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Target loads of atmospheric sulfur deposition for the protection and recovery of acid-sensitive streams in the Southern Blue Ridge Province

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A B S T R A C T

An important tool in the evaluation of acidification damage to aquatic and terrestrial ecosystems is the critical load (CL), which represents the steady-state level of acidic deposition below which ecological damage would not be expected to occur, according to current scientific understanding. A deposition load intended to be protective of a specified resource condition at a particular point in time is generally called a target load (TL). The CL or TL for protection of aquatic biota is generally based on maintaining surface water acid neutralizing capacity (ANC) at an acceptable level. This study included calibration and application of the watershed model MAGIC (Model of Acidification of Groundwater in Catchments) to estimate the target sulfur (S) deposition load for the protection of aquatic resources at several future points in time in 66 generally acid-sensitive watersheds in the southern Blue Ridge province of North Carolina and two adjoining states. Potential future change in nitrogen leaching is not considered. Estimated TLs for S deposition ranged from zero (ecological objective not attainable by the specified point in time) to values many times greater than current S deposition depending on the selected site, ANC endpoint, and evaluation year. For some sites, one or more of the selected target ANC critical levels (0, 20, 50, 100 μeq/L) could not be achieved by the year 2100 even if S deposition was reduced to zero and maintained at that level throughout the simulation. Many of these highly sensitive streams were simulated by the model to have had preindustrial ANC below some of these target values. 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 thresholds.

1. Introduction

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. Soils within these watersheds have developed from the slow breakdown of parent rock material which can be inherently low in base cations (Elwood et al., 1991). Ecosystem sensitivity to acidification and the potential effects of atmospheric sulfur (S) deposition on surface water quality have been well studied in this region (Baker et al., 1990a, b; NAPAP, 1991; Sullivan et al., 2004). Sulfur is the primary determinant of precipitation acidity and sulfate (SO_4^{2-}) is the dominant acid anion associated with acidic streams throughout most of the southern Appalachian Mountains region (Sullivan et al., 2004). 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 (NO_3^-) concentrations in streamwater are high in some high-elevation streams, especially in Great Smoky Mountains National Park, but are generally low throughout most of the region (Sullivan et al., 2004). Because SO_4^{2-} is the primary agent of acidification in most streams within the study region, the TL analyses presented here focus on S loading.

Soil and drainage water acidification developed in this region over a period of many decades in response to high levels of atmospheric S and to a lesser extent nitrogen (N) deposition. Many streams in the national forests show signs of acidification from atmospheric deposition, including streams in wilderness areas that are administered by the FS: Linville Gorge, Joyce Kilmer-Slickrock, and Shining Rock wilderness areas (Elliott et al., 2008). However, S deposition has been declining throughout the eastern United States since about the early 1980s and further decreases are

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expected in the future (U.S. EPA, 2008). More information is needed regarding the watershed responses that should be expected.

Computer models can be used to predict pollution effects on ecosystems and to perform simulations of future ecosystem response. The Model of Acidification of Groundwater in Catchments (MAGIC), a lumped-parameter, mechanistic model, has been widely used throughout North America and Europe to project streamwater and soil 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. Many streams in the national forests in this region show signs of acidification from atmospheric deposition (Sullivan et al., 2004, 2010).

Forest managers in the southern Blue Ridge Province need to know what levels of S deposition would be needed to protect those streams and soils that are not yet heavily impacted, and to restore those that are already impacted. Therefore, the principal objective of the study reported here was to use the MAGIC model to estimate for a group of 66 streams and their watersheds in the southern Blue Ridge region the loads of S deposition that would be needed to protect those streams that are not yet strongly acidified, and to restore streams that are already acidified, to specific chemical criteria values. Our focus is mainly on the more acid-sensitive streams within the region. These will likely be the subject of management actions to prevent and/or reverse acidification.

The need for emissions controls to protect natural resources has given rise to the concept of critical load (CL) of atmospheric pollutants. The CL has been 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” (Nilsson, 1986; Nilsson and Grennfelt, 1988). The basic CL concept is relatively straightforward when applied to resource protection from acidification effects. The CL is the level of atmospheric acidic deposition that is sustainable in the long-term without harmful effects on sensitive receptors. CLs, by definition, must be calculated under assumed steady-state conditions (Posch et al., 2001). However, management of natural resources often requires estimates of the deposition load that would be protective at given points in time as well as in the long-term sustainable sense. Such time-dependent estimates are called target loads (TL; Henriksen and Posch, 2001) or dynamic CLs (U.S. EPA, 2008). We use the TL terminology in this paper.

