Detailed Documentation of the National Critical Loads Database (NCLD) of Sulfur and Nitrogen, version 3.2.1

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Created for the Critical Loads of Atmospheric Deposition (CLAD) Science Subcommittee of the National Atmospheric Deposition Program (NADP)

This document is intended to provide additional database information to accompany the 12/22/2022 release of the National Critical Load Database (NCLD). Correction to this document were made on 7/6/2023

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National Critical Load Database (NCLD) Information Use Conditions

Disclaimer

The National Atmospheric Deposition Program (NADP) Critical Loads of Atmospheric Deposition (CLAD) Science Committee National Critical Loads Database (NCLD) for Nitrogen (N) and Sulfur (S) was developed cooperatively with individuals or groups sharing critical load (CL) data and is NOT intended to be comprehensive of all known CLs for the U.S. While substantial efforts are made to ensure the accuracy of data and documentation contained in the NCLD, complete accuracy of the information cannot be guaranteed. The qualities and accuracy of the CLs are best described in the associated research publication(s). It is important to review material and information in the cited papers prior to using the CL data within the NCLD. In addition, any opinions, findings, conclusions, or recommendations as part of these datasets do not necessarily reflect the views of CLAD, NADP, and/or respective members' affiliations.

Use Condition and Citation

The intended use of the NCLD is for scientific, policy-related, and/or educational purposes. Any published use of the database information must acknowledge the original source(s) of the data. Each CL value is linked to its origin source(s) through the RefID field. The proper citations for each RefID can be found in **Table 5** of the database. In addition, whenever the Data User presents and/or publishes research based on CLs in the database, NADP and CLAD must be acknowledged as well. A suggested Acknowledgement is:

"We acknowledge the Critical Loads of Atmospheric Deposition (CLAD) Science Committee of the National Atmospheric Deposition Program (NADP) for their role in making available NCLD v3.2 datasets"

and please cite:

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We request one copy of any printed publications using data from the NCLD to be sent to the NADP Program Office at the address below. Citations or electronic copies are acceptable. For online uses, we request that the author notify the Program Office of the URL address of the online publications or website that includes NCLD data. We encourage teachers and professors to send the program office a brief description of how they have used the NCLD in their curriculum. Students who use the NCLD to complete academic assignments are not required to seek permission from the Program Office but must acknowledge NADP and CLAD in any publications (e.g., a thesis).

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Questions, Errors and Corrections

Please contact NCLD manager, Jason Lynch (US EPA) with any questions about the NCLD or to report errors or corrections at lynch.jason@epa.gov or 202-343-9257.

Table of Contents

Introduction 1

This document contains information for understanding the National Critical Loads Database (NCLD) for Nitrogen (N) and Sulfur (S). It describes the origins of the NCLD, process for including studies and datasets, the database variables, how critical load (CL) values were determined, and the sources of information used in compiling the data.

Origins of the NCLD

Beginning in 2006, the primary forum for CLs research and development coordination in the U.S. has been the Critical Loads of Atmospheric Deposition (CLAD) Science Committee of the National Atmospheric Deposition Program (NADP) (http://nadp.sws.uiuc.edu/clad/).
The goals of CLAD are to:

1. Facilitate technical information sharing on CLs topics within a broad multi-agency/entity audience;
2. Fill gaps in CLs development in the U.S.;
3. Provide consistency in development and use of CLs in the U.S.;
4. Promote understanding of CLs approaches through development of outreach and communications materials.

Starting in 2010, the “FOCUS Pilot Study” project within CLAD, gathered and synthesized empirical and calculated CL data and information from dozens of regional- and national-scale projects (Blett et al., 2014). The first round of CL data synthesis was used for an informal, unofficial submission to the UNECE Coordinating Center on Effects (CCE). This unofficial submission represented the interest of a growing CLs science community in the U.S. The CLAD Science Committee members submitted data to this cooperative effort as a productive and meaningful way to share information to improve methods for estimating, calculating, mapping, interpreting, and refining CLs. Since then, the NCLD has been improved and additional CL values and information have been added.

Critical Load and Critical Load Exceedance

Critical load is a quantitative estimate 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 and Grennfelt, 1988, UNECE, 2004). Porter et al., (2005) informally defined a CL as “the threshold of deposition below which specified harmful ecological effects do not occur.” The NCLD contains three different types of CLs: Steady-State Mass Balance, Empirical, and Target loads (TL). A Steady-State Mass Balance CL is derived from mathematical mass-balance models under assumed or modeled equilibrium conditions. An empirical CL is developed using empirical approaches, which involve observed spatial or temporal gradient studies or experimental manipulations of pollutants. A TL is the deposition load that is selected or determined to provide a level of protection for or recovery of sensitive ecosystem components based on time frame for resource protection, feasibility of emissions reductions, and/or other considerations. It can be determined through management or policy considerations, or by using dynamic process-based model that calculate the deposition load that leads to a desired chemical or biological state of an ecosystem in a given future year (Posch et al., 2003). The TL may be set higher or lower than or equal to the CL. Target load has sometimes been referred to as “a dynamic critical load.”

Below is a general overview on calculating CL exceedances for all CL types. A more detail and in-depth discussion of exceedances can be found in Chapter VII: Exceedance calculation of the 2015 ICP Modelling and Mapping Manual (see http://icpmapping.org/Latest_update_Mapping_Manual) (UNECE 2004).

Critical load exceedances can be considered with respect to S, N, and combined S and N deposition depending on the type of CL. For the Forest Soil (FS) and Surface Water (SW) for acidification, these CL exceedances can be considered for S, N, and combined S and N deposition. However, the Empirical (EMP) CLs currently can only be considered with respect to N deposition.

For FS and SW, when considering only S deposition (i.e., N deposition is zero), the exceedance is expressed as the difference between the maximum CL of S (CLmaxS) and total S deposition (**Eq. 1**). If only N deposition is considered (i.e., S deposition is zero), the exceedance is expressed as the difference between maximum CL of N (CLmaxN) and total N deposition (**Eq. 2**).

$$\text{Exceedance(s)} = \text{Total S deposition} - \text{CLmaxS or CLS} \qquad (1)$$

or

$$\text{Exceedance(s)} = \text{Total N deposition} - \text{CLmaxN or CLNS} \qquad (2)$$

In most cases, deposition of both S and N contributes to the exceedance. Calculating a combined S and N CL exceedance is more complex where both CLmaxS (CLS) and CLmaxN (CLNS) are taken together with total N and S deposition and the long-term N removal processes in the soil (e.g., N uptake and immobilization) define a “minimum” CL for N (CLminN). Nitrogen deposition inputs below the CLminN do not acidify, but once CLminN is reached, the level of N deposition above CLminN contributes towards acidification. This creates a “three-node line” on a graph representing the acidity CL (**Figure 1**).

Combinations of deposition above the blue line in **Figure 1** would exceed the CL for N and S, while all areas below or on the line represent the area where CLs do not exceed. Deposition of both S and N exceedance is then a two-step calculation process (**Eq. 3** and **Eq. 4**):

When $\text{CLminN} \geq \text{Total N deposition}$, then

$$\text{Ex(N+S)} = \text{Total S deposition} - \text{CLmaxS} \qquad (3)$$

When $\text{CLminN} < \text{Total N deposition}$, then

$$\text{Ex(N+S)} = \text{Total S + N deposition} - \text{CLmaxN} \qquad (4)$$

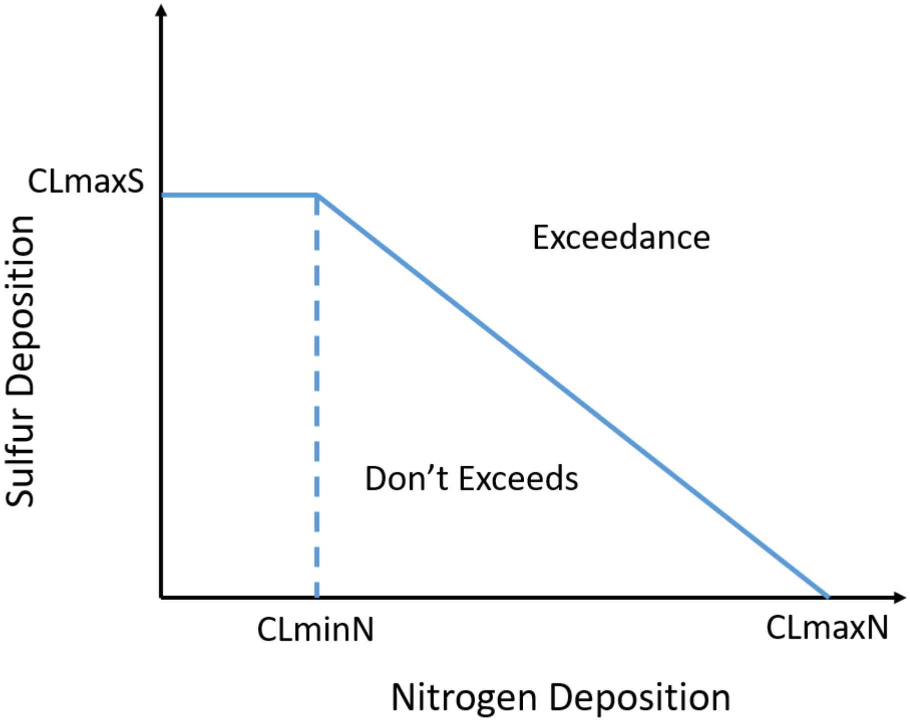

Figure 1. Three-node linear CL functions of S and N, defined by the three quantities CLmaxS (CLS), CLminN (CLNS), and CLmaxN. The area outside the blue line notes deposition pairs of N and/or S resulting in CL exceedance while the area inside notes deposition pairs that do not exceed the CL.

In the case of SW CLs for acidity, CL exceedances for both S and N deposition can also be calculated using the rate of nitrate leaching to the waterbody. The "Nleach" variable is an estimate of nitrate leaching and can be used along with Total S deposition to calculate an CL exceedance for N and S (**Eq. 5**) (Henriksen and Posch, 2001).

$$\text{Ex(A)} = (\text{Total S deposition} + N_{le}) - \text{CLmaxS} \qquad (5)$$

Where:
N_{le} = the sum of the measured concentrations of nitrate (NO_3^- µeq/L) and ammonia (NH_4^+ µeq/L) in the runoff (Q_s m/yr) as $([NO_3^-]+[NH_4^+])*Q_s$.

The exceedance for EMP CLs is simply the difference between Total N deposition and the CL itself as described in **Eq. 6**:

$$\text{Ex(E)} = \text{Total N deposition} - \text{CLN}_x \qquad (6)$$

Where:
CLN_x = empirical CLN measured as mean, max, min, etc.

Data Access, Use, and Download

All NCLD tables are in CSV format and can be accessed at the NADP CLAD webpage: https://nadp.slh.wisc.edu/clad-national-critical-load-database/ or with download links in this document. Some of the tables exceed Microsoft (MS) Excel's maximum number of rows. In addition, combined tables in a single MS Access database may exceed the maximum size of 2 gigabytes (GB). In addition, when inputting CSV files into Access, makes sure that the "Data Type" is imported correctly. Otherwise, import errors will result. Use the "Advanced" tab in Access's mode to assign the correct data type. NCLD data type are noted in the tables. It is possible to work around the size limitation in Access by linking tables between more than one Access database. Tables can also be accessed with the "Open Source" program called "Notepad++," which can be found at https://notepad-plus-plus.org/.

The NCLD also has been translated into geographic information system (GIS) files that contain a subset of the information in the data tables. Both shapefile and Esri geodatabase are provided. The GIS files can be joined to the database tables through the CLID. Additional details can be found in the metadata files provided with the GIS files.

Process for Including Studies in the NCLD

The NCLD aims to include all studies and datasets that provide estimates of CLs for terrestrial and aquatic systems in the U.S. This section provides a description of the types of studies and datasets that can be included in the NCLD, the review process, and a general description of how information is standardized when it is incorporated into the database.

Critical load studies and datasets that are published in peer-reviewed journals or reports generated by federal agencies or credible organizations can be included in the NCLD. The methods used to estimate CLs should be well documented, and the data sources should be well described. In addition, the exact location(s) (i.e., GPS coordinates) where the CL applies should be detailed (when appropriate) and the equation(s) used to generate the CLs and parameters within the CL calculations should be provided and appropriate in application. All CL studies and datasets should be submitted to the NCLD manager for consideration.

The CLAD Science Committee from time-to-time will issue a "Call-For-Data" to update the NCLD with new CL data. This "Call-For-Data" is sent to the members of CLAD and others outside of CLAD who might be interested in submitting data. Any person or entity with CL data is encouraged to submit data. All submitted datasets are reviewed by CLAD for potential inclusion in the NCLD. The author/source of the data should expect to participate in, assist with, and respond to any questions associated with the review of the CL dataset(s).

Once a dataset has been approved for inclusion in the NCLD, the dataset will be "standardized". This standardization, at a minimum, will involve converting the data to a common set of units (e.g., eq/ha-yr); a standard GIS projection; re-scaling (when appropriate); and addition of supporting information. In most cases, the dataset will maintain the original scale when included in the NCLD. See the following sections of this document for additional information regarding the standardization of CL data within the database.

Database Description and New Data Added 2

Database Description

The following tables include descriptions of the variables found in the NCLD that contains the CL values and supporting information (**Table 1**). Critical load values and supporting information have been summarized into tables based on three groupings: **Forest Soil (FS)**, **Surface Water (SW)**, and **Empirical (EMP)**. Critical loads for both FS and SW groupings are for soil and aquatic acidification impacts, respectively. The EMP grouping includes empirically determined CLs for N deposition. Each CL group has 4 tables that include: (1) Site information, (2) Critical load values, (3) Supporting information, and (4) Data source information. Combined tables contain the citations for all CL types. The below table summarizes the groupings and tables in the NCLD. See **Figure 2** for a diagram layout of the database.

NCLD_Tables_v321.zip (158 MB) (Click to download all tables)

Table 1. NCLD database overview.

Filename	Database Table	Number Table Rows
Forest Soil	Table 1A – SiteInfo	2,574,528
	Table 2A – Critical Loads and Supporting Info	2,574,528
	Table 3A – Data Source Info.	7
Surface Water		
	Table 1B – SiteInfo	172,384
	Table 2B – Critical Loads and Supporting Info	172,384
	Table 3B – Data Source Info.	39
Empirical		
	Table 1C – SiteInfo	16,847
	Table 2C – Critical Loads and Supporting Info.	16,847
	Table 3C – Data Source Info.	4
Combined		
(included in the download files above)	Table 4 - Lookup Table for Citation	220
	Table 5 - Citation	62
ALL Tables	Tables 1-3A,B,C, 4, & 5	

New Data and Changes Made in NCLDv3.1, v3.2, and v3.2.1

Below is a description of what has changes in v3.2.1 of the database. It includes need CLs added, removed CLs, and new variables added to the database.

Critical Loads	What changed in the new version of the database?
Tables	Table structure was updated to reflect a simplification of the database. The information in Table 4 was combined with Tables 2 and 3. Some supporting information has been removed because it was not being used.
Forest Soil	No new data was added to v3.1, v3.2, and v3.2.1.
Surface Water	CL datasets added to v3.1: -Lawrence.et.al.2015 – Target loads -Sullivan.et.al.2012b – Target loads -Fakhraei.et.al.2014 – Target loads -Sullivan.et.al.2005 – Target loads -McDonnell.et.al.2014 – New version replaced the old CLs -SSWC CLs from water quality data referenced in: -Lawrence.et.al.2015 -Sullivan.et.al.2012b & ALSC CL dataset added to v3.2: - McDonnell et al., 2017 - Target loads from the Sipsey and Cohutta Wilderness Areas, USFS -SSWC CLs from water quality data collected from USFS from Cherokee N.F., DeSoto N.F., Homochitto N.F., Kisatchie N.F. Sumter N.F., Talladega, N.F., etc. -New SSWC CL estimates determined by Monte Carlo Analysis and confidence intervals and rank uncertainty New dataset added to v3.2.1: - McDonnell et al., 2021 – 401 Target loads from New York Streams.
Empirical	CL datasets added to v3.1: -Herbaceous Biodiversity Ecoregion CLs based on Simkin et al. 2016 were replaced with new values for both closed and open systems for ecoregion 3 and 4 values. CLs added v3.2 include: -Lichen CLs for community level responses (Geiser et al., 2019) -Lichen species CLs Geiser et al., (2021) -New Lichen CLs replaced v3.1 values (e.g. Geiser et al., (2010) and Root et al., (2015)

	No new data in v3.2.1

Missing or Incomplete data

Missing numeric values are noted as -9999, -9999.99, -9999.999, which indicate both situations where information is not determined or does not apply. Missing text values where information is not determined are noted as “(no data)” while “(n/a)” indicates information that does not apply.

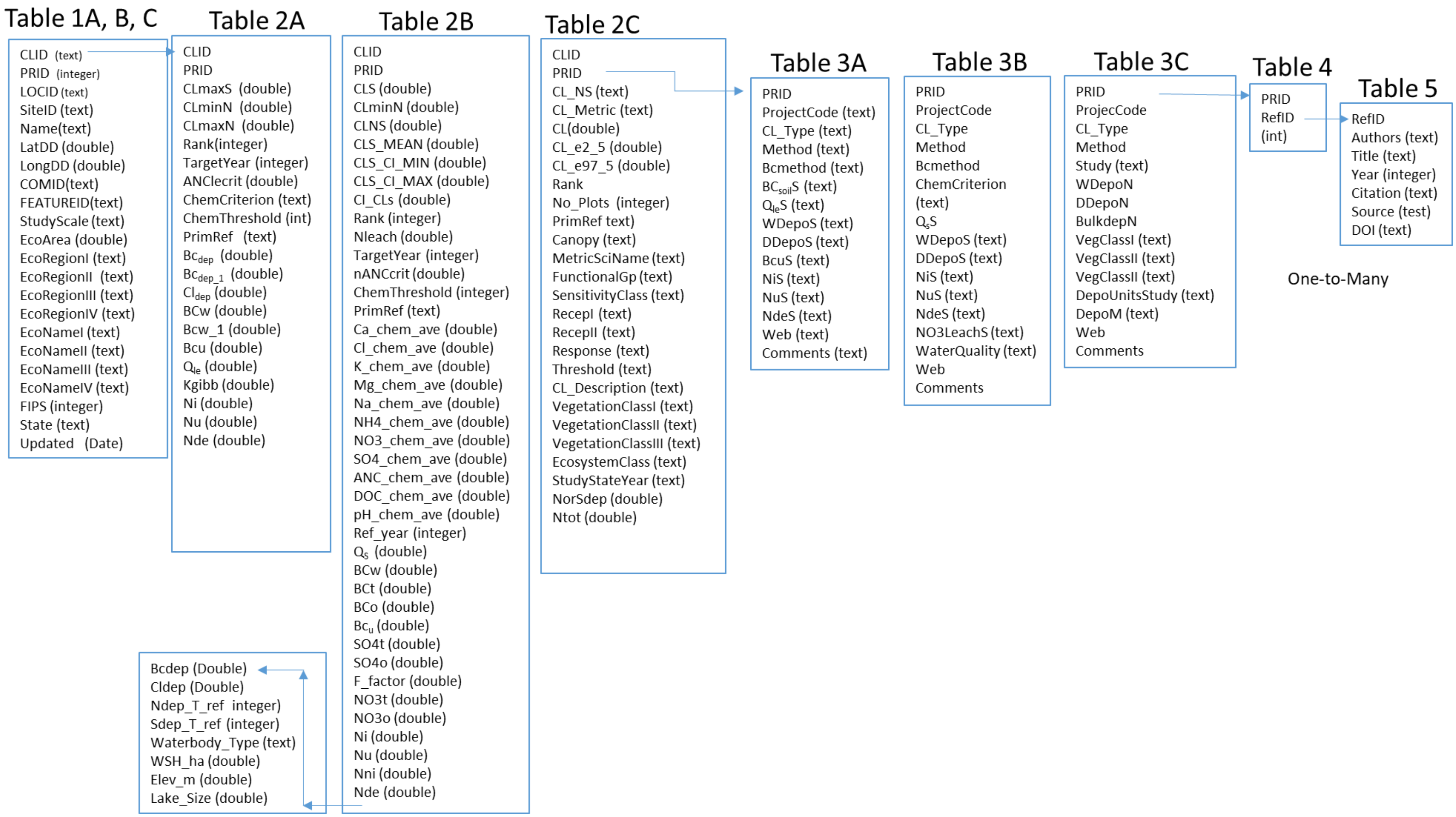

Figure 2. Diagram of the NCLD database.

Site Information

3

This section describes the site information for each CL in the NCLD. Three separate tables represent **A=Forest Soil (FS); B=Surface Water (SW); C=Empirical (EMP)**. Not all CLs have latitudes/longitudes that describe a point location. The "CLID," and "REFID" variables are unique for each CL in the database. The "REFID" through the "PRID" links each CL with the reference publication(s)/report(s) and should be cited when using the values (also see **Database Tables 3, 4 and 5**). **Database Tables 1, and 2** maintain a one-to-one relationship.

Database Tables

NCLD_Tables_v321.zip (158 MB) (Click to download all tables)

Database Table 1 A, B, C. 'SiteInfo' A=Forest Soil; B=Surface Water; C=Empirical

(This table structure is the same for all CLs)

Variable	Explanation	Note[1]
CLID (Text)	Unique(!) identifier across all three CL grouping: Forest Soil (FS), Surface Water (SW), and Empirical (EMP).	
PRID (Integer)	Unique(!) identifier of the CL project.	
LOCID (Text)	Unique(!) identifier of a particular location (e.g. lake, stream reach, or sample plot), gridded area, or Ecoregion I-IV. Lakes and stream reaches are classified by NHDPlusV2. In many cases, a single lake/stream reach or Ecoregion may have more than one CL value. The LOCID can be used to aggregate CLs for a particular location.	
SiteID (Text)	Project specific identifier of the site or species.	
Name (Text)	Specific name of site or location including waterbody name (lake and stream), national forest or national park name (e.g., Adirondack Mountains, Boundary Waters Canoe Wilderness Area, etc.). For waterbodies it's the name provided by the source of the data or NHDPlusV2.	
LatDD (Double)	Latitude (decimal degrees).	1
LongDD (Double)	Longitude (decimal degrees).	1
COMID (Text)	NHDPlusV2 unique(!) identifier for a feature in NHDPlusV2. This can be used to link the NCLD CL to NHDPlusV2 features and tables (http://www.horizon-systems.com/nhdplus/). This field is only populated for aquatic acidification.	

