae	5.0 to 5.5

^a threshold for serious adverse effects on populations (from Baker & Christensen 1991)

Although there are known differences in acid sensitivity among fish species, experimentally-determined acid sensitivities are available for only a minority of freshwater fish species. Baker and Christensen (1991) reported critical pH values for 25 species of fish. They defined critical pH as the threshold for significant adverse effects on fish populations. The range of response within species depends on differences in sensitivity among life stages, and on different exposure concentrations of calcium (Ca^{2+}) and Al. The approximate critical pH is known for eight fish species that might be expected to occur in streams within the study area for this report (Table 38). The reported range of pH values represents the authors' estimate of the uncertainty of this threshold. The ranges of response, based on multiple studies for each species, are shown in Table 38. To cite a few examples, blacknose dace (*Rhinichthys atratulus*) is regarded as very sensitive to acid stress, because population loss due to acidification has been documented in this species at pH values as high as 6.1; in field bioassays, embryo mortality has been attributed to acid stress at pH values as high as 5.9. Embryo mortality has occurred in common shiner (*Luxilus cornutus*) at pH values as high as 6.0. Although the critical pH range for rainbow trout (*Oncorhynchus mykiss*) is designated as 4.9-5.6, adult and juvenile mortality have occurred at pH values as high as 5.9. Brown trout (*Salmo trutta*) population loss has occurred over the pH range of 4.8-6.0, and brook trout fry mortality has occurred over the range of 4.8-5.9 (Baker and Christensen 1991). Relative sensitivities can be suggested by regional surveys as well, although interpretation of such data is complicated by factors that correlate with elevation. Such factors, including habitat complexity and refugia from high-flow conditions, often vary with elevation in parallel with acid sensitivity. It is the difference in acid tolerance among species that produces a gradual decline in species richness as acidification progresses, with the most sensitive species lost first. Some Blue Ridge streams can become too acidic even for brook trout, as evidenced by the absence of the species from streams with mean pH < 5.0 in Great Smoky Mountains National Park (Elwood et al. 1991). Adult brook trout are more tolerant of acidity than are adult blacknose dace. For both species, the early life stages are more sensitive than the adults, and brook trout young are actually more sensitive than blacknose dace adults (Bulger et al. 1999). Blacknose dace spawn during summer and the eggs and very young fry are therefore somewhat insulated from the most acidic episodes, which typically occur during cold-season, high-flow conditions.

A direct outcome of fish population loss as a result of acidification is a decline in species richness (the total number of species in a lake or stream). This appears to be a highly predictable

outcome of regional acidification, although the pattern and rate of species loss varies from region to region. Baker et al. (1990b) discussed 10 selected studies which documented this phenomenon, with sample sizes ranging from 12 to nearly 3,000 lakes or streams analyzed per study.

Relatively less is known about changes in fish biomass, density and condition (robustness of individual fish) which occur in the course of acidification. Such changes result in part from both indirect and direct interactions within the fish community. Loss of sensitive individuals within species (such as early life stages) may reduce competition for food among the survivors, resulting in better growth rates, survival, or condition. Similarly, competitive release (increase in growth or abundance subsequent to removal of a competitor) may result from the loss of a sensitive species, with positive effects on the density, growth, or survival of competitor population(s) of other species (Baker et al. 1990b). In some cases where acidification continued, transient positive effects on size of surviving fish were shortly followed by extirpation (Bulger et al. 1993).

The FISH Project quantified the effects of acidification on streams within Shenandoah National Park (Bulger et al. 1999). This project examined fish response on multiple levels, including condition factor for blacknose dace, increased mortality of brook trout, and fish species richness. All three indicators of biological response were closely correlated with stream acid-base chemistry. In southern Appalachian streams, local species richness of the various animal life forms depends on thermal regime, water chemistry, patterns of discharge, plus substrate type and geomorphology (Wallace et al. 1992). Acidity is only one factor among many determining species composition of Appalachian streams. This is an important consideration when evaluating the biological implications of changes in water chemistry.

Bulger et al. (1999) demonstrated a strong relationship between stream ANC and the number of fish species found in each stream (Figure 35). Presumably, streamwater acidification reduced species richness by eliminating the more sensitive species as pH and ANC declined (Baker and Christiansen 1991). In addition, however, it is likely that watershed area played a role in this observed relationship. Smaller watershed areas are often associated with fewer fish species.