Appropriate indicators of resource health or damage (criterion variables) must be selected in order to use the CL or TL approach. For each criterion variable, a critical value must be specified that defines the threshold of onset of adverse effects (Henriksen and Posch, 2001). For protection of surface waters from acidification, acid neutralizing capacity (ANC) is the most commonly used criterion variable (U.S. EPA, 2008). ANC critical values have usually been set at 0, 20, or 50 μeq/L in various European and North American applications. Selection of critical values is confounded by the existence of streams that are acidic (ANC ≤ 0 μeq/L) 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, 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 (Elwood et al., 1991; Sullivan, 2000).

Interpretation of TL estimates is complicated by differences between sites in need of protection of existing condition and those in need of recovery from previous acidification. For streams having ambient ANC below the specified critical criterion threshold (e.g., ANC = 20 or 50 μeq/L), the calculated TL represents the deposition load that will allow chemical recovery in the selected endpoint year. For streams having ambient ANC above the specified critical criterion threshold, the calculated TL represents the increased deposition load that can be tolerated without forcing stream ANC to decline to a value below the critical criterion in the selected endpoint year.

2. Methods

2.1. Site selection

The USDA Forest Service has, in recent years, collected stream chemistry samples in the Pisgah and Nantahala National Forests in western North Carolina, the 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 FS which receive special protection from air pollution damage under the Clean Air Act: Joyce Kilmer-Slickrock, Linville Gorge, and Shining Rock wildernesses. Data on additional streams in this region having available water chemistry were compiled for SAMI by Sullivan et al. (2004). Sites were selected for modeling in this project from these datasets, as described by Sullivan et al. (2010), who presented historical and future scenario modeling results for these same 66 watersheds. Site selection focused largely on inclusion of watersheds known to be acid-sensitive. Most were represented by one stream sample, collected during spring (Sullivan et al., 2010). Summary statistics for the modeled streams are given in Table 1. TLs were simulated for each of the 66 stream watersheds (Fig. 1) to which the effects model was successfully calibrated by Sullivan et al. (2010).

Table 1
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[a]	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 base saturation	%	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
S deposition[b]	kg S/ha/yr	6.51	9.96	11.21	13.36	22.17	12.00

[a] Stream chemistry data were compiled by Sullivan et al. (2004), based on samples collected during the period 1999–2005.
[b] Total wet plus dry deposition estimated for the year 2005 from interpolated wet deposition measurements by the National Atmospheric Deposition Program (NADP) and ASTRAP model estimates of dry deposition (Sullivan et al., 2004).

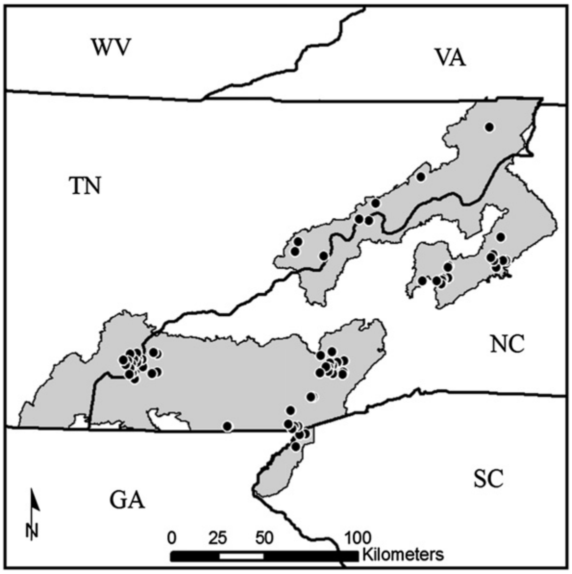

Fig. 1. Map showing locations of 66 stream sites modeled for this project. Forest Service lands within the Blue Ridge province (shaded) constitute the study area.

2.2. Modeling approach

TLs were simulated using MAGIC, a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on soil and surface water (lake or stream) chemistry (Cosby et al., 1985a, b, 2001). The model simulates soil solution chemistry and surface water chemistry to predict the monthly and annual average concentrations of major ions in solution. 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 aluminum and dissolution-speciation of inorganic carbon; 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 in response 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.

The aggregated nature of the MAGIC model requires that it be calibrated to observed data before it can be used to examine potential watershed 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, the simulated-minus-observed values of the criterion variables usually converge to zero (within some specified tolerance). The model is then considered calibrated.

Given estimates 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. (1985a, b, 1989). MAGIC has been used to reconstruct the history of acidification, to calculate CLs and TLs, and to simulate future trends on a regional basis and in a large number of individual watersheds in both North America and Europe (Cosby et al., 1989; Hornberger et al., 1989; Jenkins et al., 1990; Wright et al., 1994; Sullivan et al., 2007).

2.3. Target loads modeling

TL modeling was used to determine threshold levels of sustained atmospheric deposition of S below which adverse effects to particular sensitive aquatic receptors would not be expected to occur in specified endpoint years. In addition, interactions were evaluated between the critical ANC endpoint value specified and the time period for which the TL was calculated.

