Salting our freshwater lakes

Hilary A. Dugan[a,b,1], Sarah L. Bartlett[c], Samantha M. Burke[d], Jonathan P. Doubek[e], Flora E. Krivak-Tetley[f], Nicholas K. Skaff[g], Jamie C. Summers[h], Kaitlin J. Farrell[i], Ian M. McCullough[j], Ana M. Morales-Williams[k,2], Derek C. Roberts[l,m], Zutao Ouyang[n], Facundo Scordo[o], Paul C. Hanson[a], and Kathleen C. Weathers[b]

[a]Center for Limnology, University of Wisconsin–Madison, Madison, WI 53706; [b]Cary Institute of Ecosystem Studies, Millbrook, NY 12545; [c]School of Freshwater Sciences, University of Wisconsin–Milwaukee, Milwaukee, WI 53204; [d]Department of Biology, University of Waterloo, Waterloo, ON, N2L 3G1, Canada; [e]Department of Biological Sciences, Virginia Tech, Blacksburg, VA 24061; [f]Department of Biological Sciences, Dartmouth College, Hanover, NH 03755; [g]Department of Fisheries and Wildlife, Michigan State University, East Lansing, MI 48824; [h]Department of Biology, Queen's University, Kingston, ON, K7L 3N6, Canada; [i]Odum School of Ecology, University of Georgia, Athens, GA 30602; [j]Bren School of Environmental Science and Management, University of California, Santa Barbara, CA 93106; [k]Department of Ecology, Evolution and Organismal Biology, Iowa State University, Ames, IA 50011; [l]Department of Civil & Environmental Engineering, University of California, Davis, CA 95616; [m]UC Davis Tahoe Environmental Research Center, Incline Village, NV 89451; [n]Center for Global Change and Earth Observations, Michigan State University, East Lansing, MI 48823; and [o]Instituto Argentino de Oceanografía, Universidad Nacional del Sur–CONICET, Bahia Blanca Bs As, B8000BFW, Argentina

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The highest densities of lakes on Earth are in north temperate ecosystems, where increasing urbanization and associated chloride runoff can salinize freshwaters and threaten lake water quality and the many ecosystem services lakes provide. However, the extent to which lake salinity may be changing at broad spatial scales remains unknown, leading us to first identify spatial patterns and then investigate the drivers of these patterns. Significant decadal trends in lake salinization were identified using a dataset of long-term chloride concentrations from 371 North American lakes. Landscape and climate metrics calculated for each site demonstrated that impervious land cover was a strong predictor of chloride trends in Northeast and Midwest North American lakes. As little as 1% impervious land cover surrounding a lake increased the likelihood of long-term salinization. Considering that 27% of large lakes in the United States have >1% impervious land cover around their perimeters, the potential for steady and long-term salinization of these aquatic systems is high. This study predicts that many lakes will exceed the aquatic life threshold criterion for chronic chloride exposure (230 mg L^{-1}), stipulated by the US Environmental Protection Agency (EPA), in the next 50 y if current trends continue.

limnology | chloride | road salt | impervious surface | ecosystem services

Due to landscape position, lake ecosystems are influenced by surrounding terrestrial processes, and their generally long water residence times can contribute to the accumulation of external inputs and pollutants (1). Therefore, although lakes cover only 3% of the continental land surface (2), long-term trends in lakes are often early warning indicators of significant local, regional, or global changes (3). One such early warning indicator is change in lake chloride concentrations. Naturally occurring in freshwaters at low concentrations, chloride is a highly soluble and conservative ion that has also been shown to be a reliable proxy for chloride-based road salts (typically sodium chloride) (4, 5). Although chloride concentrations in freshwaters can vary cyclically due to climatic processes, such as extended periods of drought (6), elevated chloride concentrations in lakes often result from agricultural, industrial, and transportation practices (7). Elevated chloride concentrations can have adverse effects on water quality and aquatic ecosystems (8–11), including both immediate and long-term alterations to community structure, diversity, and productivity (12–14).