There are clear patterns in species distribution from headwater streams in the uplands to larger rivers in the lowlands. These patterns can also be seen in community comparisons among reaches at different elevations. The clearest pattern is that species richness increases in a

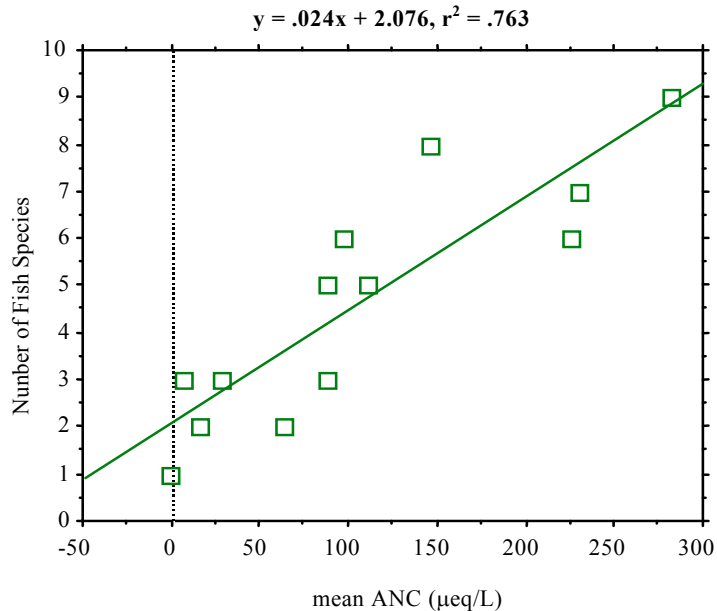


Figure 35. Number of fish species among 13 streams in SHEN. Values of ANC are means based on quarterly measurements, 1987-94. The regression analysis showed a highly significant relationship ($p \leq 0.0001$) between mean stream ANC and number of fish species. Streams having ANC consistently $< 75 \mu\text{eq/L}$ had three or fewer species. (Source: Bulger et al. 1999, Sullivan et al. 2003)

downstream direction. There is typically a rather small number of species that can tolerate the high current velocities and low pH often found in upstream reaches. In the highest headwaters, fish are absent and are replaced by salamanders. The highest-elevation fish species present is usually brook trout, typically joined downstream by dace, sculpin, and darter, and perhaps by introduced brown or rainbow trout (Wallace et al. 1992). Data from Cherokee National Forest (J. Herrig, pers. comm., October, 2006) illustrate strong correlations between fish species richness and physical stream parameters (Table 39). The number of fish species within streams in this forest is strongly associated with elevation and stream gradient. Correspondingly, the number of fish species increased with stream order, from 9 species for first order streams to 103 fish species for 6th order streams. Higher order streams (7th through 9th) do not show any further increase in number of fish species.

In most river systems in the southeastern U.S., the highest-elevation streams are the smallest, coldest, highest-gradient (steepest) streams, with fewest species of fish. There is a general pattern of increasing fish species richness and abundance from higher to lower elevation, probably resulting in part from a greater variety of habitat types (including spawning and nursery areas) and food sources in downstream reaches. Thus, many headwater streams with

Table 39. Correlation of three physical stream parameters and fish species numbers on the Cherokee National Forest. (J. Herrig, pers. comm., October, 2006)

Elevation

Elevation (feet)	Fish Species
<1000	121
1000 - 1500	82
1500 - 2000	56
2000 - 2500	30
2500 - 3000	16
3000 - 3500	11
3500 - 4000	3
>4000	0

Gradient

% Gradient	Fish Species
<2	135
2 - 4	61
4 - 6	43
6 - 8	25
8 - 10	21
10 - 12	15
12 - 14	5
14 - 16	5
16 - 18	4
18 - 20	2
>20	0

Stream Order

Stream Order	Fish Species
9	79
8	60
7	85
6	103
5	57
4	50
3	26
2	9
1	9

lower pH might be expected to have fewer fish species than lower elevation streams, regardless of pH.

The factors that affect the distribution and abundance of aquatic biota are important from an acidification standpoint because the effects of acidification interact with other habitat characteristics to determine the species and biological communities that will occur in a given stream reach. The effects of streamwater acid-base chemistry on aquatic biota were summarized by Baker et al. (1990b) and Bulger et al. (1999). Suitable streamwater acid-base chemistry is a necessary, but not necessarily sufficient, prerequisite for supporting brook trout, or any other species or biological community.

Bulger et al. (1999) developed a relationship between number of fish species in Shenandoah National Park streams and the minimum recorded ANC for each stream (Figure 35). They found a rather consistent decrease in the number of fish species observed, with decreasing minimum ANC, from 9 species at ANC of about 160 $\mu\text{eq/L}$ to 1 to 3 species at ANC near zero. The best fit regression line for this data set was also presented by Bulger et al. (1999), suggesting, on average, a loss of one species for every 21 $\mu\text{eq/L}$ decline in minimum ANC.

Bulger et al. (1999) concluded that the most important cause of the observed decline in species richness with decreasing ANC was acid stress. An additional causal factor is likely the increase in the number of available aquatic niches as you move from upstream locations (which are often low in pH and ANC in this region) to downstream locations (which are seldom low in pH and ANC). The relative importance of this latter factor, compared with the importance of acid stress, in determining this relationship, is not known.