The MAGIC model was used in an iterative fashion to calculate the S deposition that would cause the streamwater ANC of each of the modeled watersheds to either increase or decrease (depending on the ambient value in the Reference Year, 2005) to reach the specified critical levels in the specified endpoint years. For these analyses, the critical ANC levels were set equal to 0, 20, 50, and 100 μeq/L, the first two of which are believed to approximately correspond with the thresholds of chronic and episodic damage to relatively acid-tolerant brook trout populations (Bulger et al., 2000). Other more acid-sensitive species of fish and other aquatic biota may be impacted at higher ANC values (Table 2; Cosby et al., 2006). In order to conduct this TL 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. This is reflected in the generally low NO_3^- concentrations observed in streamwater (Table 1). For this analysis, both future N deposition and leaching were 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, 2040, and 2100.

In using MAGIC to estimate TLs for the ANC levels and years discussed above, it was necessary to specify the temporal pattern of deposition changes used to drive the TL calculation. Changes in deposition leading to the TL were begun in the simulations in 2009 and completed by 2018, assuming a linear decrease (or increase) to the TL value from 2009 to 2018. Deposition was then held constant at the TL value from 2018 until the end of the simulation, which was determined by the year selected for evaluation of the target ANC.

Table 2
Approximate damage thresholds for fish species expected to occur in streams within the study area and for which sensitivity to low pH has been determined.

Common name	Family	Critical pH Threshold[a]	Approximate ANC Threshold[b]
Blacknose Dace	Cyprinidae	5.6–6.2	14 to 33
Creek Chub	Cyprinidae	5.0–5.4	−4 to 9
White Sucker	Catostomidae	4.7–5.2	−17 to 3
Brook Trout	Salmonidae	4.7–5.2	−17 to 3
Brown Trout	Salmonidae	4.8–5.4	−12 to 9
Rainbow Trout	Salmonidae	4.9–5.6	−8 to 14
Rock Bass	Centrarchidae	4.7–5.2	−17 to 3
Smallmouth Bass	Centrarchidae	5.0–5.5	−4 to 11

[a] Threshold for serious adverse effects on populations (from Baker and Christensen, 1991).
[b] ANC damage thresholds were approximated from published pH thresholds and an empirical relationship between pH and ANC for streams included in this study.

3. Results

3.1. Hindcast simulations

Simulated and observed stream chemistry were in close agreement for all of the streams modeled in this study (Sullivan et al., 2010). MAGIC model simulations predicted that historical (pre-acidification) stream ANC values were above 20 μeq/L in all modeled watersheds, but below 50 μeq/L in 38% of the watersheds and below 100 μeq/L in 86% of the watersheds (Sullivan et al., 2010). These hindcast results provide context for evaluating the results presented below. The minimum simulated pre-acidification ANC among the modeled streams was 30 μeq/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 median of the modeled streams was acidified from a pre-acidification ANC = 65 μeq/L to ANC = 36 μeq/L in 2005.

3.2. Estimates of target loads for the years 2020, 2040, and 2100

The levels of S deposition were simulated that would be expected to cause streamwater ANC to increase or decrease to four specified critical levels or ANC endpoints (0, 20, 50, and 100 μeq/L) for each modeled stream. The first three of these critical levels have been utilized in CL and TL studies elsewhere (c.f., Kämäri et al., 1992; Sullivan et al., 2005). Estimated TLs for S deposition ranged from zero (ecological objective not attainable in the endpoint year regardless of deposition level) to values many times greater than current S deposition depending on the selected site, ANC endpoint, and evaluation year. For most streams, ANC = 0 was attainable in any year (2020, 2040, or 2100); quite high levels of S deposition could occur and the majority of streams would still have ANC above 0 μeq/L. In marked contrast, most of the modeled streams could not achieve ANC = 100 μeq/L in any of the future years considered, even if S deposition was reduced to zero and maintained at that level. This feature of the model output is illustrated in Fig. 2 using the years 2040 and 2100 as examples. Stream ANC above 0 was