Salt application for de-icing roadways has been recognized as a major source of chloride to groundwater (15–17), streams and rivers (5, 10, 18, 19), and lakes (7, 9, 20, 21, 22) across north temperate climates in North America and Europe. In the United States, road salting became a standard practice in the 1940s, and road salt sales over the subsequent 50 y increased from 0.15 to over 18 million metric tons per year (4). In Canada, despite its addition to the List of Toxic Substances (23) and the implementation of the Code of Practice for the Environmental Management of Road Salts in 1999, an average of 5 million metric tons of road salt per year was applied to roadways between 1995 and 2001 (23, 24). Following application, road salt quickly dissolves and is transported into rivers and lakes through leaching and runoff (5, 25). A few studies have characterized the negative short term or localized impacts of elevated road salt concentrations in freshwaters (5, 15, 25), but there have been no large-scale analyses of chloride trends in freshwater lakes.

Here, we investigate trends in lake chloride concentration, using a dataset of long-term chloride concentrations in lakes and reservoirs in North America. We identify regions of high salinization, where aquatic ecosystems may be at risk, and contrast the role of climate versus the anthropogenic practice of road salting in driving chloride variability. Lakes included in the dataset were required to have at least 10 y of chloride data, a mean chloride concentration ≤1 g L^{-1} (to exclude brackish lakes), and a surface area ≥4 ha. The median length of an individual time series was

Significance

In lakes, chloride is a relatively benign ion at low concentrations but begins to have ecological impacts as concentrations rise into the 100s and 1,000s of mg L^{-1}. In this study, we investigate long-term chloride trends in 371 freshwater lakes in North America. We find that in Midwest and Northeast North America, most urban lakes and rural lakes that are surrounded by >1% impervious land cover show increasing chloride trends. Expanding on this finding, thousands of lakes in these regions are at risk of long-term salinization. Keeping lakes "fresh" is critically important for protecting the ecosystem services freshwater lakes provide, such as drinking water, fisheries, recreation, irrigation, and aquatic habitat.

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[1]To whom correspondence should be addressed. Email: hdugan@wisc.edu.

[2]Present address: Rubenstein School of Environment and Natural Resources, University of Vermont, Burlington, VT 05405.

