Supporting Information

High-frequency data reveal deicing salts drive elevated specific conductance and chloride along with pervasive and frequent exceedances of the U.S. Environmental Protection Agency aquatic life criteria for chloride in urban streams

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MATERIALS AND METHODS

Site selection. We selected sites in small to medium watersheds (<300 km^2) with U.S. Geological Survey (USGS) gaging stations based on the availability of ≥20 discrete chloride concentration [Cl] and specific conductance (SC) samples and/or availability of ≥3 water years of high-frequency SC data (with a few exceptions). We searched for candidate study sites in Georgia, South Carolina, North Carolina, Virginia, West Virginia, Maryland, Delaware, Pennsylvania, New York, Connecticut, Rhode Island, Massachusetts, Vermont, New Hampshire, and Maine. Within the states listed above, we looked for candidate sites in watersheds that were east of the Appalachian orogeny with most watersheds in the Piedmont Province or Coastal Plains in the Southeast and Mid-Atlantic regions or in the Seaboard Lowlands in the New England region. Most watersheds are underlain primarily by silicate bedrock and surficial materials (Table S1, more detail below). We also included four watersheds in the Mid-Atlantic that are underlain by carbonate bedrock as a contrast to the SC and [Cl] patterns observed in silicate watersheds.

We included five Mid-Atlantic sites with <3 water years (defined as October–September) of high-frequency SC that added an important component to our study dataset (Table S4). Four sites (watersheds #45, 46, 56, 57) had 2 years of data but were included to ensure adequate representation of sites with low impervious surface cover (ISC) sites and higher forest cover, constituting four of the eight Mid-Atlantic sites with <5% ISC (Table S2). Watersheds #56 and #57 had substantially higher forest cover than any other Mid-Atlantic sites with 77.5% and 71.8%, respectively (Table S2). In contrast, the other low ISC sites in the Mid-Atlantic had substantial to dominant agricultural land use. We included one carbonate site with 2 water years of data (watershed #69), which was the only carbonate site with more than 1.75% ISC. One applicable Mid-Atlantic site (USGS gage #01645704) was not included because it was not discovered until late in the process.

We excluded three sites in New England (USGS gages #01104430, 01104453, and 01104480) because discharge at the sites was clearly affected by the upstream impoundments. We did include a site in New England (USGS gage #01104460) with an impoundment upstream where the majority of the watershed (69%) was unimpounded based on data from USGS StreamStats.[1] We also considered inclusion of a Mid-Atlantic watershed affected by acid mine drainage (Guest River, USGS gage #03524500) but decided that a single site would not serve as an adequate contrast to the SC and [Cl] patterns observed in watersheds underlain by silicate bedrock. A number of watersheds initially appeared to meet our criteria but were subsequently excluded because SC data were not collected in the winter (e.g., USGS gage #01480617 in the Mid-Atlantic and #01195510 in New England) or because the SC data did not meet our 85% threshold for winter data coverage in any water year (gage #01168250 in New England for all 3 years of record).

Compilation of data for watershed characteristics. Watershed boundaries were delineated using the NHDPlus Version 1 flow direction grids and the ArcGIS 10.6 watershed tool.[2,3] (Please nnote that any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.) In cases where these data were insufficient to accurately define watershed boundaries, such as for very small watersheds, boundaries were delineated with StreamStats[1] or digitized manually using topographic maps. We compiled basic watershed characteristics including USGS gage number, location, watershed area, relief, and bedrock geology (Table S1).[3,4] We also compiled climate data (temperature, precipitation, and percentage of precipitation as snow), land cover data including impervious surface cover, road length, and road crossings (Table S2).[3,5-8] We also calculated road salt application rates in each watershed for three calendar years (1999, 2011, 2015) based on annual estimates at the scale of 1 km^2 (Table S3).[9] Data for watershed characteristics are available via USGS data release.[10]

USGS data QA/QC, data download and selection for study sites. USGS samples and data were collected following well-established and vetted procedures, including a rigorous QA/QC process before public release and publication. When the USGS initially posts high-frequency data such as specific conductance on the USGS National Water Information System (NWIS) database, the data are listed as provisional.[11,12] Within several months of posting, all data undergo a QA/QC process to finalize the data as approved. The minimum [Cl] reported for any discrete sample was 0.59 mg/L, which is well above the detection limit of 0.02 mg/L listed in USGS methods.[13] We performed our final download of all data from the NWIS database in late February to early March 2019 with some water year 2018 data still provisional at that point.[14] Discrete water chemistry samples classified as “regular” or “replicate” (USGS sample type code = “9” or “7”, respectively) were included in our study. The number of high-frequency SC observations are 11, 9.0, and 8.8 million for the Southeast, Mid-Atlantic (silicate), and New England, respectively, with an additional 0.8 million for the Mid-Atlantic carbonate sites.