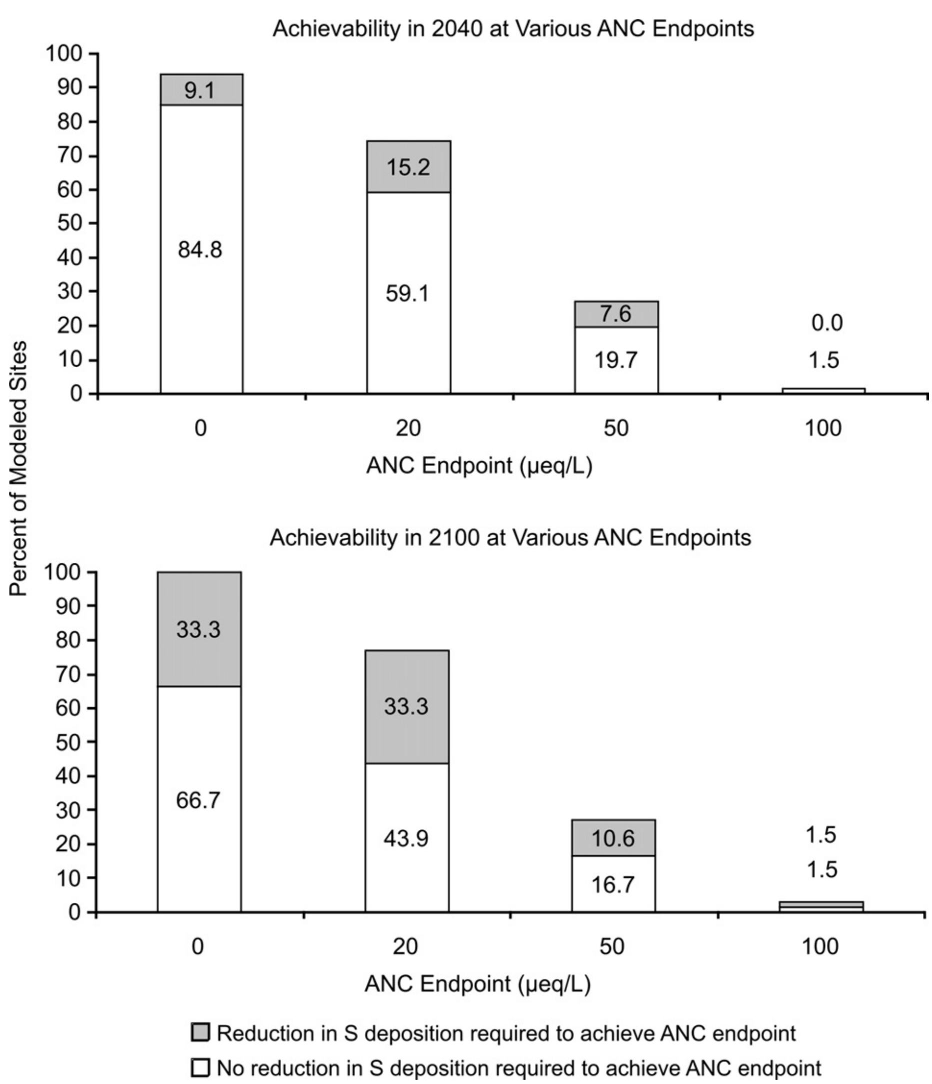

Fig. 2. Comparison of the extent to which various ANC criteria (0, 20, 50, 100 μeq/L) are simulated to be achievable in response to simulations based on the median model calibration in the year 2040 (top panel) and 2100 (bottom panel). Results are presented as percent of modeled streams. Sites for which 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.

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. In order to achieve ANC values above 0 μeq/L in 2100, more of the sites would require reductions in deposition from current levels. However, only 1.5% of the modeling sites could achieve ANC = 100 μeq/L in the year 2040 and 3% in the year 2100.

Almost all sites were simulated to maintain ANC above 0 μeq/L without reducing S deposition (Table 3). The critical criterion ANC = 100 μeq/L was not generally attainable regardless of deposition level, and most of the study streams were simulated to have had ANC below 100 μeq/L prior to the onset of acidic deposition (Sullivan et al. 2010). The ANC criteria values equal to 20 and 50 μeq/L were often achievable (Fig. 2) and are associated with lower levels of ecological harm than the criterion ANC = 0.

4. Discussion

4.1. Target loads

The calculated S deposition TLs for the modeled streams depend upon the selected chemical criterion (critical ANC value), and the future year for which the evaluation was made (Table 3). The relationships between TL, ANC criterion value, and evaluation year are affected, in turn, by the current acid-base status of the streams (as indicated by the observed reference year stream chemistry). The relationships are qualitatively different for those sites with Reference Year stream ANC below the ANC criterion value ("damaged" streams) compared with those sites for which Reference Year ANC values are above the ANC criterion ("undamaged" streams).

Consider first the effects of selection of the evaluation year for a given ANC criterion value. Higher TLs can be tolerated for damaged streams if one is willing to wait until 2100 to recover to the critical ANC target level, as compared with more stringent deposition reductions (lower TLs) required to attain recovery to specified ANC values by 2020 or 2040. For undamaged streams on the other hand, more S deposition (reflected in higher TLs) can be tolerated for short-term protection to 2020 or 2040 (Fig. 3a), versus more stringent deposition reductions (lower TLs) required to protect these streams until 2100.