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[1] This "note" field corresponds to the more detailed text, in numerical order, included in the tables that follow.

FEATUREID (Text)	NHDPlusV2 unique(!) identifier for a NHDPlusV2 catchment. This can be used to link the NCLD CL to NHDPlusV2 features and tables (http://www.horizon-systems.com/nhdplus/). Positive values in the FeatureID field indicate catchments delineated for NHDFlowlines, whereas negative FeatureID values indicate sink-related catchments. FeatureID of the Catchment which equals the ComID of an NHDFlowline feature or the SinkID of a Sink feature. This field is only populated for aquatic acidification.	
StudyScale (Text)	Critical load scale (varies by CL): Study Area Plot Watershed Ecoregion I Ecoregion II Ecoregion III Ecoregion IV.	
EcoArea (Double)	Area of the ecosystem (km^2) represented by the CL. This variable is not defined for all CLs.	2
EcoRegionI (Text)	Ecoregion code Level I.	3
EcoRegionII (Text)	Ecoregion code Level II.	3
EcoRegionIII (Text)	Ecoregion code Level III.	3
EcoRegionIV (Text)	Ecoregion code Level IV.	3
EcoNameI (Text)	Ecoregion name Level I.	3
EcoNameII (Text)	Ecoregion name Level II.	3
EcoNameIII (Text)	Ecoregion name Level III.	3
EcoNameIV (Text)	Ecoregion name Level IV.	3
FIPS (Integer)	Five digit Federal Information Pressing Standard (FIPS) code which uniquely identifies U.S. counties.	
State (Text)	State name.	
Updated (Date)	Date of when record was added or updated.	

1) These non-spatial tabular data for LatDD and LongDD are based on geographic coordinates of the North American Datum of 1983. These values were projected to “USA_Contiguous_Albers_Equal_Area_Conic_USGS_version” for representation by the available spatial feature class data. Details of this projection are listed below:

USA_Contiguous_Albers_Equal_Area_Conic_USGS_version

Projection: Albers

False_Easting: 0.00000000
False_Northing: 0.00000000
Central_Meridian: -96.00000000
Standard_Parallel_1: 29.5.00000000
Standard_Parallel_2: 45.00000000
Latitude_of_Origin: 23.00000000
Linear_Unit: Meter

Geographic Coordinate System:
GCS_North_American_1983
Datum: D_North_American_1983
Prime Meridian: Greenwich
Angular Unit: Degree

2) The EcoArea is the area of the ecosystem (km^2) represented by the CL. This variable is not defined for all CLs. For aquatic acidication it represents the watershed of the lake or stream.

3) Ecoregion level I, II, III, and IV codes, which represents the Omernik (1987) Ecoregion classification for North America. Level I Ecoregions are Arctic Cordillera, Tundra, Taiga, Hudson Plains, Northern Forests, Northwestern Forested Mountains, Marine West Coast Forests, Eastern Temperate Forests, Great Plains, North American Deserts, Mediterranean California, Southern Semi-Arid Highlands, Temperate Sierras, Tropical Dry Forests and Tropical Wet Forests (http://www.epa.gov/wed/pages/ecoregions/na_eco.htm).

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Forest Soil Critical Loads of Acidity

4

Description

This section provides a description of the Forest Soil (FS) CLs to protect against acidification of forest soils. The CLs in this dataset represent the amount of acidifying S, or N, and a combination of S and N deposition below which a forest system is not thought to experience soil acidification. The database currently consists of data from four different studies (Table 2). Therefore, care should be taken when using these data to aggregate CLs and/or calculate CL exceedances (see table below). When using these data, it is important to consider the following: (1) that these CLs were determined using different models and methods to estimate soil base cation weathering, (2) that various chemical criterions and thresholds are used, (3) that a specific location could have more than one CL, and (4) that different CL represent different spatial scales (eg. 1 km^2 to a watershed). **Database Tables 2A and 3A** describe each variable included in the Forest Soil CL database. Additional information is in **Appendix 1**.

Table 2: Forest Soil Critical Load Source Information

Source	Critical Load	Scale	Method	Location
McNulty et al., (2007, 2013)	Steady-State Mass Balance	1 km^2	SMB	Nationwide
Duarte et al., (2011, 2013)	Steady-State Mass Balance	5 km^2	SMB	New England
Phelan et al., (2014, 2016)	Steady-State Mass Balance	1 m^2	SMB	Pennsylvania
Sullivan et al., (2011a, 2011b)	Target Load (derived from a dynamic model)	Watershed	MAGIC	Virginia and New York

Forest Soil Critical Load Updates

No new data was added to the FS database for v3.1, v3.2, and v3.2.1.

Forest Soil Tables and GIS file download

NCLD_FS_GIS_Table_v321.zip (486 MB) (Click to download)

- NCLD_FS_Table_1A_v321
- NCLD_FS_Table_2A_v321
- NCLD_FS_Table_3A_v321
- NCLD_Table_4_v321
- NCLD_Table_5_v321
- NCLD_FS_GIS_Metadata_v321.pdf
- NCLD_FS_GIS_v321.gdb
  - NCLD_FS_Combined_v321
  - NCLD_FS_Duarte_v321
  - NCLD_FS_Phelan_v321
  - NCLD_FS_Sullivan_v321

NCLD_FS_McNulty_v321
NCLD_FS_Table_1A_v321
NCLD_FS_Table_2A_v321

Database Table 2A. Forest Soil Critical Load and Supporting Information

Variable	**Explanation**	**Note**
CLID (Text)	Unique(!) identifier for the CL (same as in **Database Table 1A**).	
PRID (Integer)	Unique(!) identifier of the CL project.	
CLmaxS (Double)	Maximum CL of S (eq/ha-yr).	6
ClminN (Double)	Minimum CL of N (eq/ha-yr).	6
ClmaxN (Double)	Maximum CL of N (eq/ha-yr).	6
Rank (Integer)	Uncertainty Rank from 5 to 1 with 5 the best and 1 the worst.	
TargetYear (Integer)	Target load (TL) year.	
ANC_{lecrit}[2] (Double)	Critical leaching of acid neutralizing capacity (ANC) represents the specified/desired buffering capacity of forest soils to acidic deposition.	
ChemCriterion (Text)	Chemical criterion used for acidity CL calculations: molar[Bc]:[Al] Specific percentage of base saturation (BS) molar [Ca]:[Al].	
ChemThreshold (Integer)	Critical value for the chemical criterion given in ‘ChemCriterion.’	
PrimRef (Text)	Publication citation for primary study for the CL. For some CLs, there is more than one publication. For CLs with more than one publication, use **Database Tables 4 and 5** to determine the additional reference(s).	
BC_{dep} (Double)	Total deposition of base cations (BC; Ca+Mg+K+Na) (eq/ha-yr).	
Bc_{dep_1} (Double)	Total deposition of nutrient base cations (Bc; Ca+Mg+K) (eq/ha-yr).	
Cl_{dep} (Double)	Total deposition of chloride (eq/ha-yr).	
BC_w (Double)	Weathering of base cations (BC; Ca+Mg+K+Na) (eq/ha-yr).	
Bc_{w_1} (Double)	Weathering of nutrient base cations (Bc; Ca+Mg+K) (eq/ha-yr).	
Bc_u (Double)	Net removal of nutrient base cations – removed through harvesting of trees. (Bc; Ca+Mg+K) (eq/ha-yr).	

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[2] “ANC_{lecrit}” is denoted in the database tables while “$ANC_{le,crit}$” is used throughout the text.

Q_{le} (Double)	Precipitation surplus (amount of water percolating from below the root zone) (m^3/ha-yr).	
K_{gibb} (Double)	Equilibrium constant for the Al-H relationship (log_{10}).	
N_i (Double)	Long-term average rate of N immobilization in the soil rooting zone (eq/ha-yr).	
N_u (Double)	Net removal of N – nutrients removed through harvesting of trees (eq/ha-yr).	
N_{de} (Double)	Rate of N denitrified (eq/ha-yr).	

6) Various methods for calculating the Forest Soil CLs are represented in this variable (see Appendix 1 for more detail). Critical loads are in tabular format, but a raster format of McNulty et al., (2013) is available on request.

Database Table 3A. Forest Soil Critical Load Data Source Information

Variable	**Explanation**	**Note**
PRID (Integer)	Unique(!) identifier of the CL project.	
ProjectCode (Text)	Project code. Not all studies have this variable.	
CL_Type (Text)	Critical load type: Steady-state Target-load Empirical.	
Method (Text)	Model or method used to determine the CL: Dynamic Model SMB.	
Bcmethod (Text)	Method used for base cation weathering: MAGIC (Cosby et al., 1985) PROFILE (Sverdrup, 1990) Clay correlation-substrate method.	
BcsoilS (Text)	Soil data used in the base cation weathering calculation: SSURGO STATSGO2 FIA (https://www.fia.fs.fed.us/) Site measurements NRCSSoilDataAcess (https://sdmdataaccess.nrcs.usda.gov/).	
$Q_{le}S$ (Text)	Source of the annual runoff flux: NLDAS Gebert et al., (1987) Duarte et al., (2013).	

$W_{depo}S$ (Text)	Source of the wet deposition: NADP (http://nadp.sws.uiuc.edu/) NADPPRISM (http://nadp.sws.uiuc.edu/) NADPGRIMM (Grimm and Lynch, 2005) TDEP (Schwede and Lear, 2014) CMAQ (https://www.cmascenter.org/cmaq/) ClimCalc (Ollinger et al., 1993).	
$D_{depo}S$ (Text)	Source of the dry deposition: CASTNET (https://www.epa.gov/castnet) CASTNET&AmoN TDEP (Schwede and Lear, 2014) CMAQ (https://www.cmascenter.org/cmaq/) Throughfall ClimCalc (Ollinger et al., 1993).	
Bc_uS (Text)	Source of the net removal of base cations – removed through harvesting of trees: McNulty et al., (2007, 2013) Duarte et al., (2013).	
N_iS (Text)	Source of the long-term average rate of N immobilization in the soil rooting zone: Pardo, (2010).	
N_uS (Text)	Source of the Net removal of N through harvesting of trees: Duarte.et al., (2013) McNulty et al., (2007).	
$N_{de}S$ (Text)	Source of the denitrification rate: Binkley et al., (1995) Ashby et al., (1998).	
Web (Text)	Webpage where additional information can be found.	
Comments (Text)	Additional information.	

Surface Water Critical Loads for Acidity

5

Description

This section provides a description of the aquatic or Surface Water (SW) CLs of acidity for lakes and streams. Critical loads in this dataset represent the amount of S, N, acidifying S, N, and combined S and N deposition below which a lake/stream and its watershed can receive and maintain a healthy ecosystem. The database currently consists of data from different studies (**Table 3**). Therefore, care should be taken when using these data to aggregate CLs and/or calculate CL exceedances. When using these data, it is important to consider the following: (1) the CLs were determined using different methods to estimate watershed base cation weathering (e.g. F-factor, MAGIC, etc.), (2) different chemical thresholds are used, and (3) a given specific waterbody could have more than one CL. **Database Tables 2B and 3B** describe each variable included in the SW CL database. Additional information is in **Appendix 2**.

Surface Water Critical Load Updates

Table 3. Surface Water Critical Load Source Information

Source	Method	Extent
New in v3.0		
USEPA, (2012) Scheffe et al., (2014)	Modified SSWC	Nationwide
Sullivan et al., (2012a) McDonnell et al., (2012)	Modified SSWC	Appalachian Mountains of VA, WV, MD
Miller, (2011)	Modified SSWC	Northeast
DuPont et al., (2005)	Modified SSWC	New England
Vermont Department of Environmental Conservation, (2003, 2004, 2012)	SSWC	Vermont
New in v3.1		
McDonnell et al., (2014) EMDSv4.0	Modified SSWC/MAGIC	So. Appalachian Mountains
Lynch et al., (2022)*	Modified SSWC	Nationwide
Lawrence.et.al., (2015)	MAGIC	Appalachian Mountains
Sullivan.et.al., (2005)	MAGIC	Loch Vale, Colorado
Sullivan.et.al., (2012b)	Modified SSWC/MAGIC	Adirondack Mountains, New York
Fakhraei.et.al., (2014)	PnET-BGC	New York
New in v3.2		
McDonnell et al., (2017)	MAGIC	Cohutta and Sipsey Wilderness areas in Georgia and Alabama
USFS Water Quality data	SSWC	Cherokee, De Soto, Homochitto, Kisatchie, Sumter, Talladega, Pisgah, and Nantahala

		National Forests and Joyce Kilmer, Cohutta and Sipsey Wilderness areas
New in v3.2.1		
McDonnell et al., (2021)	PnET-BGC	Adirondack Mountains, New York

*Reference to this document. Critical loads calculated from various water quality datasets using method described Appendix B.

Surface Water GIS file download

NCLD_SW_GIS_Table_v321.zip (93 MB) (Click to download)

- NCLD_SW_Table_1B_v321.csv
- NCLD_SW_Table_2B_v321.csv
- NCLD_SW_Table_3B_v321.csv
- NCLD_Table_4_v321.csv
- NCLD_Table_5_v321.csv
- NCLD_SW_GIS_Metadata_v321.pdf
- NCLD_SW_GIS_Metadata_McDonnell_v321.pdf
- NCLD_SW_GIS_v321.gdb
  - NCLD_SW_ALL_v321
  - NCLD_SW_nonMcDonnell_v321
  - NCLD_SW_Table_1B_v321
  - NCLD_SW_Table_2B_v321
- NCLD_SW_GIS_McDonnell_v321.gdb
  - NCLD_SW_pts_McDonnell_v321 (Point locations)
  - NCLD_SW_WS_McDonnell_v321 (Watersheds)

Database Table 2B. Surface Water Critical Loads and Supporting Information

Variable	**Explanation**	**Note**
CLID (Text)	Unique(!) identifier of the CL (same as in **Database Table 1**).	
PRID (Integer)	Unique(!) identifier of the CL project.	
CLS (Double)	CL of S (meq/m^2-yr).	
CLminN (Double)	Minimum CL of N (meq/m^2-yr). See Appendix 2 for a summary of the minimum CL of N.	
CLNS (Double)	CL of N+S (meq/m^2-yr).	
CLS_MEAN (Double)	Mean CL S determined by a Monte Carlo application of the SSWC model. The Monte Carlo was run 5,000 times and a mean CL S value was calculated.	
CLS_CI_MIN (Double)	CL S minimum value determined by a Monte Carlo application of the SSWC model. The Monte Carlo was run 5,000 times and a minimum CL S value was calculated.	
CLS_CI_MAX (Double)	CL S maximum value determined by a Monte Carlo application of the SSWC model. The Monte Carlo was run 5,000 times and a maximum CL S value was calculated.	

CI_CLS (Double)	Confidence intervals of CLS determined by a Monte Carlo application of the SSWC model.	
Rank (Integer)	Uncertainty Rank from 5 to 1 with 5 the best and 1 the worst.	
Nleach (Double)	Rate of nitrate leaching into the waterbody (meq/m2-yr).	
TargetYear (Integer)	Target load (TLS) year.	
nANCcrit	The quantity $ANC_{le(crit)}$ (meq/m^2-yr).	
ChemThreshold (Integer)	Value of the chemical threshold used.	
PrimRef	Publication citation for primary study for the CL. For some CLs, there is more than one publication. For CLs with more than one publication, use **Database Tables 4 and 5** to determine the additional references.	
Ca_chem_ave (Double)	Average calcium (Ca2+) water chemistry value (µeq/L).	
Cl_chem_ave (Double)	Average chloride (Cl-) water chemistry value (µeq/L).	
K_chem_ave (Double)	Average potassium (K+) water chemistry value (µeq/L).	
Mg_chem_ave (Double)	Average magnesium (Mg2+) water chemistry value (µeq/L).	
Na_chem_ave (Double)	Average sodium (Na+) water chemistry value (µeq/L).	
NH4_chem_ave (Double)	Average ammonium (NH4+) water chemistry value (µeq/L).	
NO3_chem_ave (Double)	Average nitrate (NO3-) water chemistry value (µeq/L).	
SO4_chem_ave (Double)	Average sulfate (SO4-2) water chemistry value (µeq/L).	
ANC_chem_ave (Double)	Average ANC water chemistry value (µeq/L).	
DOC_chem_ave (Double)	Average dissolved organic carbon (DOC) water chemistry value (mg/L).	
pH_chem_ave (Double)	Average pH water chemistry value (pH units).	
Ref_year (Double)	Average sampling year of water chemistry values.	
QS (Double)	Annual runoff flux (m/y). See appendix 2 for more details.	
BCw (Double)	Weathering of base cations (BCw; Ca+Mg+K+Na) (meq/m2-yr).	
BCt (Double)	Present day base cations concentration (BCt; Ca+Mg+K+Na) (µeq/L).	
BCo (Double)	Pre-acidification base cations concentration (BCo; Ca+Mg+K+Na). (µeq/L)	
Bcu	Net removal of base cation (Bcu; Ca+Mg+K) (meq/m2-yr).	

(Double)		
SO4t (Double)	Present day sulfate concentration (µeq/L).	
SO4o (Double)	Pre-acidification sulfate concentration (estimated value) (µeq/L).	
F_factor (Double)	F-factor value.	
NO3t (Double)	Present day nitrate concentration (µeq/L).	
NO3o (Double)	Pre-acidification nitrate concentration (estimated value) (µeq/L).	
Ni (Double)	Long-term average rate of N immobilization (meq/m2-yr).	
Nu (Double)	Net removal of N through harvesting of trees (meq/m2-yr).	
Nni (Double)	Nitrification rate in catchment (meq/m2-yr).	
Nde (Double)	Denitrification rate in catchment (meq/m2-yr).	
BCdep (Double)	Wet deposition of base cations (BCdep; Ca+Mg+K+Na) (meq/m2-yr). See appendix 2 for more details on the deposition sources.	
CLdep (Double)	Wet deposition of chloride (meq/m2-yr).	
Ndep_T_ref (Double)	Average total N deposition for average water chemistry value (meq/m2-yr).	
Sdep_T_ref (Double)	Average total S deposition for average water chemistry value (meq/m2-yr).	
Waterbody_Type (Text)	Type of waterbody for which the CL was determined: POND, LAKE, UNKNOWN, STREAM, RIVER, RESERVOIR.	
WSH_ha (Double)	Size of watershed (ha).	
Elev_m (Double)	Elevation of sample location (m).	
Lake_Size (Double)	Lake size (ha) provided by the data source.	

Database Table 3B. Surface Water Critical Load Source Information

Variable	**Explanation**	**Note**
PRID (Integer)	Unique(!) identifier of the CL project.	
ProjectCode (Text)	Project code.	
CL_Type (Text)	Critical load type: Steady-state Target-load Empirical.	
Method (Text)	Model or method used to determine the CL:	

	SSWC (Henriksen, 1982, 1984 and Henriksen and Posch, 2001) FAB (Henriksen et al., 1993). MAGIC (Cosby et al., 1985) PnET-BGC (Gbondo-Tugbawa et al., 2001)	
Bcmethod (Text)	Method used for base cation weathering: F-Factor (Henriksen, 1982) Statistical (McDonnell et al., 2012, 2014) MAGIC (Cosby et al., 1985) PnET-BGC (Gbondo-Tugbawa et al., 2001).	
ChemCriterion (Text)	Chemical criterion used: ANC (µeq/L).	
Q_sS (Text)	Source of the annual runoff flux. See appendix 2 for more details.	
WdepoS (Text)	Source of the wet deposition: NADPPRISM (http://nadp.sws.uiuc.edu/) NADPGRIMM (Grimm and Lynch, 2005) TDEP (Schwede and Lear, 2014) CMAQ (https://www.cmascenter.org/cmaq/) HRDM.	
DdepoS (Text)	Source of the dry deposition: TDEP (Schwede and Lear, 2014) CMAQ (https://www.cmascenter.org/cmaq/) HRDM (Miller 2000)	
NiS (Text)	Source of the long-term average rate of N immobilization.	
NuS (Text)	Source of the net removal of N through harvesting of trees. McNulty et al., (2007).	
NdeS (Text)	Source of the denitrification rate: Ashby et al., (1998).	
NO3LeachS (Text)	Source of the method to determine nitrate leaching. Henriksen et al., (2002).	
WaterQuality	Source of the water quality data used in calculating the critical load.	
Web (Text)	Webpage where additional information can be found.	
Comments (Text)	Additional information.	

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Empirical Critical Loads for Nitrogen 6

Description

This section provides a description of the Empirical (EMP) CLs for N for terrestrial ecosystems. An empirical CL is developed using empirical approaches, which involve observed spatial or temporal gradient studies or experimental manipulations of pollutants. Empirical critical loads are applied to sites or landscapes that are ecologically comparable to location(s) from which CLs were determined (cf. Pardo 2010, 2011). Empirical CLs described in the NCLD consist of values from individual site studies, regional studies, and a synthesis studies at the Ecoregion I to IV levels (**Table 4). Database Tables 2C and 3C** describe each variable included in the Empirical CL for N database. Additional information is in **Appendix 3**.

Table 4. Empirical Critical Load Source Information

Source	**Method**	**Extent**
Pardo et al., (2010, 2011)	Metadata Analysis	Nationwide
Geiser et al., (2019, 2021)	Statistical	Nationwide
Simkin et al., (2016)	Statistical	Nationwide

Empirical Critical Load Updates

A new Simkin et al., (2016) ecoregion summary is included in v3.1 of the database. See the Simkim et al., (2016) section of Appendix 3 for more information. This dataset is unchanged in v3.2. New community lichen CLs and species level CLs based on Geiser et al., (2019) and Geiser et al., (2021) have been added to v3.2. These new lichen CLs replace the GeiserRoot CLs, which have been removed from v3.2.

