Title: Natural and human drivers of salinity and major ion composition in United States lakes

Authors: Xinyu Sun[1*], Kendra Spence Cheruvelil[1,2], Patrick J. Hanly[1], Katherine E. Webster[1], & Patricia A. Soranno[3]

*Corresponding author: Xinyu Sun (sunxiny9@msu.edu)

Affiliations:

1. Department of Fisheries and Wildlife, Michigan State University, East Lansing, Michigan, USA

2. Lyman Briggs College, Michigan State University, East Lansing, Michigan, USA

3. Department of Integrative Biology, Michigan State University, East Lansing, Michigan, USA

This paper is a non-peer reviewed preprint submitted to EarthArXiv. The preprint was submitted to Limnology & Oceanography for peer review.

Article type: Research article

Title: Natural and human drivers of salinity and major ion composition in United States lakes

Authors: Xinyu Sun[1*], Kendra Spence Cheruvelil[1,2], Patrick J. Hanly[1], Katherine E. Webster[1], & Patricia A. Soranno[3]

*Corresponding author: Xinyu Sun sunxiny9@msu.edu

Affiliations:

1. Department of Fisheries and Wildlife, Michigan State University, East Lansing, Michigan, USA

2. Lyman Briggs College, Michigan State University, East Lansing, Michigan, USA

3. Department of Integrative Biology, Michigan State University, East Lansing, Michigan, USA

Author contribution statement: XS and KSC conceived the project. XS, KSC, PJH, KEW, and PAS designed the analyses and interpreted the results. XS gathered and processed data. XS and PJH performed analyses and made figures, tables, and supplements. XS wrote the first draft of the manuscript, and all authors contributed substantially to revisions.

Abstract:

Salinity and major ion composition are important for understanding and predicting lake water quality and responses to global changes. However, little is known about salinity and major ionic composition for populations of lakes at the continental scale, nor the corresponding relationships with natural and human factors operating at multiple spatial scales. To fill these knowledge gaps, we examined the spatial patterns in salinity using specific conductance as a proxy (N=9,785 lakes) and major ion concentrations (N=1,218 lakes) across the conterminous United States. We then quantified relationships between a wide range of multi-scaled natural and human factors and both salinity and ion composition. Most lakes had relatively low salinity (median=206µS/cm), although 4% were classified as saline (>1,500µS/cm) and mostly were located in the Plains, Desert Southwest, and Southeast regions. Calcium and bicarbonate were the dominant or most common ions in 61% of US lakes, with the remaining lakes dominated by magnesium or sodium and sulfate or chloride ions. Lake salinity was strongly related to natural factors (e.g., lake elevation, soil, and hydrology) and influenced by human factors including agriculture and atmospheric deposition. Major ion composition was associated with similar natural factors, but was also strongly affected by road density, urban development, agricultural activities, and atmospheric deposition. This macroscale understanding of salinity and major ions and their complex relationships to natural and human characteristics around lakes is needed to assess, predict, and manage lake impairments from human alterations of ion chemistry.

Keywords: Lake salinity; Specific conductance; Salt; Major ions; Geology; Hydrology; Road density; Macroscale

Introduction

Salinity is a critical indicator of lake water quality due to its profound effects on aquatic organisms, drinking water quality, and industrial and recreational water use (Kaushal et al. 2018; Hintz & Relyea 2019; Dugan 2024). Salinity in freshwater bodies has been fluctuating greatly in some regions of the world with negative impacts that are attributed to natural and human disturbances such as climate change, extreme climatic events, road salt application, and agricultural activities (Kaushal et al. 2019; Olson 2019; Schacht et al. 2023). Although studies have documented levels of individual ions in lakes such as chloride and their changes due to human activities (e.g., Dugan et al. 2017), we lack a macroscale understanding of the variation of overall salinity across broad ranges of lakes, how ionic composition differs among lakes, and the potential influences from multi-scaled natural environment and human activities.

Studies focussing solely on salinity are necessary but not sufficient because the impacts of salts on freshwater communities are related to major ion concentration and compositions as well as salinity levels as reflected by specific conductance (SC; Calver et al. 2009; Cañedo-Argüelles et al. 2016). Lab and microcosm experiments have shown that the same salinity levels with different ion compositions (e.g., solutions or media dominated by chloride (Cl^-) vs. sulfate (SO_4^{2-}) ions) can lead to divergent effects on organisms and communities (Nostro et al. 2005; Clements & Kotalik 2016; Van Gray & Ayayee 2024). Multiple ions can also interact to affect aquatic organisms, leading to unexpected outcomes (e.g., synergistic effects) (Elphick et al. 2011). For example, the toxicity of Cl^- on cladocerans was stronger in softer water than in hard water (Elphick et al. 2011; Rogalski et al. 2024), and the toxicities of potassium ion (K^+) could be alleviated by high sodium ion (Na^+) concentration (Mount et al. 2016). These findings underscore the necessity of understanding major ion concentrations and composition in lakes for a better assessment and prediction of salt effects on freshwaters.

Salinity and major ions in surface water are related to a variety of natural and human factors. Previous studies have suggested that salinity in streams across the conterminous United States (CONUS) ranged from extremely low (<2 μS/cm SC) to hypersaline (>10,000 μS/cm), depending on the dominant sources and/or surrounding natural factors (e.g., evaporation; magnesium (Mg^{2+}), calcium (Ca^{2+}), and SO_4^{2-} ions from weathering of rocks; and Na^+ and Cl^- ions from saline groundwater) (Gibbs 1970; Griffith 2014; Olson & Cormier 2019). Studies of lakes in south-central North Dakota revealed that within-region salinity and major ion concentrations were related to lake elevation, soil texture, and groundwater (Swanson et al. 1988). La Baugh et al. (2000) found that salinity levels in lakes and wetlands in Central North America varied with evaporation, precipitation, and groundwater fluxes. Additionally, regional studies on lakes within lake chains demonstrated the effects of landscape position, and lakes that are connected with other waterbodies (higher surface water connectivity) had higher salinity (Martin & Soranno 2006; Soranno et al. 1999). Moreover, human activities and urban development can sometimes cause long-term or permanent changes to salinity. Salt inputs from irrigation runoff, residential discharge, road salt application, wastewater effluents, and mining accumulate in the water column and increase salinity (Oswald et al. 2019; Stets et al. 2020; Dumelle et al. 2024). In particular, agricultural effluents often contain high concentrations of K^+, Cl^-, and SO_4^{2-}; road deicing salts can contribute significant amounts of Na^+, Mg^{2+}, Ca^{2+}, and Cl^-; and industrial runoff and acid rain can contribute SO_4^{2-} (Charles 1991; Dugan et al. 2017; Dugan 2024).

Previous studies have only included a limited number and type of factors (e.g., not consider human influences) and predominantly focused on individual waterbodies or watersheds, with broad-scaled studies focusing on lotic systems (e.g., Griffith 2014; Olson & Cormier 2019; Stets et al. 2020; but see Dugan 2024). Unfortunately, findings from studies on streams are not always directly applicable to lakes because streams and lakes differ in morphometry, hydrological pathways, and lakes have a range of surface water connectivity, which can lead to distinct chemical and physical characteristics (e.g., water residence time, salt retention time, and evaporation) and water chemistry (Lottig et al. 2011; Kahlert & Gottschalk 2014). Macroscale studies that incorporate a wide range of multi-scaled (local to regional) factors are therefore required to understand the spatial variation of lake salinity and major ions and influences of natural and human factors on them, and this knowledge is critical to establish reasonable thresholds and goals for management and habitat restoration, as well as to predict lake responses to future changes. In this research, we investigated two questions: 1) What are salinity and major ion concentrations and composition in lakes across the CONUS that include broad ranges of climate, hydrology, and land use? and 2) What multi-scaled natural and human factors are most strongly related to them? We answered these questions using water chemistry and ecological context data from multiple data sources for 9,785 (salinity) and 1,218 (major ions) lakes of the US, investigated their spatial variations, and conducted random forest and GLMNET analyses to examine the relationships between natural and human factors and salinity and major ions.

Methodology

Data collection

We used data from the LAGOS-US research platform (Cheruvelil et al. 2021) that includes lake, natural context, and human activity data for 479,950 lakes ≥ 1 ha surface area across CONUS. We obtained *in situ* epilimnion specific conductance (SC), individual ion concentrations for the cations Ca^{2+}, K^{+}, Mg^{2+}, and Na^{+} and the anions Cl^{-} and SO_4^{2-}, and alkalinity (as $CaCO_3$) from the LAGOS-US LIMNO module (Shuvo et al. 2023). In this study, we used SC (i.e., the electrical conductivity of one cubic centimeter of solution at 25°C), which is commonly measured in inland waters, as the proxy for salinity (Calver et al. 2009; Cañedo-Argüelles et al. 2016; Dugan 2024). The LIMNO module includes lake surface water quality data from the US Water Quality Portal (WQP; 2021), 2007, 2012, and 2017 US National Lakes Assessments (NLAs; US Environmental Protection Agency (EPA) 2010, 2016, 2022), and the US National Ecological Observatory Network (NEON; Keller et al. 2008). For natural and human factors, we used lake locational, morphometry, and surface water connectivity data from the LAGOS-US LOCUS module (Cheruvelil et al. 2021; Smith et al. 2021); data about the natural context (e.g., soil texture and climate features) and human factors (e.g., road density and atmospheric deposition) from the LAGOS-US GEO module (Smith et al. 2022); and lake reservoir designation (natural lake or reservoir) from the LAGOS-US RESERVOIR module (Polus et al. 2022; Rodriguez et al. 2023). We calculated the average wet inorganic nitrogen, nitrate, and sulfate atmospheric deposition since 2000 to indicate their current levels and the differences between before and after 2000 to indicate the change in deposition. Additionally, we acquired evapotranspiration and snowpack water equivalent storage data from Blodgett (2023) and livestock manure application data from US EPA EnviroAtlas (2015). To account for large-scale geographical variation, we used NEON regions, which are ecoregions (213,800 - 770,995 km^2) classified based primarily on climate (Hargrove & Hoffman 1999). We removed highly correlated variables, resulting in 54 multi-scaled natural and human factors (see Table S1 for a list of factors).

Data processing

We used 2000-2021 measurements of lake SC, major ion concentrations, and alkalinity. Because we had much more SC than ion concentration data, we created two datasets for further analyses: a full dataset with only SC data and a sub-dataset with SC and complete major ion data. We applied water quality QA/QC procedures from LAGOS-NE-LIMNO (v. 1.087.3; Soranno et al. 2019) to these datasets, resulting in 11,072 lakes with SC data. We further extracted the SC data from April to October (90% of data) that were taken during the most recent sampling year for each lake (median year = 2015; Figure S1a), computed the mean SC of each lake to represent lake salinity, and merged those data with multi-scaled natural and human factors, yielding 9,785 lakes with complete data (i.e., the full dataset with no missing values for any factor) that are representative of US lakes (Figure S2). For the 4,581 lakes with more than one SC sample in the latest sampling year from April to October, we calculated the coefficient of variation (CV) to represent intra-year temporal variation in salinity.

For the sub-dataset combining SC, ions, and alkalinity data, we extracted April to October data (≥85% of data) and used the most recent concurrent (i.e., taken the same day) samples available (one water sample per lake for 1,498 lakes; median year = 2016). Next, to investigate ion composition in lakes, we converted ion concentrations and alkalinity reported in mg/L units in LIMNO-US to microequivalents per liter (μeq/L; Table S2). In this study, we used bicarbonate (HCO_3^-) (in μeq/L) to represent all carbonate forms of alkalinity including CO_3^{2-} which dominates in high pH waters. Major ion data were combined with natural and human factors to generate a 1,218 lakes sub-dataset, of which about 85% were sampled by the NLAs (Figure S1).

Data analyses

Data analyses were conducted in R (v4.3.3; R Core Team, 2024). To identify spatial patterns of salinity using the full dataset, we plotted the mean and CV of salinity over the 17 NEON regions for CONUS. Then, we used Boruta feature selection ('Boruta' package, v8.0.0, Kursa & Rudnicki 2022) and random forest (RF, 'randomForest' package, v4.7-1.1, Cutler & Wiener 2022) to examine what and how multi-scaled natural and human factors affect salinity in lakes. For factors, a natural log transformation was applied to non-percent data and a generalized logit transformation was applied to percent data (Table S1). Two Boruta feature selections with a maximum of 1,000 runs were performed using salinity values as the response variable and either natural or human factors as predictors. We ranked natural and human factors separately based on Boruta importance scores, then took the first half (i.e., the top half of factors based on importance) from each and input them into an RF model with 5-fold repeated cross-validation to examine the important natural and human factors that affect lake salinity.

The effects of the important factors were assessed through 1-factor and 2-factor partial dependence plots (PDPs; ‘pdp’ package, v0.8.1, Greenwell 2022). We identified important factors for 1-factor PDPs using both the percentage increase in mean squared error (% increase MSE) and the increase in node purity. The % increase MSE represents the increase in MSE when a factor is excluded and node purity indicates the before-after change in the residual sum of squares at a splitting node (Cutler & Wiener 2022). Previous literature has suggested that natural and human factors can potentially interact to affect inland water quality (Kernan & Helliwell 2001; Nobre et al. 2020; Lin et al. 2021). Thus, although interactions are not our primary focus, we also ran an iterative random forest (iRF; Basu et al. 2018), which identifies interactions between factors and quantifies the stability of interactions to select combinations of natural and human factors to be used in PDPs. We selected natural-human-factor combinations without repetition and visualized their joint effects using 2-factor PDPs.

To study the spatial patterns of and how multi-scaled natural and human factors affect ion composition using the sub-dataset, we first applied hierarchical clustering on $\log_{10}$-transformed ion equivalent concentrations to identify common patterns among ion concentrations and composition in lakes (Ward’s method; Härdle & Simar 2019). We identified 15 clusters (Figure S3) and used them as the response variable in later analyses. Next, we ran a Boruta feature selection using all (natural and human) factors as predictors. The unimportant factors identified by Boruta were removed, and we applied a natural log transformation to numeric data (Table S1) and centered and scaled (into z-scores) all continuous factors before running a multinomial GLMNET model with LASSO regularization (glmnet package, v4.1-8, Friedman et al. 2023). Each factor was assigned to one of 11 categories: climate, hydrology, lake and watershed (morphometry), lithology, location, soil, surface connectivity, terrain, atmospheric deposition, human activities, and land use/land cover (LU/LC). We also calculated the relative importance of each factor for predicting ion cluster membership by summing the absolute value of the factor's multinomial coefficient across all clusters.

Results

Salinity and major ions in lakes across the CONUS

Using the full dataset with salinity for 9,785 lakes, we found that salinity varied within and among regions, ranging from 2.0 to 6,125 µS/cm SC (mean±standard deviation (SD)=343±511 µS/cm, median=206 µS/cm) (Figure 1a & S1j). Most lakes with high salinity (about 4% of all lakes; >1,500 µS/cm) were located in the Southeast, Northern Plains, Central Plains, Southern Plains, and Desert Southwest regions. Among the 17 NEON regions, the Desert Southwest had the highest average salinity (1,154 µS/cm), followed by the Northern Plains (897 µS/cm), and Central Plains (894 µS/cm). In contrast, the Pacific Northwest had the lowest mean salinity (72 µS/cm), followed by the Northeast (139 µS/cm) and Mid-Atlantic (146 µS/cm) regions (Figure 1a). Lakes with multiple sampling dates within a year (N=4,581) often had low intra-year temporal variation in salinity (CV: mean±SD=10%±13%, median=6%), and lakes with high CV (greater than 100%) were predominantly found in the Northeast, Southeast, and Great Lakes regions (Figure 1b).