The effects of selection of the ANC criterion value for a given evaluation year are similarly asymmetric. For a fixed period of response in damaged streams, relatively lower TLs are required to recover to higher critical ANC values (i.e., 100 μeq/L) than are needed to recover to lower (0 or 20 μeq/L) critical ANC values. However, for undamaged streams, higher TLs can be tolerated (cf., Fig. 3b) if one wishes to allow acidification to an ANC between 0 and 20 μeq/L (episodic acidification effects on brook trout likely) than if one wishes to be more restrictive and prevent acidification to ANC below 50 μeq/L (biological effects of acidification likely on biota other than brook trout). The effects described above can be illustrated by plotting the calculated TLs of S deposition required to prevent streamwater acidification to ANC values below 0 and 20 μeq/L as a function of measured Reference Year ANC (Fig. 4a) for the study sites. These model simulation results demonstrate that most modeled streams with Reference Year ANC ≤ 20 μeq/L would require low TL values to recover to an ANC of 20 μeq/L by the year 2100. At low measured ANC values, TLs were consistently near zero; at higher ANC values, TLs were more variable. Some streams having ANC above 50 μeq/L exhibited low TLs, whereas others had TLs much higher than current deposition levels (Table 1). Stronger relationships were observed when modeled TL was plotted as a function of the ratio of streamwater ANC to streamwater SO_4^{2-} concentration (Fig. 4b). Watersheds having the lowest Reference Year ANC and the highest Reference Year SO_4^{2-} concentrations in streamwater had the lowest TL. This was especially true for calculations of the TLs to achieve ANC values of 0 and 20 μeq/L. For this analysis, 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 TLs occur where SO_4^{2-} is more mobile.

Table 3
Achievability of ANC (μeq/L) endpoints in a variety of future years for 66 modeled streams.

	2020		2040		2100	
	No.	%	No.	%	No.	%
Endpoint ANC = 0						
Total achievable streams	60	91	62	94	66	100
Streams requiring reduction in S deposition[a]	3	5	6	9	22	33
Endpoint ANC = 20						
Total achievable streams	42	64	47	71	51	77
Streams requiring reduction in S deposition[a]	5	8	10	15	22	33
Endpoint ANC = 50						
Total achievable streams	14	21	18	27	18	27
Streams requiring reduction in S deposition[a]	1	2	5	8	7	11
Endpoint ANC = 100						
Total achievable streams	1	2	1	2	2	3
Streams requiring reduction in S deposition[a]	0	0	0	0	1	2

[a] 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.

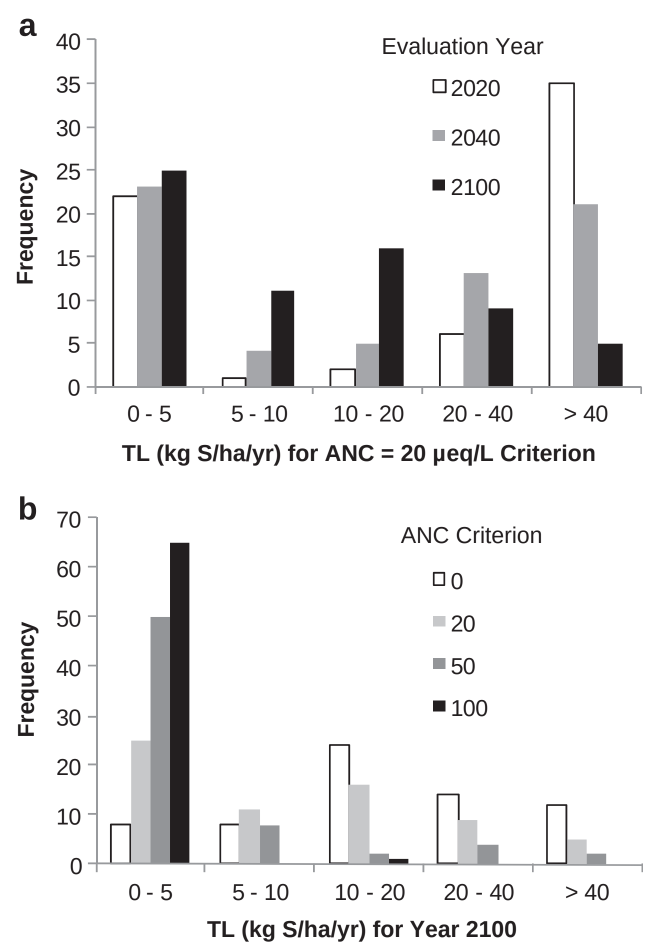

Fig. 3. Frequency distributions of TLs for 66 modeled streams, depending on selection of (a) evaluation year (2020, 2040, 2100) for the ANC = 20 μeq/L criterion and (b) ANC critical criterion (0, 20, 50, 100 μeq/L) evaluated for the year 2100.