Empirical GIS file download

Empirical CLs and tables have been made into GIS files. Click this link to download the file.

NCLD_EMP_GIS_Table_Pardo_Geiser_v321 (144 MB) (Click to download)
- NCLD_EMP_Table_1C_v321.csv
- NCLD_EMP_Table_2C_v321.csv
- NCLD_EMP_Table_3C_v321.csv
- NCLD_Table_4_v321.csv
- NCLD_Table_5_v321.csv
- NCLD_EMP_Metadata_Geiser_v321
- NCLD_EMP_Metadata_Pardo_v321
- NCLD_EMP_GIS_Pardo_v321.gdb
    - NCLD_EMP_Combined_Pardo_v321
    - NCLD_EMP_Forest_Pardo_v321
    - NCLD_EMP_Fungi_Pardo_v321
    - NCLD_EMP_HPS_Pardo_v321
    - NCLD_EMP_Lichen_Pardo_v321
    - NCLD_EMP_Nitrate_Pardo_v321
    - NCLD_EMP_Table_1C_Pardo_v321

NCLD_EMP_Table_2C_ Pardo_v321

NCLD_EMP_GIS_Geiser_v321.gdb
NCLD_EMP_Comm_Lichen_v321
NCLD_EMP_Cyanolichen_Lichen_v321
NCLD_EMP_Foragelichen_Lichen_v321
NCLD_EMP_Table_1C_Geiser_v321
NCLD_EMP_Table_2C_Geiser_v321
Ras_Cyan
Ras_Forage
Ras_Lichen

NCLD_EMP_GIS_Table_Simkin_v321.zip (1.7 GB) (Click to download)
NCLD_EMP_Table_1C_v321.csv
NCLD_EMP_Table_2C_v321.csv
NCLD_EMP_Table_3C_v321.csv
NCLD_Table_4_v321.csv
NCLD_Table_5_v321.csv
NCLD_EMP_Metadata_Simkin_v321
NCLD_EMP_pts_Simkin_v321.gdb
NCLD_EMP_pts_Simkin_v321
NCLD_EMP_Table_1C_v321
NCLD_EMP_Table_2C_v321
NCLD_EMP_ER34_Poly_Simkin_v321.gdb
NCLD_EMP_ER3_Closed_Simkin_v321
NCLD_EMP_ER3_Open_Simkin_v321
NCLD_EMP_ER4_Closed_Simkin_v321
NCLD_EMP_ER4_Open_Simkin_v321

NCLD_EMP_ER34_Rasters_Simkin_v321.zip (1.2 GB) (Click to download)
NCLD_EMP_ER34_Rasters_Simkin_v321.gdb
NCLD_ER3_Closed
NCLD_ER3_Open
NCLD_ER4_Closed
NCLD_ER4_Open

Database Table 2C. Empirical Critical Loads and Supporting Information

Variable	**Explanation**	**Note**
CLID (Text)	Unique(!) identifier of the CL (same as **Database Table 1C**).	
PRID (Integer)	Unique(!) identifier of the CL project.	
CLNS (Text)	Indication on whether this CL is for: N = Nitrogen S = Sulfur	
CL_Metric (Text)	Critical load metric that describes the CL and what it represents. CLstat – CL determine from a statical model	

	CLmin – Minimum CL CLmax – Maximum CL CLmean – Mean CL CL_q1-50 - 1th to 50th percentile Empirical CL of N (kg/ha-yr). For Ecoregions (I, II, III, or IV) based CL, a value was reported only if a statistical check was met where the sample size was considered adequate (see pages **A3-4** to **A3-5** for details on how the statistical check was determined.	
CL (Double)	Empirical CL of N or S reported (kg/ha-yr).	
CL_e2_5 (Double)	Quantile of 2.5% CL of CLN (kg/ha-yr)	
CL_e97_5 (Double)	Quantile of 97.5% CL of CLN (kg/ha-yr)	
Rank (Integer)	Uncertainty Rank from 5 to 1 with 5 the best and 1 the worst.	
No_plots (Integer)	Total number of CLs within a given Ecoregion (I, II, III, or IV) used to determine the Ecoregion based CL.	
PrimRef (Text)	Publication citation for primary study for the CL. For some CLs, there is more than one publication. For CLs with more than one publication, use **Database Tables 4** and **5** to determine the additional references.	
Canopy (Text)	Closed canopy or Open canopy. Based on the National Vegetation Classification System where woodlands, grasslands, and shrublands were "open" and forests were "closed."	
MetricSciName (text)	Metric name given by the PI or the species scientific name.	
FunctionalGp (Text)	Lichen Functional Group: Matrix lichen Forage lichen Cyanolichen.	
SensitivityClass (Text)	Sensitivity lichen class: Oligotroph S-sensitive Eutroph. S-tolerant S-intermediate Mesotroph Sensitive Intermediate Tolerant.	
RecepI (Text)	Biological and physical entity being affected: Fungi community Lichen community Lichen species Herbaceous plant and shrubs community Herbaceous plant community Forest community Surface water chemistry.	
RecepII	More specific biological and physical entity being affected:	

(Text)	Epiphytic Forage lichens Cyanolichens Mycorrhizal & Ectomycorrhizal Mycorrhizal Ectomycorrhizal Prairie Serpentine grassland Cover Alpine Coniferous Tropical & subtropical forests.	
Response (Text)	The negative response of the biological or physical entity that is to be avoided: Reduction in community composition Reduction in community richness Reduction in fungal activity Change in community structure Reduction in herb layer richness Species invasive Reduction in species growth & mortality Change in species foliar chemistry Reduction in fine root biomass Increase in soil nitrogen Increase in nitrate leaching.	
Threshold (Text)	The threshold related to the receptor and its response: No change in community composition No biodiversity loss Oligrophic lichen become dominant No change in fungal activity No change in community structure No species invasive No change in species growth & mortality No change in species foliar chemistry No change in fine root biomass No change in soil nitrogen No nitrate leaching 20% decline from peak value of metric.	
CL_Description (Text)	Text description of what the CL represents.	
VegetationClassI (Text)	Broad vegetation description based on: (1) 1997 National Vegetation Classification text name for level 1 classification (Class), or (2) USGS (2001, 2006, 2011), National Land Cover Dataset classification (NLCD) name: Forest Herbaceous Vegetation (e.g. grasslands) Shrubland Woodland.	
VegetationClassII (Text)	A description of dominant community vegetation described by: (1) 1997 National Vegetation Classification text name for level	

	3 classification (Group), (2) USFS National Forest Type Group, or (3) the USFS Land Fire classification.	
VegeationClassIII (Text)	A detail description of ecosystem as described by: (1) the referenced publication, (2) 1997 National Vegetation Classification text name for level 6 classification (Alliance), or (3) the USFS Land Fire classification.	
EcosystemClass (Text)	Specific ecosystem description based on USFS Ecological Subsections.	
StudyStartYear (Integer)	Year the study was conducted.	
NorSdep (Double)	Amount of N or S deposition at the study site.	
Nadd (Double)	Amount of N addition added to the study site.	

Database Table 3C. Empirical Critical Loads Source Information

Variable	**Explanation**	**Note**
PRID (Integer)	Unique(!) identifier of the CL project.	
ProjectCode (Text)	Project code.	
CL_Type (Text)	Critical load type: Steady-state Target-load Empirical.	
Method (Text)	Model or method used to determine the CL: Empirical Statistical.	
Study (Text)	Type of study used to determine the CL: Metadata - Analysis of multiple studies (e.g. Pardo et al., 2011) Multiple plot – Study that use multiple plots (e.g. Simkin et al., 2016, Geiser et al. 2019, 2021). Extrapolate – Analysis that extrapolates CL into Ecoregions or other regions.	
WdepoN (Text)	Source of the wet deposition: NADPPRISM (http://nadp.sws.uiuc.edu/) NADPGRIMM (Grimm and Lynch, 2005) TDEP (Schwede and Lear, 2014) CMAQ (https://www.cmascenter.org/cmaq/) HRDM (Miller 2000).	
DdepoN (Text)	Source of the dry deposition: Source of the dry deposition:	

	TDEP (Schwede and Lear, 2014) CMAQ (https://www.cmascenter.org/cmaq/) HRDM	
BulkdepN (Text)	Source of the bulk deposition:	
VegetationClassI (Text)	Source of the vegetation class I: NVC level (Class) - 1997 National Vegetation Classification text name for level 1 classification (Class) NLCD - USGS (2001, 2006, 2011), National Land Cover Dataset	
VegetationClassII (Text)	Source of the vegetation class I: NVC level 3(Group) - 1997 National Vegetation Classification text name for level 3 classification (Group) National Forest Type Group - USFS National Forest Type Group LandFire - LandFire classification.	
VegetationClassIII (Text)	Source of the vegetation class I: Referenced Publication NVC level 6 (Alliance) - 1997 National Vegetation Classification text name for level 6 classification (Alliance) LandFire - USFS Land Fire classification.	
DepoUnitsStudy (Double)	Deposition unit for study kg/ha-yr.	
DepoM (Text)	Deposition Measure. Type of deposition: Wet only Dry only Total, wet and dry Bulk.	
Web (Text)	Webpage where additional information can be found.	
Comments (Text)	Additional information.	

Critical Load Citation Tables

7

Both **Database Table 4 and 5** define the CL citation(s) for each CL. Each CL has at least one citation, but in many cases, there are more than one citation for a CL (e.g. CLID). For this reason, lookup **Database Table 4** "Lookup" links **Database Table 1-3A, B, C**, via the "PRID" variable and **Database Table 5** "Citation" and the individual "CLID."

Database Table 4. Attributes of the 'Lookup' Table

Variable	**Explanation**	**Note**
PRID (Integer)	Unique(!) identifier of the CL.	
RefID (Integer)	Unique(!) identifier of the reference from **Table 5**.	

Database Table 5. Attributes of the Database Table 'Citation'

Variable	**Explanation**	**Note**
RefID (Integer)	Unique(!) identifier of the reference.	
Authors (Text)	List of Authors.	
Title (Text)	Title of reference.	
Year (Integer)	Year of reference.	
Citation (Text)	Citation of reference.	
Source (Text)	Source of CLs.	
DOI (Text)	Digital Object Identifier.	

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9

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Appendix 1

Forest Soil Critical Loads of Acidity

Forest Soil critical loads (CL) of acidity were obtained from (1) McNulty et al., (2007, 2013), (2) Duarte et al., (2011, 2013), (3) Phelan et al., (2014, 2016), and (4) Sullivan et al., (2011a, 2011b). The models and variables used by each study are described in separate sections below. **Table A1-1**. contains a summary of key variables among the studies and sources of data.

Table A1-1. Summary of Key Variables Among the Studies and Sources of Forest Soil Critical Loads of Acidity

Source	CL / Target Load Model	Chem Threshold/Criteria	BC_w Model	Bc_u/ N_u	N_i (eq/ha-yr)	N_{de} (eq/ha-yr)	Scale (Project)
McNulty et al., (2007, 2013)	SMB	Soil solution base cation [Bc]: aluminum [Al] molar ratio of 1 for conifer forests and 10 for deciduous forests.	Clay correlation -substrate method	Y	42.86	0	1 km^2
Duarte et al., (2011, 2013)	SMB	Soil solution [Bc]:[Al] molar ratio of 10.	Clay correlation -substrate method	Y	143.0	0	5 km^2
Phelan et al., (2014, 2016)	SMB	Soil solution [Bc]:[Al] molar ratio of 10.	PROFILE	Y	42.86	0	1 m^2
Sullivan et al., (2011a, 2011b)	MAGIC	Soil base saturation of 5% and 10%; Soil solution [Bc]:[Al] and [Ca]:[Al] molar ratios of 1 and 10.	MAGIC	Y (for Bc_u, N_u not included)	Not specified	Not specified	Watershed

Forest Soil Critical Loads of Acidity as presented by McNulty et al., 2007, 2013.

The CL of acidity in McNulty et al., (2007, 2013) were modeled using the steady-state Simple Mass Balance (SMB) equation (1) outlined in CLRTAP (2004) that assumes the ecosystem is at long-term equilibrium or steady state over time and the equilibrium condition is equal to the ecosystem's CL (**Eq. A1-1**):

$CL(S+N) = BC_{dep} - Cl_{dep} + BC_w - Bc_u$[1] $+ N_i + N_u + N_{de} - ANC_{le,crit}$ (A1-1)

where (all in eq/ha-yr unless otherwise stated),

$CL(S+N)$ is the forest soil CL for S and N
BC_{dep} is total deposition of base cation (calcium [Ca]+potassium [K]+magnesium [Mg]+sodium [Na]) deposition
Cl_{dep} is total chloride deposition
BC_w is weathering of base cations (Ca+K+Mg+Na)
Bc_u is removal of nutrient base cations (Ca+K+Mg) in trees by harvesting
N_i is long-term N immobilization within the rooting zone in soil
N_u is removal of N in trees by harvesting
N_{de} is denitrification
$ANC_{le,crit}$ is the leaching of acid neutralizing capacity from the forest soil (critical level)

The critical ANC leaching rate, ANC_{lecrit}, is calculated using the following equation (**Eq. A1-2**):

$$ANC_{le,crit} = -Q^{\frac{2}{3}} \times \left(1.5 \times \frac{Bc_{dep}+Bc_w-Bc_u}{K_{gibb} \times \left(\frac{Bc}{Al}\right)_{crit}}\right)^{\frac{1}{3}} - 1.5 \times \frac{Bc_{dep}+Bc_w-Bc_u}{\left(\frac{Bc}{Al}\right)_{crit}} \qquad \text{(A1-2)}$$

where:

Q is precipitation surplus (m^3/ha-yr)
Bc_w is the weathering of nutrient base cations[2] (Ca+K+Mg) (eq/ha-yr)
Bc_{dep} is the total deposition of nutrient base cations[3] (CA+K+Mg) (eq/ha-yr)
Bc_u is removal of nutrient base cations (Ca+K+Mg) in trees by harvesting[4] (eq/ha-yr)
K_{gibb} is gibbsite equilibrium constant (m^6/eq^2)
$\left(\frac{Bc}{Al}\right)_{crit}$ is the critical soil solution base cation (Ca+K+Mg) to aluminum (Al) molar ratio

All the SMB model parameters were represented by data on a 1 km^2 grid system. All data were acquired by McNulty et al., (2007, 2013) in GIS grid format. **Table A1-2** provides a description of the parameters used in the SMB CL calculations and the data sources and assumptions adopted by McNulty et al., (2007, 2013).

Since the publication of McNulty et al., (2007), this CL dataset has been revised and updated to McNulty et al., (2013). The McNulty et al., (2013) version of the dataset includes the use of a new forest type layer and a correction was made to the pound to gram conversion in the Bc_u[4] calculation. These revised 2013 values are included in the NCLD.

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[1] In McNulty et al 2007, Bc_u in equation 1 is denoted as BC_u.

[2] McNulty et al., (2007) and Duarte et al., (2013) denoted Bc_w in equation 2 as BC_w. See Table A1-2 and A1-3 for more details.

[3] McNulty et al., (2007) and Duarte et al., (2013) denoted Bc_{dep} in equation 2 as BC_{dep}. See Table A1-2 and A1-3 for more details.

[4] McNulty et al., (2007) and Duarte et al., (2013) denoted Bc_u in equation 2 as BC_u. See Table A1-2 and A1-3 for more details.

Table A1-2. Description of Variables in the Database for Forest Soil Critical Load of Acidity Calculations (for McNulty et al., 2007, 2013)

Variable	**Units**	**Description of the Variable**	**Data Source**	**Dates of Data**	**Assumptions**
$ANC_{le,crit}$	eq/ha-yr	Critical leaching of forest soil acid neutralizing capacity (ANC) represents the buffering capacity of forest soils to acidic deposition.	This variable is calculated as shown in Equation 6 on page 285 in McNulty et al., (2007).	n/a	Critical leaching of ANC is calculated as according to Equation 2. See other variables in this table for associated assumptions. Based on the description in McNulty et al., (2007), the base cation deposition Bc_{dep} was equal to (Bc; Ca+Mg+K) but was denoted as BC_{dep}. Within the $ANC_{le,crit}$ term, base cation weather is denoted as BCw and assumed to be equal to (Bc; Ca+Mg+K+Na). It is unclear if McNulty et al., (2007) restricted the $ANC_{le,crit}$ estimates to the rooting zone.
ChemCriteria	n/a	Chemical criterion used for CL of acidity calculations: soil solution [Bc] (Ca+Mg+K):[Al] molar ratio. This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).	Page 286 in McNulty et al., (2007).	n/a	This is used in calculating $ANC_{le,crit}$ and indicates the sensitivity of the biological receptor to the soil conditions created by acidifying deposition.

ChemThreshold	n/a	Critical (or threshold) value for the chemical criterion given in 'ChemCriteria.'	Page 285 of the McNulty et al., (2007).	n/a	The critical value for the chemical criterion, molar [Bc]:[Al] ratio[1], was set at 10 for broadleaf forests and 1 for coniferous forests. If the [Bc]:[Al] ratio declines below the critical value, there is an increased likelihood of adverse impacts on trees.
BC_{dep}	eq/ha-yr	Total deposition of base cations (BC; Ca+Mg+K+Na). This term is included in the calculation of CL (Eq. A1-1).	Pages 282-283 of McNulty et al., (2007).	1994-2000	Base cation deposition was the sum of annual wet Ca, Mg, K, and Na deposition (i.e., dry deposition not included). The data source was the National Atmospheric Deposition Program/National Trends Network.
Bc_{dep}	eq/ha-yr	Total deposition of nutrient base cations (Bc; Ca+Mg+K). This term is included in the calculation of ANCle,crit (Eq. A1-2).	n/a	n/a	McNulty et al., (2007) denotes this as BC_{dep} in the $ANC_{le,crit}$ (equation 2).
Cl_{dep}	eq/ha-yr	Total deposition of chloride. This term is included in the calculation of CL (Eq. A1-1).	Page 283 of McNulty et al., (2007).	1994-2000	McNulty et al., (2007) used uncorrected wet deposition data, acknowledging that the SMB may have overestimated forest soil CL within 70 km of the coast due to sea salt. The data source was the National Atmospheric Deposition.
BC_w	eq/ha-yr	Weathering of base cations (BC; Ca+Mg+K+Na). This term is included in the calculation of CL (Eq. A1-1).	Described on pages 283-284 of McNulty et al., (2007, 2013). CONUS-SOIL from the Earth System Science Center (ESSC) at Pennsylvania State University. Key soil data inputs included map units	n/a	Base cation weathering rates were calculated using the clay correlation–substrate methods for CLs for soils. This method uses a combination of parent material and clay percent to determine the weathering rate. Parent material dictates which of the following equations is used to determine the weathering rate.

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[1] McNulty et al., (2007) denoted [Bc]:[Al] ratio as BC/AL

			and clay fraction. (CONUS – Continental United States).		- Acid substrate: $BC_w = (56.7 \times \%clay) - (0.32 \times \%clay^2)$ - Intermediate substrate: $BC_w = 500 + (53.6 \times \%clay) - (0.18 \times \%clay^2)$ - Basic substrate: $BC_w = 500 + (59.2 \times \%clay)$ Parent material for each soil map unit was classified using the dominant mineralogy based on the STATSGO map unit component (comp) and taxonomic (tax) classifications tables. This can be found in Table 3 of McNulty et al. (2007). The percent clay was derived from a weighted average of 11 standard soil layers per map unit using the following equation: Clay fraction for soil = Sum of s_{1-11} x (t/depth to bedrock layer) where s is the percent of clay in each soil map unit, t is the thickness of each soil layer in centimeters, and depth to bedrock is the mean depth to bedrock for each map unit in centimeters. Soil depth in meters was obtained from the CONUS-SOIL “depth to bedrock” layer. This layer identified map units with bedrock less than 1.52 m below the soil surface (ie., map units coded 1.52 m did not encounter bedrock).

					Based on the description in McNulty et al., (2007), it appears that the BCw estimates were for the whole soil profile (i.e, were not restricted to the forest rooting zone).
Bc_w	eq/ha-yr	Weathering of nutrient base cations (Bc; Ca+Mg+K). This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).			It is not clear if Bcw was estimated in McNulty et al., (2007) and used in the calculations of ANCle,crit
Bc_u	eq/ha-yr	Net removal of nutrient base cations (Bc; Ca+Mg+K) through removal of trees with harvesting. This term is included in the calculation of CL (Eq. A1-1) and $ANC_{le,crit}$ (Eq. A1-2).	Page 285, Table 4 in McNulty et al., (2007, 2013). The following databases were used to create Table 4: USDA Forest Service Forest Inventory and Analysis database (http://www.fia.fs.fed.us). Tree Chemistry Database (Pardo et al., 2005). Forest Cover Types came from the National Atlas (nationalatlas.gov).	n/a	Base cation and N uptake/removal were included in soil CL calculations in non-Wilderness areas (in Wilderness areas, uptake was set to 0 eq/ha-yr). It was assumed that trees would be harvested and removed in non-Wilderness areas. Base cation and N uptake were calculated using the following equation: Uptake (eq/ha-yr) = AVI x NC x SG x %bark x 0.65. AVI is the average forest volume increment annually (growth), NC is the base cation or N nutrient concentration in bark and bole, SG is the specific gravity of the bark and bole wood, percent bark is the volume growth that is allotted to bark, and it is assumed that 65 percent of the tree volume is removed from the site when harvested. Forest uptake of base cations and N was calculated for 21 forest types and the results can be found in Table 4 of the

					McNulty et al., (2007). The National Atlas forest cover types were used to map associated uptake for each grid cell. McNulty et al., (2007) denotes this parameter as BC_u in the (equations 1 and 2).
Q_{le}	m^3/ha-yr	Precipitation surplus percolating/leaching below the rooting zone [Defined as Annual Run-off in McNulty et al., (2007). This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).	Page 284 in McNulty et al., (2007).	1951 to 1980	Water leaching below rooting zone was estimated using values from interpolated isopleth maps representing average annual runoff in inches per year for the U.S. from 1951 to 1980 produced by Gebert et al., (1987). It is unclear whether this runoff datalayer estimated water leaching below the rooting zone. Values were assigned to the 1 km^2 grid cells through a geospatial technique called “linegrid”.
K_{gibb}	m^6/eq^2	Gibbsite equilibrium constant. This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).	Page 286 (Table 5) in McNulty et al., 2007).	n/a	This varies partly as a function of the organic matter content of the soil. The value for K_{gibb} ranges from 950 m^6/eq^2 in mineral soil to 9.5 m^6/eq^2 in peaty and organic soils.
N_i	eq/ha-yr	Long-term net N immobilization in the rooting zone of the soil. This term is included in the calculation of CL (Eq. A1-1).	Page 285 in McNulty et al., (2007).	n/a	Set at 42.86 eq/ha-yr based on an average of the colder and warmer climate N immobilization rates reported for European countries.