The relationship between fish species richness and ANC developed by Bulger, et al. (1999) was observed for streams in the mountainous watersheds of Shenandoah National Park, where the minimum recorded ANC accounted for 82% of the variance in fish species richness. However, Bulger et al. (1999) investigated both very small headwater streams and larger rivers, some of which contained many species of fish; those containing more than five species of fish generally are not small headwater streams (Table 40). Small headwater streams contain one species (brook trout) or in some cases a few species, and as you move higher in the stream system, eventually contain no fish species at all.

The observed relationship between fish species richness and ANC does not prove that ANC is solely responsible for fish species richness. Correlation provides one line of evidence

Table 40. Median streamwater ANC and watershed area of streams used by Bulger et al. (1999) to evaluate the relationship between ANC and fish species richness.

Site ID	Watershed Area (mi ²)	Median ANC (µeq/L)	Number of Fish Species ^a
Smaller Watersheds (< 4 mi²)			
North Fork Dry Run	0.9	48.7	2
Deep Run	1.4	0.3	N.D. ^b
White Oak Run	1.9	16.2	3
Two Mile Run	2.1	10.0	2
Meadow Run	3.4	-3.1	1
Brokenback Run	3.9	74.4	3
Larger Watersheds (4-10 mi²)			
Staunton River	4.1	76.8	5
Piney River	4.8	191.9	7
Paine Run	4.9	3.7	3
Hazel River	5.1	86.8	6
White Oak Canyon	5.4	119.3	7
N. Fork Thornton River	7.3	249.1	9
Jeremy's Run	8.5	158.5	6
Rose River	9.1	133.6	8

^a Data regarding number of fish species were provided by A. Bulger, University of Virginia.

^b Data were not available regarding the number of fish species in Deep Run.

that a causal relationship may exist. It is always possible that the true causal agent co-varies with the variable under study (in this case ANC).

Median streamwater ANC values and watershed areas are shown in Table 40 for the 14 streams used by Bulger et al. (1999) to develop the relationship between ANC and fish species richness. These study streams include several much larger streams, which are actually called “rivers” (North Fork Thornton River, Piney River, Rose River, Staunton River, Hazel River). All of the “rivers” have watersheds larger than 4 mi² and ANC higher than 75 µeq/L. In contrast, the majority (but not all) of the “runs” (or streams) have watershed area smaller than 4 mi² and ANC less than 20 µeq/L. All of the streams that have watershed areas smaller than 4 mi² have 3 or fewer known species of fish present. The ANC of the smaller streams is determined largely by the underlying geology. All of the streams having larger watersheds (> 4 mi²) have 3 or more known fish species; 7 of 8 have 5 or more species; and the average number of fish species is 6. There is no clear distinction between river and run, but it is clear that as small streams in this region combine and flow into larger streams and eventually to rivers, two things happen: acid-sensitivity generally declines, and habitat generally becomes suitable for additional fish species.

Watershed area can be important in this context because smaller watersheds generally contain smaller streams having less diversity of habitat, more pronounced impacts on fish from high flow periods, and often lower food availability. Such issues interact with other stresses, including acidification, to determine habitat suitability.

4. Acidification Effects on Aquatic Invertebrates

It has been well-documented that low streamwater pH can be associated with reductions in benthic invertebrate density (Hall et al. 1980, Townsend et al. 1983, Aston et al. 1985, Burton et al. 1985, Kimmel et al. 1985), and also species richness or diversity (Townsend et al. 1983, Raddum and Fjellheim 1984, Kimmel et al. 1985, Burton et al. 1985, Hall and Ide 1987, Rosemond et al. 1992, Peterson and van Eeckhaute 1992, Sullivan et al. 2003). Effects on invertebrate density are not universal; a number of studies have found no density effects (Harriman and Morrison 1982, Simpson et al. 1985, Ormerod et al. 1987, Winterbourn and Collier 1987). However, a decrease in species richness with decreasing pH has been found in almost all such studies (Rosemond et al. 1992), and this finding has been especially pronounced for order Ephemeroptera (mayflies) and the grazer feeding group. Trichoptera are also highly sensitive.

Porak (1981) found that the caddisfly *Diplectrona* and the stoneflies *Leuctra*, *Acroneuria*, and *Peltoperla* appeared to be tolerant of *Anakeesta* leachates, with associated acidification in receiving streams. All species of mayfly were intolerant of the acid condition in these streams.