ENVIRONMENTAL SCIENCES

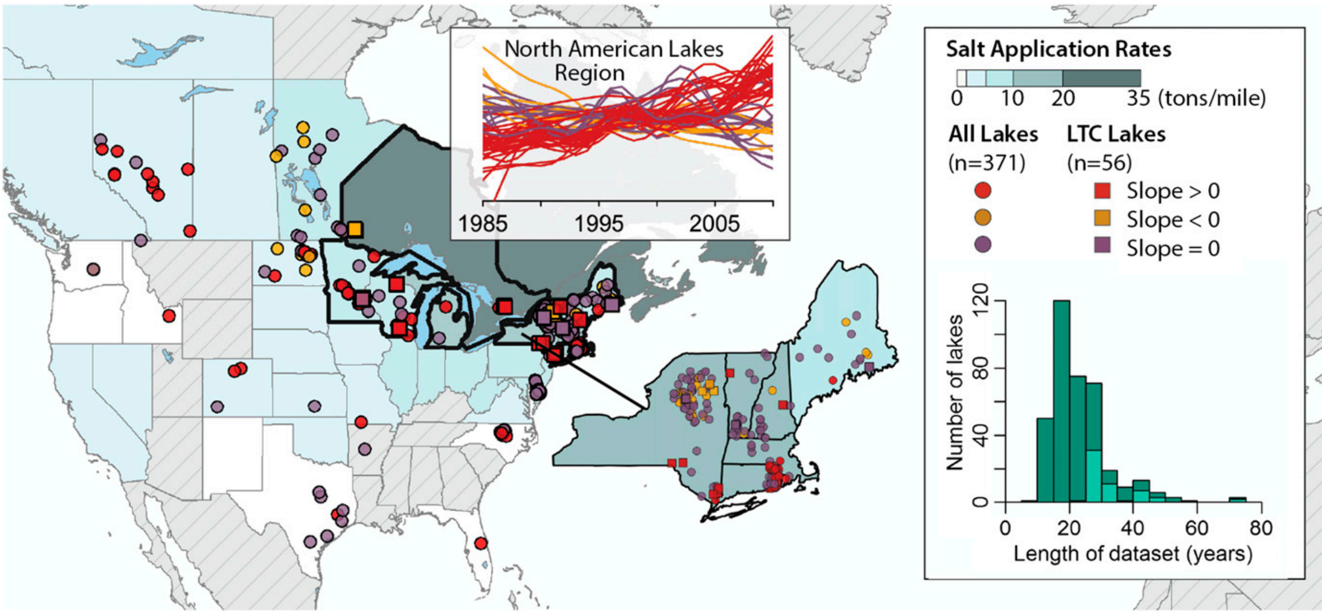

Fig. 1. Chloride trends for North American freshwater lakes (circles and squares, $n = 371$). The states and province included in the NALR are outlined in black. Points are colored by the slope value of linear regression models (red, positive slope; yellow, negative slope; purple, zero or nonsignificant slope). Squares denote lakes with at least biennial chloride concentrations recorded from 1985 to 2010 ($n = 56$). These LTC datasets are a subset of lakes in the NALR, which is a region of dense sampling ($n = 284$). *Upper Inset* of chloride time series from 1985 to 2010 are colored by slope value. Road salt application rates for North American provinces and states range from 0 to 35 US tons per mile and are shown in blue. No salt application rates were available in areas with hatched lines. The lengths of all individual datasets (dark green) as well as the lengths of LTC datasets (light green) are shown in the *Inset* histogram.

21 y. The dataset included lake morphometric characteristics, climate statistics on temperature and precipitation, and atmospheric sea salt deposition. As a proxy for road salt application, land cover metrics were calculated, including road density (26) [length of road in a given area ($km\ km^{-2}$)] and percent impervious land cover (25) within a 100- to 1500-m buffer surrounding each lake. Road density and impervious land cover represent the best proxies for road salt application, given that variability in road salt application, both spatially and on a year-to-year basis, prevents application rates from being calculated at spatial and temporal scales relevant to lakes.

Lakes in this dataset were not randomly sampled and thus do not necessarily represent the distribution of lakes within each state or province. To limit sampling bias in this dataset, we focused our analyses on a geographic area with dense sampling coverage: a North American lakes region (NALR), which includes Connecticut, Maine, Massachusetts, Michigan, Minnesota, New Hampshire, New York, Ontario, Rhode Island, Vermont, and Wisconsin (Fig. 1). We excluded North Dakota lakes from this grouping, as many are part of the Devil's Lake watershed, an endorheic (closed-basin) system where water levels have risen ~10 m since 1992, and therefore, the hydrology is vastly different from exorheic (open) lakes (27). Likewise, Manitoba lakes were excluded, as many were enlarged or drained during hydroelectric construction along the Churchill and Nelson Rivers (28). Of the 371 North American lakes in our dataset, 284 were in the NALR (Fig. 1). Mean chloride concentrations in lakes across the NALR ranged from 0.18 to 240.8 $mg\ L^{-1}$, with a median value of 6.0 $mg\ L^{-1}$.

Chloride time series for each lake differed in the frequency, duration, and depth of sampling. We pooled all depth samples for analyses, based on observations that chloride concentrations track similar trends throughout the water column of most lakes and that previous studies of long-term chloride trends have shown similarity with depth (7, 29). To reduce autocorrelation due to seasonality, we reduced all time series to annual averages. To enable comparison of chloride trends across lakes, a linear model was fit to the annual data, where chloride (standardized to a distribution with mean = 0 and SD = 1) was a function of time. Lakes were classified by simple linear regression models into three possible long-term trends: decreasing ($n = 42$, slope < 0, $P <= 0.01$), stationary ($n = 204$, slope $= 0$, $P > 0.01$), or increasing ($n = 125$, slope > 0, $P <= 0.01$). Of the 125 lakes with a positive trend in chloride, 99 were in the NALR (Fig. 1).

To investigate both linear trends and time-series patterns over a comparable period, any site in the NALR that had at least biennial data from 1985 to 2010 was included in a subset of long-term continuous (LTC) data. Clustering the 56 LTC lakes into three groupings using a hierarchical clustering analysis revealed three characteristic trends in chloride concentrations: neutral/decreasing (cluster 1, $n = 16$), oscillating (cluster 2, $n = 4$), and increasing (cluster 3, $n = 36$) (Fig. 2*A*). Cluster 1 was a geographical mix of lakes with both decreasing and neutral slope trends, cluster 2 lakes were exclusively in Maine and had neutral slope trends, and cluster 3 lakes, 21 of which were in Minnesota, had predominantly increasing slope trends (35 of 38) (Fig. 2*B*).