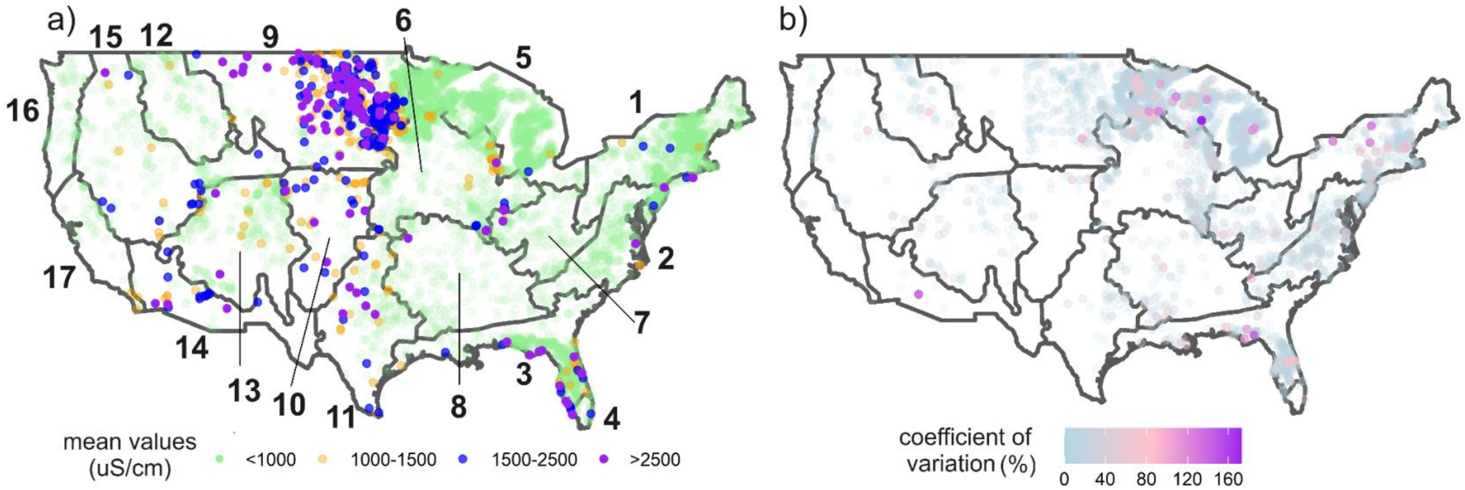


Figure 1. Maps showing mean April-October specific conductance (SC) as a proxy for salinity (a; N=9,785) and the intra-year SC coefficient of variation (b; N=4,581) of each lake by NEON region. Each dot represents a lake. NEON regions: 1 = Northeast, 2 = Mid-Atlantic, 3 = Southeast, 4 = Atlantic Neotropical, 5 = Great Lakes, 6 = Prairie Peninsula, 7 = Appalachians & Cumberland Plateau, 8 = Ozarks Complex, 9 = Northern Plains, 10 = Central Plains, 11 = Southern Plains, 12 = Northern Rockies, 13 = Southern Rockies & Colorado Plateau, 14 = Desert Southwest, 15 = Great Basin, 16 = Pacific Northwest, 17 = Pacific Southwest.

For the sub-dataset with salinity and major ions, we classified the 1,218 lakes into 15 well-distinct clusters with divergent ion concentrations and compositions (Figures 2&3, Table 1). In most clusters (clusters 13, 15, 11, 4, 2, 9, 14, and 3), despite different salinity, Ca^{2+} and HCO_3^- were the most abundant cation and anion, respectively. However, about 8% of lakes were dominated by Mg^{2+} and SO_4^{2-} (cluster 8); Ca^{2+} and Mg^{2+} were almost equally abundant in 8% of lakes (clusters 12 and 5); Na^+ and Cl^- were the most abundant cation and anion, respectively, in 5% of lakes (clusters 6 and 1); and in 17% of lakes, Ca^{2+} and Na^+ had similar proportions among cations (clusters 7 and 10). Among the clusters, cluster 8 had the highest salinity, followed by clusters 6 and 9, and cluster 4 had the lowest, followed by clusters 11 and 2. Saline lakes

(SC>1,500 µS/cm; DeVilbiss et al. 2022) were found in clusters 8 (55 lakes, 56%), 6 (19 lakes, 46%), and 5 (1 lake, 1%).

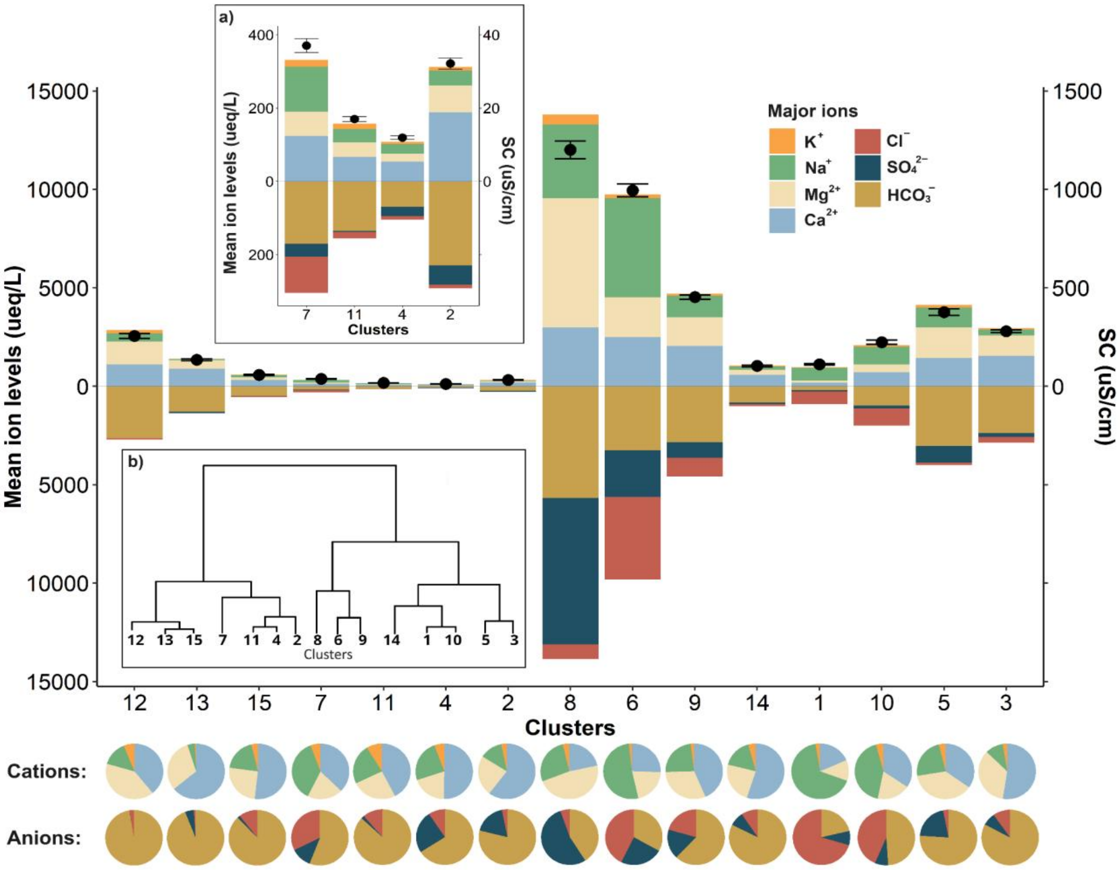


Figure 2. The mean equivalent concentrations (µeq/L) of major ions, mean salinity levels (SC, uS/cm), and proportions of cations and anions of each cluster. Stacked bars represent ion concentrations with cations above and anions below the zero line, respectively, with corresponding values shown on the left y-axis; black dots represent mean cluster salinity levels and correspond to the right y-axis; error bars represent 1 standard deviation; and pie charts at the bottom represent the proportions of each cation and anion within a cluster. To aid in visualization, subplot a) shows ion and salinity levels in clusters 7, 11, 4, and 2. Subplot b) shows a simplified dendrogram of the hierarchical clustering (a complete dendrogram is in Figure S3).

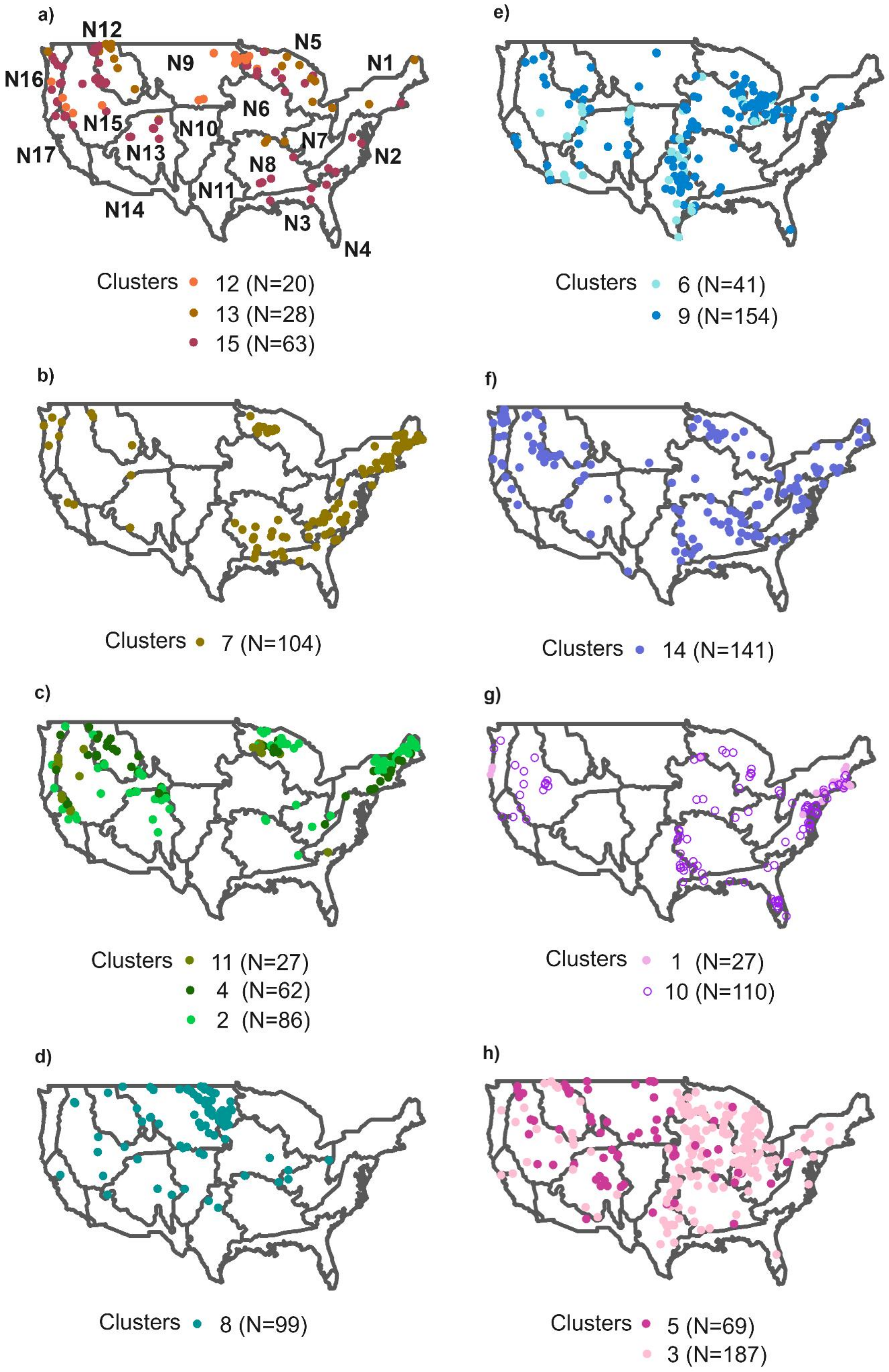


Figure 3. Maps showing the spatial distributions of lakes in each major ion composition cluster by NEON region (N1-N17 in plot a). Each set of clusters are more similar in ion composition to each other than to other clusters according to the hierarchical clustering procedure. The numbers in the parentheses indicate the number of lakes in each cluster. See Figure 1 caption for NEON region names.

Table 1. Summary of each cluster's dominant cation(s) and anion(s), percentage of each cluster among all lakes, and average salinity (as SC) of each cluster. Clusters are ordered according to dominant ions and average salinity. (See Figure S4 for lake-specific relationships between SC and salinity calculated using major ion concentrations.)

Cluster	Dominant cation(s)	Dominant anion(s)	Percentage	Average salinity±SD (μS/cm)
4	Ca^{2+}	HCO_3^-	5%	12±4
11	Ca^{2+}	HCO_3^-	2%	17±7
2	Ca^{2+}	HCO_3^-	7%	32±15
15	Ca^{2+}	HCO_3^-	5%	57±26
14	Ca^{2+}	HCO_3^-	12%	103±40
13	Ca^{2+}	HCO_3^-	2%	134±44
3	Ca^{2+}	HCO_3^-	15%	279±75
9	Ca^{2+}	HCO_3^-	13%	453±111
12	Ca^{2+}, Mg^{2+}	HCO_3^-	2%	255±119
5	Ca^{2+}, Mg^{2+}	HCO_3^-	6%	377±172
7	Ca^{2+}, Na^+	HCO_3^-	8%	37±18
10	Ca^{2+}, Na^+	Cl^-, HCO_3^-	9%	224±113
8	Mg^{2+}	SO_4^{2-}	8%	1,201±448
1	Na^+	Cl^-	2%	111±40
6	Na^+	Cl^-	3%	995±330

Among lakes dominated by Ca^{2+} and HCO_3^-, most (clusters 11, 15, 14, and 3) were widely distributed across multiple NEON regions, with others clumped together in some regions, including clusters 4 (60% in Northeast and Northern Rockies), 2 (53% in Northeast and Great

Lakes), 13 (64% in Great Lakes and Northern Rockies), and 9 (70% in Great Lakes, Prairie Peninsula, Southern Plains) (Figure 3). Among lakes with other ion compositions, those in cluster 12 (cations co-dominated by Ca^{2+} and Mg^{2+}) were mostly located in the Northern Plains and Great Basin (Figure 3a); lakes in cluster 7 (cations co-dominated by Ca^{2+} and Na^{+}) were commonly found in the Northeast, Mid-Atlantic, and Ozarks Complex regions (Figure 3b); lakes in cluster 8 (dominated by Mg^{2+} and SO_4^{2-}) were mostly abundant in the Northern Plains (Figure 3d); more than half of the lakes in cluster 6 (dominated by Na^{+} and Cl^{-}) were in the Prairie Peninsula and Southern Plains; and lakes in cluster 1 (dominated by Na^{+} and Cl^{-}) were predominantly in the Northeast and Mid-Atlantic regions (Figure 3g). Generally, NEON regions with more sampled lakes tended to have higher numbers of clusters, except for the Northern Plains where the majority of the lakes were in cluster 8.

Multi-scaled natural and human factors related to salinity and major ions

Salinity was related to both natural and human factors (RF out-of-bag variance explained=60%, N=9,785) although more natural than human factors were among the most important factors (top 10). Using the increase in node purity (using the Gini coefficient), groundwater recharge was the most important factor, followed by runoff and percent clay in the soil (Figure 4a). Based on the RF model % increase in MSE, lake elevation was the most important factor, followed by the watershed percent agricultural land use and runoff (Figure 4b). We observed a range of relationships between factors and lake salinity using partial dependence plots (PDPs) of selected important factors. Lakes at the lowest elevations had high salinity, followed by a steep decline for lakes located above 120 m, then salinity gradually declined and eventually became stable as lake elevation increased (Figure 5a). A positive relationship was observed between watershed agricultural land use and salinity (Figure 5b). Salinity was lower in lakes with higher watershed forest land cover (which can represent areas with no or little human disturbance) until forest cover reached about 80%, then salinity became higher (Figure 5c). The percentage of clay in soil and salinity were negatively related until clay got to 2%, and the consistently low salinity was higher when clay was greater than 12% (Figure 5d). Lake salinity was lower with higher annual runoff and became stable at a low level when runoff was greater than 20 inches/year (Figure 5e). Similarly, a negative relationship was found between salinity and groundwater recharge until about 150 mm/year, at which point salinity stabilized at a low level (Figure 5f).