N_u	eq/ha-yr	Net removal of N through removal of trees with harvesting. This term is included in the calculation of CL (Eq. A1-1).	Page 284-285 in McNulty et al., (2007).	n/a	See BC_u.
N_{de}	eq/ha-yr	Amount of N denitrified. This term is included in the calculation of CL (Eq. A1-1).	Page 285 in McNulty et al., (2007).	n/a	This was set to 0 eq/ha-yr because all forests were assumed to be on upland soils where denitrification is minimal.

Forest Soil Critical Loads of Acidify as calculated by Duarte et al., (2011, 2013)

The CL of acidity estimates within Duarte et al., (2011, 2013) were modeled using the steady-state the CLRTAP (2004) Simple Mass Balance (SMB) equation (1) and ANC equation (2) that were also applied in McNulty et al., (2007). However, based on the equation outlined by Duarte et al., (2011, 2013), it is uncertain if the deposition of chloride (Cl_{dep}) was included in the calculations of CLs. The SMB equation assumes the ecosystem is at long-term equilibrium or steady state and the equilibrium condition is equal to the ecosystem's CL. The Duarte et al., (2011, 2013) CL estimates represent data from 2,565 forested plots in seven states in the Northeastern U.S. These forested plots are from regional and national forest health surveys. **Table A1-3** provides a description of the parameters used in the SMB CL calculations (and included in the NCLD) and the data sources and assumptions adopted by Duarte et al., (2011, 2013).

Table A1-3. Description of Variables in the Database for Forest Soil Critical Load of Acidity Calculations (for Duarte et al., 2011, 2013)

	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
$ANC_{le,crit}$	eq/ha-yr	Critical leaching of forest soil acid neutralizing capacity (ANC) represents the buffering capacity of forest soils to acidic deposition. This term is included in the calculation of CL (Eq. A1-1).	This variable is calculated as shown in Equation 2 on page 6 in Duarte et al., (2011) and page 4 in Duarte et al., (2013).	n/a	Critical leaching of ANC is calculated as according to Equation 2. See other variables in this table for associated assumptions. Based on the description in Duarte et al., (2011, 2013), the base cation terms, BC_{dep} and BC_w, within the $ANC_{le,crit}$ estimates appear to be for total base cations (BC; Ca+Mg+K+Na) and not nutrient base cations (Bc; Ca+Mg+K).
ChemCriteria		Chemical criterion used for CL of acidity calculations: soil solution [Bc] (Ca+Mg+K):[Al] molar ratio. This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).		n/a	This is used in calculating $ANC_{le,crit}$ and indicates the sensitivity of the biological receptor to the soil conditions created by acidifying deposition.
ChemThreshold		Critical value for the chemical criterion given in 'ChemCriteria'.	Page 5 in Duarte et al., (2011), page 4 in Duarte et al., (2013), NEG/ECP 2003.	n/a	The critical value for the chemical criterion, molar [Bc]:[Al] ratio, was 10. This value was selected with the objective of having no decrease in soil base saturation. If the [Bc]:[Al] ratio declines below the critical value, there is an increased likelihood of adverse impacts on trees.
BC_{dep}	eq/ha-yr	Total deposition of base cations (BC; Ca+Mg+K+Na). This term is included in the calculation of CL (Eq. A1-1).	Page 9 and 17 in Duarte et al. (2011), page 4 in Duarte et al. (2013).	1980-1991	Base cation deposition was included as the sum of wet and dry Ca, Mg, K, Na deposition. The data for the Northeast were calculated using ClimCalc model which calculates wet + dry deposition based on NADP data (1980-1991), National Dry Deposition Network, NDDN (1989-1990) and the Digital Elevation Model

	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
					(Ollinger et al., 1993; http://www.pnet.sr.unh.edu/climcalc).
Bc_{dep}	eq/ha-yr	Total deposition of nutrient base cations (Bc; Ca+Mg+K). This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).			It is not clear if Bc_{dep} was estimated in Duarte et al., (2011, 2013) and used in the calculations of $ANC_{le,crit}$.
Cl_{dep}	eq/ha-yr	Total deposition of chloride. This term is included in the calculation of CL (Eq. A1-1).	Page 9 and 17 in Duarte et al., (2011).	1980-1991	Cl deposition was included as the sum of wet +dry deposition from ClimCalc (see above). ClimCalc deposition is corrected for sea salt contribution. However, based on CL equation outlined in Duarte et al., (2011, 2013), it is uncertain if Cl deposition was included in the calculations.
BC_w	eq/ha-yr	Weathering of base cations (BC; Ca+Mg+K+Na). This term is included in the calculation of CL (Eq. A1-1). The mid-point value was used for the data in the NCLD.	Page 11 and 20 in Duarte et al., (201), page 6 in Duarte et al., (2013). SSURGO http://soils.usda.gov/survey/geography/ssurgo/.	n/a	This is explained in more detail in Duarte et al., (2011, 2013). Key information is included here. Soil base cation weathering rates in the rooting zone were calculated using the clay correlation–substrate method for soil CLs (same method as used in McNulty et al., (2007)). This method uses a combination of parent material and clay percent to determine the weathering rate. Parent material dictates which of the following equations is used to determine the weathering rate. • Acid substrate: BCw = (56.7 x %clay) – (0.32 x %clay2) • Intermediate substrate: BCw = 500 + (53.6 x %clay) – (0.18 x %clay2)

	Units	**Description of the Variable**	**Data Source**	**Dates of Data**	**Assumptions**
					• Basic substrate: BCw = 500 + (59.2 x %clay) Digitized soil county survey maps (Soil Survey Geographic (SSURGO) Database) were used to identify soil series. The Official Soil Series were then used to assign mean values for the required soil parameters (depth, clay percent, texture, moisture, substrate type).
Bc_w	eq/ha-yr	Weathering of nutrient base cations (Bc; Ca+Mg+K). This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).			It is not clear if Bc_w was estimated in Duarte et al., (2011, 2013) and used in the calculations of $ANC_{le,crit}$.
Bc_u	eq/ha-yr	Net removal of nutrient base cations (Bc; Ca+Mg+K) through removal of trees with harvesting. This term is included in the calculation of CL (Eq. A1-1) and ANCle,crit (Eq. A1-2).	Page 10 and 19 in Duarte et al. (2011), page 5 in Duarte et al. (2013). USDA Forest Service Forest Inventory and Analysis database (http://www.fia.fs.fed.us). Tree Chemistry Database (Pardo et al. 2005).	n/a	Base cation (Bc) and N uptake/removal were included in soil CL calculations in non-Wilderness areas (in Wilderness areas and areas without vegetation (NRCS sites), uptake was set to 0 eq/ha-yr). It was assumed that trees would be harvested and removed in non-Wilderness areas only. In order to calculate Bc and N uptake and removal for each site, biomass (kg) by above-ground compartment (stem, branch, bark) were multiplied by nutrient concentration (%) by compartment. All of the required data for calculating annual merchantable removals of growing stock trees on timberland are available in the publicly accessible FIA database

	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
					(http://www.fia.fs.fed.us/tools-data/data/). For the P2 plots, the sample years used varied by state due to FIA's sampling cycles. (Table 2 in Duarte et al., (2011)). The data for the P3 plots in the New England states were provided by FIA and are from 1994-1995 (prior to the 1999 merger of FHM and FIA). Data for the New York P3 plots were from years 1993 and 2004. The algorithms outlined in Table 5 of Chapter 4 in the FIA Database Documentation (Algorithms for summarizing data, http://nrcs2.fs.fed.us/4801/fiadb/fiadb documentation/fiadb chapter 4.htm) were used. These current removal rates were combined with chemistry data from the Tree Chemistry Database (Pardo et al., 2005) in order to calculate the annual nutrient removal rates. The Tree Chemistry Database includes information on nutrient concentration by compartment (stem, branch, bark, foliage) for tree species found in the northeastern U.S. Although removal rates were not available for sites other than FIA, species composition and DBH data (used for calculating biomass) were available for the other vegetation survey sites and additional research plots. Saw timber harvest was considered to be the dominate harvest type for New England and New York and estimates of tree wood and bark were made using allometric equations (Jenkins et al., 2003; Vermont Site-Specific Report; http://www.fs.fed.us/ne/durham/4352/critical_loa

	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
					ds/Critical_loads_webs/home.htm). The estimates of tree wood and bark biomass were combined with compartment-specific nutrient concentrations in order to estimate the nutrient content via saw timber harvest. Annual biomass extraction rates for saw timber compiled from FIA data and tabulated by county, land-ownership category (public, private) and gross forest type (softwood, hardwood, mixed) were used to estimate the nutrient removal from these non-FIA sites.
Q_{le}	m^3/ha-yr	Precipitation surplus percolating/leaching below the rooting zone. This term is included in the calculation of ANCle,crit (Eq. A1-2).	Page 10 in Duarte et al., (2011), page 4 in Duarte et al. (2013) (Section 2.3.1), 2013 (page 4).	1950 to 1980	Calculated from precipitation volume (from ClimCalc model, Ollinger et al. 1993) minus evapotranspiration (E.K. Miller, personal communication).
K_{gibb}	m^6/eq^2	Gibbsite equilibrium constant. This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).	NEG/ECP, (2001).	n/a	K_{gibb} was set at 8.8 m^6/eq^2.
N_i	eq/ha-yr	Long-term net N immobilization in the rooting zone of the soil. This term is included in the calculation of CL (Eq. A1-1).	Page 7 in Duarte et al., (2011).	n/a	Set at 143 eq/ha-yr (2 kg/ha-yr) (Pardo, 2010).
N_u	eq/ha-yr	Net removal of N through removal of trees	Page 10 and 19 in Duarte et al.,	n/a	See Bc_u.

	Units	**Description of the Variable**	**Data Source**	**Dates of Data**	**Assumptions**
		with harvesting. This term is included in the calculation of CL (Eq. A1-1)	(2011), pages 5-6 in Duarte et al., (2013).		
N_{de}	eq/ha-yr	Amount of N denitrified. This term is included in the calculation of CL (Eq. A1-1).	Page 8 in Duarte et al., (2011).	n/a	Set at 0 eq/ha-yr based on assumption that denitrification rates in upland forests are low and negligible (Binkley et al., 1995).

Forest Soil Critical Loads of Acidity as presented by Phelan et al., (2014, 2016)

The CL of acidity estimates within Phelan et al., (2014) were modeled using the steady-state the CLRTAP (2004) Simple Mass Balance (SMB) equation (1) and ANC equation (2) that were also applied in McNulty et al., (2007) and Duarte et al., (2011, 2013). The SMB equation assumes the ecosystem is at long-term equilibrium or steady state and the equilibrium condition is equal to the ecosystem's CL. The Phelan et al., (2014) CL estimates represent data from fifty-one 1 m^2 points within forests in Pennsylvania. These points are the sampling locations of the U.S. Geological Survey Landscapes project (USGS, 2013 - https://minerals.cr.usgs.gov/projects/geochemical_landscapes/); the USGS Landscapes project dataset was the source of soil mineralogy data that were used to estimate soil base cation weathering (Bc_w) by Phelan et al., (2014). All other data for the SMB equation parameters and Bc_w estimates were acquired as GIS raster or polygon datasets that were extracted for data for each of the fifty-one 1 m^2 forest points. **Table 1A-4** provides a description of the parameters used in the SMB CL calculations (and included in the NCLD) and the data sources and assumptions adopted by Phelan et al. (2014).

Since the publication of Phelan et al., (2014), the CL estimates were revised in 2016 to restrict BC_w and Bc_w rates to the forest rooting zone. This revised values are included in the CLAD NCLD and described in **Table A1-4**.

Table A1-4. Description of Variables in the Database for Forest Soil Critical Load of Acidity Calculations (for Phelan et al, 2014, 2016)

Variable	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
ANC_{crit}	eq/ha-yr	Critical leaching of forest soil acid neutralizing capacity (ANC) represents the buffering capacity of forest soils to acidic deposition. This term is included in the calculation of CL (Eq. A1-1).	This variable is calculated using equation 2 and as shown on page 9 in Phelan et al., (2014).	n/a	Critical leaching of $ANC_{le,crit}$ is calculated as according to Equation 2. See other variables in this table for associated assumptions. The nutrient base cation (Bc_w) estimates used in the ANC calculations by Phelan et al., (2014) were for the full soil profile (i.e., were not restricted to the forest tree rooting zone). However, since the publication of Phelan et al., (2014), the Bc_w estimates have been revised (in 2016) for restriction of weathering to the rooting zone, and these revised estimates were used to re-calculate $ANC_{le,crit}$.
ChemCriteria	n/a	Chemical criterion used for CL of acidity calculations: soil solution [Bc] (Ca+Mg+K):[Al] molar ratio. This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2)	n/a	n/a	This is used in calculating $ANC_{le,crit}$ and indicates the sensitivity of the biological receptor of interest to the soil conditions created by acidifying deposition.
ChemThreshold	Molar ratio	Critical value for the chemical criterion given in 'ChemCriteria'.	Page 9 in Phelan et al., (2014).	n/a	Soil CLs were based on soil solution [Bc]:[Al] molar ratio of 10 for deciduous and mixed forests. If the ratio declines to below the critical value, there is an increased likelihood of adverse impacts on trees.
BC_{dep}	eq/ha-yr	Total deposition of all base cations (BC; Ca+Mg+K+Na). This term is included in the calculation of CL (Eq. A1-1).	Page 7 in Phelan et al., (2014).	2002	Base cation deposition was the sum of annual wet Ca, Mg, K, and Na deposition (i.e., dry deposition not included). Data was from the National Atmospheric Deposition Program/National Trends Network.

Variable	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
Bc_{dep}	eq/ha-yr	Total deposition of nutrient base cations (Bc; Ca+Mg+K). This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-2).	Page 7 in Phelan et al., (2014).	2002	Base cation deposition was the sum of annual wet Ca, Mg, K, and Na deposition (i.e., dry deposition not included). Data was from the National Atmospheric Deposition Program/National Trends Network.
Cl_{dep}	eq/ha-yr	Total deposition of chloride. This term is included in the calculation of CL (Eq. A1-1).	Page 7 in Phelan et al., (2014).	2002	Chloride deposition was annual wet Cl deposition from the National Atmospheric Deposition Program/National Trends Network. Phelan et al., (2014) did not correct Cl deposition for sea-salt. Therefore, the CL for sites within 70 km of the coast may have been overestimates. Atmospheric Deposition Program/National Trends Network.
BC_w	eq/ha-yr	Weathering of all base cations (BC; Ca+Mg+K+Na). This term is included in the calculation of CL (Eq. A1-1).	Page 3-9 in Phelan et al., (2014).	n/a	Total BC_w was estimated using the PROFILE model. See Phelan et al., (2014) for more details on model parameters, calibration, and simulations. Phelan et al., (2014) estimated BC_w for the full soil profile (i.e., not restricted to forest tree rooting zone). However, since the publication of Phelan et al. (2014), the BC_w estimates have been revised (in 2016) for restriction of weathering to the rooting zone.
Bc_w	eq/ha-yr	Weathering of nutrient base cations (Ca+Mg+K). This term is included in the calculation of CL (Eq. A1-2).	Pages 3-9 of Phelan et al., (2014).	n/a	Nutrient Bc_w was estimated using the PROFILE model. See Phelan et al. (2014) for more details on model parameters, calibration, and simulations. Phelan et al. (2014) estimated Bc_w for the full soil profile (i.e., not restricted to forest tree rooting zone). For consistency with the other parameters of the SMB equation and representing soil conditions in the portion of the soil where the roots of the biological indicator are located, Bc_w should be restricted to the rooting zone.

Variable	**Units**	**Description of the Variable**	**Data Source**	**Dates of Data**	**Assumptions**
Bc_u	eq/ha-yr	Net removal of nutrient base cations (Bc; Ca+Mg+K) through removal of trees with harvesting. This term is included in the calculation of CL (Eq. A1-1) and $ANC_{le,crit}$ (Eq. A1-2).	Page 9 in Phelan et al., (2014).	n/a	Based on Bc_u estimates from McNulty et al. (2007). See Tables 2 and 4 in Phelan et al., (2014) for more details.
Q_{le}	m^3/ha-yr	Precipitation surplus percolating/leaching below the rooting zone. This term is included in the calculation of ANCle,crit (Eq. A1-2).	Page 7 in Phelan et al., (2014).	1951-1980, 2002.	Precipitation surplus that leached below the rooting zone was calculated using a combination of surface run-off, subsurface run-off, and soil moisture content.
K_{gibb}	m^6/eq^2	Gibbsite equilibrium constant. This term is included in the calculation of $ANC_{le,crit}$ (Eq. A1-1)	Page 10 in Phelan et al., (2014).	n/a	This varies partly as a function of the organic matter content of the soil. The value for Kgibb was 950 m6/eq-2 due to the low organic matter contents of the soils.
N_i	eq/ha-yr	Long-term net N immobilization in the rooting zone of the soil. This term is included in the calculation of CL (Eq. A1-1).	Page 10 in Phelan et al., (2014).	n/a	Set at 42.86 eq/ha-yr based on an average of colder and warmer climate N immobilization rates reported for European countries.
N_u	eq/ha-yr	Net removal of N through removal of trees with harvesting. This term is included in the calculation of CL	Page 10 in Phelan et al., (2014).	n/a	See Bc_u.

Variable	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
		(Eq. A1-1).			
N_{de}	eq/ha-yr	Amount of N denitrified. This term is included in the calculation of CL (Eq. A1-1).	Page 10 in Phelan et al., (2014).	n/a	This was set to 0 eq/ha-yr because all forests were assumed to be on upland soils were denitrification is minimal.

Forest Soil Target Loads of Acidity as presented by Sullivan et al., (2011a, 2011b)

The target loads (TLs) of acidity in Sullivan et al., (2011a, 2011b) were modeled using the dynamic MAGIC model. As described by the authors, model simulations were based on one source of acidic deposition (S), two sensitive soil receptors (soil and soil solution), and one or two chemical indicators for each receptor (soil base saturation, soil solution molar [Bc/Al] and [Ca/Al] ratios). The TLs were estimated based on two end-point years of model simulation (2050 and 2100) for a total of 97 watersheds in the Adirondack mountains in New York. MAGIC model input data consisted of water and soil chemistry, deposition and base cation uptake parameters. Soil and water parameters were measured in 76 representative watersheds with the soils data being from three soil pits within each watershed (Sullivan et al., 2006). In addition, this subset of data includes 65 TLs for watersheds in the Southern Blue Ridge Province in VA. These 65 watershed used the same methods as described in Sullivan et al., (2011a, 2011b) using base saturation receptor only. **Table 1A-5** describes the variables used to estimate TLs of acidity using the MAGIC model, and how these parameters were represented by Sullivan et al., (2011a, 2011b) in their estimates of TLs. This table also includes additional variables required by the MAGIC model to estimate TLs.

Table A1-5. Description of Variables in the Database for Forest Soil Target Loads of Acidity Calculations (for Sullivan et al., 2011a, 2011b)

Variable	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
$ANC_{le,crit}$	meq/m^2-yr	Critical leaching of forest soil acid neutralizing capacity (ANC) represents the buffering capacity of forest soils to acidic deposition.	n/a	n/a	
ChemCriteria		Chemical criteria used for target load of acidity calculations: soil base saturation (%) and soil solution [Ca]:[Al] and [Bc] (Ca+Mg+K)/[Al] molar ratios.	Page 5 in Sullivan et al., (2011a).	n/a	These were selected to explore a variety of criterion to protect against biological impact of acidifying S deposition. The [Ca]:[Al] and [Bc]:[Al]criteria are commonly used to protect forest resources.
ChemThreshold		Critical value for the chemical criterion given in 'ChemCriteria'.	Page 5 in Sullivan et al., (2011a).	n/a	The critical values for soil base saturation were 5% and 10% (which bracket the base saturation of the upper B soil horizon in many acid-sensitive Adirondack watersheds). The critical values for the soil solution chemical criterions were 1 and 10, which are commonly used threshold critical values to protect forest resources (see references in Sullivan et al., 2011a, 2011b). If the [Ca]:[Al] and [Bc]:[Al] ratio declines below the critical value, there is an assumed increased likelihood of adverse impacts on trees.
BC_{dep}	meq/m^2-yrr	Total deposition of base cations (BC; Ca+Mg+K+Na).	Page 4 in Sullivan et al., (2011a).	1850-1978	Base cation deposition was estimated by assuming that: background pre-industrial deposition was 10% of current deposition, deposition after 1850 increased linearly to 1950 estimated deposition,

Variable	**Units**	**Description of the Variable**	**Data Source**	**Dates of Data**	**Assumptions**
					wet deposition from 1950-1978 was derived from empirical relationships between deposition and emissions of particulate matter (PM-10), and assumed dry to wet ratio of 0.5.
Bc_{dep}	meq/m^2-yr	Total deposition of nutrient base cations (Bc; Ca+Mg+K).	n/a	n/a	
Cl_{dep}	meq/m^2-yr	Total deposition of chloride.	Page 4 in Sullivan et al., (2011a).	1850-1978	Base cation deposition was estimated by assuming that: background pre-industrial deposition was 10% of current deposition, deposition after 1850 increased linearly to 1950 estimated deposition, wet deposition from 1950-1978 was derived from empirical relationships between deposition and emissions of particulate matter (PM-10), and assumed dry to wet ratio of 0.25.
BCw	meq/m2-yr	Weathering of total base cations (BC; Ca+Mg+K+Na).	Not specified in Sullivan et al., (2011a).	n/a	BCw is calculated as a mass-balance difference within MAGIC. However, BCw values are not provided.
Bc_w	meq/m^2-yr	Weathering of nutrient base cations (Bc; Ca+Mg+K).	n/a	n/a	
Bc_u	meq/m^2-yr	Net removal of nutrient base cations (Bc; Ca+Mg+K) through removal of trees with harvesting.	See page 4 in Sullivan et al., (2011a). Appeared to us Bc_u estimates from McNulty et al., (2007)	n/a	Base cation uptake/removal was only included in target load calculations for non-wilderness/non-protected areas. In wilderness/protected areas, uptake was set to 0 meq/m^2/yr based on the assumption that trees would not be removed and nutrients would remain on site. See Table 2 and he description for McNulty et al., (2007) for a greater description of the method and assumptions of the Bc_u estimates.