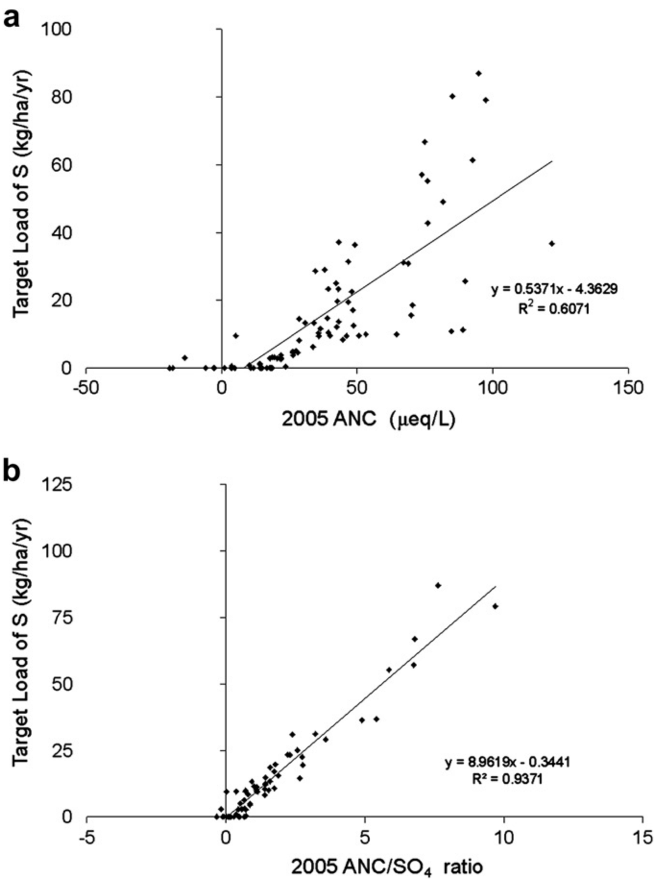

Fig. 4. Target loads of S for 66 modeled sites to achieve streamwater ANC of 20 μeq/L, evaluated in 2100 as a function of (a) Reference Year ANC and (b) Reference Year ANC divided by streamwater SO_4^{2-} concentration. Target load implementation was begun in 2009 and completed in 2018. Target loads are depicted as median simulated values at each site, calculated from multiple (5–10) calibrations of MAGIC for each site.

An advantage of using a dynamic model like MAGIC to calculate TLs is that the model can provide estimates of streamwater ANC and SO_4^{2-} for the study sites prior to the onset of acidic deposition ("preindustrial" estimates). It is reasonable to expect that these preindustrial estimates would reflect the underlying watershed sensitivity to acidification with respect to background ANC and SO_4^{2-} values in the streams. Whereas the Reference Year ANC and SO_4^{2-} values examined above will be correlated with these preindustrial estimates, the Reference Year values also reflect the extent of current acidification and thus are a mixed function of watershed sensitivity and deposition history. The preindustrial streamwater chemistry values derived from the model are presumably a function only of the watershed sensitivity and can be used to examine the variations in TLs calculated for the study region.

The simulated TL was related to the baseline preindustrial ANC of the stream, but only weakly so (Fig. 5a). All streams having TL equal to 0 kg/ha/yr (target not attainable even without any atmospheric S deposition) to protect ANC from falling below 20 μeq/L in the year 2100 had simulated preindustrial ANC below 50 μeq/L. Nevertheless, many streams having higher preindustrial ANC (50–140 μeq/L) had relatively low (<15 kg S/ha/yr) simulated TL. This result was attributable to differences in the dynamics of S retention in watershed soils. The simulated TL was also related to the current Reference Year SO_4^{2-} concentration (Fig. 5b). Streams having the highest TL generally had fairly low concentrations of SO_4^{2-}, suggesting near total retention of atmospherically deposited S at the present time. As a consequence of these dynamics, the TL was strongly correlated with the ratio of preindustrial ANC divided by current Reference Year SO_4^{2-} concentration (Fig. 5c). Streams having low TL were those that had low preindustrial ANC and that currently leach substantial amounts of SO_4^{2-} to streams.

More sites could achieve ANC of 20 μeq/L in the year 2100 (51 modeled sites) as compared with the year 2020 (42 modeled sites; Table 3). Nevertheless, deposition would have to be reduced from ambient values at a third of the modeled sites to achieve ANC = 20 μeq/L in 2100, versus just 8% of the modeled sites to achieve ANC = 20 μeq/L in 2020. For streams that do not have deposition higher than a given TL, higher loads can be tolerated over the shorter term. For streams that are in exceedance, higher loads can be tolerated over the longer term.