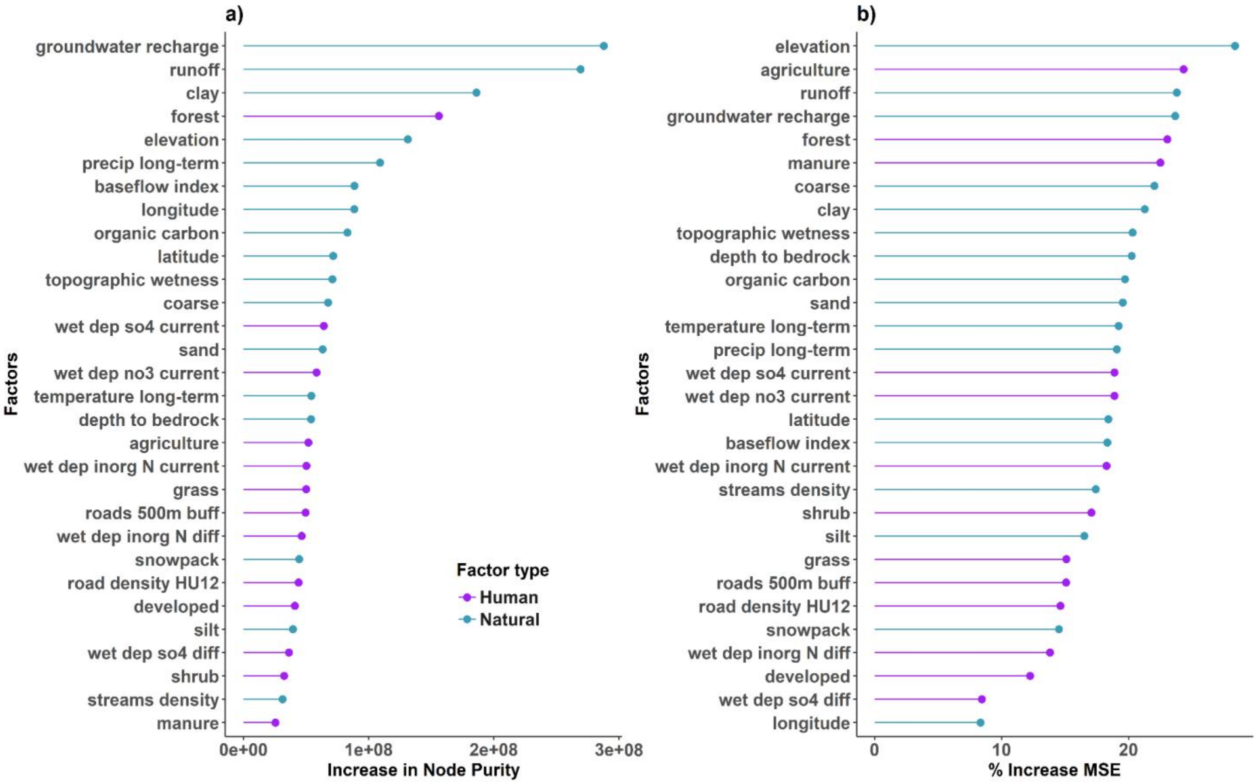


Figure 4. Results from random forest model (N=9,785) of natural and human factors and their relationships to lake salinity: a) importance scores of important natural and human factors based on the increase in node purity; b) importance scores of important natural and human factors based on the percentage increase in mean squared error.

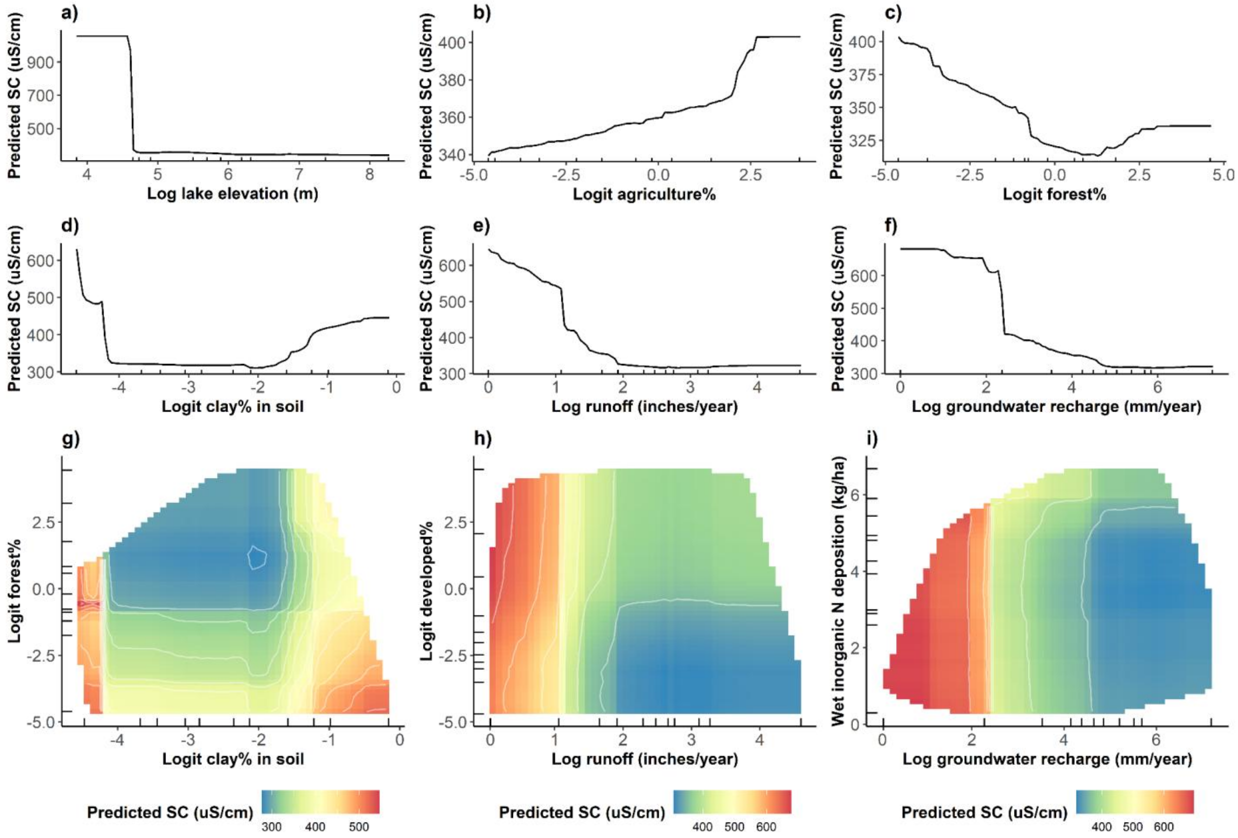


Figure 5. 1-factor PDPs show the relationships between predicted salinity (as SC) and the most important natural and human factors (plots a-f), and 2-factor PDPs show the joint influence of important natural and human factors on predicted salinity (plots g-i). Tick marks on the axes represent the deciles of the natural and human factor values. ‘Log’ indicates natural log transformation.

The PDP showing two-way relationships among factors suggested that salinity is higher in lakes located in areas with greater percent clay in the soil and lower watershed forest cover (highest salinity at 31% clay and 1% forest) (Figure 5h). Areas with lower groundwater recharge and wet inorganic nitrogen (N) deposition also tended to have greater salinity (highest salinity at 0 groundwater recharge and 1 kg/ha inorganic N deposition) (Figure 5i). Moreover, although high salinity was found in lakes with less runoff and more developed watersheds (highest salinity at 0 runoff and more than 65% developed land use), we observed that for lakes with high runoff (>7 inches/year), those with developed watershed (>35% of the watershed) had higher salinity (Figure 5j).

Both natural and human factors determined ion cluster assignment for the 1,218 lakes, and the direction of factor influence (positive or negative) varied by cluster (Figure 6). Ion clusters showed stronger associations with factors in the categories of soil, terrain, and climate than those in other categories, and hydrology, particularly groundwater features, and lake locations were moderately associated with major ions. Most lake and watershed morphometry and lithology factors had weak associations with clusters. Among natural factors, the percentage of clay in the soil had the highest importance for ion cluster membership, followed by long-term average precipitation and annual runoff (Figure 6a). Soil clay had high positive associations with clusters 6, 8, and 9 and was strongly negatively associated with clusters 4, 11, and 10. Both long-term average precipitation and runoff were positively associated with clusters with relatively low salinity (e.g., clusters 15, 7, 11, 4, 14, and 1) and negatively associated with clusters with greater salinity (e.g., clusters 9, 8, and 6).

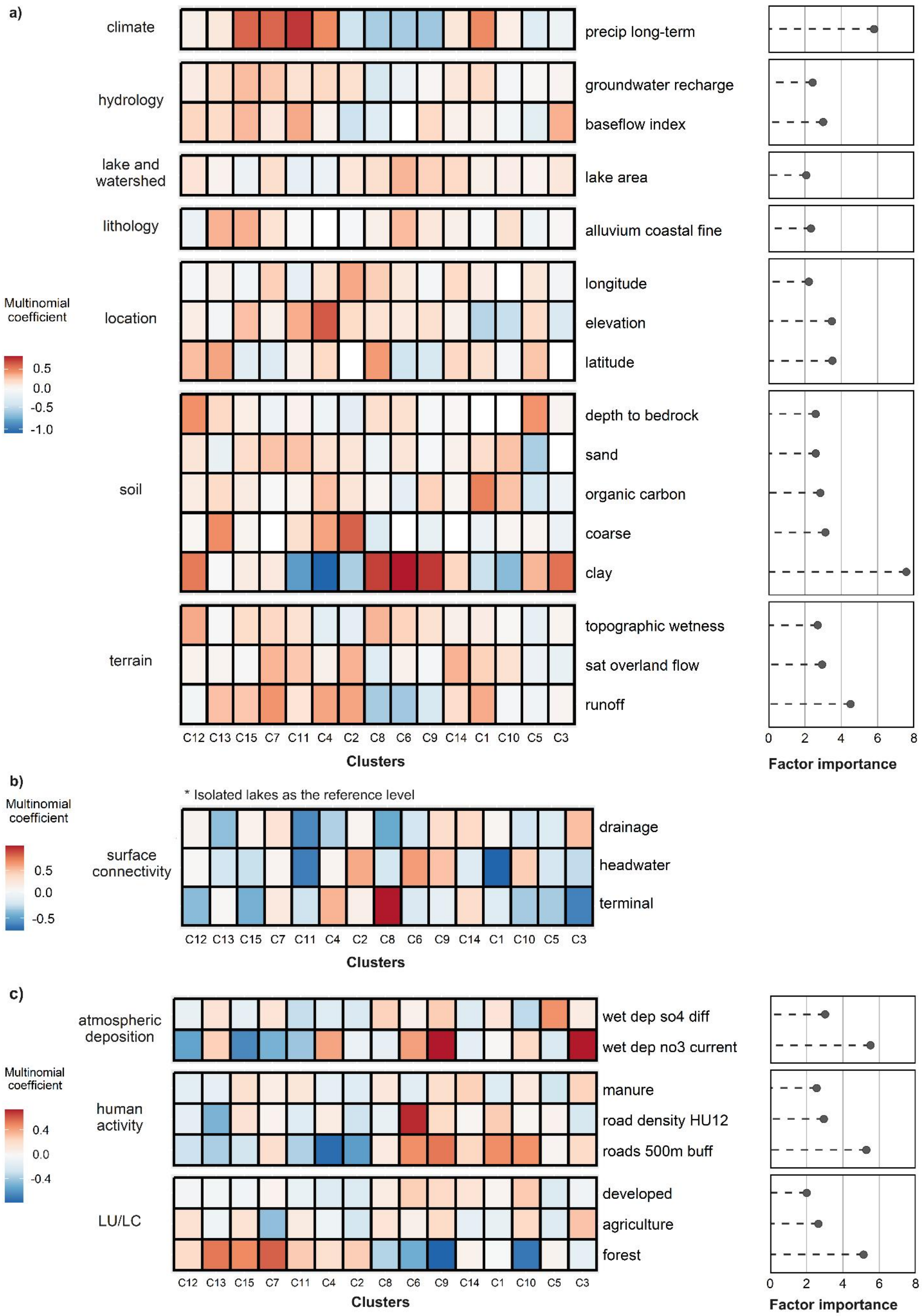


Figure 6. Heatmaps showing multinomial coefficients from a GLMNET model predicting ion

composition cluster assignment based on each natural (a,b) and human factor (c), with the factor importance plotted on the right (lollipop plots) that was calculated as the absolute sum of the cluster-specific coefficients. Numeric factors in plots a) and c) were centered and scaled. The text on the left of the heatmaps indicates factor categories, and the text on the right indicates factor names. Surface connectivity (plot b) was input as a categorical natural variable in the model; we plotted this factor separately to avoid confusion and did not calculate its importance score. Only factors with importance values ≥2 were plotted (17 of 34 inputted natural factors, surface connectivity was counted as one factor, 8 of 20 inputted human factors). A full table of coefficients can be found in Table S3.

When comparing other surface water connectivity classes to those with no inflow and no outflow (i.e., isolated lakes), our results suggested that lakes with only inflow (terminal lakes) were more likely to be assigned to cluster 8 and less likely to cluster 3; lakes with only outflow (headwater lakes) were more likely to be assigned to clusters 6 and 2 and had low probabilities to be in clusters 1 and 11; and lakes with inflow and outflow (drainage lakes) were more likely of being in clusters 3, 14, and 9 and less likely to be in clusters 11, 8, and 13 (Figure 6b, Table S4).

Across human factors, atmospheric deposition, human activities, and LU/LC all influenced ion compositions (Figure 6c). The recent wet deposition of nitrate (NO_3^-), road density within 500m of the lake, and watershed forest land cover were the strongest factors associated with ion clusters. NO_3^- deposition was highly positively associated with clusters 9 and 3 and negatively associated with clusters 15, 12, and 7. Road density had positive associations with clusters 9, 6, 1, and 10 and negative associations with clusters 4 and 2. We found watershed forest land cover had positive associations with clusters with relatively low amounts of ions (e.g., clusters 7, 13, and 15) and negative associations with clusters with higher total ion amounts (e.g., clusters 9, 10, 6, and 8).

Discussion

Salinity and major ion composition varied across US lakes, as did the natural and human factors related to them. Although most lakes had low salinity, 4% of lakes across the US were saline and distributed broadly across the US. Across the CONUS, 61% of lakes were dominated by Ca^{2+} and HCO_3^-, and the remaining lakes were dominated by Mg^{2+} or Na^+ and SO_4^{2-} or Cl^-. Salinity was related to natural factors primarily and human factors secondarily, while ionic composition and concentration were strongly associated with both human and natural factors.

Salinity and major ions in US lakes

Most lakes had relatively low salinity (median SC=206 µS/cm), and the high salinity lakes (SC>1,500 µS/cm) were mainly in the Plains, Desert Southwest, and Southeast regions. Previous studies on macroscale stream conductivities also observed relatively high salinity in most of these regions (Griffith 2014; Olson & Cormier 2019) but did not find similar high salinity in the Southeast region. This difference could demonstrate the water chemistry differences between lakes and streams in this region, be attributed to the large difference in the number of systems studied between previous and our research (1,521 lakes in the Southeast region), or be due to temporal variations in salinity, as some lakes in the Southeast had relatively high CVs. Regardless, this new knowledge about high-salinity lakes occuring in the southeastern US provides new insights into macroscale salinity prediction and management, particularly in the Southeast region.