Potential drivers of increasing lake chloride were first assessed by relating slope values to lake, climate, and landscape characteristics of lakes in the NALR (Fig. 3 *A–C*). Due to the prevalence of zero-values in the data, it was not possible to build robust log-linear models for most of the landscape characteristics. Therefore, we used both classification/regression trees and random forests to build predictive models for the NALR data. A classification/regression tree and a random forest were created for each of three response variables: linear slope, tested as both continuous numerical and categorical (positive, zero, negative) variables, and hierarchical cluster grouping (1, 2, or 3). Categorical slope was used as a response variable to further remove any bias in our linear model application by removing magnitude. The motivation for using two approaches and three response variables was to improve the accuracy of our analytics, in much the same way as ensemble modeling.

Results of the three classification/regression trees and three random forests revealed that impervious land cover and road

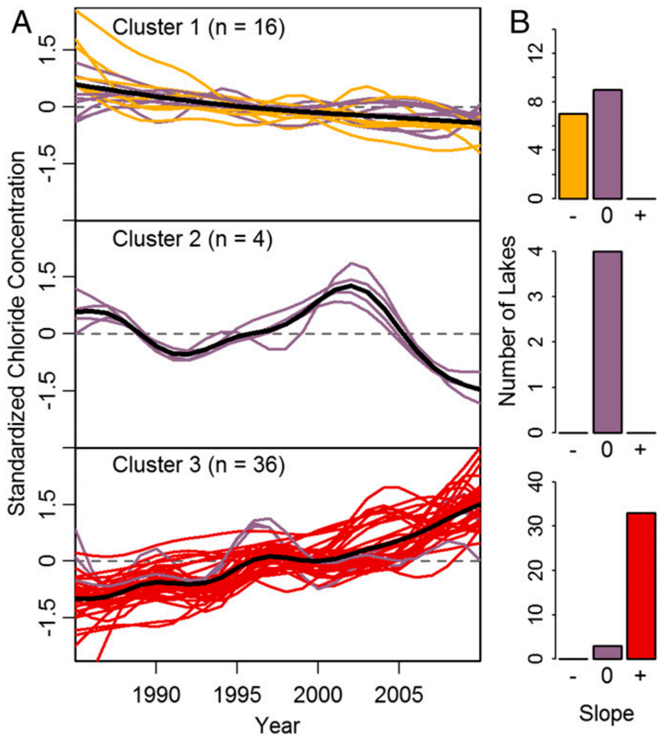

Fig. 2. (*A*) LTC lakes (n = 56) with biennial chloride data from 1985 to 2010 grouped into three clusters using a hierarchical cluster analysis. In general, the three clusters show a neutral/decreasing (cluster 1), oscillating (cluster 2), or increasing (cluster 3) pattern. Thick black lines are GAMs fit to all lakes within each cluster, to represent the average pattern. (*B*) Histograms display the number of lakes in each cluster by linear slope (yellow, negative slope; purple, zero slope; red, positive slope).

density surrounding each lake were the primary classification splits and the most important predictors for lake chloride trends and cluster grouping (Table 1).

The predictors used in the tree-based models were all static variables, meaning values did not vary with time. This limitation may misrepresent relationships between chloride concentrations and drivers that vary on a subannual basis (e.g., precipitation). Monthly precipitation data were obtained from the PRISM high-resolution spatial climate dataset, which covers the United States at a spatial resolution of 4 km (30). To account for the lag in chloride retention in a watershed (19), a LOESS curve was fit to mean monthly precipitation (mm/d) from 1985 to 2010 at each LTC site. A correlation between precipitation and chloride concentration at each LTC lake was calculated from annual data predicted from the LOESS precipitation curve and the generalized additive model (GAM) of chloride concentration. There was a strong negative correlation (r^2 = 0.71–0.87) between precipitation and chloride concentration for the four Maine lakes that group into cluster 2 (oscillating pattern). These four lakes are all less than 0.25 km^2 and receive ∼1.25 m of precipitation per year and have no impervious land cover within 500 m. Without knowledge of the groundwater hydrology of these lakes, it may be that precipitation controls the chloride balance, with heavy rains and large snowfalls diluting the chloride concentrations. A strong relationship between precipitation and chloride is not evident for lakes that group into cluster 1 or 3 (median r^2 = 0.12, range = 0–0.61).