Inclusion of SC (and [Cl]) data from water years with <85% winter data coverage. At four Mid-Atlantic sites, we included data from water years where the winter high-frequency data were <85% complete: 2011 for site #60 where 82.1% of SC data were recorded for 1 of the 3 years of data (otherwise the site would have been excluded), 2017 for site #48 where 75.6% recorded for 1 of the 2 years of high-frequency data for this site, 2016 for site #57 where 74.6% recorded for 1 of the 2 years of high-frequency data for this site, and 2014 for sites #51 and #52 where because the 2 years of high-frequency data at this site did not line up with water years (high-frequency data from mid-2014 to mid/end-2016). As mentioned previously, sites #51 and 52 are the only Mid-Atlantic sites with >70% forest cover (and only three others had forest >50%, Table S2) with a maximum [Cl] of 26 mg/L recorded from high-frequency data (Table S6) so we are not concerned that that [Cl] peaks were missed.

Regression models for discrete [Cl] versus SC and generating high-frequency [Cl] using SC as a surrogate. We calculated single linear regression (SLR) models for each site that included all discrete [Cl] and SC data from NWIS using the *lm* function in R. While only the discrete data from 1998 to 2018 were reported in the statistical summaries (Table S5), all discrete data were used in the regression models and are publicly available via a USGS data release.[10] We used the SLR models and the *segmented* library to calculate piecewise regression models for each site. S*egmented* compares the SLR and the data to generate two slopes and a breakpoint between the slopes with associated uncertainty. We used piecewise regression models in cases where the fit had a R^2 value that was at least 0.02 higher than the SLR model. In some cases, the piecewise models did not converge. In a few cases, we discarded the piecewise models because they were not plausible geochemically, which generally occurred when only one or two data points were above the breakpoint.

After calculating SLR and piecewise models using all discrete data, we examined the [Cl] versus SC SLR models for outliers. We identified outliers, or apparent outliers, as samples with standardized residuals >±4 (with 1 exception where <–3) and among the top 3 or 4 samples for Cook’s distance and/or leverage. Apparent outlier samples were retained in our regression models in the following cases. First, the samples were among the high SC and [Cl] samples for the site, the piecewise model was a better fit, and the slope of the second (higher) regression was geochemically plausible. Second, we retained outlier samples where the R^2 for the SLR models with all samples ≥0.99 and the site had >100 discrete samples. All samples excluded from SLR and piecewise regression models for individual watersheds also were excluded from the regional piecewise regression models. The Mid-Atlantic regional model uses data from only the silicate watersheds. Only 31 discrete samples, out of 6432 in total, and no more than two per site were excluded from the regressions (Table S14). Outlier samples also were excluded from the statistical summaries of the discrete samples (Table S5, S6, S8, S9). The final versions of the regression models are reported here (Table S10) as well as in a USGS data release.[10]

Predicted [Cl] based on watershed-specific models are a good match for observed discrete [Cl] with consistently high R^2 values (median = 0.93) and small median concentration differences (see Supporting Information, Table S10). We did not validate the models. The foremost reason for not validating the models is that a number of watersheds had few samples (<30), which would have made it challenging to withhold samples to use for validation. A major focus of this work was including as many watersheds as possible.

When we used the regional piecewise regression models to calculate [Cl] for sites with little or no discrete [Cl] data, occasional very low SC values in the high-frequency data predicted "negative" [Cl]. To address that issue, in place of all predicted "negative" [Cl] values, we substituted a [Cl] value that was one half of the lowest measured discrete value for each region. The minimum [Cl] measured in the discrete samples in our dataset for the Southeast, Mid-Atlantic, and New England were 0.59, 1.4, and 5.5 mg/L, respectively (Table S5), resulting in the minimum possible values for high-frequency [Cl] of 0.30, 0.7, and 2.8 mg/L, respectively (Table S6, S8, S9). All predicted [Cl] values are publicly available in a USGS data release, including values predicted with regional single-linear and piecewise regression models for all watersheds and watershed-specific single-linear and piecewise regression models where applicable.[10]

A few items of note about the data and the relationship between [Cl] and SC. First, discrete SC data collected by the USGS are frequently reported as the average of the SC measured at several points across the stream. In contrast, high-frequency SC data are collected at a single point by an *in-situ* sensor.