Although salinity is an important measure of water quality, major ion concentration and composition are also critical for monitoring because the same salinity levels can be made up of very different major ion compositions and concentrations. For example, for clusters 14 and 1 that had similar salinity levels, we observed different ion concentrations and compositions with cluster 1 being dominated by Na^+ and Cl^- and cluster 14 being dominated by Ca^{2+} and HCO_3^-. These differences are important because the chemical composition of salt can determine the aquatic species present in water, the impacts of salts on organisms (Elphick et al. 2011; Mount et al. 2016; Huber et al. 2024), the usage of water (Tiri et al. 2018; Zaman et al. 2018), and management strategies. Unfortunately, in the US, management agencies responsible for monitoring lake water quality (which is our primary data source) are more likely to measure salinity than major ions, perhaps due to the high costs associated with sampling and measuring ions compared to SC. Government agencies responsible for lake water quality monitoring should consider approaches to fill this data gap and, in the meantime, researchers should develop predictive models for salt chemical composition in lakes to enhance our understanding of major ion levels and their potential impacts on lake ecosystems. Our models, which will be discussed later, that documented relationships between major ion composition and major natural and human features could be an important first step in this direction.

We demonstrated that ion concentration and composition varied greatly within and among regions. As expected (Dugan 2024), Ca^{2+}, and sometimes Na^+, was the dominant cation in many lakes. However, in about 16% of lakes, Mg^{2+} was the predominant cation or equally abundant to Ca^{2+}, mostly in the Northern Plains, with a few others located in the Prairie Peninsula region and West US. We also found within-region variation in ion levels, which is intriguing because earlier studies suggested that the composition of salt in streams was controlled by natural mechanisms (e.g., precipitation, rock dominance, and evaporation), which often cluster regionally (Griffith 2014; Olson & Cormier 2019). However, previous regional lake studies found heterogeneity in ion composition within study regions, which was attributed to natural processes and human activities (e.g., groundwater, soil texture, and acid deposition) (Baker et al. 1991; DeSellas et al. 2023). Our research finding that each NEON region included multiple ion clusters further supports the idea that major ions are influenced by multiple multi-scaled natural and human factors that vary both locally and regionally.

Multi-scaled natural and human factors related to salinity and major ions

The important factors related to salinity and ion composition differed by response variable. Salinity was related more to natural than human factors. This result suggests that natural factors (e.g., geology, hydrology, climate) may determine salt inputs from natural sources and that, in combination with the water balance in the lake, may control background salinity levels, whereas human disturbances (e.g., road salt applications; Solomon et al. 2023) that export ions to lakes, may cause salinity to deviate from those background levels and vary across lakes. However, human factors were as important as natural factors in determining major ion concentrations and compositions in lakes, particularly for Cl^- which were strongly and mostly influenced by human activities (Figure S5). Therefore, effective management strategies will vary depending on targeted response variables.

We found a variety of multi-scaled natural factors were important for understanding salinity and major ions. At the lake scale, elevation was negatively related to salinity, which is a pattern also reported by other regional studies (e.g., Müller et al. 1998; Borowiak et al. 2020) attributed to increasing rock weathering and dissolution rates (Müller et al. 1998), declined relative water inputs from rainfall, as well as greater urbanization and human salt inputs (Larned et al. 2004) with decreasing elevation. We anticipated that surface connectivity would be important for understanding differences in salinity and major ions because it measures the water and salt inputs from direct tributaries to the lake. Although surface connectivity did not stand out as one of the top factors in salinity models, it was related to lake membership in some ion clusters. For instance, 41% of lakes that only received inflows (i.e., terminal lakes) and 20% of lakes that had no inflow or outflow (i.e., isolated lakes) were the high-salinity lakes in cluster 8. Previous research found that these two types of lakes contain saline waters due to being salt sinks for the catchment, retention, and/or evaporation (Saleem et al. 2015; Cotner et al. 2022; Ding et al. 2024). Interestingly, drainage lakes did not show high proportions in the clusters characterized by low salinity (e.g., 10%, 1%, 4%, and 7% of drainage lakes were in clusters 7, 11, 4, and 2, respectively) compared with other clusters. This result implies that the direct surface water inflow of these low-salinity lakes contained low ion levels and, compared with direct tributaries, freshwater inputs from watersheds and precipitation had a more pronounced influence on major ions in these lakes (Bennett et al. 2007; Dumelle et al. 2024).

At the watershed scale, soil composition variables and measures of surface and groundwater were important in models of lake salinity and ion composition. These results make sense ecologically. For example, the percentage of clay is related to land use and affects ion movement through subsurface flow paths and release to waterbodies (Dugan 2024). The negative relationship between watershed groundwater recharge and salinity may indicate greater domination of hydrologic and chemical budget by inputs from precipitation and surface water streams (Webster et al. 2006) compared to the high-salinity groundwater inputs that can be a dominant source of salts to lakes in many regions (Li et al. 2020; Dugan 2024). Similarly, watershed runoff and regional precipitation were negatively related to salinity, suggesting they exert more control on salt budgets in lakes where precipitation inputs are high.

Lake salinity and major ions were related to multi-scaled human factors (and those that indicate the lack of disturbance), including watershed land use/cover, road density, and wet deposition of sulfate and NO_3^-. For example, forest land cover, which can be used to identify lakes with no or little human disturbances, was negatively related to salinity, and agricultural land use had a positive relationship with salinity. Moreover, one of the most crucial factors that affected ion composition was road density, which had strong positive relationships with clusters 9, 6, 1, and 10. These same clusters, which are located in regions where road salt applications happen (e.g., the Northeast, Mid-Atlantic, Great Lakes, Prairie Peninsula, and Pacific Southwest regions; Dugan et al. 2017), also had higher concentrations and proportions of Na^+ and Cl^- than other clusters. Therefore, this pattern is likely due to road deicing salts that were applied in winter and retained in lakes, causing increases in salt concentrations throughout the year (Kaushal et al. 2021). Combining our results with those of a long-term study that found 125 of 371 lakes in North America have increasing temporal trends of Cl^- concentrations (Dugan et al. 2017), demonstrates the importance of reducing road salt applications in these regions where there is high impervious surface area and road density.

Atmospheric deposition, which has a complex history in the US, is a regional-scale variable that can be important for understanding broad-scale variation in lake salinity and major ions. We found that the individual components of atmospheric deposition varied in their relationships with salinity and major ions. For example, deposited sulfate can replace HCO_3^- and carbonate in water while building up SO_4^{2-} concentration (Charles 1991; Shammas et al. 2020). Although we observed an overall negative relationship between wet sulfate deposition and salinity (Figure S6), some studies suggested more complex relationships and the existence of other influential factors such as ion inputs from rock weathering (Charles 1991; Kaushal et al. 2018). Deposited sulfate and NO_3^- also can influence surrounding soil pH and mobilize cations (e.g., Ca^{2+} and Mg^{2+}) within soil and into groundwater (Driscoll et al. 2001; Shammas et al. 2020), which can be then flushed into lakes by the rain or get into lakes through groundwater discharge, thus their impacts on lake salinity are also jointly affected by climate and hydrology. Our results presented complex impacts of atmospheric deposition on lake salinity and major ions, demonstrating the significance of it on lake salt contamination and the necessity of incorporating multi-scaled natural factors to better understand how deposition affects salts in lakes.

Our findings suggest that the effects of soil composition, groundwater, and runoff on salinity can be modulated by forest land cover, atmospheric deposition, and urban development. Although our RF model considered the effects of all factors, we only examined the joint modeled effects of selected natural and human factors. There are likely more complex interactive effects that are beyond the scope of this study. Very few studies have looked at the combined effects of factors on other water quality parameters (Kernan & Helliwell 2001; Nobre et al. 2020; Lin et al. 2021), and no or limited information exists on conductivity and major ions. However, our results and previous studies demonstrate that human and natural factors can jointly affect lake salinity and water quality, thus, future research efforts should consider this area to disentangle the complex underlying mechanisms affecting ion concentrations and compositions in lakes.

Conclusion

We documented spatial variation in salinity and major ion concentration and composition in 9,785 and 1,218 lakes, respectively, across the CONUS. We found that ion composition varied substantially and that similar salinity levels can be caused by different ion compositions, which have different effects on lake biota, may change differently in the future, and require different strategies to manage. These findings highlight the importance of considering all major ions when studying lake salinity. At a continental scale, we also found that variation in salinity was related to a wide range of local, watershed, and regional factors in lakes, such as lake elevation, soil texture, hydrology, precipitation, agricultural land use, and atmospheric deposition. Major ions were strongly associated with both human and natural factors. In particular, elevated Cl^- levels were predominantly related to road density and urban development, which can be managed to address issues such as freshwater salinization. Our results suggest that while geology, combined with hydrology, surface connectivity, and climate, controls the salt inputs from natural sources and pathways of water flowing into the lake, human disturbances can export ions to lakes, causing salinity and major ions to deviate from their background levels.

Conflict of interest: The authors declare no conflict of interest.

Acknowledgment: This work was supported by the US National Science Foundation (NSF) Macrosystems Biology & NEON-Enabled Science Program (DEB #1638679) and the United States Department of Agriculture (USDA) National Institute of Food and Agriculture, Hatch project 1013544.

References:

Baker, L. A., Eilers, J. M., Cook, R. B., Kaufmann, P. R., & Herlihy, A. T. (1991). Interregional Comparisons of Surface Water Chemistry and Biogeochemical Processes. In D. F. Charles (Ed.), *Acidic Deposition and Aquatic Ecosystems* (pp. 567–613). Springer New York. https://doi.org/10.1007/978-1-4613-9038-1_23

Basu, S., Kumbier, K., Brown, J. B., & Yu, B. (2018). Iterative random forests to discover predictive and stable high-order interactions. *Proceedings of the National Academy of Sciences*, *115*(8), 1943–1948. https://doi.org/10.1073/pnas.1711236115

Bennett, D. M., Fritz, S. C., Holz, J. C., Holz, A. A., & Zlotnik, V. A. (2007). Evaluating climatic and non-climatic influences on ion chemistry in natural and man-made lakes of

Nebraska, USA. *Hydrobiologia*, *591*(1), 103–115. https://doi.org/10.1007/s10750-007-0798-z

Blodgett, D. L. (2023). *Twelve-digit hydrologic unit soil moisture, recharge, actual evapotranspiration, and snowpack water equivalent storage from the National Hydrologic Model Infrastructure with the Precipitation-Runoff Modeling System 1980-2016*. https://doi.org/10.5066/P9W148A1

Borowiak, M., Borowiak, D., & Nowiński, K. (2020). Spatial Differentiation and Multiannual Dynamics of Water Conductivity in Lakes of the Suwałki Landscape Park. *Water*, *12*(5), 1277. https://doi.org/10.3390/w12051277

Calver, M., Lymbery, A., McComb, J., & Bamford, M. (Eds.). (2009). *Environmental Biology* (1st ed.). Cambridge University Press. https://doi.org/10.1017/CBO9780511841569

Cañedo-Argüelles, M., Hawkins, C. P., Kefford, B. J., Schäfer, R. B., Dyack, B. J., Brucet, S., Buchwalter, D., Dunlop, J., Frör, O., Lazorchak, J., Coring, E., Fernandez, H. R., Goodfellow, W., Achem, A. L. G., Hatfield-Dodds, S., Karimov, B. K., Mensah, P., Olson, J. R., Piscart, C., … Timpano, A. J. (2016). Saving freshwater from salts. *Science*, *351*(6276), 914–916. https://doi.org/10.1126/science.aad3488

Charles, D. F. (Ed.). (1991). *Acidic Deposition and Aquatic Ecosystems: Regional Case Studies*. Springer New York. https://doi.org/10.1007/978-1-4613-9038-1

Cheruvelil, K. S., Soranno, P. A., McCullough, I. M., Webster, K. E., Rodriguez, L. K., & Smith, N. J. (2021). LAGOS-US LOCUS v1.0: Data module of location, identifiers, and physical characteristics of lakes and their watersheds in the conterminous U.S. *Limnology and Oceanography Letters*, *6*(5), 270–292. https://doi.org/10.1002/lol2.10203

Clements, W. H., & Kotalik, C. (2016). Effects of major ions on natural benthic communities: an experimental assessment of the US Environmental Protection Agency aquatic life benchmark for conductivity. *Freshwater Science*, *35*(1), 126–138. https://doi.org/10.1086/685085

Cotner, J. B., Anderson, N. J., & Osburn, C. (2022). Accumulation of recalcitrant dissolved organic matter in aerobic aquatic systems. *Limnology and Oceanography Letters*, *7*(5), 401–409. https://doi.org/10.1002/lol2.10265

Cutler, F. original by L. B. and A., & Wiener, R. port by A. L. and M. (2022). *randomForest: Breiman and Cutler's Random Forests for Classification and Regression* (4.7-1.1). https://cran.r-project.org/web/packages/randomForest/index.html

DeSellas, A. M., Paterson, A. M., Rühland, K. M., & Smol, J. P. (2023). Lake water chemistry and its relationship to shoreline residential development and natural landscape features in Algonquin Provincial Park, Ontario, Canada. *Canadian Journal of Fisheries and Aquatic Sciences*, cjfas-2023-0103. https://doi.org/10.1139/cjfas-2023-0103

DeVilbiss, S. E., Steele, M. K., Brown, B. L., & Badgley, B. D. (2022). Stream bacterial diversity peaks at intermediate freshwater salinity and varies by salt type. *Science of The Total Environment*, *840*, 156690. https://doi.org/10.1016/j.scitotenv.2022.156690

Ding, M., Wang, J., Song, C., Sheng, Y., Shawn Hutchinson, J. M., Langston, A. L., & Marston, L. (2024). A framework of freshwater and saline lake typology classification through leveraging hydroclimate, spectral, and literature evidence. *Journal of Hydrology*, *632*, 130704. https://doi.org/10.1016/j.jhydrol.2024.130704

Driscoll, C. T., Lawrence, G. B., Bulger, A. J., Butler, T. J., Cronan, C. S., Eagar, C., Lambert, K. F., Likens, G. E., Stoddard, J. L., & Weathers, K. C. (2001). Acidic Deposition in the Northeastern United States: Sources and Inputs, Ecosystem Effects, and Management

Strategies. *BioScience*, *51*(3), 180. https://doi.org/10.1641/0006-3568(2001)051[0180:ADITNU]2.0.CO;2

Dugan, H. A. (2024). Chapter 12 - Salinity and Ionic Composition of Inland Waters. In I. D. Jones & J. P. Smol (Eds.), *Wetzel's Limnology (Fourth Edition)* (pp. 275–299). Academic Press. https://doi.org/10.1016/B978-0-12-822701-5.00012-4

Dugan, H. A., Bartlett, S. L., Burke, S. M., Doubek, J. P., Krivak-Tetley, F. E., Skaff, N. K., Summers, J. C., Farrell, K. J., McCullough, I. M., Morales-Williams, A. M., Roberts, D. C., Ouyang, Z., Scordo, F., Hanson, P. C., & Weathers, K. C. (2017). Salting our freshwater lakes. *Proceedings of the National Academy of Sciences*, *114*(17), 4453–4458. https://doi.org/10.1073/pnas.1620211114

Dumelle, M., Ver Hoef, J. M., Handler, A., Hill, R. A., Higham, M., & Olsen, A. R. (2024). Modeling lake conductivity in the contiguous United States using spatial indexing for big spatial data. *Spatial Statistics*, *59*, 100808. https://doi.org/10.1016/j.spasta.2023.100808