Of our NALR sites, 44% of freshwater lakes have undergone long-term salinization. Positive chloride trends were present in lakes with as little as 1% impervious coverage. This finding is consistent with studies of US streams that found increased chloride concentrations associated with any urban land cover (31) or roads (32, 33) and substantiates findings of ecological community thresholds associated with low levels of catchment urbanization (34). Across the NALR, lakes with mean chloride concentrations >1 mg L^{-1} (mean value of the time series) were more likely to be associated with positive trends in chloride (Fig. 3*D*). This suggests that high chloride concentrations in this region may be an indicator and warning sign of recent salinization.

If impervious land cover surrounding a lake is a robust predictor of water quality, it is important to understand the probability of its occurrence across all lakes within a region or country. Using national hydrography and land cover datasets for the continental United States, we found that the median percent impervious land cover within 500 m of all lakes greater than 4 ha is 0.31% (n = 149,350; Fig. 4). Of these US lakes, 28% had greater than 1% impervious land cover in a 500-m buffer zone. The density of roads and other impervious surfaces surrounding lakes in US regions where road salt is applied should therefore be of high concern. In the NALR, 70% (94 out of 134) of lakes with > 1% impervious land cover in the 500-m

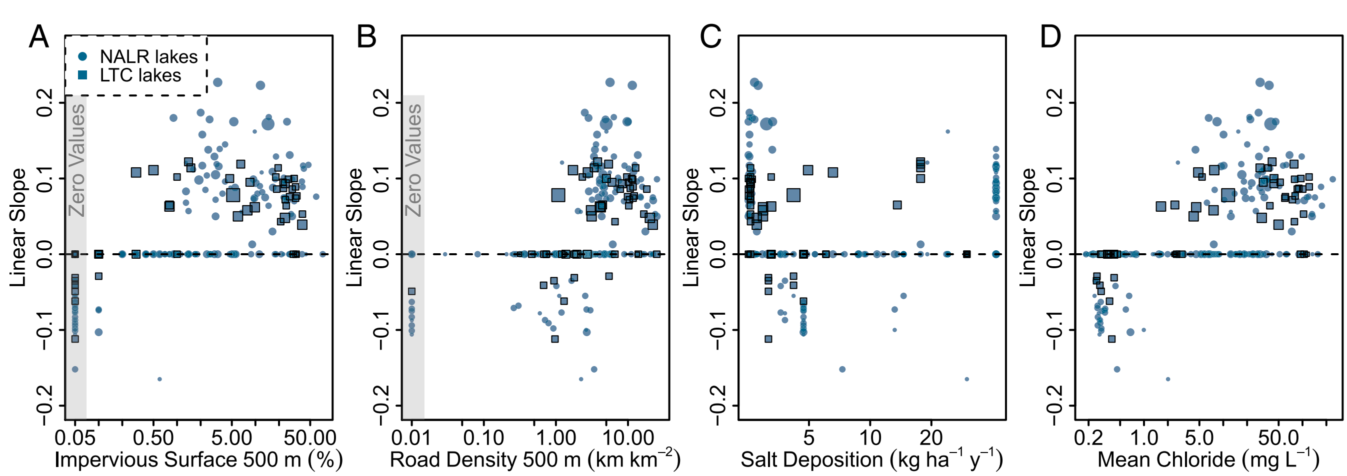

Fig. 3. Scatterplots of linear regression slope values versus (*A*) impervious surface within a 500-m buffer, (*B*) road density within a 500-m buffer, (*C*) rate of atmospheric salt deposition, and (*D*) mean in-lake chloride concentration over the entire time series for all NALR sites (n = 284). In all plots, the size of the symbol is scaled by lake area. Squares with black borders denote LTC lakes. In *A* and *B*, zero values have been adjusted to fit on the x axis and are highlighted in gray.