Kaufmann et al. (1999) concluded that “documented biological changes due to acid deposition in invertebrate communities have been limited to northern states, Canada, and Scandinavian countries.” This was attributed to northern waters being generally more sensitive to acidification because of loss of soils from glaciation. However, it may also be partially due to a paucity of studies in southeastern states. Nevertheless, some very useful aquatic invertebrate dose-response data are available for a number of streams in the southeastern United States, including Shenandoah National Park, the St. Marys River in George Washington National Forest, Virginia, and GRSMNP. Such studies do, in fact, suggest that biological changes have occurred in invertebrate communities in some areas.

Benthic macroinvertebrates have been monitored in Shenandoah National Park streams since 1986 as part of the Long-Term Ecological Monitoring System (LTEMs). Moeykens and Voshell (2002) examined these data, comparing them with streamwater chemistry in the park.

Their analysis was based on interpretation of 10 chemical and physical variables measured at 89 sites (28 low-ANC sites and 61 higher-ANC sites) for which macroinvertebrate data were available. They compared their results for streams in the park with similar analyses for 45 sites (13 low-ANC sites and 32 higher-ANC sites) elsewhere in the Blue Ridge ecoregion of Virginia. The macroinvertebrate communities in both data sets were characterized with 12 robust variables thought to represent the ecological function and composition of these communities. Moeykens and Voshell (2002) concluded that the higher-ANC streams in the park had “superior ecological condition” which was comparable to the best that can be found among the streams in the broader Blue Ridge ecoregion. However, they also concluded that acidification of streamwater causes the only conspicuous degradation of macroinvertebrate communities in some low-ANC streams. Other disturbances, such as fire and flood, did not appear to have had noticeable long-term effects on the streams. Moeykens and Voshell (2002) concluded that acidified streams in the park host fewer invertebrate taxa and fewer functional groups than streams with higher pH and ANC. Similar findings were reported earlier for Shenandoah National Park streams by Feldman and Connor (1992).

As described by Kauffman et al. (1999), the record for St. Marys River provides a unique opportunity to compare reliable macroinvertebrate data on an acidified stream over a 60-year time span. Surber (1951) collected the earliest benthic data for St. Marys River. Starting in August of 1935, and continuing for two years, he collected 20 samples per month from the river’s main stem. Subsequent data were collected by the Virginia Department of Game and Inland Fisheries (VDGIF) in 1976 and then biennially beginning in 1986 (Kauffman et al. 1999) using methods comparable to those used for the 1930s collections. The VDGIF data were collected at six evenly spaced locations extending the length of the main stem above the wilderness boundary. The later collections were made in June, and only June data are used in the following comparisons.

The total abundance of mayfly (Ephemeroptera) larva in the St. Marys River has dramatically decreased over the 60-year period, and two of the mayfly genera, *Paraleptophlebia* and *Epeorus*, were last collected in 1976. Mayflies are known to decline in species abundance and richness with increasing acidity (Peterson and Van Eeckhaute 1992, Kobuszewski and Perry 1993). The total abundance of caddisfly (Trichoptera) larva also declined dramatically over the 60-year period of record. Baker et al. (1990b) indicated that caddisflies exhibit a wide range of response to acidity, with some species affected by even moderate acidity levels. The total

abundance of the larva of the stonefly (Plecoptera) genera *Leuctra/Alloperla* has dramatically increased over the 60-year period. Increased abundance of these stoneflies in acidified waters has been well documented (Kimmel and Murphy 1985). Another insect family that has prospered in St. Marys River is the midge (Chironomidae), whose larval population has increased tenfold since the 1930s collections. Increased midge abundance in acidified waters has also been well documented (Kimmel and Murphy 1985, Baker et al. 1990b).

The St. Marys River watershed is underlain by siliceous bedrock and had measured ANC in the late 1990s generally between about -5 and 15 $\mu\text{eq/L}$, with a 10-year median value of 4 $\mu\text{eq/L}$ (Webb 2003). In 1936 and 1937, the numbers of benthic invertebrate taxa collected were 32 and 29, respectively. The number of taxa declined to 23 in 1976. During the period 1986 through 1998, the average number of benthic invertebrate taxa collected was down to 17, varying from 13 to 22 in a given year (Webb 2003). Acid-sensitive mayflies and caddisflies decreased in abundance, and some more acid-tolerant invertebrate taxa increased in abundance (Webb 2003), probably due to reduced competition.

Effects of acidification on aquatic invertebrates were investigated in streams within GRSMNP by Rosemond et al. (1992). They determined patterns in benthic invertebrate community structure in four streams, with baseflow pH ranging from 4.5 to 6.8. A number of studies and analyses were conducted at these stream locations, including identification of sensitive species, toxicity (*in situ* transplant and exposure) tests, and assessment of differences in species richness, diversity, and density.