Elphick, J. R. F., Bergh, K. D., & Bailey, H. C. (2011). Chronic toxicity of chloride to freshwater species: Effects of hardness and implications for water quality guidelines. *Environmental Toxicology and Chemistry*, *30*(1), 239–246. https://doi.org/10.1002/etc.365

EnviroAtlas. (2015). *Manure application (kg N/ha/yr) -- [ManureMean]*. EnviroAtlas. https://catalog.data.gov/dataset/enviroatlas-manure-application-to-agricultural-lands-from-confined-animal-feeding-operatio-2006

Friedman, J., Hastie, T., Tibshirani, R., Narasimhan, B., Tay, K., Simon, N., Qian, J., & Yang, J. (2023). *glmnet: Lasso and Elastic-Net Regularized Generalized Linear Models* (4.1-8). https://cran.r-project.org/web/packages/glmnet/index.html

Gibbs, R. J. (1970). *Mechanisms controlling world water chemistry*. https://www.jstor.org/stable/1730827

Greenwell, B. M. (2022). *pdp: Partial Dependence Plots* (0.8.1). https://cran.r-project.org/web/packages/pdp/index.html

Griffith, M. B. (2014). Natural variation and current reference for specific conductivity and major ions in wadeable streams of the conterminous USA. *Freshwater Science*, *33*(1), 1–17. https://doi.org/10.1086/674704

Härdle, W. K., & Simar, L. (2019). *Applied Multivariate Statistical Analysis*. Springer International Publishing. https://doi.org/10.1007/978-3-030-26006-4

Hargrove, W. W., & Hoffman, F. M. (1999). Using multivariate clustering to characterize ecoregion borders. *Computing in Science & Engineering*, *1*(4), 18–25. https://doi.org/10.1109/5992.774837

Hintz, W. D., & Relyea, R. A. (2019). A review of the species, community, and ecosystem impacts of road salt salinisation in fresh waters. *Freshwater Biology*, *64*(6), 1081–1097. https://doi.org/10.1111/fwb.13286

Huber, E. D., Hintz, L. L., Wilmoth, B., McKenna, J. R., & Hintz, W. D. (2024). Coping with stress: Salt type, concentration, and exposure history limit life history tradeoffs in response to road salt salinization. *Science of The Total Environment*, *949*, 174998. https://doi.org/10.1016/j.scitotenv.2024.174998

Kahlert, M., & Gottschalk, S. (2014). Differences in benthic diatom assemblages between streams and lakes in Sweden and implications for ecological assessment. *Freshwater Science*, *33*(2), 655–669. https://doi.org/10.1086/675727

Kaushal, S. S., Likens, G. E., Pace, M. L., Haq, S., Wood, K. L., Galella, J. G., Morel, C., Doody, T. R., Wessel, B., Kortelainen, P., Räike, A., Skinner, V., Utz, R., & Jaworski, N. (2019). Novel ‘chemical cocktails’ in inland waters are a consequence of the freshwater salinization syndrome. *Philosophical Transactions of the Royal Society B: Biological Sciences*, *374*(1764), 20180017. https://doi.org/10.1098/rstb.2018.0017

Kaushal, S. S., Likens, G. E., Pace, M. L., Reimer, J. E., Maas, C. M., Galella, J. G., Utz, R. M., Duan, S., Kryger, J. R., Yaculak, A. M., Boger, W. L., Bailey, N. W., Haq, S., Wood, K. L., Wessel, B. M., Park, C. E., Collison, D. C., Aisin, B. Y. 'aaqob I., Gedeon, T. M., … Woglo, S. A. (2021). Freshwater salinization syndrome: from emerging global problem to managing risks. *Biogeochemistry*, *154*(2), 255–292. https://doi.org/10.1007/s10533-021-00784-w

Kaushal, S. S., Likens, G. E., Pace, M. L., Utz, R. M., Haq, S., Gorman, J., & Grese, M. (2018). Freshwater salinization syndrome on a continental scale. *Proceedings of the National Academy of Sciences*, *115*(4). https://doi.org/10.1073/pnas.1711234115

Keller, M., Schimel, D. S., Hargrove, W. W., & Hoffman, F. M. (2008). A continental strategy for the National Ecological Observatory Network. *Frontiers in Ecology and the Environment*, *6*(5), 282–284. https://doi.org/10.1890/1540-9295(2008)6[282:ACSFTN]2.0.CO;2

Kernan, M. R., & Helliwell, R. C. (2001). Partitioning the variation within the acid neutralizing capacity of surface waters in Scotland in relation to land cover, soil and atmospheric depositional factors. *Science of The Total Environment*, *265*(1–3), 39–49. https://doi.org/10.1016/S0048-9697(00)00648-3

Kursa, M. B., & Rudnicki, W. R. (2022). *Boruta: Wrapper Algorithm for All Relevant Feature Selection* (8.0.0). https://cran.r-project.org/web/packages/Boruta/index.html

La Baugh, J. W., Winter, T. C., & Rosenberry, D. O. (2000). Comparison of the variability in fluxes of ground water and solutes in lakes and wetlands in central North America. *SIL Proceedings, 1922-2010*, *27*(1), 420–426. https://doi.org/10.1080/03680770.1998.11901266

Larned, S. T., Scarsbrook, M. R., Snelder, T. H., Norton, N. J., & Biggs, B. J. F. (2004). Water quality in low-elevation streams and rivers of New Zealand: Recent state and trends in contrasting land-cover classes. *New Zealand Journal of Marine and Freshwater Research*, *38*(2), 347–366. https://doi.org/10.1080/00288330.2004.9517243

Li, C., Gao, X., Li, S., & Bundschuh, J. (2020). A review of the distribution, sources, genesis, and environmental concerns of salinity in groundwater. *Environmental Science and Pollution Research*, *27*(33), 41157–41174. https://doi.org/10.1007/s11356-020-10354-6

Lin, J., Compton, J. E., Hill, R. A., Herlihy, A. T., Sabo, R. D., Brooks, J. R., Weber, M., Pickard, B., Paulsen, S. G., & Stoddard, J. L. (2021). Context is Everything: Interacting Inputs and Landscape Characteristics Control Stream Nitrogen. *Environmental Science & Technology*, *55*(12), 7890–7899. https://doi.org/10.1021/acs.est.0c07102

Lottig, N. R., Stanley, E. H., Hanson, P. C., & Kratz, T. K. (2011). Comparison of regional stream and lake chemistry: Differences, similarities, and potential drivers. *Limnology and Oceanography*, *56*(5), 1551–1562. https://doi.org/10.4319/lo.2011.56.5.1551

Martin, S. L., & Soranno, P. A. (2006). Lake landscape position: Relationships to hydrologic connectivity and landscape features. *Limnology and Oceanography*, *51*(2), 801–814. https://doi.org/10.4319/lo.2006.51.2.0801

Mount, D. R., Erickson, R. J., Highland, T. L., Hockett, J. R., Hoff, D. J., Jenson, C. T., Norberg-King, T. J., Peterson, K. N., Polaske, Z. M., & Wisniewski, S. (2016). The acute toxicity of major ion salts to *Ceriodaphnia dubia* : I. influence of background water chemistry. *Environmental Toxicology and Chemistry*, *35*(12), 3039–3057. https://doi.org/10.1002/etc.3487

Müller, B., Lotter, A. F., Sturm, M., & Ammann, A. (1998). Influence of catchment quality and altitude on the water and sediment composition of 68 small lakes in Central Europe. *Aquatic Sciences*, *60*(4), 316. https://doi.org/10.1007/s000270050044

Nobre, R. L. G., Caliman, A., Cabral, C. R., Araújo, F. D. C., Guérin, J., Dantas, F. D. C. C., Quesado, L. B., Venticinque, E. M., Guariento, R. D., Amado, A. M., Kelly, P., Vanni, M. J., & Carneiro, L. S. (2020). Precipitation, landscape properties and land use interactively affect water quality of tropical freshwaters. *Science of The Total Environment*, *716*, 137044. https://doi.org/10.1016/j.scitotenv.2020.137044

Nostro, P. L., Ninham, B. W., Nostro, A. L., Pesavento, G., Fratoni, L., & Baglioni, P. (2005). Specific ion effects on the growth rates of *Staphylococcus aureus* and *Pseudomonas aeruginosa*. *Physical Biology*, *2*(1), 1–7. https://doi.org/10.1088/1478-3967/2/1/001

Olson, J. R. (2019). Predicting combined effects of land use and climate change on river and stream salinity. *Philosophical Transactions of the Royal Society B: Biological Sciences*, *374*(1764), 20180005. https://doi.org/10.1098/rstb.2018.0005

Olson, J. R., & Cormier, S. M. (2019). Modeling spatial and temporal variation in natural background specific conductivity. *Environmental Science & Technology*, *53*(8), 4316–4325. https://doi.org/10.1021/acs.est.8b06777

Oswald, C. J., Giberson, G., Nicholls, E., Wellen, C., & Oni, S. (2019). Spatial distribution and extent of urban land cover control watershed-scale chloride retention. *Science of The Total Environment*, *652*, 278–288. https://doi.org/10.1016/j.scitotenv.2018.10.242

Polus, S. M., Hanly, P. J., Rodriguez, L. K., Wang, Q., Díaz Vázquez, J., Webster, K. E., Tan, P.-N., Zhou, J., Danila, L., Soranno, P. A., & Cheruvelil, K. S. (2022). *LAGOS-US RESERVOIR: Data module classifying conterminous U.S. lakes 4 hectares and larger as natural lakes or reservoirs*. Environmental Data Initiative. https://doi.org/10.6073/PASTA/F9AA935329A95DFD69BF895015BC5161

R Core Team. (2024). *R: A language and environment for statistical computing.* R Foundation for Statistical Computing, Vienna, Austria. https://www.R-project.org/

Rodriguez, L. K., Polus, S. M., Matuszak, D. I., Domka, M. R., Hanly, P. J., Wang, Q., Soranno, P. A., & Cheruvelil, K. S. (2023). LAGOS-US RESERVOIR : A database classifying conterminous U.S. lakes 4 ha and larger as natural lakes or reservoir lakes. *Limnology and Oceanography Letters*, *8*(2), 267–285. https://doi.org/10.1002/lol2.10299

Rogalski, M. A., Baker, E. S., Benadon, C. M., Tatgenhorst, C., & Nichols, B. R. (2024). Lake water chemistry and local adaptation shape NACL toxicity in *Daphnia ambigua*. *Evolutionary Applications*, *17*(3), e13668. https://doi.org/10.1111/eva.13668

Saleem, M., Jeelani, G., & Shah, R. A. (2015). Hydrogeochemistry of Dal Lake and the potential for present, future management by using facies, ionic ratios, and statistical analysis. *Environmental Earth Sciences*, *74*(4), 3301–3313. https://doi.org/10.1007/s12665-015-4361-3

Schacht, J. R. S., MacKeigan, P. W., Taranu, Z. E., Huot, Y., & Gregory-Eaves, I. (2023). Agricultural land use and morphometry explain substantial variation in nutrient and ion concentrations in lakes across Canada. *Canadian Journal of Fisheries and Aquatic Sciences*, *80*(11), 1785–1797. https://doi.org/10.1139/cjfas-2023-0109

Shammas, N. K., Wang, L. K., & Wang, M.-H. S. (2020). Sources, Chemistry and Control of Acid Rain in the Environment. In Y.-T. Hung, L. K. Wang, & N. K. Shammas, *Handbook of Environment and Waste Management* (Vol. 03, pp. 1–26). WORLD SCIENTIFIC. https://doi.org/10.1142/9789811207136_0001

Shuvo, A. K., Lottig, N. R., Webster, K. E., Delany, A., Reinl, K., Gries, C., Smith, N. J., Poisson, A. C., McCullough, I. M., Collins, S. M., King, K. B. S., Phillips, E., Cheruvelil, K. S., & Soranno, P. A. (2023). *LAGOS-US LIMNO: Data module of surface water chemistry from 1975-2021 for lakes in the conterminous U.S.* Environmental Data Initiative. https://doi.org/10.6073/PASTA/2C58F5A50AB813919F99CC1F265F271C

Smith, N. J., Webster, K. E., Rodriguez, L. K., Cheruvelil, K. S., & Soranno, P. A. (2021). *LAGOS-US LOCUS v1.0: Data module of location, identifiers, and physical characteristics of lakes and their watersheds in the conterminous U.S.* Environmental Data Initiative. https://doi.org/10.6073/PASTA/E5C2FB8D77467D3F03DE4667AC2173CA

Smith, N. J., Webster, K. E., Rodriguez, L. K., Cheruvelil, K. S., & Soranno, P. A. (2022). *LAGOS-US GEO v1.0: Data module of lake geospatial ecological context at multiple spatial and temporal scales in the conterminous U.S.* Environmental Data Initiative. https://doi.org/10.6073/pasta/0e443bd43d7e24c2b6abc7af54ca424a

Solomon, C. T., Dugan, H. A., Hintz, W. D., & Jones, S. E. (2023). Upper limits for road salt pollution in lakes. *Limnology and Oceanography Letters*, *8*(6), 859–866. https://doi.org/10.1002/lol2.10339

Soranno, P. A., Lottig, N. R., Delany, A. D., & Cheruvelil, K. S. (2019). *LAGOS-NE-LIMNO v1.087.3: A module for LAGOS-NE, a multi-scaled geospatial and temporal database of lake ecological context and water quality for thousands of U.S. Lakes: 1925-2013*. Environmental Data Initiative. https://doi.org/10.6073/PASTA/08C6F9311929F4874B01BCC64EB3B2D7

Soranno, P. A., Webster, K. E., Riera, J. L., Kratz, T. K., Baron, J. S., Bukaveckas, P. A., Kling, G. W., White, D. S., Caine, N., Lathrop, R. C., & Leavitt, P. R. (1999). Spatial Variation among Lakes within Landscapes: Ecological Organization along Lake Chains. *Ecosystems*, *2*(5), 395–410. https://doi.org/10.1007/s100219900089

Stanley, E. H., Collins, S. M., Lottig, N. R., Oliver, S. K., Webster, K. E., Cheruvelil, K. S., & Soranno, P. A. (2019). Biases in lake water quality sampling and implications for macroscale research. *Limnology and Oceanography*, *64*(4), 1572–1585. https://doi.org/10.1002/lno.11136

Stets, E. G., Sprague, L. A., Oelsner, G. P., Johnson, H. M., Murphy, J. C., Ryberg, K., Vecchia, A. V., Zuellig, R. E., Falcone, J. A., & Riskin, M. L. (2020). Landscape Drivers of Dynamic Change in Water Quality of U.S. Rivers. *Environmental Science & Technology*, *54*(7), 4336–4343. https://doi.org/10.1021/acs.est.9b05344

Swanson, G. A., Winter, T. C., Adomaitis, V. A., & La Baugh, J. W. (1988). *Chemical characteristics of prairie lakes in south-central North Dakota: Their potential for influencing use by fish and wildlife*. U.S. Department of the Interior, Fish and Wildlife Service.

Tiri, A., Belkhiri, L., & Mouni, L. (2018). Evaluation of surface water quality for drinking purposes using fuzzy inference system. *Groundwater for Sustainable Development*, *6*, 235–244. https://doi.org/10.1016/j.gsd.2018.01.006

U.S. Environmental Protection Agency. (2010). *National Aquatic Resource Surveys. National Lakes Assessment 2007*. Available from U.S. EPA website: http://www.epa.gov/national-aquatic-resource-surveys/data-national-aquatic-resource-surveys.

U.S. Environmental Protection Agency. (2016). *National Aquatic Resource Surveys. National Lakes Assessment 2012*. Available from U.S. EPA website: http://www.epa.gov/national-aquatic-resource-surveys/data-national-aquatic-resource-surveys.