Table 1. Primary node splits from regression/classification tree models and the top predictor from random forest models

Response variable	Subset of data	Regression/classification tree primary node split	Random forest top predictor, variance explained
Linear slope, numerical	*NALR*	Impervious land cover 100 m	Road density 500 m, 50%
Linear slope, categorical	*NALR*	Impervious land cover 500 m	Impervious land cover 500 m
Cluster, categorical	*NALR LTC*	Impervious land cover 100 m	Impervious land cover 200 m

Models were built using linear slope (both as a number and a category), and cluster category as response variables for NALR lakes (all data $n = 284$ and LTC $n = 56$). Predictors included lake surface area, road density and impervious land cover (100-, 200-, 300-, 400-, 500-, 1,000-, 1500-m buffers), mean January air temperature, annual precipitation, wet/dry chloride deposition, and distance from the coast. For random forest models using a numerical response variable, % variance explained by the model is provided.

buffer had increasing chloride trends. If this result is extrapolated to all lakes in the US NALR (CT, MA, ME, MI, MN, NH, NY, RI, VT, and WI), ~7,770 lakes may be experiencing elevated chloride concentrations, likely due to road salt runoff. This is calculated as 70% of the 11,104 out of 38,603 lakes in the US NALR greater than 4 ha that have >1% impervious land