Second, the relationship between [Cl] and SC varies among regions with median slope values of 0.07, 0.24, and 0.29 for watershed-specific regression models for the Southeast, Mid-Atlantic, and New England, respectively. Thus regionally appropriate relationships for [Cl] and SC should be used when using SC to estimate [Cl] in streams. Many of the study watersheds were suburban and urban with frequent application of deicing salts, meaning that [Cl] could expected be a substantial contributor to the SC. In other settings, e.g., watersheds where water chemistry is affected by acid-mine drainage or mountain-top mining, this relationship would be expected to be much different with other ions contributing much more to SC. The relationship between [Cl] and SC might be expected to be most variable in watersheds with low ISC, perhaps less than 2–3%, where many ions contribute. The [Cl]–SC relationship also will likely vary to some degree across different types of bedrock though that variation is likely to be largely masked by deicing salt contributions in watersheds with higher ISC and when [Cl] exceeded 100 mg/L. Additionally, if discrete samples for [Cl] and SC are only collected in a particular watershed in non-winter months, then the slope of the [Cl]-SC is likely to be underestimated at [Cl] greater than 100 mg/L with the result that [Cl] exceedances may be underestimated. In such a case, the slope of a regional [Cl]-SC relationship may be better suited to estimating [Cl] exceedances until a watershed-specific relationship can be established.

Third, the largest mismatch between predicted and observed [Cl] on a percentage basis occurs at concentrations less than 50–75 mg/L where non-Cl ions frequently contribute more to the SC, particularly for the regional regression models (Figure S2). Predicted [Cl] often are overestimated using the regional relationship at these lower concentrations but match well at [Cl] >100 mg/L. Thus the use of regional relationships to calculate high-frequency [Cl] at concentrations <50–75 mg/L should be viewed as semi-quantitative where [Cl] may be within 20-30 mg/L of measured values, which could represent up to 60–80% difference on a percentage basis, e.g., observed [Cl] of ~10 mg/L and predicted [Cl] of ~18 mg/L or observed [Cl] of ~30 mg/L and predicted [Cl] of ~50 mg/L.

Fourth, for the Southeast and Mid-Atlantic regions, no more than 13% of the discrete data in the regional relationships are from a single watershed. For the New England region, a substantial plurality of the discrete SC and [Cl] in the regional relationship are from one watershed (watershed #81, 490 of 1636 or 30%, Table S10).

Fifth, the [Cl] versus SC slope for watershed-specific and regional models in the Mid-Atlantic and New England ranges from 0.1 – 0.23 when SC values are <300–400 μS/cm and [Cl] <50–60 mL, but the slopes increase to ~0.3 at higher SC and [Cl] values. [Cl] versus SC slopes remain at ~0.3 up to concentrations of approximately 14,000 μS/cm. In New England where the highest [Cl] and SC values were measured, the [Cl] versus SC slope increased further at very high concentrations with a slope of 0.39 for samples with SC >14,000 μS/cm when discrete samples with SC >20,000 μS/cm were included in the regional regression (Figure S5, Table S10).

Discrete SC and [Cl] data from 1997 and earlier were excluded from the main statistical summary (Table S5) and plots (Figure 3, S1A) to facilitate the comparison of discrete and high-frequency data over a similar period. For several watersheds, a substantial number of samples (as many as 360) were collected before 1997 when SC and [Cl] concentrations were generally lower than 1998–2018, e.g., watershed #81 with median [Cl] of 28 mg/L before 1997 versus 48.6 mg/L from 1998–2018 (Table S5, S15).

Calculating exceedances of U.S. Environmental Protection Agency (EPA) chloride criteria. We calculated exceedances of the EPA chronic criterion for Cl at 230 mg/L as follows. First, we calculated daily average (mean) [Cl] from high-frequency [Cl]. Second, we calculated rolling 4-day averages based on the daily average [Cl] using the *rollmean* function from the *zoo* library in *R* with right-aligned rolling averages. Third, we calculated the sum of 4-day periods for which the average [Cl] exceeded 230 mg/L in each water year. Fourth, we extracted the data for each exceedance event with an event being constituted of consecutive 4-day periods exceeding the EPA chronic criterion. We used those data to characterize each event, including event duration, maximum [Cl] from the unaveraged high-frequency data, and mean [Cl] during the event from the unaveraged high-frequency data. For each water year, we calculated the number of events per year, the time between events when multiple events occurred, and the end date of the last event in each year. We calculated statistical summaries of exceedance events and annual statistics for chronic exceedances (Table S12).

As an example, the daily average [Cl] for watershed #55 (from Mid-Atlantic) was 107, 80.5, 25.6, 63.6, 910, and 722 mg/L for December 5–10, 2013. The 4-day [Cl] average for December 5–8, 2013 was 69.2 mg/L, which was reported as the 4-day average for December 8, 2013 using a right-aligned rolling average, and the 4-day averages ending on December 9 and 10, 2013 were 270 and 430 mg/L, respectively. Thus, for this 6-day period, we reported two exceedances of the EPA chronic criteria. We calculate the average [Cl] for the period of exceedance from the high-frequency [Cl], as would be the case for an entire event, from December 6 at 0:00 to December 10 at 23:55 as 318 mg/L, which is somewhat lower than the average [Cl] of 360 mg/L based on the daily [Cl] values for those days.