U.S. Environmental Protection Agency. (2022). *National Lakes Assessment 2017 Datafiles for Report "National Lakes Assessment 2017: The third collaborative survey of lakes in the United States"*. U.S. Environmental Protection Agency. https://doi.org/10.23719/1529585

Van Gray, J. B., & Ayayee, P. (2024). Examining the impacts of salt specificity on freshwater microbial community and functional potential following salinization. *Environmental Microbiology*, *26*(5), e16628. https://doi.org/10.1111/1462-2920.16628

Water Quality Portal. (2021). *Washington (DC): National Water Quality Monitoring Council, United States Geological Survey (USGS), Environmental Protection Agency (EPA)*. https://doi.org/10.5066/P9QRKUVJ

Webster, K. E., Bowser, C. J., Anderson, M. P., & Lenters, J. D. (2006). Understanding the lake-groundwater system: Just follow the water. In *Long-term Dynamics of Lakes in the Landscape: Long-term Ecological Research on North Temperate Lakes* (pp. 19–48). Oxford University Press.

Zaman, M., Shahid, S. A., & Heng, L. (2018). Irrigation Water Quality. In M. Zaman, S. A. Shahid, & L. Heng, *Guideline for Salinity Assessment, Mitigation and Adaptation Using Nuclear and Related Techniques* (pp. 113–131). Springer International Publishing. https://doi.org/10.1007/978-3-319-96190-3_5

Supplementary Tables

Paper title: Natural and human drivers of salinity and major ion composition in United States lakes

Authors: Xinyu Sun, Kendra Spence Cheruvelil, Patrick J. Hanly, Katherine E. Webster, & Patricia A. Soranno

Table S1. Table of natural and human factors with variable sources, spatial scale, group, explanation, and data transformation information.

Factors_name_ in_figure	Factors_original_ name	Spatial scale of measurement	Group	Natural_ Human	Data sources	Explanation	Transformation in RF	Transformation in GLMNET
connectivity	lake_connectivity _class	lake	surface connectivity	N	LAGOS-US LOCUS	Hydrologic connectivity class of the focal lake	NA	NA
elevation	lake_elevation_m	lake	location	N	LAGOS-US LOCUS	Mean elevation in the zone	natural log	natural log
latitude	lake_lat_decdeg	lake	location	N	LAGOS-US LOCUS	Latitude of the lake polygon centroid	NA	NA
longitude	lake_lon_decdeg	lake	location	N	LAGOS-US LOCUS	Longitude of the lake polygon centroid	NA	NA
reservoir	lake_rsvr_class	lake	lake and watershed	N	LAGOS-US RESERVOIR	The classification of a lake into either natural lake or reservoir	NA	NA
shoreline dev factor	lake_shorelinedev factor	lake	lake and watershed	N	LAGOS-US LOCUS	A measure of the deviation of lake shape from 1, which is a perfect circle, calculated from the lake water area and perimeter	natural log	natural log
lake area	lake_waterarea_ha	lake	lake and watershed	N	LAGOS-US LOCUS	The water area of the lake contained within the outer shoreline	natural log	natural log
wet dep inorg N current	NA, calculated, kg/ha	HU12	atmospheric deposition	H	LAGOS-US GEO	Wet inorganic nitrogen deposition since year 2000 measured in 12-digit	NA	NA

						hydrological unit area (local sub-watershed level)		
wet dep inorg N diff	NA, calculated, kg/ha	HU12	atmospheric deposition	H	LAGOS-US GEO	Percentage decrease in wet inorganic nitrogen deposition after year 2000 compared with before year 2000 (1985-2000)	NA	NA
wet dep no3 current	NA, calculated, kg/ha	HU12	atmospheric deposition	H	LAGOS-US GEO	Wet nitrate deposition since year 2000	NA	NA
wet dep no3 diff	NA, calculated, kg/ha	HU12	atmospheric deposition	H	LAGOS-US GEO	Percentage decrease in wet nitrate deposition after year 2000 compared with before year 2000 (1985-2000)	NA	NA
wet dep so4 diff	NA, calculated, kg/ha	HU12	atmospheric deposition	H	LAGOS-US GEO	Percentage decrease in wet sulfate deposition after year 2000 compared with before year 2000 (1985-2000)	NA	NA
wet dep so4 current	NA, calculated, kg/ha	HU12	atmospheric deposition	H	LAGOS-US GEO	Wet sulfate deposition since year 2000	NA	NA
manure	ManureMean	HU12	human activity	H	EnviroAtlas	Manure application (kg N/ha/yr)	natural log	natural log
evapotranspiration	NA, calculated	HU12	climate	N	Blodgett 2023	Long-term average evapotranspiration data by HU12 region	NA	NA
snowpack	NA, calculated	HU12	climate	N	Blodgett 2024	Long-term average snowpack water equivalent storage data by HU12 region	NA	NA
NEON region	neon_zoneid	NEON region	location	N	LAGOS-US LOCUS	National Ecological Observatory Network (NEON) region id	NA	NA
precip long-term	NA, calculated	HU12	climate	N	LAGOS-US GEO	Long-term average precipitation data (5 years before sampling year)	NA	NA
roads 500m buff	road.500buff	500m buffer	human activity	H	LAGOS-US GEO	Road density in 500 m buffer around the shoreline of a LAGOS US lake polygon	natural log	natural log
temperature long-term	NA, calculated	HU12	climate	N	LAGOS-US GEO	Long-term average air temperature data (5 years before sampling year)	NA	NA
baseflow index	value.baseflowindex_pct	HU12	hydrology	N	LAGOS-US GEO	Mean within the zone of the percentage of streamflow that can be attributed to groundwater discharge into streams, calculated as baseflow divided by total flow using data from	logit, generalized (-0.01 to 1.01)	NA

						1951-1980		
groundwater recharge	value.groundwaterrecharge_mmperyr	HU12	hydrology	N	LAGOS-US GEO	Mean within the zone of annual groundwater recharge calculated as baseflow multiplied by mean annual runoff using data from 1951-1980	natural log	natural log
alluvium coastal fine	value.lith_alluviumcoastalfine_pct	HU12	lithology	N	LAGOS-US GEO	Percent of zone with lithology classified as alluvium and fine textured coastal zone sediment	logit, generalized (-0.01 to 1.01)	NA
carbonate residual	value.lith_carbonateresid_pct	HU12	lithology	N	LAGOS-US GEO	Percent of zone with lithology classified as carbonate residual material	logit, generalized (-0.01 to 1.01)	NA
coastal zone coarse	value.lith_coastalzonecoarse_pct	HU12	lithology	N	LAGOS-US GEO	Percent of zone with lithology classified as coastal zone sediment, coarse textured	logit, generalized (-0.01 to 1.01)	NA
glacial outwash coarse	value.lith_glacialoutwashcoarse_pct	HU12	lithology	N	LAGOS-US GEO	Percent of zone with lithology classified as glacial outwash and glacial lake sediment, coarse textured	logit, generalized (-0.01 to 1.01)	NA
glacial till clayey	value.lith_glacialtillclayey_pct	HU12	lithology	N	LAGOS-US GEO	Percent of zone with lithology classified as glacial till, clayey	logit, generalized (-0.01 to 1.01)	NA
glacial till coarse	value.lith_glacialtillcoarse_pct	HU12	lithology	N	LAGOS-US GEO	Percent of zone with lithology classified as glacial till, coarse textured	logit, generalized (-0.01 to 1.01)	NA
saline lake sediment	value.lith_salinelake_pct	HU12	lithology	N	LAGOS-US GEO	Percent of zone with lithology classified as saline lake sediment	logit, generalized (-0.01 to 1.01)	NA
mines heavy metal	value.mines_heavymetal_n	HU12	human activity	H	LAGOS-US GEO	Count of heavy metal mines active in 2003 within the zone	natural log	natural log
CAFO	value.npdes_cafo_n	HU12	human activity	H	LAGOS-US GEO	Count of concentrated animal feed operations within the zone	natural log	natural log
major discharge sites	value.npdes_majordischarge_n	HU12	human activity	H	LAGOS-US GEO	Count of facilities with major discharges within the zone	natural log	natural log
stormwater from industrial	value.npdes_stormwaterindustrial_n	HU12	human activity	H	LAGOS-US GEO	Count of industrial storm water sewers within the zone	natural log	natural log
stormwater from municipal	value.npdes_stormwatermunicipal_n	HU12	human activity	H	LAGOS-US GEO	Count of Phase I MS4: Municipal separate storm sewer systems within the zone	natural log	natural log
road density HU12	value.roads_mperha	HU12	human activity	H	LAGOS-US GEO	Road density within each HU12 region	natural log	natural log

runoff	value.runoff_inperyr	HU12	terrain	N	LAGOS-US GEO	Mean within the zone of annual runoff; data from 1951 to 1980	natural log	natural log
sat overland flow	value.satoverlandflow_pct	HU12	terrain	N	LAGOS-US GEO	Mean within the zone of the average percentage of saturation overland flow in total streamflow using data from 1951-1980	logit, generalized (-0.01 to 1.01)	NA
clay	value.soil_clay_pct	HU12	soil	N	LAGOS-US GEO	Average percentage mass fraction of clay, 0 to 2 micrometers, in the 0 to 5 cm depth soil layer within the zone	logit, generalized (-0.01 to 1.01)	NA
coarse	value.soil_coarse_pct	HU12	soil	N	LAGOS-US GEO	Average percentage by volume of coarse fragments in the 0 to 5 cm soil depth layer within the zone	logit, generalized (-0.01 to 1.01)	NA
depth to bedrock	value.soil_depthtobedrock_cm	HU12	soil	N	LAGOS-US GEO	Average absolute depth to bedrock within the zone	natural log	natural log
k factor	value.soil_kffact	HU12	soil	N	LAGOS-US GEO	Average soil erodibility factor, not adjusted for the effect of rock fragments, within the zone	natural log	natural log
organic carbon	value.soil_orgcarbon_gperkg	HU12	soil	N	LAGOS-US GEO	Average organic carbon content, fine earth fraction, in the 0 to 5 cm soil layer within the zone	natural log	natural log
sand	value.soil_sand_pct	HU12	soil	N	LAGOS-US GEO	Average percentage mass fraction of sand, 50 to 200 micrometers, in the 0 to 5 cm depth soil layer within the zone	logit, generalized (-0.01 to 1.01)	NA
silt	value.soil_silt_pct	HU12	soil	N	LAGOS-US GEO	Average percentage mass fraction of silt, 2 to 50 micrometers, in the 0 to 5 cm depth soil layer within the zone	logit, generalized (-0.01 to 1.01)	NA
streams density	value.streams_all_mperha	HU12	hydrology	N	LAGOS-US GEO	Density of all streams within the zone, calculated as the sum of the stream length divided by the zone area	natural log	natural log
topographic wetness	value.topographicwetness	HU12	terrain	N	LAGOS-US GEO	Mean topographic wetness index of cells within the zone	natural log	natural log
watershed area	ws_area_ha	watershed	lake and watershed	N	LAGOS-US LOCUS	Area of zone polygon	natural log	natural log
watershed:lake area ratio	ws_lake_arearatio	lake	lake and watershed	N	LAGOS-US LOCUS	Ratio between watershed area and lake water area	natural log	natural log

agriculture	NA, combined nlcd_cultcrop82_pct with nlcd_past81_pct	watershed	LU/LC	H	LAGOS-US GEO	Percent of zone classified as cultivated crops or pasture and hay using 2006 data	logit, generalized (-0.01 to 1.01)	NA
grass	nlcd_grass71_pct	watershed	LU/LC	H	LAGOS-US GEO	Percent of zone classified as grassland or herbaceous using 2006 data	logit, generalized (-0.01 to 1.01)	NA
shrub	nlcd_shrub52_pct	watershed	LU/LC	H	LAGOS-US GEO	Percent of zone classified as shrub and scrub using 2006 data	logit, generalized (-0.01 to 1.01)	NA
developed	NA, combined nlcd_devopen21_pct, nlcd_devlow22_pct, nlcd_devmed23_pct, and nlcd_devhi24_pct	watershed	LU/LC	H	LAGOS-US GEO	Percent of zone classified as developed, including open space and low, medium, and high intensity, using 2006 data	logit, generalized (-0.01 to 1.01)	NA
forest	NA, combined nlcd_forcon42_pct, nlcd_fordec41_pct, and nlcd_formix43_pct	watershed	LU/LC	H	LAGOS-US GEO	Percent of zone classified as evergreen forest, deciduous forest, or mixed forest using 2006 data	logit, generalized (-0.01 to 1.01)	NA
wetland	NA, combined nlcd_wetemerg95_pct with nlcd_wetwood90_pct	watershed	LU/LC	H	LAGOS-US GEO	Percent of zone classified as emergent herbaceous or woody wetlands using 2006 data	logit, generalized (-0.01 to 1.01)	NA

Table S2. Convert factors used for converting ion concentrations from mg/L to ueq/L. Convert factors were extracted from Hem (1985).

Ions	Convert factor (from mg/L to ueq/L)
Ca	49.9
Mg	82.29
Na	43.5
K	25.58
Cl	28.21
SO4	20.82
Alkalinity (as CaCO3)	19.98

Hem, J.D. 1985. Study and Interpretation of the Chemical Characteristics of Natural Water. U.S. Geological Survey, Water Supply Paper 2254. Retrieved from https://pubs.usgs.gov/wsp/wsp2254/html/pdf.html

Table S3. Multinomial coefficients from a GLMNET model predicting ion composition cluster assignment. C1-C15 indicate ion clusters. Group indicates natural and human factor category.

Factors	Group	Natural_ Human	C1	C10	C11	C12	C13	C14	C15	C2	C3	C4	C5	C6	C7	C8	C9
evapotranspiration	climate	N	0.06	0.03	0.12	0.06	0.17	0.05	-0.04	0.10	-0.05	0.00	0.09	-0.26	0.23	-0.09	-0.47
snowpack	climate	N	-0.05	-0.06	-0.05	-0.04	0.16	-0.11	0.12	0.06	-0.09	0.07	0.06	-0.01	0.03	-0.04	-0.07
precip long-term	climate	N	0.47	-0.05	0.72	-0.05	0.02	0.04	0.59	-0.46	-0.23	0.47	-0.33	-0.59	0.59	-0.58	-0.61
temperature long-term	climate	N	0.00	0.00	0.02	-0.09	-0.07	-0.01	0.04	-0.13	0.09	-0.08	0.00	0.15	0.01	-0.11	0.17
baseflow index	hydrology	N	-0.07	-0.19	0.35	0.18	0.14	-0.07	0.30	-0.46	0.33	-0.07	-0.31	0.00	0.03	-0.34	0.16
groundwater recharge	hydrology	N	0.15	-0.18	0.09	-0.02	0.15	0.04	0.28	0.10	-0.10	0.17	-0.13	-0.25	0.23	-0.41	-0.12
streams density	hydrology	N	-0.02	0.13	-0.14	-0.07	-0.23	0.06	-0.18	0.16	-0.06	-0.09	0.18	0.08	-0.09	0.00	0.27
shoreline dev factor	lake and watershed	N	0.30	0.11	-0.11	-0.25	-0.03	-0.06	-0.30	-0.02	0.05	0.01	-0.04	0.11	-0.01	0.13	0.13
lake area	lake and watershed	N	-0.06	-0.04	-0.28	0.06	-0.08	0.16	-0.24	0.05	0.03	-0.25	-0.09	0.31	0.12	0.11	0.20
watershed area	lake and watershed	N	-0.03	0.17	-0.14	-0.10	0.04	-0.02	-0.12	-0.05	0.03	-0.23	-0.03	0.24	-0.02	0.11	0.13
watershed:lake area ratio	lake and watershed	N	0.00	0.15	0.01	-0.10	0.07	-0.08	0.01	-0.06	0.01	-0.08	0.02	0.06	-0.07	0.04	0.02
alluvium coastal fine	lithology	N	-0.16	0.11	-0.15	-0.24	0.32	-0.05	0.33	-0.16	-0.13	0.00	-0.22	0.29	0.08	-0.05	0.03
carbonate residual	lithology	N	-0.03	-0.03	-0.04	0.02	0.15	0.10	0.08	0.04	0.09	-0.04	-0.12	-0.13	-0.08	-0.04	0.02
coastal zone coarse	lithology	N	0.26	0.24	-0.05	-0.02	0.00	-0.15	-0.14	-0.02	-0.05	0.00	0.00	0.00	-0.02	0.00	-0.05
glacial outwash coarse	lithology	N	0.14	-0.11	0.03	0.04	0.03	-0.06	0.04	0.04	0.03	-0.05	-0.14	-0.06	-0.04	-0.13	0.23
glacial till clayey	lithology	N	0.04	0.00	-0.04	-0.05	0.13	-0.06	0.07	-0.05	-0.08	-0.01	-0.10	0.29	-0.11	-0.13	0.09
glacial till coarse	lithology	N	0.16	0.17	-0.07	-0.08	-0.19	-0.08	-0.16	0.04	-0.18	0.02	0.15	-0.01	0.23	0.04	-0.04
saline lake sediment	lithology	N	0.00	0.02	0.09	-0.04	-0.01	-0.04	-0.05	-0.05	0.04	-0.02	-0.08	0.19	-0.01	0.01	-0.06
elevation	location	N	-0.54	-0.49	0.34	-0.03	-0.19	-0.05	0.27	0.16	-0.38	0.63	0.14	0.02	-0.06	0.08	0.11
latitude	location	N	0.11	-0.17	-0.06	0.28	0.37	0.17	-0.32	0.00	0.00	0.16	0.25	-0.45	-0.36	0.41	-0.39
longitude	location	N	-0.02	0.00	-0.29	-0.17	-0.06	0.16	-0.20	0.36	-0.19	0.10	0.04	0.02	0.21	0.21	-0.18