Neither ANC = 0 nor ANC = 100 μeq/L seem to be particularly useful targets for land management. Almost all sites can maintain ANC above 0 μeq/L without reducing S deposition below current values (Table 3), and it has been well demonstrated that a variety of

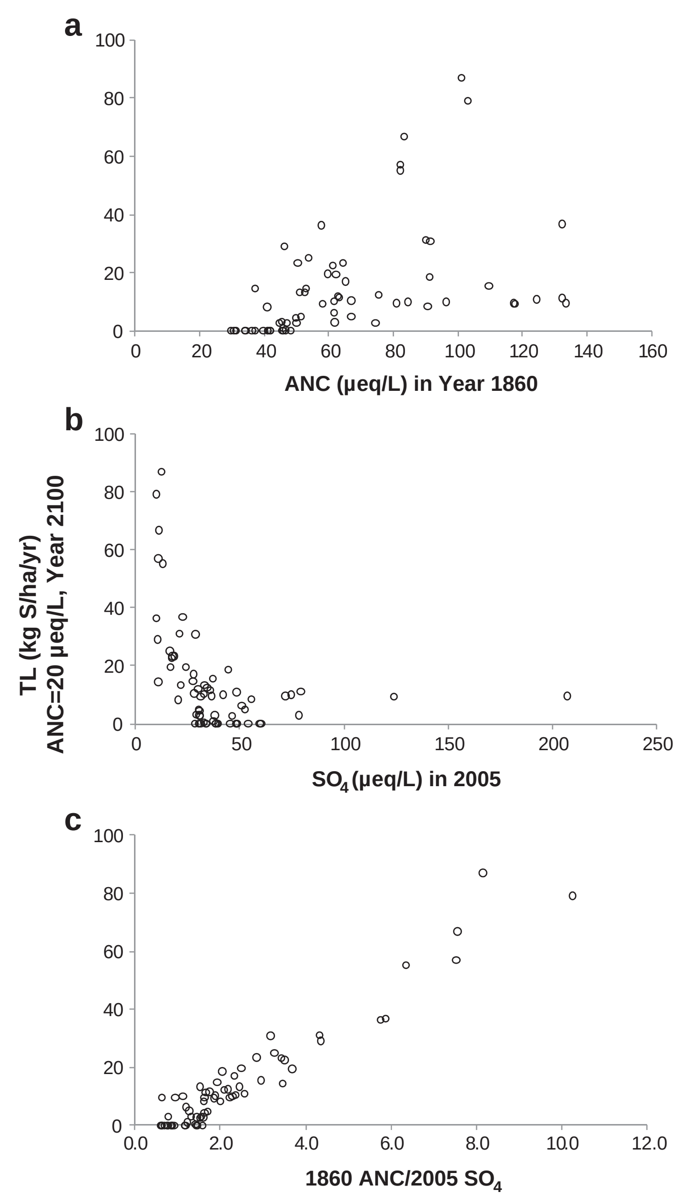

Fig. 5. Target loads of S for 66 modeled streams to achieve streamwater ANC of 20 μeq/L, evaluated in 2100 as a function of (a) preindustrial ANC, (b) Reference Year SO_4^{2-} concentration, and (c) preindustrial ANC divided by Reference Year SO_4^{2-} concentration. Target loads are depicted as described for Fig. 4.

adverse ecological effects occur at ANC near 0 μeq/L (Cosby et al., 2006). ANC of 100 μeq/L or greater is generally not attainable within 100 years, irrespective of the level of S deposition (Table 3), and most of the study streams were simulated to have had ANC below 100 μeq/L prior to the onset of acidic deposition (Sullivan et al. 2010). The ANC criteria values 20 and 50 μeq/L are often achievable within a reasonable timeframe and are associated with lower levels of ecological harm than ANC = 0 μeq/L.

4.2. Uncertainty in model simulations and target loads estimates

The MAGIC model was calibrated 10 times at each of the 66 sites as part of a fuzzy optimization procedure (Cosby et al., 2006; Sullivan et al., 2003) designed to provide estimates of simulation uncertainty. The model results presented here 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.

Uncertainty estimates were derived from the multiple calibrations at each site provided by the fuzzy optimization procedure employed in MAGIC simulations. 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 uncertainty in the assumptions made in calibrating the model and the inherent uncertainties in the available data can be assessed by using all successful calibrations for a site when calculating a TL estimate.

When implemented with the ensemble parameter sets at each site, the model produces an ensemble of TL 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 as standard output are the median values from the ensemble of calibrations for each of the 66 modeled sites. The simulated values in the ensemble can also be used to estimate the magnitude of uncertainty in the projection. Specifically, the difference in any year between the maximum and minimum simulated TL values from the ensemble of calibrated parameter sets at a site can be used to define a confidence width for the simulation at any point in time for that individual site.