Rosemond et al. (1992) transplanted and placed into flow-through chambers three species of acid-sensitive mayfly between high and low pH streams. A transplant of *Drunella conestee* from pH 6.4 to 5.0 did not show a statistically-significant increase in mortality. In contrast, transplants of *Stenonema* sp. and *Epeorus pleuralis* from pH 6.4 to 5.0 showed 100% mortality of *Epeorus pleuralis* (20% for control) and 18% mortality for *Stenonema* sp. (0% for control) after 8 and 4 day exposures, respectively. Similarly, Mackay and Kersey (1985) found that *Stenonema* sp. was restricted to pH greater than 5.3 in streams in Ontario, and similar results have been found for *Epeorus* in the northeastern United States (Hall et al. 1980, Simpson et al. 1985).

Rosemond et al. (1992) found increasing species richness of Ephemeroptera (Richness = $2.09 \times \text{pH} - 8.5$; $r^2=0.96$; $P < 0.05$) and Trichoptera (Richness = $1.52 \times \text{pH} - 4.1$; $r^2=0.96$; $P < 0.05$) with increasing pH, but no significant relationship with pH for Plecoptera. In both the

Ephemeroptera and Trichoptera evaluations, there was found about 1 ½ to 2 additional insect species of a given order for a rise in pH of 1 pH unit. Mayflies of the family Ephemerellidae appeared to be especially acid-sensitive, and are often restricted to streams having pH above about 5.0 (Fiance 1978, Harriman and Morrison 1982, Simpson et al. 1985, Rosemond et al. 1992).

Many stream invertebrate communities are dominated by early life stages of insects that have great dispersal abilities as flying adults. In all likelihood, currently-acidified streams hosted more diverse invertebrate communities in pre-industrial times. Given the relatively rapid recovery time (about 3 years) of stream invertebrate communities from disturbance, more productive and diverse invertebrate communities might be among the first positive results of lower acid deposition. On the other hand, if streamwater ANC declines further, we can expect macroinvertebrate diversity to decrease.

5. Species – ANC Relationships for Aquatic Invertebrates

Quantitative relationships between invertebrate communities and streamwater quality in SHEN streams were analyzed by Sullivan et al. (2003). The objective was to describe and quantify the correlations between streamwater ANC and various measures of invertebrate community status in the streams. There are 14 SWAS streams in the park that have quarterly water quality data extending back to 1988. The means, maxima, and minima of solute concentrations in these streams were calculated for the period 1988 to 2001 for use in the analyses (Table 41).

The LTEMs benthic invertebrate data for the period June 1988 through June 2000 were selected for comparison with water quality data. There are five phyla of benthic macroinvertebrates represented in the samples (Annelida, Arthropoda, Mollusca, Nematoda, and Platyhelminthes). Because of their importance to park streams and known sensitivity of many taxa to acidification, this analysis was limited to the data collected on aquatic insects (class Insecta of the phylum Arthropoda).

There are nine orders of aquatic insects present in the Shenandoah National Park LTEMs samples: Coleoptera, Collembola, Diptera, Ephemeroptera, Hemiptera, Megaloptera, Odonata, Plecoptera, and Trichoptera. From these nine orders of aquatic insects, 79 families have been collected. Not all families are present in each stream. The total number of insect families found in a given stream during the sampling period varied from 21 to 56. Of the nine orders of aquatic

Table 41. Minimum, average and maximum ANC values in the 14 SHEN study streams during the period 1988 to 2001 for all quarterly samples. The data cover 14 water years except for VT75 (11 years).

Site ID	Watershed	ANC ($\mu\text{eq/L}$)		
		Minima	Mean	Maxima
Siliciclastic Bedrock Class				
DR01	Deep Run	-9.5	2.9	24.4
VT35 (PAIN)	Paine Run	-1.3	7.0	19.5
VT36	Meadow Run	-11.4	-1.3	6.2
VT53	Twomile Creek	2.8	15.2	38.6
WOR1	White Oak Run	3.6	27.7	58.6
Granitic Bedrock Class				
NFDR	North Fork Dry Run	22.5	65.6	187.8
VT58	Brokenback Run	44.0	87.9	155.4
VT59 (STAN)	Staunton River	46.1	87.3	189.4
VT62	Hazel River	54.4	95.6	163.6
Basaltic Bedrock Class				
VT51	Jeremys Run	93.7	217.2	542.5
VT60 (PINE)	Piney River	118.7	228.4	382.9
VT61	North Fork Thornton River	156.2	286.6	452.9
VT66	Rose River	94.4	150.2	229.2
VT75	White Oak Canyon Run	81.2	138.6	237.2

insects found in SHEN streams, there were three which were most abundant both in terms of frequency of occurrence in samples and total numbers of individuals collected: Ephemeroptera (mayflies); Plecoptera (stoneflies); and Trichoptera (caddisflies). The use of these three orders as indicators of acidification response in streams is well established. A combined metric based on all three families, the Ephemeroptera-Plecoptera-Trichoptera (EPT) index, is one measure of stream macroinvertebrate community integrity. This is the total number of families in the three insect orders present in a collection. These orders contain families of varying acid sensitivity so the index value (the number of families) is lower at acidified sites (c.f., SAMAB, 1996). In general, mayflies (Ephemeroptera) are most sensitive to acidity, and stoneflies (Plecoptera) are least sensitive. Caddisflies (Trichoptera) are intermediate (Peterson and Van Eeckhaute 1992).