Variable	**Units**	**Description of the Variable**	**Data Source**	**Dates of Data**	**Assumptions**
Q_{le}	m^3/ha-yr	Precipitation surplus percolating/leaching below the rooting zone.	Not specified in Sullivan et al., (2011a).		Q is an input parameter to MAGIC.
K_{gibb}	m^6/eq^2	Gibbsite equilibrium constant.	Not specified in Sullivan et al., (2011a).	n/a	K_{gibb} is an input parameter to MAGIC.
N_i	meq/m^2-yr	Long-term net N immobilization in the rooting zone of the soil.	n/a	n/a	
N_u	meq/m^2-yr	Net removal of N through removal of trees with harvesting.	n/a	n/a	
N_{de}	meq/m^2-yr	Amount of N denitrified.	n/a	n/a	
pH		Soil pH (determined on 0.01 M $CaCl_2$ extractions) of O and upper B horizon (10 cm).	Based on soil samples collected from three locations in each watershed (Sullivan et al., 2006).	2003	Assumed that pH from soils collected at three locations within watershed were representative of watershed.
exchageable acidity	$cmol_c$/kg	Exchangeable acidity (determined on 1M KCl extractions) of O and upper B horizon (10 cm).	Based on soil samples collected from three locations in each watershed	2003	Assumed that exchangeable acidity from soils collected at three locations within watershed were representative of watershed.

Variable	Units	Description of the Variable	Data Source	Dates of Data	Assumptions
			(Sullivan et al., 2006).		
Exchangeable aluminum (Al)	$cmol_c$/kg	Exchangeable aluminum (determined on 1M NH_4Cl extractions) of O and upper B horizon (10 cm).	Based on soil samples collected from three locations in each watershed (Sullivan et al., 2006).	2003	Assumed that exchangeable Al from soils collected at three locations within watershed were representative of watershed.
Effective cation exchange capacity (CEC_e)	$cmol_c$/kg	Calculated as sum of exchangeable base cations plus exchangeable acidity.	Based on soil samples collected from three locations in each watershed (Sullivan et al., 2006).	2003	Assumed that CEC_e from soils collected at three locations within watershed were representative of watershed.
Base saturation	%	Calculated as the percentage of the CEC_e occupied by exchangeable cations.	Based on soil samples collected from three locations in each watershed (Sullivan et al., 2006).	2003	Assumed that base saturation from soils collected at three locations within watershed were representative of watershed.
total N and C content	%	Determined by combustion analysis on samples from O and upper B horizon (10 cm).	Based on soil samples collected from three locations in each watershed (Sullivan et al., 2006).	2003	Assumed that total N and C content from soils collected at three locations within watershed were representative of watershed.

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Appendix 2

Surface Water Critical Loads of Acidity

The NCLD is comprised of critical loads (CLs) calculated from a common mass-balance approach with different ways for estimating watershed base cation weathering (e.g., F-Factor or dynamic model). The following section describes the models and components for calculating CLs of acidifying N and S deposition. Surface water CLs of acidity are listed in **Table A2-1**. See **Tables A2-2** to **A2-8** below for more detail information.

Table A2-1. Summary of Key Variables Among the Sources

Source	**CL Model**	**ChemCriterion/ ChemThreshold**	**BC_w Model**	**Bc_u/ N_u**	**N_i (eq/ha)**	**N_{de} (eq/ha)**	**Location**
USEPA, (2012) Scheffe et al., (2014)	Modified SSWC	ANC/50 or 20 µeq/L	F-Factor	No/No	Yes	Yes	Nationwide
Sullivan et al., (2012a) McDonnell et al., (2012)	Modified SSWC	ANC/50 µeq/L	MAGIC; Regression	Yes/Yes	Yes	Yes	Appalachian mountains of Virginia
Miller, (2011)	Modified SSWC	ANC/50 µeq/L	F-Factor	No/No	Yes	Yes	Northeast
DuPont, et al. (2005)	SSWC	ANC/40 µeq/L	F-Factor	No/No	Yes	Yes	New England
Vermont Department of Environmental Conservation, VDEC (2003, 2004, 2012)	SSWC	ANC/50 µeq/L	F-Factor	No/No	Yes	Yes	Vermont
McDonnell et al., (2014)	Modified SSWC	ANC/50 µeq/L	MAGIC; Regression	Yes/n/a	n/a	n/a	So. Appalachian mountains
Lynch et al., (2022)*	Modified SSWC	ANC/50 or 20 µeq/L	F-Factor	No/No	Yes	Yes	Nationwide
Lawrence.et.al., (2015)	MAGIC	ANC/20, 50, 100 µeq/L	MAGIC	Yes/Yes	n/a	n/a	Appalachian Mountains
Sullivan.et.al., (2005)	MAGIC	ANC/0, 20, 50 µeq/L	MAGIC	No/No	n/a	n/a	Loch Vale, Colorado
Sullivan.et.al., (2012b)	Modified SSWC/MAGIC	ANC/ 0, 20, 50 µeq/L	MAGIC; Regression	Yes/Yes	n/a	n/a	Adirondack Mountains, New York
Fakhraei.et.al., (2014)	PnET-BGC	ANC/11 µeq/L	PnET-BGC	No/No	n/a	n/a	New York
McDonnell et.al., (2021)	PnET-BGC	ANC/ 20, SS µeq/L	PnET-BGC	No/No	n/a	n/a	New York

*Reference to this document. Critical loads calculated from various water quality datasets using method described in Appendance B.

Steady-State Water Chemistry (SSWC) model and F-Factor (Scheffe et al., 2014, Miller, 2011, DuPont et al., 2005, VDEC, 2003, 2004, 2012, Lynch et al. 2022)

Critical loads were derived from present-day water chemistry and are based on the principle that excess base cation production within a catchment area should be equal to or greater than the acid anion input, thereby maintaining the ANC above a pre-selected level (Reynolds and Norris, 2001). This model assumes a mass balance and that all SO_4^{2-} in runoff originates from sea salt spray and anthropogenic deposition.

In the SSWC model, CL of acidity, CL(A), is a calculated form the principle that the acid load should not exceed the non-marine, base cation inputs minus a nutrient base cation uptake and ANC buffer to protect selected biota from being damaged (**Eq. A2-1**):

$$CL(A) = BC^{*}_{dep} + BC_{w} + - Bc_{u} - nANC_{crit} \tag{A2-1}$$

Where:

BC^{*}_{dep} (BC; Ca+Mg+K+Na) = the sea-salt corrected non-anthropogenic deposition of base cations;
BC_{w} (BC; Ca+Mg+K+Na) = the average watersed weathering flux;
Bc_{u} (Bc: Ca+Mg+K) = the net long-term average uptake of base cations in the biomass (i.e., the annual average removal of base cations due to harvesting);
$nANC_{crit}$ = the lowest ANC-flux that protects the biological communities.

Bc_{u} for these studies was set to zero.

For these CLs, the SSWC model was modified to incorporate simplified N framework whereby N components that account for nitrogen removal from long-term nitrogen immobilization and denitrification were included in the model (**Eq. A2-2**):

$$CL(A) = BC^{*}_{dep} + BC_{w} + N_{u} + N_{i} + N_{de} - Bc_{u} - nANC_{crit} \tag{A2-2}$$

Where:

N_{u} = N removal through removal of trees with harvesting;
N_{i} = N removal from long-term N immobilization;
N_{de} = N removal from the soil through microbial denitrification;

N_{i} was set equal to 4.30 meq/m^{2}-yr (McNulty et. al., 2007) and N_{de} was set equal to 7.14 meq/m^{2}-yr (Ashby et al., 1998) for sites in the east. For western states, a combined value of $N_{i}+N_{de}$ = 11.0 eq/ha-yr was used based on Nanus et al. (2012). N_{u} value varies depending on CL project.

See above Calculating Exceedance of Critical Loads of S, N and Combined S and N Deposition chapter for how to calculate CL exceedance Ex(A). In addition, exceedance for these CLs can be determined using the N_{le} (Henriksen and Posch, 2001) after removing the N terms from (**Eq. A2-3**):

$Ex(A) = S_{dep} + N_{le} - CL(A)$ (A2-3)

Where:

N_{le} = the sum of the measured concentrations of nitrate (NO_3^- μeq/L) and ammonia (NH_4^+ μeq/L) in the runoff (Q_s m/yr) as $([NO_3^-]+[NH_4^+])*Q_s$.

Equation A2-3 determines the CL exceedance based on S deposition while incorporating the present day measured (or assumed future) extent of N leaching.

F-factor and Pre-Acidification Base Cation Concentration for Input to SSWC

Since the average flux of base cations weathered in a catchment and reaching the water body is difficult to measure or compute from available information, the average flux of base cations and the resulting CL estimates were derived from water quality data (Sverdrup et al., 1990; Henriksen and Posch, 2001). Weighted annual mean water chemistry values were used to estimate average base cation fluxes, which were calculated from water chemistry data.

The pre-acidification non-marine flux of base cations for each lake or stream, BC^*_o, is (**Eq. A2-4**)

$BC^*_o = BC^*_{dep} + BC_w - Bc_u$ (A2-4)

Thus, CL for acidity can be re-written as (**Eq. A2-5)**

$CL(A) = BC^*_o - ANC_{limit} = Q_s.([BC^*]_o - nANC_{crit})$ (A2-5)

where **Eq. A2-5** expresses the CL for acidity in terms of catchment runoff Q_s (m/yr) and concentration ($[x] = X/Q_s$).

Present-day surface water concentrations of base cations are elevated above their steady-state concentrations because of base cation leaching through ion exchange in the soil due to anthropogenic inputs of SO_4^{2-} to the watershed. For this reason, present-day surface water base cation concentrations are higher than natural or pre-acidification levels, which if not corrected for, would result in CL values outside of natural conditions. To estimate the pre-acidification flux of base cations, we started by calculating the present flux of base cations, BC^*_t, given by (**Eq. A2-6**)

$BC^*_t = BC^*_{dep} + BC_w - Bc_u + BC_{exc}$ (A2-6)

where

BC_{exc} = the release of base cations due to ion-exchange processes.

Assuming that deposition, weathering rate, and net uptake have not changed over time, BC_{exc} was obtained by subtracting **Eq. A2-4** from **Eq. A2-2** to get **Eq. A2-7**:

$BC_{exc} = BC^*_t - BC^*_o$ (A2-7)

This present-day excess production of base cations in the catchment was related to the long-term changes in inputs of non-marine acid anions ($\Delta SO_4^{2-} + \Delta NO_3^-$) by the F-factor (see below) (**Eq. A2-8**):

$BC_{exc} = F\ (\Delta SO^*{}_4^{2-} + \Delta NO_3^-)$ (A2-8)

For the pre-acidification base cation flux, we get from **Eq. A2-9**:

$BC^*_o = BC^*_t – F\ (SO^{*\,2-}_{4\ \ t} - SO^*_{42-,o} + NO_{3,t} - NO_{3,o})$ (A2-9)

The pre-acidification nitrate concentration, $NO_{3,o}$, was assumed to be zero. An F-factor was used to correct the concentrations and estimate pre-acidification base concentrations. An F-factor is a ratio of the change in non-marine base cation concentration due to changes in strong anion concentrations (Henriksen, 1984; Brakke et al., 1989, 1990) (**Eq A2-10**):

$F = [BC^*]_t - [BC^*]_o / [SO^{*}_4{}^{2-}]_t - [SO^{*}_4{}^{2-}]_o + [NO_3^-]_t - [NO_3^-]_o$ (A2-10)

where the subscripts t and “O” refer to present and pre-acidification concentrations, respectively.
If F=1, all incoming protons are neutralized in the catchment (only soil acidification occurs); at F=0, none of the incoming protons are neutralized in the catchment (only water acidification occurs). The F-factor was estimated empirically to be in the range 0.2–0.4 based on the analysis of historical data from Norway, Sweden, the United States, and Canada (Henriksen, 1984). Brakke et al., (1989, 1990) later suggested that the F-factor should be a function of the base cation concentration (**Eq. A2-11**):

$F = \sin (\pi/2\ Qs.[BC^*]t/[S])$ (A2-11)

where

Q_s = the annual runoff (m/yr)
[BC] = the base cation concentration at which F=1; and for $[BC^*]_t>[S]$ F is set to 1.

For Norway [S] has been set to 400 μeq/m^3 (ca. 8 mg Ca/L) (Brakke et al., 1989, 1990).

The pre-acidification sulfate concentration in lakes, $[SO^{*}_4{}^{2-}]_o$, is assumed to consist of a constant atmospheric contribution and a geologic contribution proportional to the concentration of base cations (Brakke et al., 1989, 1990). The $[SO^{*}_4{}^{2-}]_o$, is determined using the relationship between Sum of Base Cations (SBC) and pre-acidification sulfate concentration, derived from different methods based on the study. Scheffe et al., (2014) used the MAGIC model estimates $[SO^{*}_4{}^{2-}]_o$. The resulting linear regression was used to calculate $[SO^{*}_4{}^{2-}]_o$, for each water body (**Figure A2-1**) for CLs using the methodology in Scheffe et al., (2014) (**Eq. A2-12**). **Tables A2-2** to **A2-5** description the variables used for the studies in this group.

$[SO4^*]o, = 0.0229*SBC + 9.0341$ (A2-12)

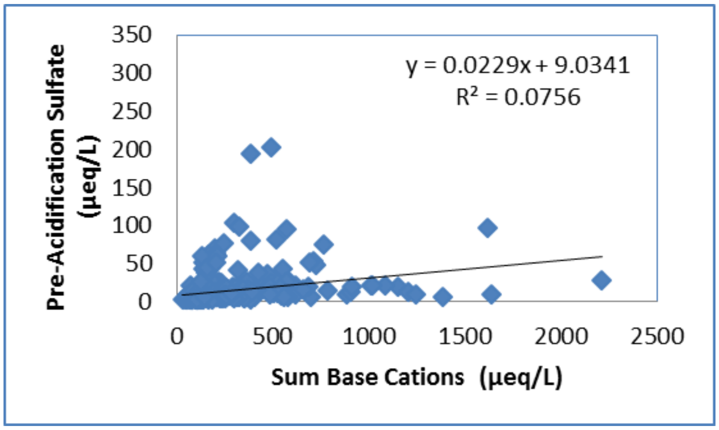

Figure A2-1. The relationship between Sum of Base Cations (SBC) and pre-acidification sulfate concentration as derived from MAGIC model estimates.

Table A2-2. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations (for EPA, 2012 and Scheffe et al., 2014, Lynch et al. 2020)

Variable	**Criterion (Unit)**	**Description of the Variable**	**Data Source**	**Data**	**Assumptions and value**
ChemCriterion/ ChemThreshold	ANC (µeq/L)	Critical value for chemical criterion	n/a	n/a	For Western states (AZ, CA, CO, OR, MT, NM, NV, UT, WA, WY) an ANC value of 20 µeq/L used while all other used 50 µeq/L.
BC_{dep}	meq/m^2-yr	BC; Ca+Mg+K+Na	NADP deposition corrected with Prism precipitation OR NADP deposition model by Grimm	Reference year of water quality data (1982 to 2012)	[BC_t] from water quality used. However, wet BC_{dep} was calculated.
CL_{dep}	meq/m^2-yr	Wet deposition of chloride	NADP deposition corrected with Prism precipitation OR NADP deposition model by Grimm and Lynch, (2005)	Reference year of water quality data (1982 to 2012)	[Cl] from water quality used. However, wet CL_{dep} was calculated.
BC_w	eq/ha-yr or meq/m^2-yr	BC$_;$ Ca+Mg+K+Na	Henriksen A. 1984	F-Factor	
Bc_u	meq/m^2-yr	Bc_u = Ca+Mg+K	n/a	n/a	Was not calculated.
N_i	meq/m^2-yr	Acceptable nitrogen	McNulty et. al. (2007) or Nanus et al., (2012)	n/a	Set to 4.3 meq/m^2-yr for all states except AZ,

		immobilized in soil			CA, CO, OR, MT, NM, NV, UT, WA, and WY. For western states a combined values of Ni+Nde = 11.0 eq/ha-yr was used based on Nanus et al. (2012).
N_{de}	meq/m^2-yr	Denitrification rate in catchment	Ashby et al. (1998) or Nanus et al., (2012)	n/a	Set to 7.14 meq/m^2-yr for all states except AZ, CA, CO, OR, MT, NM, NV, UT, WA, and WY. For western states a combined values of Ni+Nde = 11.0 eq/ha-yr was used based on Nanus et al. (2012).
N_{leach}	meq/m^2-yr	N leaching from the watershed as $[NO_3]*Q_s$	Henriksenet.al., (2002)	Reference year of water quality data (1982 to 2012)	This can also be used to calculate N+S exceedance
Q_S	m/yr	Annual runoff flux	Gebert, et al., (1987).	1950-1980 Normal	n/a

Table A2-3. Water Quality Data Sources for Surface Water Critical Loads from EPA, 2012 and Scheffe et al., 2014.

Program Name	**Collecting Org**	**Web Link**	**Reference**
EPA Long term Monitoring (LTM)- Adirondacks - Annual average from 1992-2007	ALSC	http://www.epa.gov/airmarkt/assessments/TIMELTM.html	Stoddard.et.al.WR R.1996
EPA Long term Monitoring (LTM)- Maine - Annual average 1992-2007	UNH	http://www.epa.gov/airmarkt/assessments/TIMELTM.html	Stoddard.et.al.WR R.1996

EPA Long term Monitoring (LTM)-Vermont - Annual average 1992-2007	VT	http://www.epa.gov/airmarkt/assessments/TIMELTM.html	Stoddard.et.al.WRR.1996
EPA Long term Monitoring (LTM)-Catskills - Annual average 1992-2007	USGS	http://www.epa.gov/airmarkt/assessments/TIMELTM.html	Stoddard.et.al.WRR.1996
EPA Long term Monitoring (LTM)-Pennsylvania - Annual average 1992-2007	PSU	http://www.epa.gov/airmarkt/assessments/TIMELTM.html	Stoddard.et.al.WRR.1996
EPA Long term Monitoring (LTM)-VTSSS- Annual average 1992-2007	UVA	http://www.epa.gov/airmarkt/assessments/TIMELTM.html	Stoddard.et.al.WRR.1996
EPA Long term Monitoring (LTM)-Upper Midwest-	EPA	http://www.epa.gov/airmarkt/assessments/TIMELTM.html	Stoddard.et.al.WRR.1996
EPA Long term Monitoring (LTM)-Colorado	EPA	http://www.epa.gov/airmarkt/assessments/TIMELTM.html	Stoddard.et.al.WRR.1996
Eastern Lakes Survey dataset (ELS) 1984	EPA	http://www.epa.gov/emap2/html/data/surfwatr/data/els.html	EPA/600/4-88/032
EPA-EMAP Northeast Lake Survey 1991-94	EPA	http://www.epa.gov/emap2/html/data/surfwatr/data/nelakes.html	EPA/620/R-93/009
EPA REGIONAL EMAP (RMAP) PROGRAM 1993	EPA	http://www.epa.gov/emap2/remap/html/one/data/index.html	
EPA-EMAP Mid-Appalachian Highland Assessment (MAHA) 1994-1996	EPA	http://www.epa.gov/emap2/html/data/surfwatr/data/mastreams/9396/index.html	EPA/903/R-00/015
EPA-EMAP Mid-Atlantic Integrated Assessment (MAIA) 1997-1998	EPA	http://www.epa.gov/emap2/html/data/surfwatr/data/mastreams/9798/index.html	EPA/R-06/XX
EPA National Stream Survey (NSS) 1986	EPA	http://www.epa.gov/emap2/html/data/surfwatr/data/nss.html	EPA-600-388-021a
Virginia Trout Stream Sensitivity Study (VTSSS) Surveys 1987 & 2000	UVA	http://swas.evsc.virginia.edu/	
EPA National Wadeable Stream Survey (WSS) 2007	EPA	http://www.epa.gov/owow/streamsurvey/web_data.html	EPA 841-B-06-002
EPA Western Lake Survey (WLS) 1985	EPA	http://www.epa.gov/emap2/html/data/surfwatr/data/wls.html	Stoddard.et.al.WRR.1996
EPA-EMAP Western Stream & River Survey 2000-2004	EPA	http://www.epa.gov/esd/land-sci/water/streams.htm	EPA 620-R-05-005
EPA National Lake Survey 2010	EPA	http://www.epa.gov/lakessurvey	EPA 841-F-09-007
NPS Vital Signs water Quality Data	NPS	http://www.nature.nps.gov/water/infoanddata/index.cfm	

USFS Forest Service Water Quality Data	USFS	http://views.cira.colostate.edu/web/SiteBrowser/fswq.aspx	
USGS Water-Quality Data for the Nation	USGS	http://waterdata.usgs.gov/nwis/qw	

Table A2-3. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations (for Miller, 2011)