NEON region	location	N	0.00	-0.05	0.08	0.05	0.03	-0.01	-0.04	0.02	0.16	-0.06	-0.07	0.01	-0.10	-0.03	0.03
clay	soil	N	-0.47	-0.67	-0.82	0.52	-0.15	0.18	-0.01	-0.58	0.55	-1.08	0.31	0.79	0.02	0.70	0.72
coarse	soil	N	-0.24	-0.11	0.11	-0.12	0.46	-0.01	-0.10	0.60	-0.25	0.38	-0.10	0.01	-0.01	-0.37	-0.27
depth to bedrock	soil	N	-0.01	0.00	-0.08	0.45	0.17	-0.13	-0.04	-0.31	-0.10	-0.22	0.44	0.11	-0.24	0.12	-0.16
k factor	soil	N	0.09	0.24	-0.10	-0.19	0.06	0.20	0.11	-0.23	-0.06	-0.23	0.08	0.07	-0.04	0.05	-0.07
organic carbon	soil	N	0.50	0.26	-0.08	-0.02	0.15	-0.12	-0.01	0.05	-0.23	0.26	-0.42	-0.30	-0.15	-0.10	0.20
sand	soil	N	0.14	0.26	0.26	0.05	-0.26	-0.07	0.14	0.05	0.01	0.09	-0.57	0.05	0.26	-0.23	-0.16
silt	soil	N	-0.05	-0.06	0.00	-0.06	0.07	0.02	-0.02	0.01	0.06	0.01	0.02	-0.01	-0.02	0.05	-0.01
drainage	surface connectivity	N	0.15	-0.15	-0.60	0.09	-0.36	0.40	0.18	0.08	0.50	-0.27	-0.09	-0.16	0.32	-0.46	0.37
headwater	surface connectivity	N	-0.76	0.45	-0.66	0.08	-0.15	-0.07	-0.20	0.58	-0.22	0.04	-0.14	0.63	0.05	-0.14	0.50
terminal	surface connectivity	N	-0.06	-0.30	-0.18	-0.38	0.02	0.38	-0.43	0.20	-0.63	0.55	-0.30	-0.10	0.27	0.99	-0.02
runoff_zone	terrain	N	0.35	-0.17	0.07	-0.18	0.27	0.14	0.24	0.37	-0.10	0.37	-0.21	-0.60	0.44	-0.59	-0.39
sat overland flow	terrain	N	0.20	0.09	0.23	-0.08	-0.10	0.32	-0.14	0.32	-0.27	-0.05	-0.25	-0.07	0.31	-0.34	-0.17
topographic wetness	terrain	N	-0.02	-0.13	0.09	0.35	-0.14	-0.06	0.14	-0.30	-0.10	-0.31	-0.29	0.19	0.18	0.31	0.09
wet dep inorg N current	atmospheric deposition	H	-0.06	0.01	0.10	-0.11	0.01	-0.35	-0.19	0.03	0.15	0.09	-0.05	0.15	-0.05	0.09	0.17
wet dep no3 current	atmospheric deposition	H	-0.01	0.23	-0.41	-0.55	0.26	-0.15	-0.66	-0.11	0.73	0.40	-0.22	0.41	-0.50	-0.14	0.73
wet dep no3 diff	atmospheric deposition	H	0.17	0.24	0.18	0.12	-0.23	0.11	-0.04	-0.01	-0.11	-0.11	-0.10	-0.10	-0.05	-0.13	0.04
wet dep so4 diff	atmospheric deposition	H	0.14	-0.33	-0.31	-0.13	0.15	-0.20	-0.15	-0.22	0.08	-0.18	0.46	0.03	0.15	0.25	0.27
manure	human activity	H	-0.20	0.10	0.13	-0.16	-0.14	0.25	0.17	-0.19	0.25	-0.14	-0.27	-0.17	0.06	0.12	0.19
roads 500m buff	human activity	H	0.47	0.45	-0.37	-0.29	-0.38	0.24	-0.27	-0.58	0.22	-0.76	0.01	0.46	0.19	0.09	0.53
major discharge sites	human activity	H	0.00	-0.01	-0.04	-0.03	-0.04	-0.06	-0.01	-0.05	0.05	0.00	-0.12	0.23	-0.02	0.05	0.06
stormwater from industrial	human activity	H	0.13	-0.05	-0.06	-0.04	-0.02	0.19	-0.01	-0.06	-0.01	0.11	-0.08	-0.09	-0.13	0.00	0.11

road density HU12	human activity	H	0.27	0.00	-0.18	-0.15	-0.49	0.00	0.16	-0.28	-0.24	0.08	0.02	0.71	0.02	-0.12	0.22
agriculture	LU/LC	H	-0.12	0.23	0.08	0.16	-0.09	-0.14	0.16	-0.26	0.30	-0.09	-0.19	0.04	-0.44	0.14	0.22
grass	LU/LC	H	0.04	-0.18	-0.06	-0.17	0.00	0.07	-0.08	0.06	-0.06	0.10	0.10	0.11	0.10	0.04	-0.05
shrub	LU/LC	H	-0.02	0.35	0.08	-0.04	-0.01	0.04	-0.11	-0.11	-0.11	0.14	0.01	-0.02	-0.03	-0.09	-0.07
developed	LU/LC	H	0.01	0.28	-0.12	-0.07	-0.06	0.14	-0.07	-0.21	-0.08	-0.19	-0.21	0.26	0.02	0.07	0.22
forest	LU/LC	H	-0.03	-0.72	0.28	0.21	0.53	0.03	0.48	0.27	-0.14	0.18	0.01	-0.52	0.59	-0.36	-0.79
wetland	LU/LC	H	0.02	-0.10	-0.08	0.13	-0.13	0.03	0.10	0.01	-0.03	-0.12	0.06	-0.04	0.02	0.01	0.12

Table S4. Number of lakes in each connectivity class in each cluster.

Cluster	Drainage	Headwater	Isolated	Terminal
12	10	2	8	0
13	16	4	7	1
15	43	6	13	1
7	84	12	7	1
11	12	2	12	1
4	34	17	8	3
2	60	17	6	3
8	44	4	34	17
6	23	5	10	3
9	125	12	14	3
14	118	8	11	4
1	23	0	4	0
10	80	11	18	1
5	52	2	12	3
3	145	10	31	1

Supplementary Figures

Paper title: Natural and human drivers of salinity and major ion composition in United States lakes

Authors: Xinyu Sun, Kendra Spence Cheruvelil, Patrick J. Hanly, Katherine E. Webster, & Patricia A. Soranno

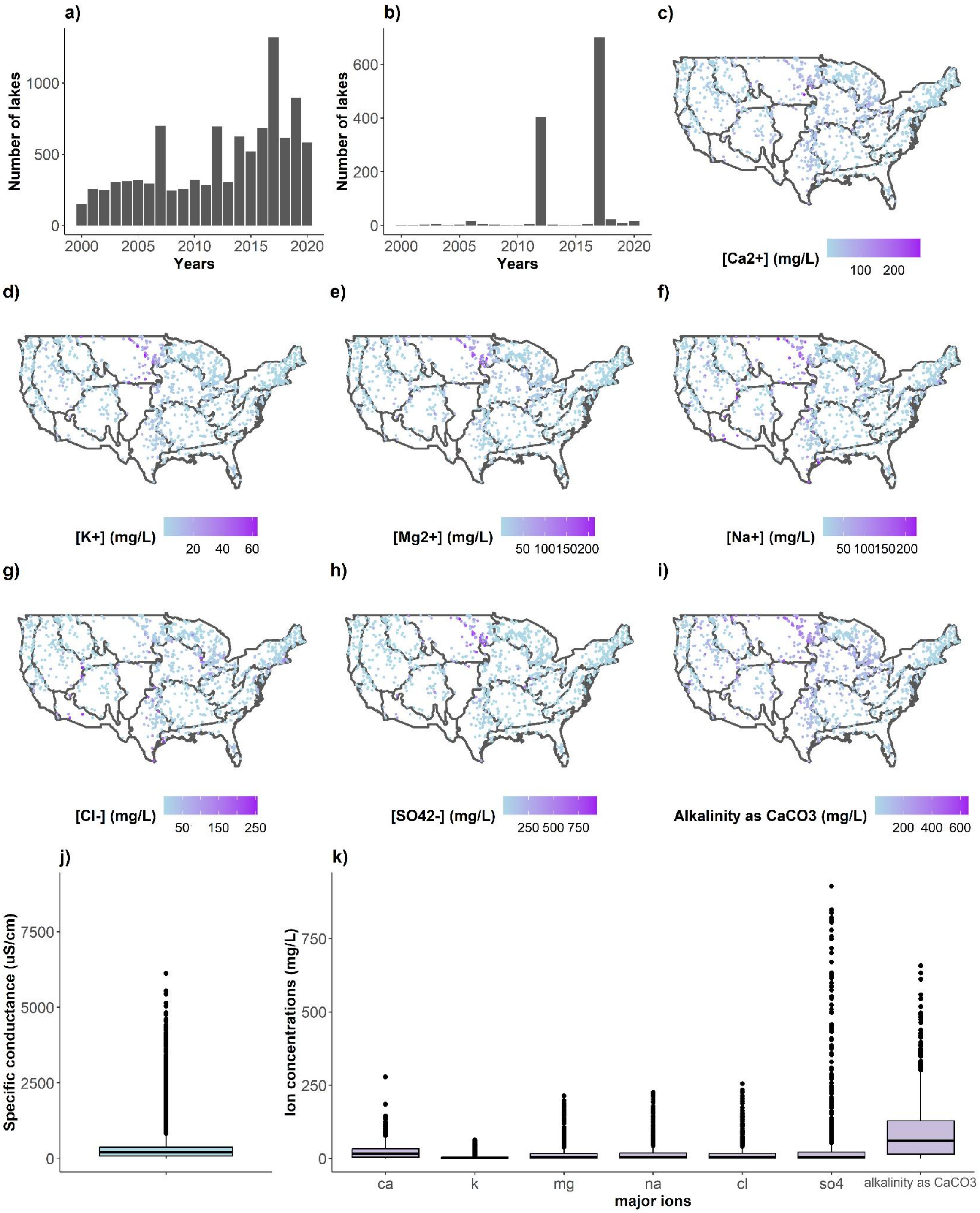


Figure S1. Summary of specific conductance (SC) and major ion data: a) number of lakes sampled in each latest SC sampling year; b) number of lakes sampled in each latest major ion sampling year; c) - i) maps showing major ion and alkalinity levels (mg/L) by NEON region; j) boxplot showing the distribution of SC data; and k) boxplots showing the distribution of major ion concentrations. See Figure 1 caption for NEON region names.

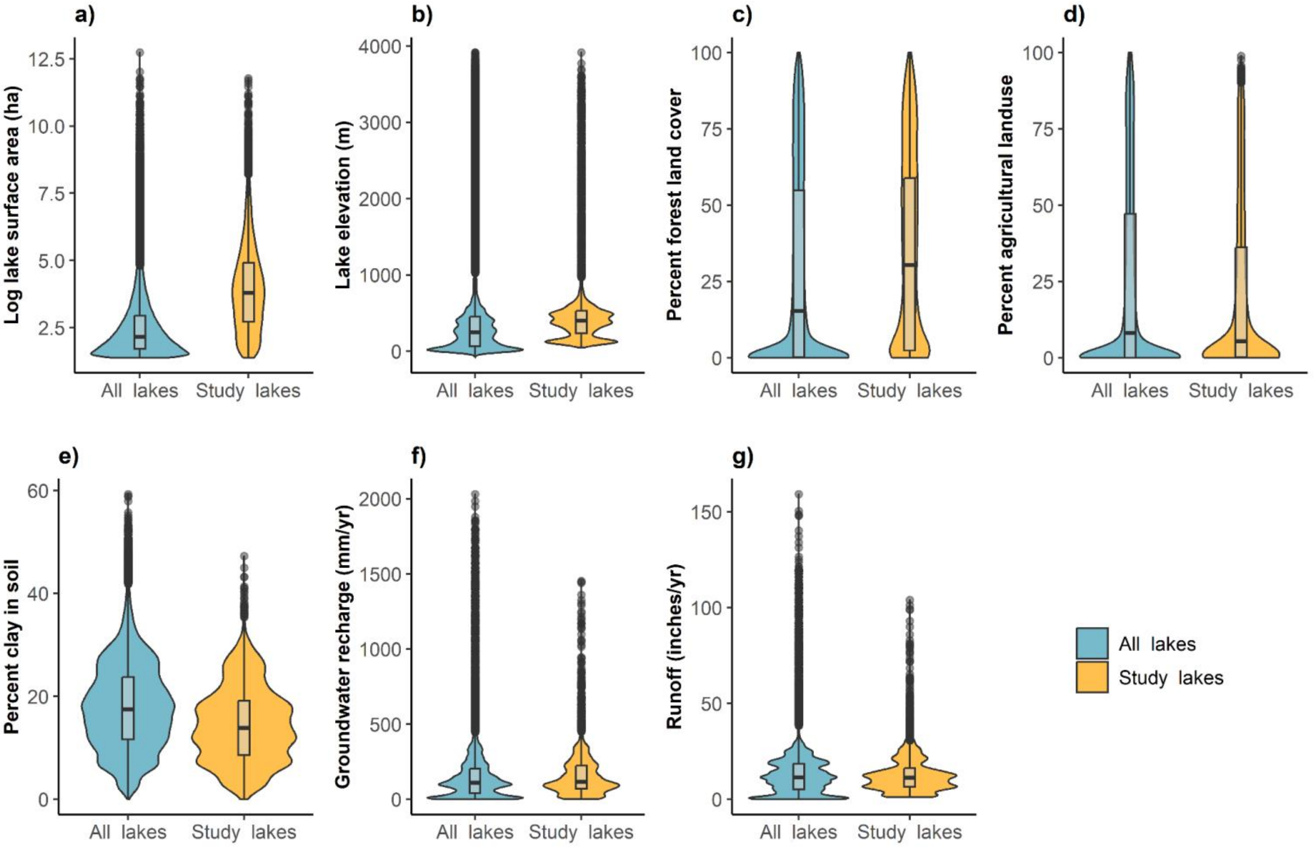


Figure S2. Comparison of all lakes > 4 ha in the U.S. (All lakes, blue) relative to the study lake population (Study lakes, orange). The violin plots show all data for the two groups of lakes; the black dots are the median values; and, the boxes within the violin plots are the interquartile ranges. The study lakes are included in the all-lake population derived from the LAGOS lake population > 4 ha.