Positive relationships were observed between mean and minimum streamwater ANC and the number of families in the orders Ephemeroptera and Plecoptera, but less so for Trichoptera (Figure 36). The total numbers of individuals in the orders Ephemeroptera and Trichoptera were also related to the mean and minimum ANC values of the 14 streams (Figure 37). The EPT index provides a single measure of all three orders and was, as expected, also related to mean and minimum streamwater ANC (Figure 38). These data can be used to estimate the increase in the number of individuals of the orders Ephemeroptera or Trichoptera, or the number of families of all three orders, that might be expected to occur in response to a given increase or decrease in stream ANC.

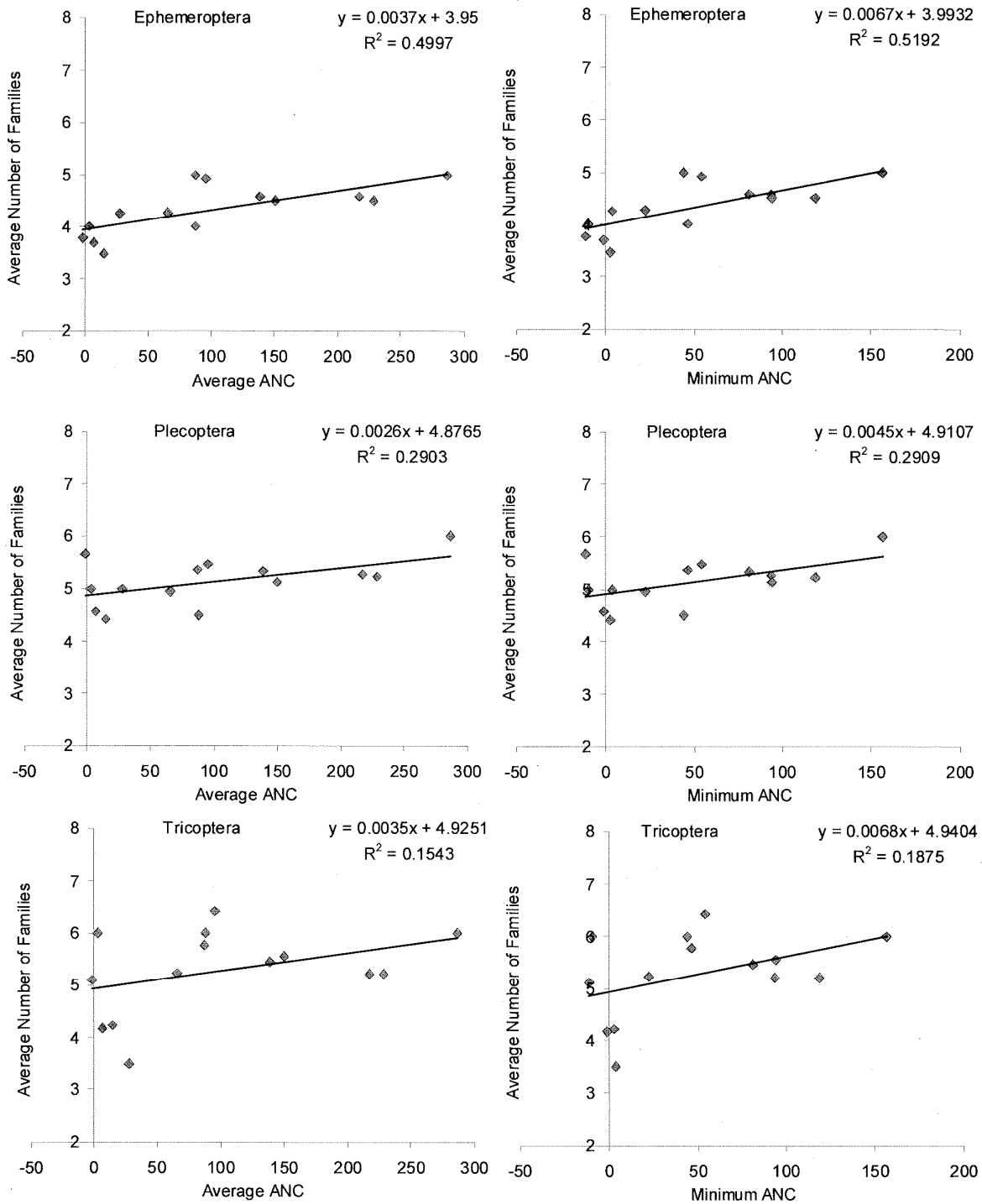


Figure 36. Average number of families of aquatic insects in a sample for each of 14 streams in SHEN versus the mean (left) or minimum (right) ANC of each stream. The stream ANC values are based on quarterly samples from 1988 to 2001. The invertebrate samples are contemporaneous. Results are presented for the orders Ephemeroptera (top), Plecoptera (center), and Tricoptera (bottom). The regression relationship and correlation are given on each diagram. (Source: Sullivan et al. 2003)