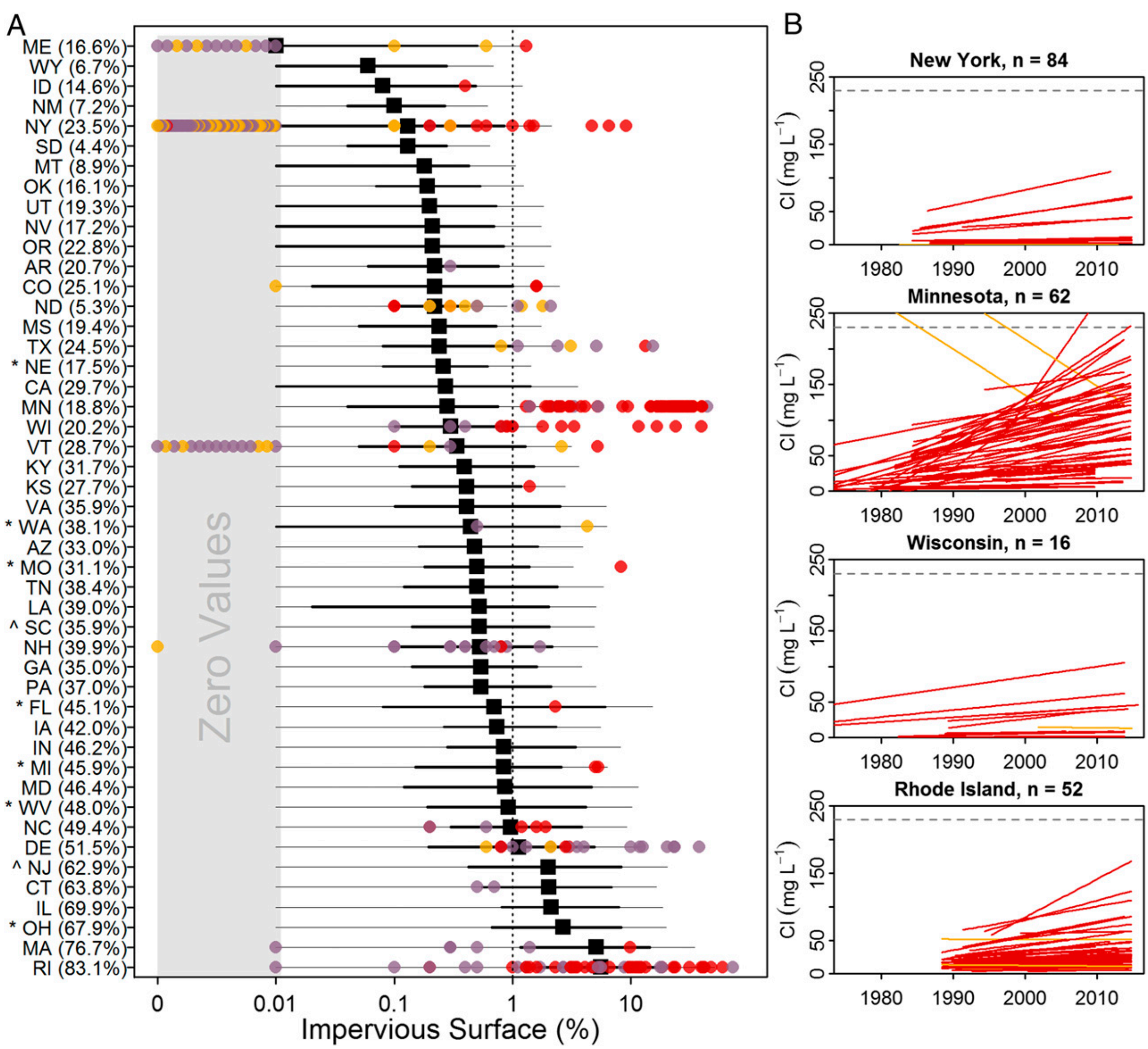

Fig. 4. (*A*) Distribution of impervious land cover within a 500-m buffer of all lakes >4 ha in the lower 48 United States ($n = 149{,}350$). Black squares represent the median impervious land cover percentage in each state. Thick horizontal black lines denote the interquartile range of the distribution, and thin black lines extend to 1.5 times the interquartile range. The vertical dashed line is shown at impervious land surface = 1%. Circles represent lakes included in this study, colored by slope (yellow, negative slope; purple, zero slope; red, positive slope). Due to the frequency of zero values on the *x* axis, circles are spread out within the gray rectangle. Percentages following *y* axis labels represent the percent of lakes in that state with greater than 1% impervious land cover within a 500-m buffer. In states with >10 lakes present in the dataset, an asterisk denotes that the sampling distribution in our dataset was significantly different from statewide distribution (Mann–Whitney test, $P < 0.05$), and ^ denotes that the sampling distribution was not significantly different from statewide distribution. (*B*) Chloride trends, as represented by linear regression model fits, are shown for four states with relatively large sample sizes (New York, Minnesota, Wisconsin, and Rhode Island). The dotted gray line demarcates the EPA's aquatic life criterion of 230 mg L^{-1}.

cover within 500 m. We note that data from Wisconsin and Minnesota are heavily biased toward urban lakes, whereas data from Maine, New York, and Vermont are heavily biased toward lakes in remote areas. This dataset (Fig. 4) includes lakes from all environments and should be representative of the Midwest and Northeast US region as a whole.

In North America, specifically in the Midwest and Northeast, local salt application leaves freshwater lakes vulnerable to salinization. Of the 284 lakes in the NALR, 26 already have a chloride concentration above 100 mg L^{-1} at their last sampling date. The median impervious land cover within a 500-m buffer surrounding these 26 lakes is 24.8%, compared with the US mean 0.31%. If a linear relationship between time and chloride concentration is extrapolated, 47 lakes are on track to reach 100 mg L^{-1} by the year 2050, and 14 are expected to surpass the EPA's aquatic life criterion concentration of 230 mg L^{-1} by 2050 (Fig. 4*B*). This is also the concentration at which a deterioration in drinking water taste is perceptible.

Elevated chloride concentrations in lakes can alter the composition and function of phytoplankton, zooplankton, macroinvertebrate, and fish communities (10–12, 35). As a consequence of salinization, aquatic species richness and abundance may decline, which could result in trophic cascades and altered water quality and ecosystem structure and function (36). In extreme cases, salinization can generate density gradients within the lake water column that prevent vertical mixing. Permanent stratification can result in anoxia and internal nutrient and metal resuspension, which decreases lake habitability and water quality (37). All of these ecosystem alterations can significantly affect lake water quality, which has millions of dollars in economic value (38, 39).

Our estimate that 7,770 lakes in the US NALR may be at risk for elevated chloride concentrations is likely an underestimate, as it does not consider regions of heavy road salt application where no long-term lake data were available, such as Québec or the Maritime provinces of Canada. Many states and municipalities are aware of the importance of shoreline management for maintaining healthy lakes; however, many shoreline zoning regulations are only enforced within 300 m or less of a lake (e.g., Wisconsin and Minnesota regulate 300 m, whereas Vermont and Maine only regulate 76 m). Because impervious surfaces and road density within at least 500 m of a lake are associated with increased chloride in areas that apply road salt, best management practices should recognize that lakeshore management extends well beyond the lake perimeter. Further, many jurisdictions lack consistent long-term monitoring programs, which provide data for predictive models and can be used to raise awareness and inform policy and management decisions used to curtail the threat of lake salinization. Clearly, keeping lakes "fresh" is critically important for protecting the ecosystem services freshwater lakes provide, such as drinking water sources, commercial fisheries, tourism, recreation, irrigation, and aquatic habitat.