Variable	Criterion (Unit)	Description of the Variable	Data Source	Data	Assumptions and value
ChemCriterion/ ChemThreshold	ANC (μeq/L)	Critical value for chemical criterion	n/a	n/a	Target pH = 6.6, Implied 50 μeq/L at DOC= 0 mg/L
BC_{dep}	meq/m^2-yr	BC; Ca+Mg+K+Na	High Resolution Deposition Model (HRDM).	Average of 1999---2003	Total deposition
CL_{dep}	meq/m^2-yr	Wet deposition of chloride	High Resolution Deposition Model (HRDM).	Average of 1999---2003	Wet deposition only.
BC_w	eq/ha-yr or meq/m^2-yr	BC; Ca+Mg+K+Na	Henriksen A. 1984	F-Factor	
Bc_u	meq/m^2-yr	Bc; Ca+Mg+K	n/a	n/a	Was not calculated.
N_i	meq/m^2-yr	Acceptable nitrogen immobilized in soil	McNulty et. al., (2007)	n/a	Set to 4.3 meq/m^2-yr
N_{de}	meq/m^2-yr	Denitrification rate in catchment	Ashby et al., (1998)	n/a	Set to 7.14 meq/m^2-yr
N_{leach}	meq/m^2-yr	N leaching from the watershed as $[NO_3]*Q_R$	Henriksen et.al., (2002)	Reference year of water quality data	This can also be used to calculate N+S exceedance
Q_S	m/yr	Annual runoff flux	HRDM Climate module using 30-year (1971-2000) Normal precipitation and temperature	1971-2000	

			data from the National Climatic Data Center and Environment Canada		

Table A2-4. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations (for DuPont et al., 2005)

Variable	Criterion (Unit)	Description of the Variable	Data Source	Data	Assumptions and value
ChemCriterion/ ChemThreshold	ANC (μeq/L)	Critical value for chemical criterion	n/a	n/a	50 μeq/L
BC_{dep}	meq/m^2-yr	BC; Ca+Mg+K+Na	n/a	n/a	[BC_t] from water quality used
CL_{dep}	meq/m^2-yr	Wet deposition of chloride	n/a	n/a	[Cl] from water quality used
BC_w	eq/ha-yr or meq/m^2-yr	BC; Ca+Mg+K+Na	Henriksen A. 1984	F-Factor	
Bc_u	meq/m^2-yr	Bc; Ca+Mg+K	n/a	n/a	Was not calculated.
N_i	meq/m^2-yr	Acceptable nitrogen immobilized in soil	McNulty et. al., (2007)	n/a	Set to 4.3 meq/m^2-yr
N_{de}	meq/m^2-yr	Denitrification rate in catchment	Ashby et al., (1998)	n/a	Set to 7.14 meq/m^2-yr
Nl_{each}	meq/m^2-yr	N leaching from the watershed as [NO_3]*Q_R	Henriksen et.al., (2002)	Reference year of water quality data	This can also be used to calculate N+S exceedance
Q_S	m/yr	Annual runoff flux	Randall, (1996)	1951-1980 normal	

Table A2-5. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations (for VDEC2003, 2004, 2012)

Variable	Criterion (Units)	Description of the Variable	Data Source	Data	Assumptions and value
ChemCriterion/ ChemThreshold	ANC μeq/L	Critical value for chemical criterion	n/a	n/a	Target pH = 6.6, Implied 50 μeq/L
BC_{dep}	meq/m^2-yr	BC; Ca+Mg+K+Na	n/a	n/a	[BC_t] from water quality used
CL_{dep}	meq/m^2-yr	Wet deposition of chloride	n/a	n/a	[Cl] from water quality used

BC_w	eq/ha-yr or meq/m^2-yr	$BC_{;}$ Ca+Mg+K+Na	Henriksen A. 1984	F-Factor	
Bc_u	meq/m^2-yr	$Bc_{;}$ Ca+Mg+K	n/a	n/a	Was not calculated.
N_i	meq/m^2-yr	Acceptable nitrogen immobilized in soil	McNulty et. al., (2007)	n/a	Set to 4.3 meq/m^2-yr
N_{de}	meq/m^2-yr	Denitrification rate in catchment	Ashby et al., (1998)	n/a	Set to 7.14 meq/m^2-yr
N_{leach}	meq/m^2-yr	N leaching from the watershed as $[NO_3]*Q_R$	Henriksen et.al., (2002)	Reference year of water quality data	This can also be used to calculate N+S exceedance
Q_R	m/yr	Annual runoff flux	Krug et al., (1990)	1951-1980 normal	

MAGIC Model and Regional Linear Regression Models for Estimating BC_w Input to SSWC (Sullivan et al., 2012 and McDonnell et al., 2012a)

For this study, CLs were derived using a modified form of the SSWC model (see **Eq. A2-2**)[1]. Additionally, base cation weathering was derived using a new method based on MAGIC model estimates of BC_w and regional linear regression models (see Sullivan et al., 2012a and McDonnell et al., 2012), rather than the F-factor method for estimating BC_w.

The MAGIC model was used to calculate watershed-specific BC_w for input to regional linear regression models that estimated BC_w in watersheds without MAGIC values. The BCw estimates were then used as input to the SSWC model. MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on surface water chemistry (Cosby et al., 1985). The model simulates soil solution chemistry, weathering rates, and surface water chemistry to predict the monthly and annual average concentrations of the major ions in these waters (see Cosby et al., 1985 for more details about the model itself). The base cation weathering terms in MAGIC represent the catchment-average weathering rates for the soil compartments. In a one soil-layer application of MAGIC (such as here) the weathering rates in MAGIC thus reflect the catchment-average net supply of base cations to the surface waters draining the catchment. The sum of the MAGIC weathering rates for the individual base cations is therefore identical in concept to the base cation weathering term, BC_w, in the SSWC CL model (**Eq. A2-1**). Base cation weathering rates in MAGIC are calibrated parameters. The calibration procedure uses observed deposition of base cations, observed (or estimated) base cation uptake in soils, observed stream water base cation concentrations, and runoff (Q_S). These observed input and output data provide upper and lower limits for internal sources of base cations in the catchment soils. The two most important internal sources of base cations in catchment soils are modeled explicitly by MAGIC: primarily mineral weathering and soil cation exchange. During the calibration process, observed soil base saturation for each base cation and observed soil chemical characteristics are combined with the observed input and output data to partition the inferred net internal sources of base cations between weathering and base cation exchange.

Sullivan et al., (2012a) and McDonnell et al., (2012) used the watershed-specific BC_w to develop a regional regression model for calculating watershed specific BC_w for 500+ monitoring locations in the Appalachian Mountains of Virginia and West Virginia. Water chemistry and landscape variables were used as the predictor variables in regression analyses to extrapolate BC_w. Each of the calibrated MAGIC study watersheds was placed in an Ecoregion category based on which Ecoregion contained the maximum watershed area and three separate regression models were developed for each Ecoregion (**Table A2-6**). Watershed averages were used to represent the spatial variability within each watershed for the landscape characteristics, except for watershed area. Regression models were established using stepwise linear regression using ‘best subsets’ to evaluate candidate models and constrain the number of independent predictor variables during model selection. Water quality predictor data was collected during several regional surveys, as compiled by Sullivan and Cosby, (2004). One water quality sample, generally collected during the spring between 1985 and 2001, was used to characterize each watershed. Water quality data were derived from several regional surveys, including the National Stream Survey (NSS), Environmental Monitoring and Assessment Program (EMAP), Virginia Trout Stream Sensitivity Study (VTSSS), and stream surveys conducted in Monongahela National Forest. Equations used to estimate BC_w for input to the SSWC model are listed in **Table A2-6** and a summary of model parameters are included in **Table A2-7**.

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[1] Variable names were used consistently throughout the document and often different from that of the study publication.

Table A2-6. Multiple Regression Equations to Estimate BC_w from Either Water Chemistry and Landscape Variables or from Landscape Variables Alone, Stratified by Ecoregion. (McDonnell et al. 2012).

Ecoregion	**n**	**Equation**[1]	**r^2**
Central Appalachian	24	BC_w = -37.5 + 0.6 (SBC) + 0.9 (NO3) + 0.006 (WS Area)	0.93
Ridge and Valley	42	BC_w = 107.0 + 0.5 (SBC) - 0.06 (Elevation) - 3.2 (Slope)	0.86
Blue Ridge	26	BC_w = 27.1 + 0.6 (CALK) + 0.6 (NO3)	0.90

[1] SBC is the sum of base cations; CALK is calculated ANC.

Table A2-7. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations (for Sullivan et al. 2012a and McDonnell et al. 2012)

Variable	**Criterion (Units)**	**Description of the Variable**	**Data Source**	**Data**	**Assumptions and value**
ChemCriterion/ ChemThreshold	ANC µeq/L	Critical value for chemical criterion	n/a	n/a	50 µeq/L
BC_{dep}	meq/m^2-yr	BC; Ca+Mg+K+Na	NADP (wet) and Baker (1991) (dry) using dry:wet ratio	5-year average centered at 2002	
CL_{dep}	meq/m^2-yr	Wet deposition of chloride	NADP (wet) and Baker 1991 (dry) using dry:wet ratio	5-year average centered at 2002	
BC_w	eq/ha-yr or meq/m^2-yr	BC; Ca+Mg+K+Na	McDonnell et al. (2012)	Regionalization of BC_w	
Bc_u	meq/m^2-yr	Bc; Ca+Mg+K	McNulty et. al., (2007)	n/a	
N_i	meq/m^2-yr	Acceptable nitrogen immobilized in soil	McNulty et. al., (2007)	n/a	Set to 4.3 meq/m^2-yr
N_{de}	meq/m^2-yr	Denitrification rate in catchment	Ashby et al., (1998)	n/a	Set to 7.14 meq/m^2-yr
N_{leach}	meq/m^2-yr	N leaching from the watershed as $[NO_3]*Q_S$	n/a	n/a	
Q_S	m/yr	Annual runoff flux	HRDM Climate	n/a	

			module using 30-year (1971-2000) Normal precipitation and temperature data from the National Climatic Data Center and Environment Canada		

MAGIC model and Hurdle Modeling for Estimating BC_w Input to SSWC (McDonnell et al., 2014 and Povak et al., 2014)

For this study, CLS were derived using a modified form of the SSWC model that excluded the N terms. Building on the framework of Sullivan et al., (2012) and McDonnell et al., (2012), McDonnell et al. (2014) and Povak et al. (2014) expanded the study area and developed new statistical models to better predict BC_w and evaluate CLs of S. Their study expanded the area to include the full Southern Appalachian Mountain (SAM) region and surrounding terrain from northern Georgia to southern Pennsylvania, and from eastern Kentucky and Tennessee to central Virginia and western North Carolina. As with Sullivan et al., (2012a) and McDonnell et al., (2012), the MAGIC model was used to calculate watershed-specific BC_w for 140 stream locations containing both measured soil chemistry and water chemistry data (see section above on page A-14 for a description of MAGIC). In addition, McDonnell et al., (2014) aggregated all known water quality data that totaled 933 sample locations in order to develop a statistical model to predict ANC and BC_w for all streams in the SAM region. Water chemistry data were collected between 1986 and 2009, with stream ANC calculated as the equivalent sum of the base cation concentrations (Ca^{2+}, Mg^{2+}, K^+, Na^+, ammonium [NH_4^+]) minus the sum of the mineral acid anion concentrations (chloride [Cl_-], NO_3^-, SO_4^{2-}). BC_w was estimated as the net internal source of base cations between weathering and base cation exchange for the watershed based on the MAGIC model calibrations, which used observed stream, soil, and atmospheric deposition data to match current observed stream and soil chemistry conditions. With the use of a random forest regression modeling technique, a continuous BC_w layer was regionalized using a suite of initial candidate predictor variables chosen to represent potential broad- to fine-scale climatic, lithologic, topoedaphic, vegetative, and S deposition variables that have the potential to influence ANC and BC_w. To represent the landscape conditions that influence specific locations along a stream, all candidate landscape predictor variables were expressed on a 30 m grid basis across the SAM's domain and were upsloped averaged based on the technique described in McDonnell et al., (2012). This resolution allowed for the creation of "flowpaths" for the development of a topographically determined stream network. This approach allowed for a total of 140,504 watersheds which were represented (i.e. delineated) with the use of a hydrologically conditioned DEM. CLs were calculated for all grid cells that had a predicted "low" ANC value (<300 µeq/L). If the grid value was predicted to be greater than 300 µeq/L, then the grid cell was considered well-buffered and assigned an arbitrarily large ANC and CL (e.g. 8888) value. CLs were then calculated with SSWC (Henriksen and Posch, 2001) with estimates of BCdep, BC_w , Bc_u, Q_s and an ANC chemical criteria set to an value of 50 µeq/L for each stream node.. See McDonnell et al., (2014) and Povak et al., (2014) for additional methods detail. A summary of model parameters is included in **Table A2-8**.

CLs exceedances can be calculated as total S deposition minus the CL. In McDonnell et al., (2014), estimates of S deposition were calculated for two time periods as three-year averages centered on 2001 and 2011. These deposition rates were used to evaluate changes in CL exceedance over the period 2001 to 2011.

Table A2-8. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations for McDonnell et al., 2014.

Variable	**Criterion (Units)**	**Description of the Variable**	**Data Source**	**Data**	**Assumptions and value**
ChemCriterion/ ChemThreshold	ANC µeq/L	Critical value for chemical criterion	n/a	n/a	50 µeq/L
BC_{dep}	meq/m^2-yr	BC; Ca+Mg+K+Na	NADP-Grimm (wet) Grimm and Lynch, (2005). Baker, (1991) (dry), using dry:wet ratio.	3-year average centered at 2002.	Wet and dry deposition.
CL_{dep}	meq/m^2-yr	Chloride deposition	NADP-Grimm (wet) Grimm and Lynch, (2005). Baker, (1991) (dry), using dry:wet ratio.	3-year average centered at 2002.	
BC_w	eq/ha-yr or meq/m^2-yr	BC; Ca+Mg+K+Na	Povak et al. (2014).	Regionalization of BCw based on Random Forest (RF) modeling techniques.	
Bc_u	meq/m^2-yr	Bc; Ca+Mg+K	McNulty et al., (2007).		
N_i	meq/m^2-yr	Acceptable nitrogen immobilized in soil	n/a	n/a	
N_{de}	meq/m^2-yr	Denitrification rate in catchment	n/a	n/a	
N_{ni}	meq/m^2-yr	Nitrification rate in the catchment	n/a	n/a	
N_{leach}	meq/m^2-yr	N leaching from the watershed as $[NO_3]*Q_R$	n/a	n/a	

Q_S	m/yr	Annual runoff flux	McCabe & Wolock, (2011)	n/a	
Watershed/stream reaches	km^2	Watersheds	Watersheds were delineated based on hydrologically conditioned DEM derivatives from the National Hydrography Dataset (NHDPlus; USEPA and U. S. Geological Survey [USGS], 2005). This process delineated a total of 140,504 watersheds within the study region, with an average size of approximately 1 km^2.		

MAGIC model and Target Loads (Lawrence et. al., 2015)

For this study, Target Loads (TLs) were derived using the MAGIC model for ANC endpoints of 20, 50, and 100 µeq/L for years of 2050, 2100, and 3000. MAGIC scenarios and TL calculations to estimate acidification caused by S deposition within a management timeline generally assume constant N retention, as a percent of input, from the Reference year into the future. A target load calculated for the year 3000 is considered long-term steady state CL, roughly analogous to the steady-state CL calculated by the Simple Mass Balance (SMB) and Steady-State Water Chemistry (SSWC) models (Henriksen and Posch 2001). The estimate of the TL of S+N were generated by determining the TL of S without any future N deposition and the TL of N without any future S deposition. However, TLs for N were not included in the database at this time.

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. 1985). It simulates monthly and annual average concentrations of major ions in drainage 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. Central to the MAGIC calculations is the size of the pool of exchangeable base cations on 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. 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. The only sources of S to the soils are assumed to be atmospheric deposition and, in some cases, underlying geology. Aluminum dissolution and precipitation are assumed to be controlled by equilibrium with a solid phase of Al(OH)3. Aluminum speciation is calculated by considering hydrolysis reactions as well as complexation with SO_4^{2-} and F^-. Effects of carbon dioxide (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. The rate constants are typically not varied during the simulation period. Weathering rates for base cations are assumed to be constant.

The MAGIC model calibrates soil N retention such that estimates of N deposition correspond with vegetative N uptake and observed stream water NO_3^- concentrations. For watersheds with significantly higher depositional N inputs relative to output from vegetative uptake and stream flow, N retention is often more than 90%, indicating that the vast majority of incoming N is immobilized by the soil environment. Under the N saturation paradigm, all forest soils will eventually become N saturated in response to continued human-caused N inputs and will eventually begin to leach more NO_3^- to streams.

Section 3.6.1.2 of Lawrence et. al., (2015) describes in detail the input data used in MAGIC. Calibration of the MAGIC model was achieved by setting the values of certain parameters within the model that can be directly measured or observed in the system of interest (called fixed parameters). The model is then run (using observed and/or assumed atmospheric and hydrologic inputs) and the outputs (stream water and soil chemical variables, called criterion variables) are compared to observed values of these variables. If the observed and simulated values differ, the values of another set of parameters in the model (called optimized parameters) are adjusted to improve the fit. After a number of iterations 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. See Table A2-8 for additional description of model parameters.

Table A2-8. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations for Lawrence et. al., 2015.

Variable	Criterion (Units)	Description of the Variable	Data Source	Data	Assumptions and value
ChemCriterion/ ChemThreshold	ANC µeq/L	Critical value for chemical criterion	n/a	n/a	20, 50, 100 µeq/L
BC_{dep}	meq/m^2-yr	BC; Ca+Mg+K+Na	NADP, CASTNET	2 km, interpolated wet + est. dry/wet	Wet and dry deposition.
CL_{dep}	meq/m^2-yr	Chloride deposition	NADP, CASTNET	2 km, interpolated wet + est. dry/wet	
BC_w	meq/m^2-yr	BC; Ca+Mg+K+Na	Cosby et al. 1985	MAGIC	
Q_S	m/yr	Mean annual	McCabe & Wolock (2011)	n/a	4 km, 1900-2008 period
Soils	units, mm, %, %, µm/s	Mean pH, depth, clay content, organic matter, saturated hydraulic conductivity	SSURGO	n/a	1:12K -1:63K
Watershed drainage area	m^2	Total area	National Elevation Dataset	10 m DEM	

MAGIC model and Target Loads (Sullivan.et.al., 2005)

The MAGIC model was used to calculate the TLs for the Loch Vale watershed, Colorado. MAGIC was calibrated to The Loch and to its alpine tributary, Andrews Creek. Wet deposition was estimated from the NADP/NTN wet deposition monitoring site located at Loch Vale. Dry deposition of S to the forest in the watershed was estimated to be near zero. Biological uptake of NO_3^- and NH_4^+ were estimated using the following assumptions: (1) uptake of both NO_3^- and NH_4^+ in the soil compartment was set to 100%; (2) uptake of both NO_3^- and NH_4^+ in the talus compartment was set to 0%; (3) NH_4^+ deposited to the talus was nitrified and leached to the lake as NO3; and (4) uptake of NO_3^- and NH_4^+ in the lake was adjusted such that the estimated catchment output flux of each ion matched observed output fluxes (i.e., lake uptakes were calibrated to observed data). Target year was 2046. See above for description of the MAGIC model. Also, see Sullivan et al. 2005 for a complete description of the method, model inputs, and values. See Table A2-9 for additional description of model parameters.

Table A2-9. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations for Sullivan.et.al., 2005.

Variable	**Criterion (Units)**	**Description of the Variable**	**Data Source**	**Data**	**Assumptions and value**
ChemCriterion/ ChemThreshold	ANC µeq/L	Critical value for chemical criterion	n/a	n/a	0, 20, 50 µeq/L
S_{dep}	Kg/ha/yr	S	NADP wet / Dry	NTN collected at Loch Vale, CO	Wet deposition/ dry = equal to 10% of measured wet deposition
N_{dep}	Kg/ha/yr	N	NADP wet / Dry by Baron and Campbell (1997)	NTN collected at Loch Vale, CO	Wet deposition/ dry = equal to 50% of measured wet deposition
BC_{dep}	Kg/ha/yr	BC; Ca+Mg+K+Na	NADP/Dry	NTN collected at Loch Vale, CO	Wet deposition / dry = equal to 50% of measured wet deposition
CL_{dep}	Kg/ha/yr	Chloride deposition	NADP	NTN collected at Loch Vale, CO	
Deposition scenarios	Kg/ha/yr	S, N, BC	n/a	n/a	Constant deposition at 1996 levels 2.2 kg S ha/yr, 4.2 kg N ha/yr
BC_w	meq/m^2-yr	BC; Ca+Mg+K+Na	Cosby et al. 1985	MAGIC	Based on 10 calibrations
Q_S	m/yr	Mean annual	n/a	n/a	4 km, 1900-2008 period

Soils		Exchangeable Ca, Mg, Na, K %, Base saturation, %	Soils data derived from Baron (1992)	n/a	

MAGIC model and Extrapolation of Target Loads to the ALSC lake dataset (Sullivan.et.al., 2012b)

For this study, Target Loads (TL) were calculated using the MAGIC model at 115 Adirondack lakes and their watersheds. See **Lawrence et. al., 2015** above for description of the MAGIC model. These 115 TLs of S deposition were then extrapolated to a larger lake dataset of 1469 ALSC lakes. Of those these lakes, 1136 were larger than 1 ha in area and located within the Adirondack ecoregion. Target loads were calculated for ANC endpoints of 0, 20, 50 µeq/L and for target years of 2050, and 2100. The extrapolation to a larger ALSC lake dataset was based on relating ANC, as a predictor variable, to the TL determined by the MAGIC model. This regression was then used along with measured water quality ANC values to estimate the TL for a given lake. The regression r^2 for this relationship was ranged from 0.72 (to protect ANC to 0 µeq/L in the year 2050) to 0.92 (to protect ANC to 50 µeq/L) (see Table A2-10). See Table A2-11 for additional description of model parameters.