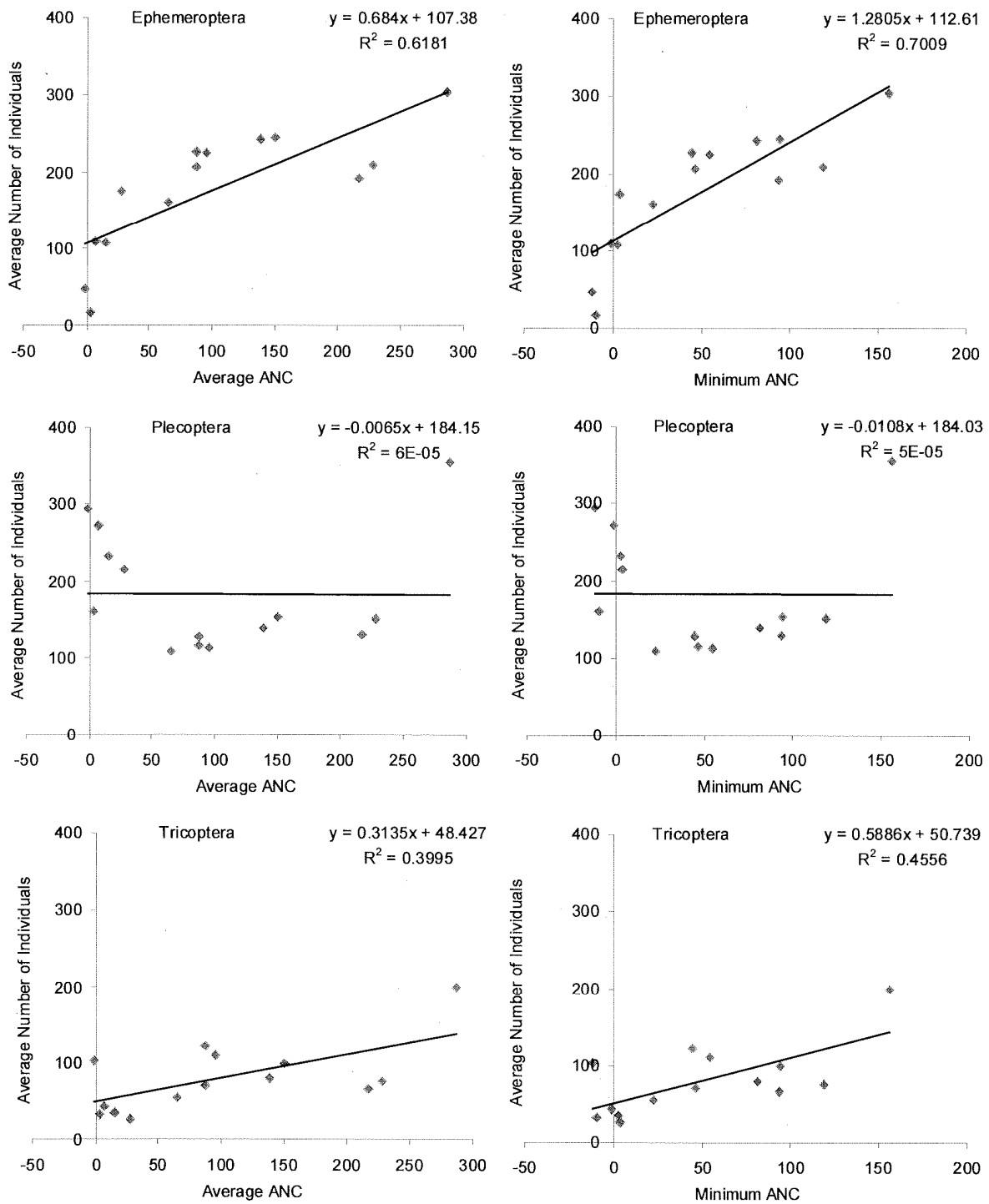


Figure 37. Average total number of individuals of aquatic insects in a sample for each of 14 streams in SHEN versus the mean (left) or minimum (right) ANC of each stream. The stream ANC values are based on quarterly samples from 1988 to 2001. The invertebrate samples are contemporaneous. Results are presented for the orders Ephemeroptera (top), Plecoptera (center), and Trichoptera (bottom). The regression relationship and correlation are given on each diagram. (Source: Sullivan et al. 2003)