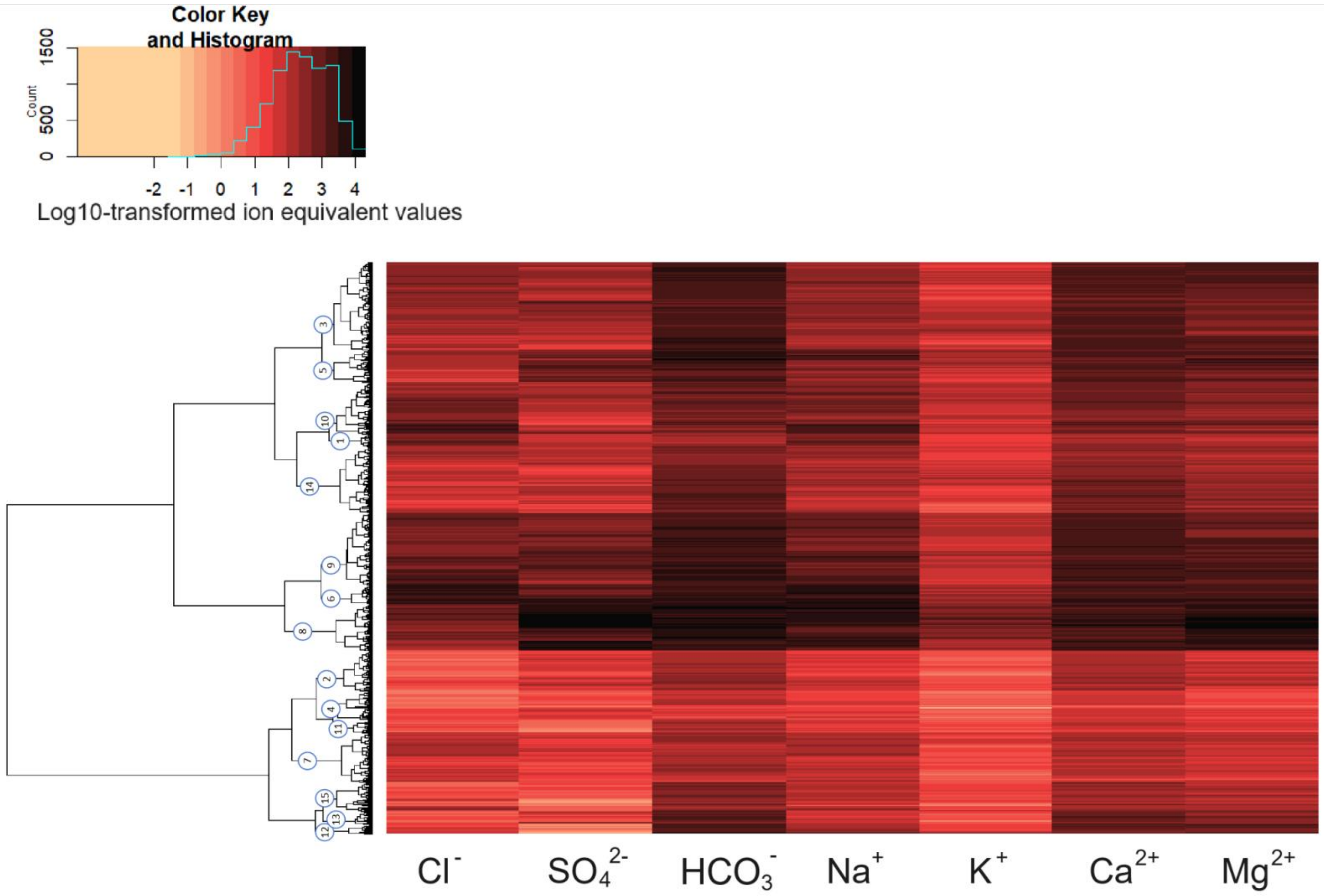
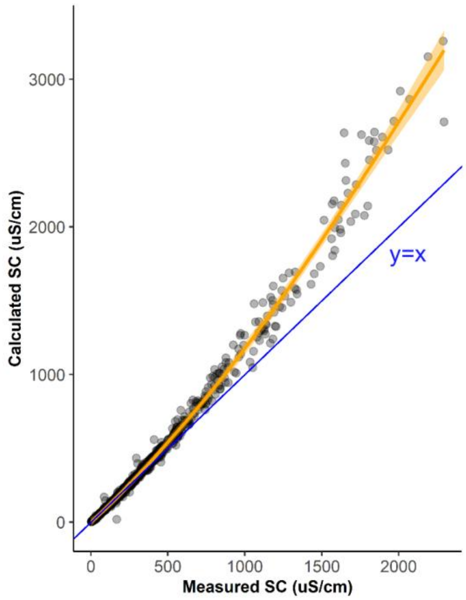
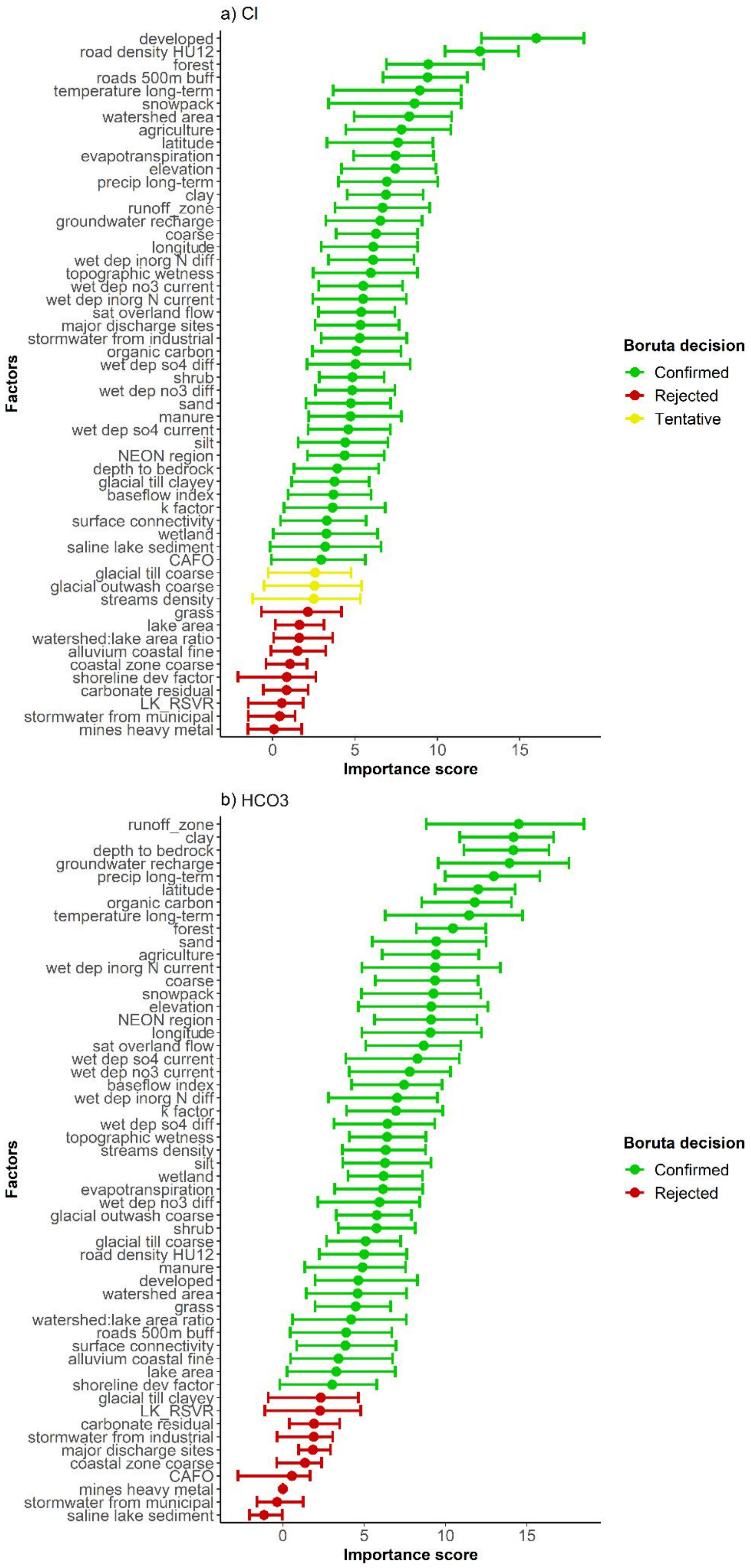


Figure S3. A Heatmap showing log10-transformed major ion equivalent values grouped by clusters with a dendrogram on the left edge. The histogram at the top shows the color key and distribution of data.



Ions	Conversion factors
Ca^{2+}	52.0
Mg^{2+}	46.6
Na^{+}	48.9
K^{+}	72.0
Cl^{-}	75.9
SO_4^{2-}	73.9
HCO_3^{-}	43

Figure S4. Lake-specific relationships between SC and salinity values calculated using ion equivalent concentrations (in µeq/L). The ion conversion factors used in the calculation are listed in the table on the right. The orange curve is the general additive model fitted regression line and the shaded area represents 1 SD.


a) Cl
developed
road density HU12
forest
roads 500m buff
temperature long-term
snowpack
watershed area
agriculture
latitude
evapotranspiration
elevation
precip long-term
clay
runoff_zone
groundwater recharge
coarse
longitude
wet dep inorg N diff
topographic wetness
wet dep no3 current
wet dep inorg N current
sat overland flow
major discharge sites
stormwater from industrial
organic carbon
wet dep so4 diff
shrub
wet dep no3 diff
sand
manure
wet dep so4 current
silt
NEON region
depth to bedrock
glacial till clayey
baseflow index
k factor
surface connectivity
wetland
saline lake sediment
CAFO
glacial till coarse
glacial outwash coarse
streams density
grass
lake area
watershed:lake area ratio
alluvium coastal fine
coastal zone coarse
shoreline dev factor
carbonate residual
LK_RSVR
stormwater from municipal
mines heavy metal
Factors
0
5
10
15
Importance score
Boruta decision
Confirmed
Rejected
Tentative
b) HCO3
runoff_zone
clay
depth to bedrock
groundwater recharge
precip long-term
latitude
organic carbon
temperature long-term
forest
sand
agriculture
wet dep inorg N current
coarse
snowpack
elevation
NEON region
longitude
sat overland flow
wet dep so4 current
wet dep no3 current
baseflow index
wet dep inorg N diff
k factor
wet dep so4 diff
topographic wetness
streams density
silt
wetland
evapotranspiration
wet dep no3 diff
glacial outwash coarse
shrub
glacial till coarse
road density HU12
manure
developed
watershed area
grass
watershed:lake area ratio
roads 500m buff
surface connectivity
alluvium coastal fine
lake area
shoreline dev factor
glacial till clayey
LK_RSVR
carbonate residual
stormwater from industrial
major discharge sites
coastal zone coarse
CAFO
mines heavy metal
stormwater from municipal
saline lake sediment
Factors
0
5
10
15
Importance score
Boruta decision
Confirmed
Rejected

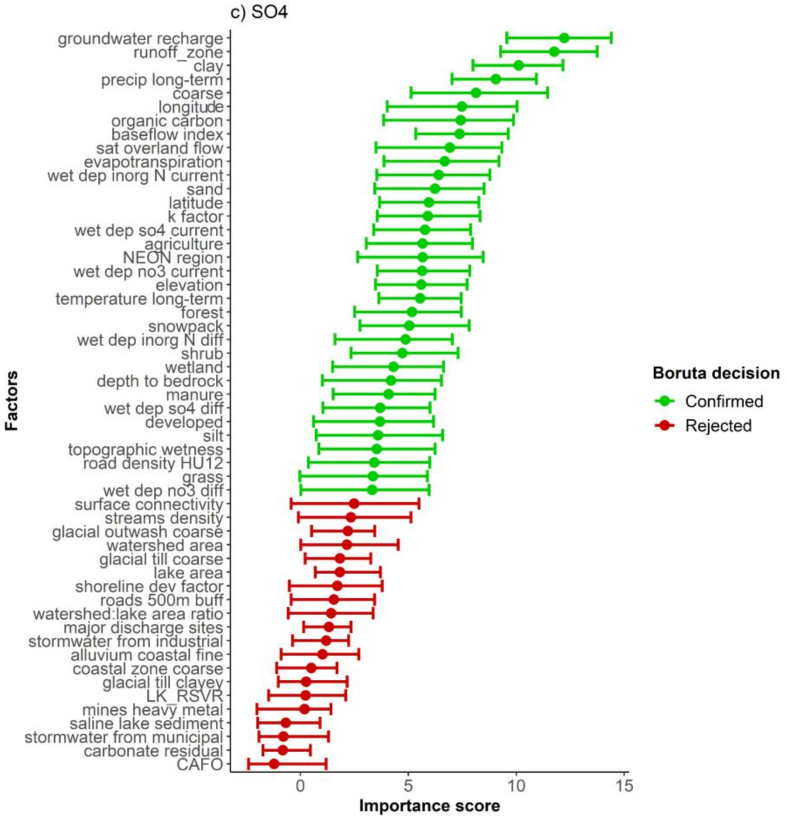

c) SO4

Figure S5. Importance scores from Boruta feature selection for the three anions: Cl^- (a), HCO_3^- (b), and $SO4^{2-}$ (c). Green dots and bars indicate that the factors were identified as ‘important’ by Boruta; yellow dots and bars indicate that the factors were identified as ‘tentative’ by Boruta; and red dots and bars indicate that Boruta rejected the factors. Dots are the mean Boruta importance values and bars are the minimum and maximum Boruta importance values.

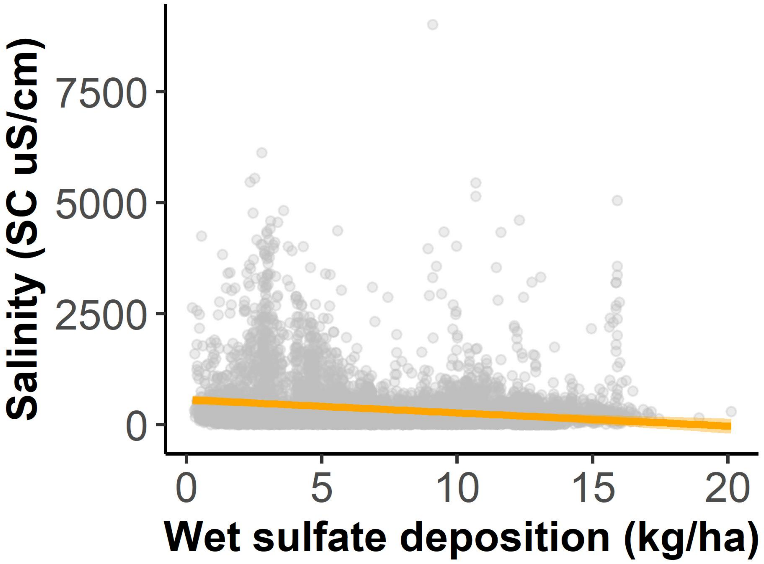


Figure S6. Scatterplot showing the relationship between wet sulfate deposition and salinity (using SC as a proxy). An orange linear regression line was added to show the overall trend.