Table A2-10. Regression Equations to Estimate the Target Load (TL) of Sulfur Deposition to Protect Lake Water ANC From Acidifying Below Designated Threshold Criteria in Designated Future Years. Regressions are based on charge balance ANC determined by the ALS during the 1980s. From Sullivan.et.al., (2012b).

Critical Threshold ANC Value (µeq/L)	Endpoint Year	Equation to Predict Target Load (meq S /m^{2-}yr)	r2
0	2050	TL = 67.9 + 0.790 ANC	0.72
	2100	TL = 61.7 + 0.719 ANC	0.80
20	2050	TL = 43.4 + 0.848 ANC	0.81
	2100	TL = 41.1 + 0.760 ANC	0.86
50	2050	TL = 11.8 + 0.852 ANC	0.90
	2011	TL = 13.7 + 0.765 ANC	0.92

Table A2-11. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations for Sullivan.et.al., 2012b.

Variable	**Criterion (Units)**	**Description of the Variable**	**Data Source**	**Data**	**Assumptions and value**
ChemCriterion/ ChemThreshold	ANC µeq/L	Critical value for chemical criterion	n/a	n/a	0, 20, 50 µeq/L
MAGIC simulations		Target Year	n/a	n/a	2050 and 2100
S_{dep}	Kg/ha/yr	S	NADP wet / CMAQ Dry	Grimm and Lynch (1997)/CMAQ	For each study watershed, area-

					weighted total wet plus dry S and N deposition values were calculated using the interpolated NADP wet deposition and the CMAQ dry to wet ratios.
N_{dep}	Kg/ha/yr	N	NADP wet / CMAQ Dry	Grimm and Lynch (1997)/CMAQ	For each study watershed, area-weighted total wet plus dry S and N deposition values were calculated using the interpolated NADP wet deposition and the CMAQ dry to wet ratios.
BC_{dep}	Kg/ha/yr	BC; Ca+Mg+K+Na	NADP/ CMAQ Dry	Grimm and Lynch (1997)	For each study watershed, area-weighted total wet plus dry S and N deposition values were calculated using the interpolated NADP wet deposition and the CMAQ dry to wet ratios.

CL_{dep}	Kg/ha/yr	Chloride deposition	NADP wet/ CMAQ Dry	Grimm and Lynch (1997)/ CMAQ	For each study watershed, area-weighted total wet plus dry S and N deposition values were calculated using the interpolated NADP wet deposition and the CMAQ dry to wet ratios.
BC_w	meq/m^2-yr	BC; Ca+Mg+K+Na	Cosby et al. 1985	MAGIC	
Deposition scenarios	Kg/ha/yr	S, N, BC	n/a	n/a	For both sets of simulations, deposition of the strong acid anion not being considered for determination of the TL (i.e., N load for determination of target S load) was set to follow future trajectories anticipated by the U.S. EPA in the Clean Air Interstate Rule (CAIR).
Q_S	m/yr	Mean annual	Not indicated	n/a	4 km, 1900-2008 period
Soils		Exchangeable Ca, Mg, Na, K %, Base saturation, %	Sullivan et al. (2006)	n/a	

PnET-BGC Model (Fakhraei.et.al., 2014)

For this study, Target Loads (TL) were calculated using the PnET-BGC model for 128 lakes impaired due to elevated acidity under Section 303(d) of the Clean Water Act, in 2010 within the Adirondack Forest Preserve of New York. Water quality data was from New York State Department of Environmental Conservation (NYSDEC) and the Adirondack Long-Term Monitoring (ALTM) program (Driscoll et al., 2007, 2003). PnET-BGC is a biogeochemical model that uses meteorological, atmospheric deposition and historical land disturbance data to simulate hydrology and major ion chemistry in vegetation, soil and water (see Gbondo-Tugbawa et al., 2001 for more details about the model). PnET-BGC was span-up for a 1000 years to achieve steady state conditions before anthropogenic disturbances. The model is run on a monthly time step using monthly values of atmospheric deposition and meteorological data. Historical emission estimates were used for values of wet deposition of major solutes, using linear regression models to relate national emissions and measured air concentrations of wet deposition at an NADP site (NY20). Dry deposition of chemical constituents was based on user inputs of dry to wet deposition ratios. National Land Cover Data were used to define the forest composition for each study watershed. PRISM model (http://www.prism.oregonstate.edu/) beginning in the year 1895 was used for the national climatic data for the model. Following calibration and confirmation, TLs were calculated for each lake for 2200. For more details about the modeling see Fakhraei.et.al., 2014 and Table A2-12.

Table A2-12. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations for Fakhraei.et.al., 2014.

Variable	**Criterion (Units)**	**Description of the Variable**	**Data Source**	**Data**	**Assumptions and value**
ChemCriterion/ ChemThreshold	ANC μeq/L	Critical value for chemical criterion	n/a	n/a	20, μeq/L
MAGIC simulations		Target Year	n/a	n/a	2200
S_{dep}	Kg/ha/yr	S	NADP wet / CASTNET Dry	From NY20	Dry to wet deposition ratios. Spatial patterns in dry to wet deposition for the S were calculated based on spatial models developed by Ollinger et al. (1993) and then modified by Reynolds and Driscoll (2004) to incorporate effects of forest

					composition (Cronan, 1985).
N_{dep}	Kg/ha/yr	N	NADP wet / CASTNET Dry	From NY20	Dry to wet deposition ratios. Spatial patterns in dry to wet deposition for the N were calculated based on spatial models developed by Ollinger et al. (1993) and then modified by Chen and Driscoll (2004) to incorporate effects of forest composition (Cronan, 1985).
BC_{dep}	Kg/ha/yr	BC; Ca+Mg+K+Na	NADP wet / CASTNET Dry	From NY20	Dry to wet deposition ratios. Spatial patterns in dry to wet deposition for the BCwere calculated based on spatial models developed by Ollinger et al. (1993) and then modified by Chen and Driscoll (2004) to incorporate effects of forest composition (Cronan, 1985).

CL_{dep}	Kg/ha/yr	Chloride deposition	NADP wet / CASTNET Dry	From NY20	Dry to wet deposition ratios. Spatial patterns in dry to wet deposition for the BCwere calculated based on spatial models developed by Ollinger et al. (1993) and then modified by Chen and Driscoll (2004) to incorporate effects of forest composition (Cronan, 1985).
BC_w		BC; Ca+Mg+K+Na	Gbondo-Tugbawa et al., 2001	PnET-BGC	
Climatic data		Maximum and minimum monthly temperature and precipitation	PRISM model	n/a	
Q_S		Modeled in PnET-BGC	n/a	n/a	n/a
Soils		Exchangeable Ca, Mg, Na, K %, Base saturation, %	Not indicated	n/a	n/a

Regional Target Loads for Streams in Adirondack Mountains (McDonnell.et.al., 2021)

For this study, Target Loads (TL) were calculated using the Pnet-BGC model at 25 Adirondack streams and their watersheds (Shao et al., 2020), then spatially extrapolated to 401 streams included in the Western Adirondack Stream Survey (WASS) and Eastern Adirondack Stream Survey (ECASS), collectively "WECASS". Regionalization of the 25 modeled streams/watersheds was based on characteristics of the stream watersheds sampled in the WECASS. Given the importance of discharge on regional stream chemistry, model parameters calibrated to specific stream chemistry data were corrected for streamflow conditions at the time of sample collection (see section 2.4 McDonnell et al. 2021). TLs were based on a fixed criteria of ANC = 20 µeq/L and a second approach using a site-specific ANC value that varied by stream/watershed. Site-specific ANC values were estimated by hindcast simulations of the 25 modeled watersheds for ANC in 1850 and extrapolated to the 401 streams. Site-specific ANC was defined as preindustrial ANC minus 20 µeq L^{-1}. The ANClimit for these TLs are noted as SS – "Site-Specific" since the ANC values are not included in the database.

Table A2-13. Regression Equations to Estimate the Target Load (TL). The three predictors most selected across all models were current ANC, latitude, and longitude. See section 3.3 and Supplemental Material 9 of McDonnell et al., 2021. See Supplemental Material 6 for description of parameters within the regression model.

Critical Threshold ANC Value (µeq/L)	Endpoint Year	Equation to Predict Target Load (meq S+N /m^{2-}yr)	r2
20	2050	57.69 + 0.67*Ambient ANC – 0.95*Ambient N+S Deposition	0.69
20	2150	TLANC=20, 2050 = 57.69 + 0.67*Ambient ANC – 0.95*Ambient N+S Deposition	0.88
SS	2050	−962.29 + 0.14∗Ambient ANC +21.95∗Latitude – 0.03∗Elevation +33.30∗Deciduous	0.77
SS	2150	-962.21 + 0.11*Ambient ANC + 23.15*Longitude – 0.05*Elevation	0.59

Table A2-14. Description of Variables in the Database for Surface Water Critical Load of Acidity Calculations for McDonnell.et.al., 2021.

Variable	Criterion (Units)	Description of the Variable	Data Source	Data	Assumptions and value
ChemCriterion/ ChemThreshold	ANC µeq/L	Critical value for chemical criterion	n/a	n/a	20 and Site-specific (SS) µeq/L
Pnet-BGC simulations		Target Year	n/a	n/a	2050 and 2150
S_{dep}	Kg/ha/yr	S	NADP wet / CASTNET Dry	From NY20	Dry to wet deposition ratios. Spatial patterns in dry

					to wet deposition for the BCwere calculated based on spatial models developed by Ollinger et al. (1993) and then modified by Chen and Driscoll (2004) to incorporate effects of forest composition (Cronan, 1985).
N_{dep}	Kg/ha/yr	N	NADP wet / CASTNET Dry	From NY20	Dry to wet deposition ratios. Spatial patterns in dry to wet deposition for the BCwere calculated based on spatial models developed by Ollinger et al. (1993) and then modified by Chen and Driscoll (2004) to incorporate effects of forest composition (Cronan, 1985).
BC_{dep}	Kg/ha/yr	BC; Ca+Mg+K+Na	NADP wet / CASTNET Dry	From NY20	Dry to wet deposition ratios. Spatial patterns in dry to wet deposition for the BCwere calculated based

					on spatial models developed by Ollinger et al. (1993) and then modified by Chen and Driscoll (2004) to incorporate effects of forest composition (Cronan, 1985).
CL_{dep}	Kg/ha/yr	Chloride deposition	NADP wet / CASTNET Dry	From NY20	Dry to wet deposition ratios. Spatial patterns in dry to wet deposition for the BCwere calculated based on spatial models developed by Ollinger et al. (1993) and then modified by Chen and Driscoll (2004) to incorporate effects of forest composition (Cronan, 1985).
BC_w		BC; Ca+Mg+K+Na	Gbondo-Tugbawa et al., 2001	PnET-BGC	
Climatic data		Maximum and minimum monthly temperature and precipitation	PRISM model	n/a	
Q_S		Modeled in PnET-BGC	n/a	n/a	n/a
Soils		Exchangeable Ca, Mg, Na, K	Not indicated	n/a	n/a

		%, Base saturation, %			

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Appendix 3

Empirical Critical Loads of Nitrogen

Empirical (observation-based) or statically determined CLs of N are described in this section. Three different studies are included: (1) Pardo et al., (2011, 2010), (2) Geiser et al., (2019, 2021), and (3) Simkin et al., (2016).

Empirical Critical Loads of Nitrogen as presented by Pardo et al., (2011, 2010)

The empirical CLs of N developed by Pardo et al., (2011, 2010) included minimum and maximum values for Ecoregion I level. Five different biological receptors are included. The NCLD contains the minimum and maximum CL for each receptor (mycorrhizal fungi, lichens, herbaceous species and shrubs, and forest ecosystems) mapped to the Level I Ecoregions (**Table A3-1**). Because a range of responses was reported for each receptor, using the low end of the range provides somewhat conservative CLs when mapped to a gridded system. See Pardo et al., (2011, 2010) for how these CLs were determined and for more details.

Table A3-1. Minimum empirical Critical Loads of Nitrogen reported in Pardo et al., (2011).

Ecoregion Level I	**Range of Critical Loads for Nitrogen (kg/ha-yr)**				
	Mycorrhizal fungi	**Lichens**	**Herbaceous species and shrubs**	**Forest ecosystems**	**Nitrate Leaching**
Northern Forests	5 - 7	4 – 6	7 – 21[2]	3 – 26[4]	8
Northwest Forested Mtns	5 – 10	2.5 – 7.1	4 – 10	4 – 17	4 – 17
Marine West Coast	5[1]	2.7 – 9.2	n/a	5[1]	n/a
Eastern Temperate Forests	5 – 12	4 – 8	17.5[1,3]	3 – 8[5]	8
Great Plains	12[1]	n/a	5 – 25	n/a	10 – 25
North American Deserts	n/a	3	3 – 8.4	n/a	n/a
Mediterranean California	7.8 – 9.2	3.1 – 6	6 – 33	17 – 39	10 – 17
Temperate Sierras	n/a	4 – 7	n/a	n/a	n/a
Tropical Wet Forests	n/a	n/a	n/a	5 – 10[6]	n/a

[1] Single value reported; minimum and maximum set equal to each other.
[2] Published range is >7 to <21.
[3] Published value is <17.5.
[4] Published range is >3 to <26
[05] Published range is >3 to 8
[6] Published range is <5 to 10

Empirical Critical Loads of Nitrogen as presented by Geiser et al., (2010) and Root et al., (2015)

These critical loads have been removed from the NCLDv3.2. Please use CL values determined by Geiser et al. 2019 (see below). This section has been left for reference.

A second set of lichen-based CL estimates were calculated following a method developed by Geiser et al., (2010) that used "air score," by Ecoregion Level I with average annual precipitation using the following equation (**Eq. A3-1**):

$$\text{CL (kg/ha-yr)} = [\text{Air Score} + 0.091756 + (0.002388 \times \text{precip})] / 0.1493339 \qquad \text{(Eq. A3-1)}$$

Geiser et al. (2010) developed air scores based on quantitative measures of lichen community composition (**Table A3-2**). The mean air scores combined with annual precipitation, mapped at a 4x4 km grid resolution, were used to calculate the CLs. This analysis used the 30-year annual average precipitation for the period 1961-1990 (http://prism.oregonstate.edu/products/matrix.phtml?vartype=tmax&view=data) (**Table A3-2**).

Table A3-2. Air Scores for Ecoregions Level I (Geiser et al., 2010)

Ecoregion Level 1	EcoCode 1	Minimum Air Score	Maximum Air Score
Northern Forests	5	0.21	0.21
Marine West Coast Forests	7	0.21	0.21
Northwest Forested Mtns	6	0.21	0.49
Eastern Temperate Forest	8	0.33	0.33
Mediterranean CA	11	0.33	0.49
Temperate Sierras	13	0.49	0.49

Geiser et al., (2010) CLs were modified by Root et al., (2015) for selected regions in the Pacific Northwest. Geiser et al., (2010) CLs were replaced with the Root et.al., (2015) CL minimum and maximum value of 1.54 and 2.51 Kg N/ha-yr for the following five Ecoregion level III: (1) North Cascades, (2) Northern Rockies, (3) Cascades, (4) Eastern Cascades Slopes and Foothills, and (5) Blue Mountains.

Root et al., (2015) based their CLs on two lichen-based indicators of depositional N for interior forested mountain ecosystems of the Pacific Northwest. Lichen community composition and concentration of elemental N in lichen thalli are proven approaches to biomonitoring N deposition patterns in many regions (Geiser et al., 2010, Root et al., 2013). As N deposition increases, N-loving eutrophic lichens become dominant over oligotrophic lichens that thrive in nutrient-poor habitats. Based on the lichen communities shift, CLs of 1.54 and 2.51 kg N/ha-yr of through-fall dissolved inorganic N deposition were determined for lichen communities and lichen N concentration, respectively. Please see Geiser et al., (2010) and Root et al., (2015) for more details.

Empirical Critical Loads of Nitrogen and Sulfur as presented by Geiser et al., (2019, 2021)

Critical Load Introduction and Background

Epiphytic macrolichens are sensitive receptors that inhabit trees, shrubs and other plants in US forests and beyond. Epiphytic lichens are not parasites but rather depend directly on atmospheric sources of water and nutrients for their metabolism and survival. Therefore, the gain or loss of lichen species is directly linked to air quality because different species have different pollution tolerances. Lichens may be assigned to functional groups (**Fig. 1**) based on ecological functions: *oligotrophs* are associated with clean air and low-nutrient conditions; *cyanolichens* are nitrogen-fixing lichens associated with clean air conditions, *forage lichens* are important for wildlife nesting and winter forage and are also associated with clean air, and *matrix lichens* include all others. The relative abundances and changes of each of these groups give insights into the status of forest air-quality.

Figure 1. Epiphytic macrolichens exemplifying (**a**) pendant forage lichens, (**b**) shrubby forage lichens, (**c**) large cyanobacterial lichens (cyanolichens), (**d**) small to medium cyanolichens, (**e**) medium to large matrix lichens, and **(f)** small matrix lichens. Near Philomath, western Oregon. Photo credit: (**f**) Jim Riley.

Critical Load Determination

Definitions: Critical loads for epiphytic lichens were defined as the deposition value at which a 20% decrease of peak diversity of lichen communities occurs. "Diversity" comprises five separate lichen metrics: 1) total lichen species richness, 2) "oligotroph/sensitive" species richness, 3) "forage lichen" abundances, 4) "cyanolichen" abundances, and 5) "community composition" changes. Community composition changes are defined by community airscores – these are the abundance-weighted mean value of species' sensitivities, where sensitivities are defined as the "optimum" deposition value at which each species was most frequently detected among 10,000+ survey plots across the United States. Airscores are the mean of these sensitivities across all species in a given site. The intuition is that most species at a clean-air site will tend to have lower optimal deposition values (on average), while most species at a polluted site will tend to have higher optimal deposition values, due to their inherent tolerances.

Models: The 20% change in lichen diversity was estimated from a fitted regression curve of lichen diversity in response to atmospheric deposition (**Fig. 2**). The model was nonlinear quantile regression estimated for the 90th-percentile. Uncertainty of the 90th-percentile regression estimate was based on bootstrapped confidence intervals. Geiser et al. (2019) describe analysis methods in fine detail.

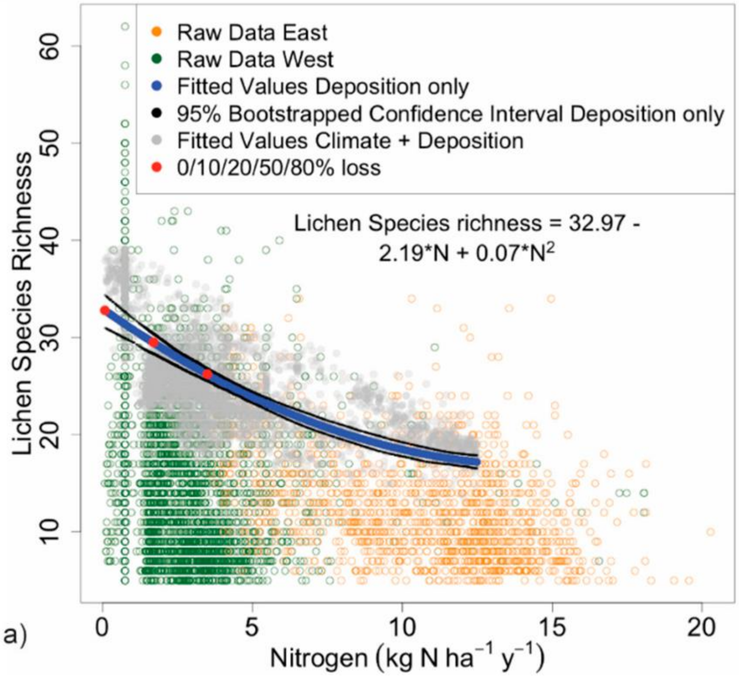

Figure 2. Example of one lichen-based critical loads model. The critical load is the deposition value at which a 20% decrease in the lichen metric occurs, based on a nonlinear quantile regression model for the 90th percentile. In this example, the lichen metric is simply lichen species richness. Bootstrapped confidence intervals are provided.

Masks: Estimates for lichen-based critical loads were restricted to locations presumed to be habitable by epiphytic macrolichens or lichen functional groups. Locations were therefore masked according to the joint occurrence of 1) habitable climates and 2) available forest cover. For the masks, climate data (mean annual temperature, precipitation, moisture deficit and continentality) were sourced from the ClimateNA database (https://adaptwest.databasin.org/pages/adaptwest-climatena), forest cover data were sourced from the National Land Cover Database (NLCD; https://www.mrlc.gov/data), and lichen occurrences were from the joint set of all FIA and ARM lichen surveys combined with data from the Consortium of

North American Lichen Herbaria (CNALH; https://lichenportal.org/cnalh/). All geospatial data were converted to identical projections, resolutions, origins, and extents: Albers equal-area projection at 1-km^2 resolution for all of North America.

The climate mask employed an inclusive climate-envelope algorithm called BIOCLIM, which compares environmental values at any location to the distribution of values at known locations of occurrence – suitable sites have climates close to the 50th percentile, relative to less-suitable climates at the 10th or 90th percentiles. Thresholds (to include or exclude locations from the mask) were set based on maximizing accuracy (Cohen's kappa) and fine-tuned by supervised visual inspection to reduce errors of omission. Separate BIOCLIM models for the East, West and Arctic were merged into one aggregate climate mask. Finally, the joint intersection of the climate mask and the forest cover mask resulted in a final "suitable habitats" mask beyond which lichen-based critical loads estimates should not be made. This process was iterated for 1) all lichens, 2) cyanolichens, and 3) forage lichens separately because of differing habitat requirements. **Fig. 3** depicts one such example.