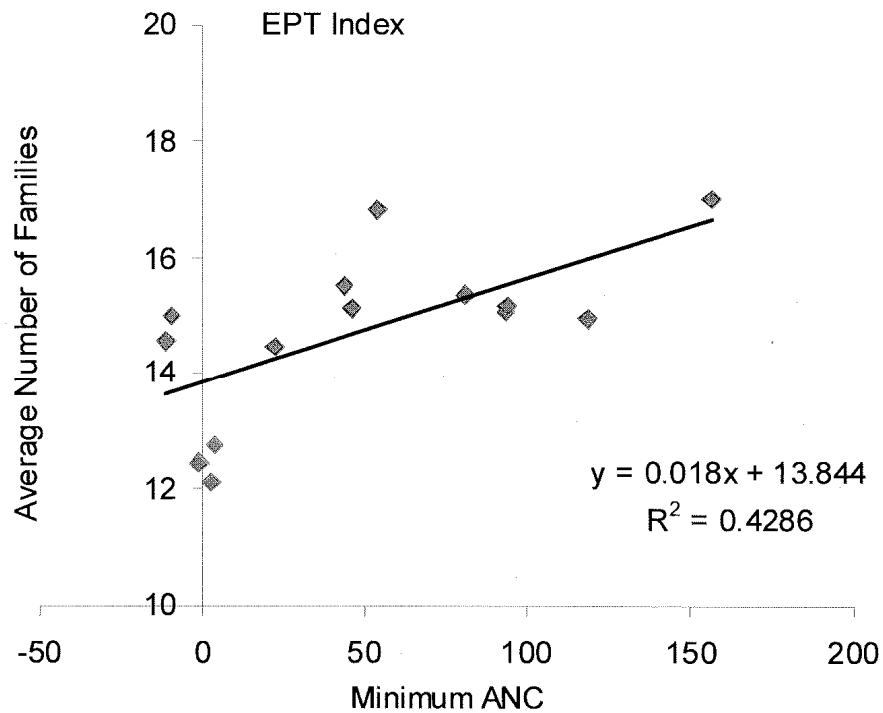
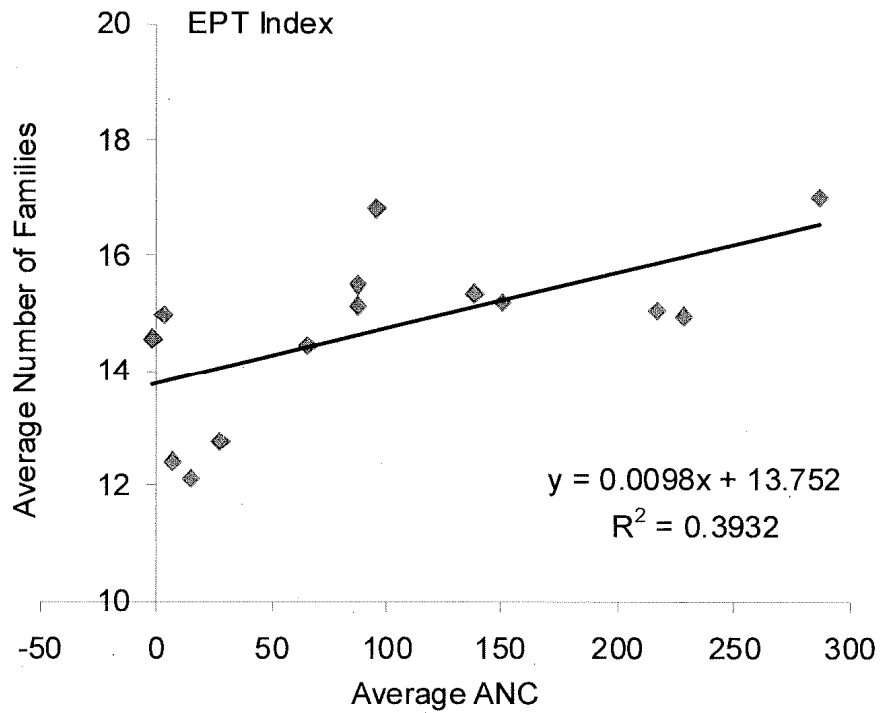


Figure 38. Average EPT index in a sample for each of 14 streams in SHEN versus the mean (top) or minimum (bottom) ANC of each stream. The stream ANC values are based on quarterly samples from 1988 to 2001. The invertebrate samples are contemporaneous. The regression relationship and correlation are given on each diagram. (Source: Sullivan et al. 2003)

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