Methods and Materials

Impervious land coverage at 20- to 30-m resolution was available for lakes in the United States as the degree of impervious surface per pixel (0–100%) and for Canadian and US lakes as a boolean value (0 or 1) representing whether the majority of each pixel was impervious surface. We adjusted Canadian values to match US values by using a conversion constructed from pooled impervious surface data from the United States ($r^2 = 0.91$, $P \sim 0$, Eq. **1**), assuming that the relationship between boolean and percent impervious classifications would be similar in the United States and Canada:

$$\text{Revised Impervious Surface} = \text{Impervious Surface as Boolean}(\text{Canada}) * 0.388 \quad \textbf{[1]}$$

Using log-transformed, nonzero values ($n = 302$), we found that the means of impervious land cover across seven buffer sizes (100–500, 1,000, and 1,500 m) were statistically equal (Bartlett Test for homogenous variance $P = 1$; ANOVA $F = 0.18$, $P = 0.98$). For road density ($n = 435$), this was only true for buffer sizes of 400 m through 1,500 m (Bartlett Test $P = 0.11$; ANOVA $F = 2.58$, $P = 0.052$). Median road density across our 371 lakes decreased from 3.2 km km^{-2} to 1.9 km km^{-2} as the buffer size increased from 100 to 1,500 m. Because the variability in road density and impervious land cover was much greater between lakes than for a single lake within a range of buffer sizes, the choice of buffer size was not a determining factor in this analysis. Therefore, for most analyses we present road density and impervious land cover estimates within a 500-m buffer of each lake, and these generally represent average conditions.

Road salt (as sodium chloride) application rates were difficult to find at the local or regional level. If available, the rates were typically published as single values of average annual use or only included data for a single year. The best available data were at the state, provincial, or county level. In the United States, state-level highway data were obtained from the 1991 National Research Council published report on salt use (40), individual Department of Transportation reports [CT (41), KS (42), NC (43), PA (44), RI (45)], and by contacting individual states (ND). Many of these estimates were conservative, with much higher values being cited in recent years for some states, including IA (46), ME (47), and WI (48). Canadian provincial salt application rates were calculated by dividing metric tonnage per year (49) by the number of lane miles per province (50). All road salt data are presented in units of US tons per lane mile. State- and provincial-level application rates were multiplied by road density to give an approximation of potential road salt loading for North American lakes.

LTC lakes were fit with a GAM to predict chloride trends from 1985 to 2010 at a regularly spaced time interval. GAMs were fit using the mgcv package in R [v.1.8–12 (51)] using standardized chloride data and allowing k (basis dimension for smoothing term) to vary for each penalized thin-plate regression spline. A hierarchical cluster analysis was performed on the LTC time series to test if similar temporal patterns in chloride concentrations were present across multiple lakes. We used Ward clustering, under which dissimilarities were squared before clustering, on a dissimilarity matrix constructed from Euclidean distances [R package: TSclust v.1.2.3 (52)]. We performed a k-means clustering on the LTC data and visually identified the optimal number of clusters to be three, based on a sum of squares screen plot. No distinct trends were exposed by moving beyond three clusters.

Two statistical techniques were used to build predictive models:

i) Classification/regression trees [R package: rpart v.4.1–10 (53)]. Regression trees were split using the ANOVA method, which maximizes the sum of squares between groups. Classification trees were used only when cluster group was the response variable and used the Gini index as the splitting criterion.

ii) Random forest [R package: randomForest v.4.6–12 (54)].

Static predictor variables sourced from the dataset were lake area, road density, and percent impervious land cover (100-, 200-, 300-, 400-, 500-, 1,000-, and 1,500-m buffer) surrounding each lake, January mean monthly air temperatures, mean annual precipitation, distance to the coast, and mean annual sea salt deposition.

To assess the potential for salinization of lakes at the country scale, we calculated the percent impervious land cover in 500-m and 1,000-m buffers for all lakes ≥4 ha in the United States, using shapefiles from the National Hydrography Dataset ($n = 152{,}199$) and the 2011 US National Land Cover Database Percent Developed Impervious layer (55).

Analytical scripts are available from the corresponding author.

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