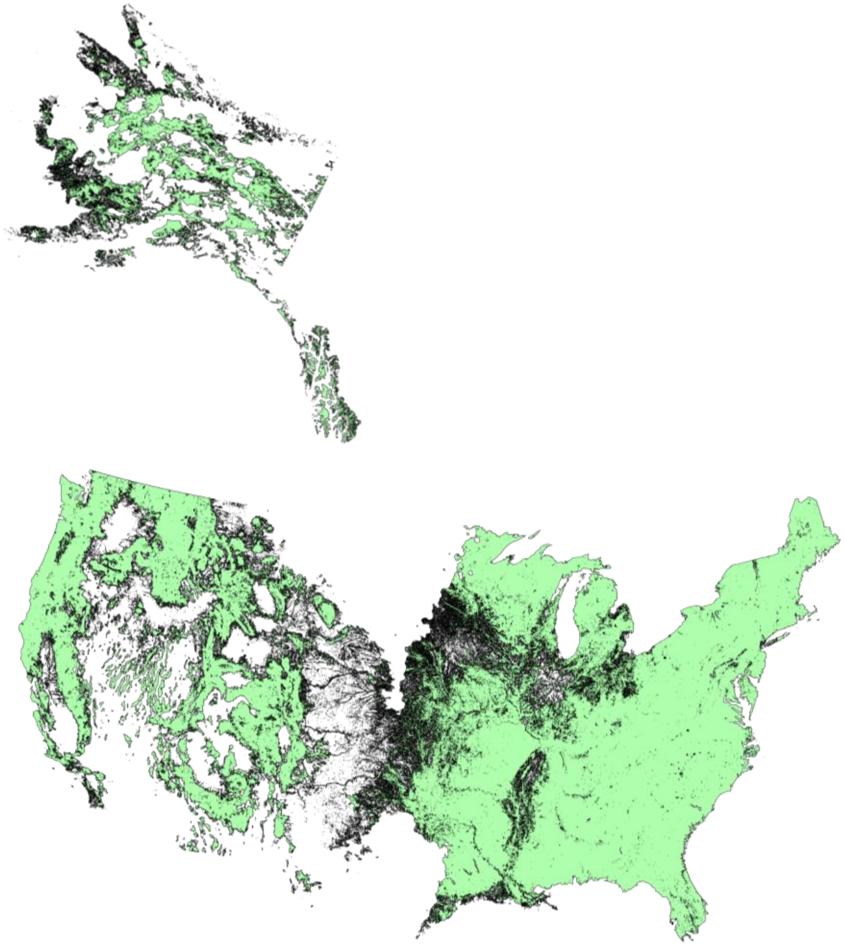

Figure 3. Example of a habitat suitability mask for epiphytic cyanolichens of North America. Green areas are habitable for this group of lichens, based on the intersection of suitable climates and forested lands.

Empirical Critical Loads of Nitrogen as presented by Simkin et al., (2016)

This section describes the background and procedures for determining the herbaceous biodiversity CLs from Simkin et al., (2016). These CLs are for N deposition and describe the level of N deposition above which decreases in herbaceous plant species biodiversity are observed. They were derived from Simkin et al., (2016) which included a nationwide statistical analysis of 15,136 plots (**Figure 3A-1**) assembled from 12 distinct datasets, and represent empirical CLs of N.

The CLs from Simkin et al., (2016) are calculated separately for "Open Canopy" and "Closed Canopy" systems based on Level 1 of the National Vegetation Classification (USNVC, 2016) where the former includes grasslands, shrub lands, and woodlands, and the latter includes forested understories. This was done because light-limited herbaceous systems (Closed Canopy) function differently from systems where light is not limiting (Open Canopy) (Neufeld and DR, 2014). The CLs were derived statistically, using multiple regression models relating the species richness of a plot to up to eight factors: N deposition (Ndep), temperature (T), precipitation (P), soil pH (pH), Ndep*T, Ndep*P, Ndep*pH, and Ndep^2.

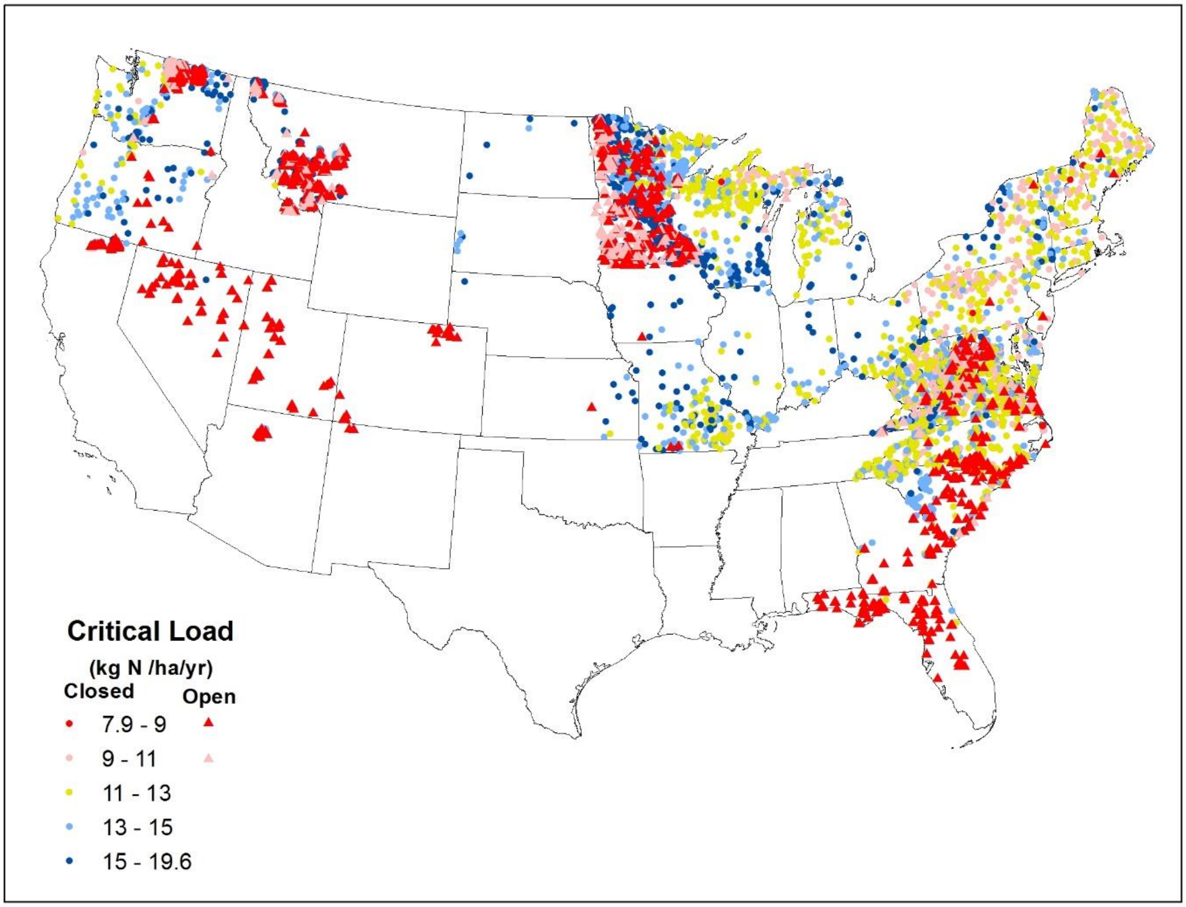

Figure 3A-1: Critical loads for nitrogen deposition based on total graminoid plus forb species richness (colored symbols) from Simkin et al., (2016). The 3,317 open canopy sites (combined grassland, shrub land, and woodland vegetation types) are portrayed with triangles, and the 11,819 closed canopy sites (deciduous, evergreen, and mixed forests) are portrayed with circles. Modified from Simkin et al., (2016).

The CLs were calculated using the partial derivative with respect to N (∂/∂N, equations **3A-2** to **3A-5**) of the best statistical model for species richness and solving for N deposition. This expression (**Eq. 3A-5**) represents the rate of N deposition (kg/ha-yr) above which reductions in herbaceous biodiversity begins to occur, using local values for soil pH, temperature, and precipitation.

The full statistical model with all possible terms is (some may drop out if they are not significant):

$$SR = \beta_0 + \beta_1(Ndep) + \beta_2(P) + \beta_3(T) + \beta_4(pH) + \beta_5(Ndep)(pH) + \beta_6(Ndep)(T) + \beta_7(Ndep)(P) + \beta_8(Ndep^2) \qquad \text{(Eq. 3A-2)}$$

which after one takes the partial derivative with respect to N becomes,

$$\frac{\partial}{\partial N}(SR) = \beta_1 + \beta_5(pH) + \beta_6(T) + \beta_7(P) + 2\beta_8(Ndep) \qquad \text{(Eq. 3A-3)}$$

To obtain the CL, we set Eqn. 2 to less than zero (i.e. species loss with N deposition) and solve for Ndep,

$$\frac{\partial}{\partial N}(SR) = 0, \qquad \text{(Eq. 3A-4)}$$

which becomes in terms of Ndep,

$$CL(Ndep) = \frac{\beta_1 + \beta_5(pH) + \beta_6(T) + \beta_7(P)}{-2\beta_8} \qquad \text{(Eq. 3A-5)}$$

Using the statistical results from Simkin et al. (2016), these equations for open (**Eq. 3A-6**) and closed (**Eq. 3A-7**) canopy systems are:

$$CL(Ndep) = 4.75 + 0.481 * pH + 0.00182 * precip - 0.0739 * temp \qquad \text{(Eq. 3A-6)}$$

$$CL(Ndep) = 1.80 + 2.17 * pH \qquad \text{(Eq. 3A-7)}$$

Further details on the original CLs are available in Simkin et al. (2016). We converted these point estimates of CLs into two different versions for the NCLD that are described below: (1) point based CLs, and (2) Ecoregion (Level I-IV) area based area CLs. There is a third version under development that will be gridded values of the herbaceous biodiversity CL based on Eq. 4 that were not available at the time of this update to the NCLD v3.0.

Location Based Critical loads

Location (point) based CLs are directly from the "Data Dryad" public database from Simkin et al., (2016) (e.g. Simkin_et_al_2016_data_from_PNAS_Div_and_N_dep.csv). No modifications were made to the CLs, although additional information was added to the NCLD database. Please note that both LatDD and LongDD have been "fuzzed" (accuracy reduced from 8 to 4 digits), as required by the data owner(s). For this reason, some plot locations will fall outside of their correct Ecoregion. Use caution when comparing the plot locations in this dataset with other GIS spatial data. Coordinates values may be obtained by contacting Chris Clark, USEPA (Clark.Christopher@epa.gov). This fuzzing is only true for the point-based version of the database, true locations were used prior to averaging at larger Ecoregions levels for the area-based versions (below).

Ecoregion (Area) based Critical loads

The Ecoregion area-based CLs used the unfuzzed point/plot estimates from Simkin et al. (2016) to extrapolate plot-based CLs to similar ecosystem types. This extrapolation was limited to Ecoregion levels III and IV to maintain confidence in the extrapolation. A statistical check was performed to determine if the sample sizes within each Ecoregion (III and IV) were adequate to calculate statistical summary CL values (e.g. the mean, 10th percentile, etc.), given a predefined error rate and confidence described below. If the statistical checks were met within Ecoregion IV, then the sample size was considered adequate for the given CL statistic, and thus the value derived from the sample Ecoregion IV was used. If the criteria did not meet the confidence level, then the statistic CL derived for Ecoregion III (i.e., coarser scale) was determined for the Ecoregion. If the criteria were not met for either Ecoregion III or IV, no CL was extrapolated for the Ecoregion. This method was used to calculate CLs for the: (1) average, (2) minimum, (3) 1st quantile (Q1), (4) 5th quantile (Q5), (5) 10th quantile (Q10), (6) 50th quantile (Q50), and (7) the maximum. The details of the calculations are provided below.

The standard statistical equation for a recommended sample size is given by:

$$n = \left(\frac{t_{\propto/2} * \sigma}{E}\right)^2, \qquad \text{(Eq. 3A-8)}$$

where n is the recommended minimum sample size, given the desired error rate E, standard deviation from the sample σ, and desired confidence as specified by the two-tailed t-statistic $t_{\alpha/2}$. In the NCLD v3.0, we assume an error rate of 0.5 kg N/ha-yr (i.e. +/- 0.5 kg N/ha-yr, within 1 kg N/ha-yr of the true CL), and a 95% confidence for t based on the number of plots in the sample. Given that the true deposition is not known to this degree of accuracy in many areas, constraining the CL to an accuracy higher than +/- 0.5 kg or 95% was not considered necessary. However, individual users can input different values of E or t if they desire using standard statistical tables for t. Thus, Ecoregions that were more variable would have a higher σ, and therefore require a larger sample size for a given error rate and confidence. Also, Ecoregions that had fewer samples, would have a larger t, and also require a larger sample size for the given error rate and standard deviation.

Equation 3A-8 is valid whether one is estimating the sample size for the mean, or any quantile - what changes is the value of σ. For estimating the sample size required for the mean, σ is the standard deviation of the mean, that is readily calculated with any statistical package. For estimating the sample size required for a specific quantile, σ is the standard deviation around that quantile. This estimate is not commonly available, but is readily estimated using a delta method argument and the “density” function in R[1]. This function uses a standard procedure (fast Fourier Transform) to estimate the density of points around any quantile specified by the user. That density is then used to estimate the standard deviation of the sample around that quantile. The script is below:

```
# x is your data
# q is your desired quantile level, i.e. 0.01 or 0.05

quant = function(input) {quantile(input, prob = q, type = 1, names = FALSE)}
```

---

[1] https://www.rdocumentation.org/packages/stats/versions/3.4.1/topics/density

```
xi.hat = quant(x)
f.hat.temp = density(x, from = xi.hat, to = xi.hat, n = 1)
f.hat = f.hat.temp$y
se = sqrt(q*(1-q)/n)/f.hat
sd = sqrt(q*(1-q))/f.hat
```

These estimates are only available from the NCLD v3.1 and v3.2 for the specified statistics above, but interested users are welcome to use the R code above to estimate σ for any quantile between Q1 and Q99. More extreme quantiles require additional assumptions and procedures that are not readily available.

A few caveats and additional comments are warranted. First, the expectation that more data is required the further on the tail (e.g. Q10 versus Q50) depends heavily on the distribution of the data. An earlier draft of this database used a simpler approach for estimating the sample size needed for quantiles – it assumed that the sample size for any quantile needed to be triple that of the mean. This approach was usually conservative (i.e. was a larger sample than from using the density estimator function above), but is not statistically robust. That assumption, that more data is needed further on the tail, depends on the assumption of normality. Although logical, closer inspection of the critical loads identified many Ecoregions where the data is not normal, either strongly skewed left, right, or even bimodal (i.e. included sensitive and non-sensitive areas). In these cases, the assumption that more data is needed for the tails is not necessarily valid. For example, in a skewed-right distribution (i.e. a long right tail, **Figure 3A-2**), there is more data on the low end than the average, and thus fewer sample may be needed for the Q10 than the average. In an extreme case, for a bimodal distribution (**Figure 3A-2**) there are few samples near the mean, thus there is a larger sample size required to estimate the average with the same error rate and confidence than either of the modes. If users want to calculate the sample size requirements for quantiles not offered here, we recommend using the R script above. Users may use the “triple the requirement for the average” rule, as it is often more conservative, but in non-normal cases which are common at smaller scales it is not advised.

Second, we required there to be a minimum sample of 10 to perform these checks for any statistic (mean, Q1, etc.). A robust estimate of σ, whether for the average or any quantile, requires a reasonable number of points to sample the underlying (unknown) population. Commonly, for normally distributed data, the recommended minimum sample size is 30. Also common is a recommendation based on which quantile you’re trying to estimate, where you want 3-5 samples on either side of a given quantile. Using a central value of 4, that translates to a recommendation of 8 for the median, and 40 for the Q10. Generalizing, the recommended sample size for any quantile (irrespective of the data) is 4/min(q, 1-q) (where q is the quantile being estimated, so for Q50, that is 4/0.5 = 8 samples; for Q1 or Q99, that is 4/0.01 = 400 samples. This assumption, however, ignores how variable your samples actually are, and the acceptable error rate and confidence. Since our acceptable error rate is high relative to the variation in most Ecoregions, and we were interested in providing estimates for as many Ecoregions as is defensible, we consulted with statisticians on relaxing these guidelines and determined that a minimum of 10 would be acceptable. Users are not encouraged to consider any estimate, especially the tails, as reliable if N<10 since much of these data are non-normal, especially at smaller Ecoregions. For example, with an N=5 and left skewed data, the average of your sample is probably closer to Q80 than the mean, and the minimum of your sample may be closer to the average of the true population.

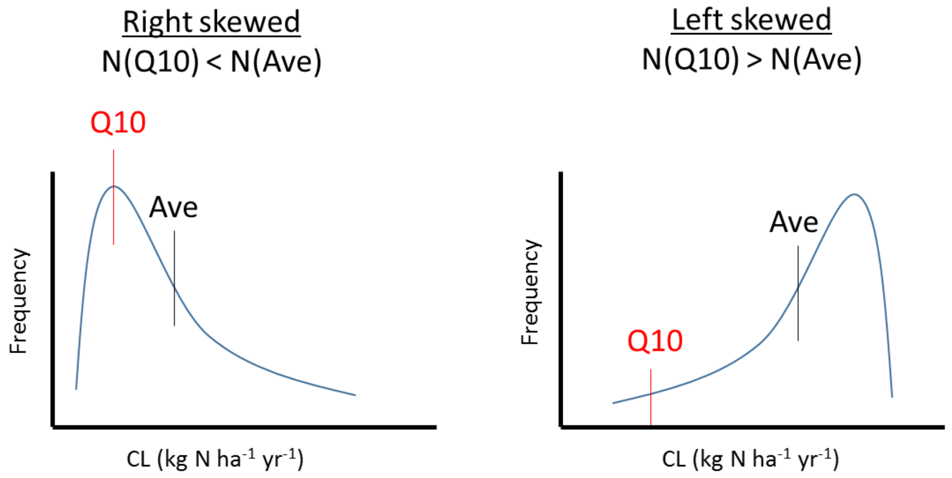

Figure 3A-2. Examples of non-normal distributions affecting sample size requirements. In right skewed distribution (and bimodal), fewer samples may be required for lower quantiles than for the average. The opposite is predicted for left skewed (and normal) distributions.

Third, there are some instances where the recommended sample size is very low (e.g. 1-2 is not uncommon). This is not unexpected because in many cases the standard deviation among samples (esp. for Open canopy systems) is very low relative to the acceptable error rate. Thus, only a few points are needed to estimate the statistics within the relatively large acceptable error rate. This does not mean that one should use an estimate of the Q10 based on a sample of one, which is why the minimum sample size to perform these checks at all is 10. Rather, it means that the sample of 1 is all you actually needed to be within the specified quantile given the acceptable error rate. One would not have known that without the original sample of 10.

Fourth, there is no way to estimate the sample size for the minimum or the maximum. These are at the extremes of the data and thus theoretically require sampling the entire population ($n = \infty$). Thus, as an approximation, we assumed that if the Q1 criteria was met, then we could estimate the minimum and maximum CL with some confidence.

Fifth, there were cases where n could not be calculated with **Eq. 3A-8** because there was zero variability in the sample ($\sigma=0$; N=11 and 108 individual Ecoregions at Level III, and IV, respectively). This could occur two ways: if there was a sample size of one, or if there was no variation among individual samples. If the sample size was one, the sample size under the threshold of 10 and the next highest level is recommended for the CL statistics. If the sample had no variation but 10 or more points, the CL was estimated to not vary and the required sample size is predicted to be 1. This occurred solely because for Closed Canopy systems in the Temperate Sierras Level I Ecoregion (N=42) and all nested sub Ecoregions, all CL values were the same. Here, soil pH had the same value in all sites and was the only modifying factor in the equation for the CL. In these cases, only a mean value is presented since it is not appropriate to infer a quantile when all values are identical. Furthermore, soil pH likely varies across sites in this Ecoregion, and updated soil pH values will be used once they become available.

This approach is valid for setting minimum sample sizes for other critical loads as well (e.g. for sulfur, aquatic acidification) with appropriate modifications, and will be explored in future versions of the NCLD. Below are the results for all Ecoregion levels (**Table 3A-3**).

Table 3A-3. Sample Size Results for All Ecoregions

				Mean		Q50		Q10		Q5		Q1	
Canopy Type	**Ecoregion Level**	**# Ecoregions in US**	**# Ecoregions with Plots**	**# Yes**	**%**	**# Yes**	**%**	**# Yes**	**%**	**# Yes**	**%**	**# Yes**	**%**
Closed	III	84	52	29	56%	25	48%	23	44%	23	44%	30	58%
	IV	967	340	93	27%	77	23%	76	22%	77	23%	96	28%
Open	III	84	39	27	69%	26	67%	26	67%	27	69%	27	69%
	IV	967	162	78	48%	76	47%	75	46%	74	46%	78	48%

Development of Area Based Herb Richness CL rasters

The Ecoregion III and IV CLs were made into a Raster GIS format based on Q5 statistic. Polygons for Ecoregion IV were used in both sets of analyses as they provide a more refined coastline. Plots were grouped into open and closed datasets and analyzed separately. For each Ecoregion polygon where the sample size criteria were met, the Q5 CL value was assigned to those polygons. The Ecoregion IV boundaries are subsets of Ecoregion III boundaries. Thus, the Ecoregion IV polygons with CLs were subsituted for the Ecoregion III areas.

The National Land Cover Dataset (NLCD) from 2011 was used to define the spatial extent of "open "or "closed" canopies ecosystems across the conterminous US (Homer et al. 2011). The Open Canopy Layer are defined by the land cover classes of Shrub/Scrub (52) and Grassland/Herbaceous (71). The Closed Canopy Layer are defined as Deciduous Forest (41), Evergreen Forest (42), and Mixed Forest (43) classes. All other land cover classes were excluded from the layer. The combined polygon Ecoregion CL layer was converted to a 30m raster to be aligned with the National NLCD raster. The Ecoregion raster was then merged separately with the Open Canopy Layer and the Closed Canopy Layer, creating two distinct rasters that define herb richness for appropriate open and closed canopy ecosystems.

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