Uncertainty widths (maximum minus minimum TL value obtained from multiple calibrations) for TL estimates were derived for 12 different TL analysis protocols: four different target ANC values in three different years (Table 4). The number of sites contributing to an average uncertainty width for a particular TL analysis protocol was variable. Uncertainty widths could only be calculated for sites where a particular TL analysis was possible (target ANC less than historical ANC at the site), and where at least one of the ensemble estimates of the TL at the site was greater than zero. For instance, there were only nine modeled sites that had an historical ANC greater than 100 μeq/L. Therefore, only those nine sites could be analyzed for a TL producing the target ANC of 100 μeq/L. Of those nine sites, only one site gave at least one of the ensemble TL estimates as greater than zero. Therefore, in Table 4, the uncertainty width estimate for target ANC = 100 μeq/L 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 TL protocol was for a target ANC = 0 μeq/L in year 2020, for which 60 sites produced at least one TL estimate greater than zero. The average uncertainty widths presented in Table 4 for target ANCs of 0 and 20 μeq/L are based on between 41 and 60 sites for all years, and are considered robust estimates of uncertainty. Average uncertainty widths for ANC = 50 μeq/L are based on 15–16 samples in a given year and are probably reliable. The result for target ANC of 100 μeq/L ($n = 1$) will not be discussed further.

The average uncertainty widths for TL estimates were not excessively large, ranging from ±17% to ±36% depending on the analysis conditions. The estimated dry and occult deposition of S to sites in this region ranges from 80% to 270% of wet deposition (Sullivan et al., 2004). The uncertainty in estimated TL (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 CL or TL exceedance for a given site is obtaining a reasonable estimate of ambient atmospheric deposition.

The average uncertainty widths for TL estimates are greatest (±31% to 36%) for target year 2020, and least (±17% to 21%) for target year 2100. For a given target year, there is no apparent variation in the uncertainty width across the target ANC values (Table 4). Contrary to the pattern in average uncertainty widths for simulated variables (which increased into the future; Sullivan et al., 2010), the uncertainty in TLs estimates seems to be greatest for target years nearest to the present.

The pattern of larger uncertainty in TL calculations in the short term is due, at least in part, to uncertainty in the timing of future changes in S adsorption in the soils of the study watersheds. The model formulation requires that the S adsorption sites in the soil become increasingly saturated (Turner et al., 1990) in response to increases in S deposition. These adsorption sites desorb and leach previously adsorbed SO_4^{2-} in response to decreases in S deposition. Such soil adsorption–desorption processes can affect the calculation of TLs.

For damaged streams where the calculated TL calls for a reduction in S deposition, SO_4^{2-} appearing in the stream from soil desorption decreases the amount of S that can be derived from atmospheric deposition while maintaining a particular ANC criterion (i.e., the calculated TL will be lower). Similarly, for undamaged streams where the calculated TL allows for an increase in S deposition, some of the increased S deposition will be adsorbed in the soils. This increases the amount of additional S deposition (and thus the TL) that can be tolerated.

The dynamics of S adsorption and the time scale of response as soils adjust to new S deposition levels are poorly known for this region. Uncertainty in TL values would be greatest, therefore, in the short term as the soil adsorption adjusts to the new TL levels. Thus, the lack of knowledge of adsorption dynamics is most crucial in the shorter term. Over longer periods of time, however, the time scale of this transient adjustment in S adsorption becomes moot as the

Table 4
Uncertainty widths (UW, % ±) for estimated target loads, expressed as the difference between the maximum and minimum simulated TL values obtained from the multiple calibrations of the model.

	Uncertainty width for target 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

The uncertainty widths are the average widths for all sites for which a given target load could be estimated. The number of sites averaged (*n*) for each UW is indicated. The different target load analyses are based on target stream ANC values (left column) in target years (top row).

adsorption process has time to reach a new steady state with the TL S deposition level.

4.3. Conclusions and recommendations

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

- Choice of environmental response indicator.
- Selected critical endpoint criterion value(s) for the response indicator.
- Determination of what constitutes "recovery" in the context of this indicator.
- Time period in the future for evaluating the TL.
- Representativeness of the water bodies selected for modeling.
- Major sources and levels of uncertainty.

Thus, there are a number of decisions needed in implementing a TL analysis, and the choices that are made will affect interpretation of the modeling results and their utility with respect to natural resource management.

This is the first regional aquatic TL study conducted in the Southern Blue Ridge, one of the most acid-sensitive and acid-impacted regions in the United States. Results of model simulations and TL calculations presented here will help to inform the development of the CL and TL approaches as potential assessment and policy tools 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 TLs 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 TL and CL values that would sustain sensitive aquatic and terrestrial life forms.

It must be recognized that streamwater chemistry in this region 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 (Turner et al., 1990). The latter process can cause streamwater base cation concentrations and ANC to decrease over time even while SO_4^{2-} and NO_3^- concentrations maintain relatively constant levels (Sullivan et al., 2004). 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.

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