We calculated exceedances of the EPA acute criterion for Cl at 860 mg/L similarly to our approach for chronic exceedances. First, we calculated hourly average (mean) [Cl] from high-frequency [Cl]. Second, we calculated the sum of hours for which the average [Cl] exceeded 860 mg/L in each water year. We calculated statistics for acute exceedance events and for each water year following our approach for chronic exceedance events (Table S13).

Results and Discussion

Description of watershed characteristics. The Southeast sites are located in the Piedmont physiographic province and underlain primarily by felsic crystalline bedrock with no watershed having <79% felsic bedrock and only three with >10% mafic bedrock (Table S1). Most Mid-Atlantic silicate sites are located in the Piedmont and Coastal Plain physiographic provinces (13 and 10, respectively), including 2 watersheds with >20% of their area in both provinces based on the bedrock geology (Table S1). Piedmont watersheds are primarily underlain by felsic silicate crystalline bedrock, and Coastal Plain watersheds are underlain by unconsolidated sediments that were sourced from the Piedmont and other geologic provinces to the west of the Coastal Plain. Three Mid-Atlantic sites are underlain by silicate sedimentary bedrock along with some mafic bedrock indicating the watersheds are in Triassic rift basins. The four Mid-Atlantic carbonate sites are underlain by carbonate bedrock with silicate sedimentary rock constituting 38.7% of the watershed area in one case (Table S1). The New England sites are dominantly underlain by felsic bedrock with the exception of two sites (Table S1). One site is underlain by silicate sedimentary bedrock in a Triassic rift basin and another has 17.3% carbonate bedrock (marble).

Land cover for the Southeast sites is mostly developed land and forest with 7 of 39 sites having >10% of the watershed area in pasture and no sites with >0.5% of cultivated crops (Table S2). In the Mid-Atlantic, pasture and cropland are more common with several sites having >5% cultivated crops (Table S2). Land cover in New England is primarily developed land and forest with less pasture or cultivated cropland cover than the Southeast or Mid-Atlantic (Table S2).

Best subset regression. Best subset regression generates and compares the correlation of median annual [Cl] with a set of response variables using single- and multiple-linear regression models. Our response variables are compiled in a USGS data release and include median SC values for water year, winter, and non-winter; median [Cl] values for water year, winter, and non-winter; the 5% and 95% quartiles along the difference between those quartiles; median days and hours for chronic and exceedance events, respectively, along with the median duration of each type of event, the number of events per year, and the median number of days since the last event.[10] We used the *leaps* package in R[15] to generate and display the models with the highest R^2 values based on five or six response variables: mean annual temperature, watershed area, developed land (as defined by the National Land Cover Database 2011)[6], ISC, road length (km / km^2 of watershed), (estimated) average salt application rate (kg / km^2). We performed three subset regressions with all six response variables: all 93 sites, Mid-Atlantic sites, and New England sites (Figure S4A, S4C, S4E). We performed another set of three subset regressions for the same geographic areas but did not include the average salt application rate (Figure S4B, S4D, S4F). Across all 93 sites, a regression model with median annual [Cl] and only average salt application rate had a R^2 of 0.69 (Figure S4A). The inclusion of additional response variables only minimally increased the R^2 to 0.72. When salt application rates are excluded from the best subset regression, the median [Cl] across all sites was explained reasonably well with mean annual temperature and road length (Figure S4B).

[Cl] exceedances, additional discussion. In Southeast streams from 2008 to 2018, 20 of the 23 periods when [Cl] was greater than 230 mg/L occurred in January and February with two additional events in December or March. Ten of the 23 periods occurred in clusters with [Cl] >230 mg/L measured at 4–5 sites on January 12–14, 2011 and on January 22, 2018 following snowfall at Atlanta Hartsfield International Airport within the previous 3–5 days. Although occasional inputs of Cl from sodium hypochlorite used to treat water in combined sewer overflows in Southeast watersheds (#1 and #9) have been proposed to explain high [Cl] in discrete samples collected in 2003–2007,[16] if such inputs occurred during the study period, they do not seem to drive sustained elevation of [Cl] or exceedances of [Cl] criteria.

Temporal changes, additional discussion. The highest [Cl] watershed in the Mid-Atlantic (#46) is an unusual watershed in that it has been defined as a "sewershed" with much of the water entering the watershed via sewer systems and storm pipes. Additionally, 30% of the watershed is classified as having industrial land cover. Thus, temporal dynamics for [Cl] in this watershed are likely to have different controls than most Mid-Atlantic sites.

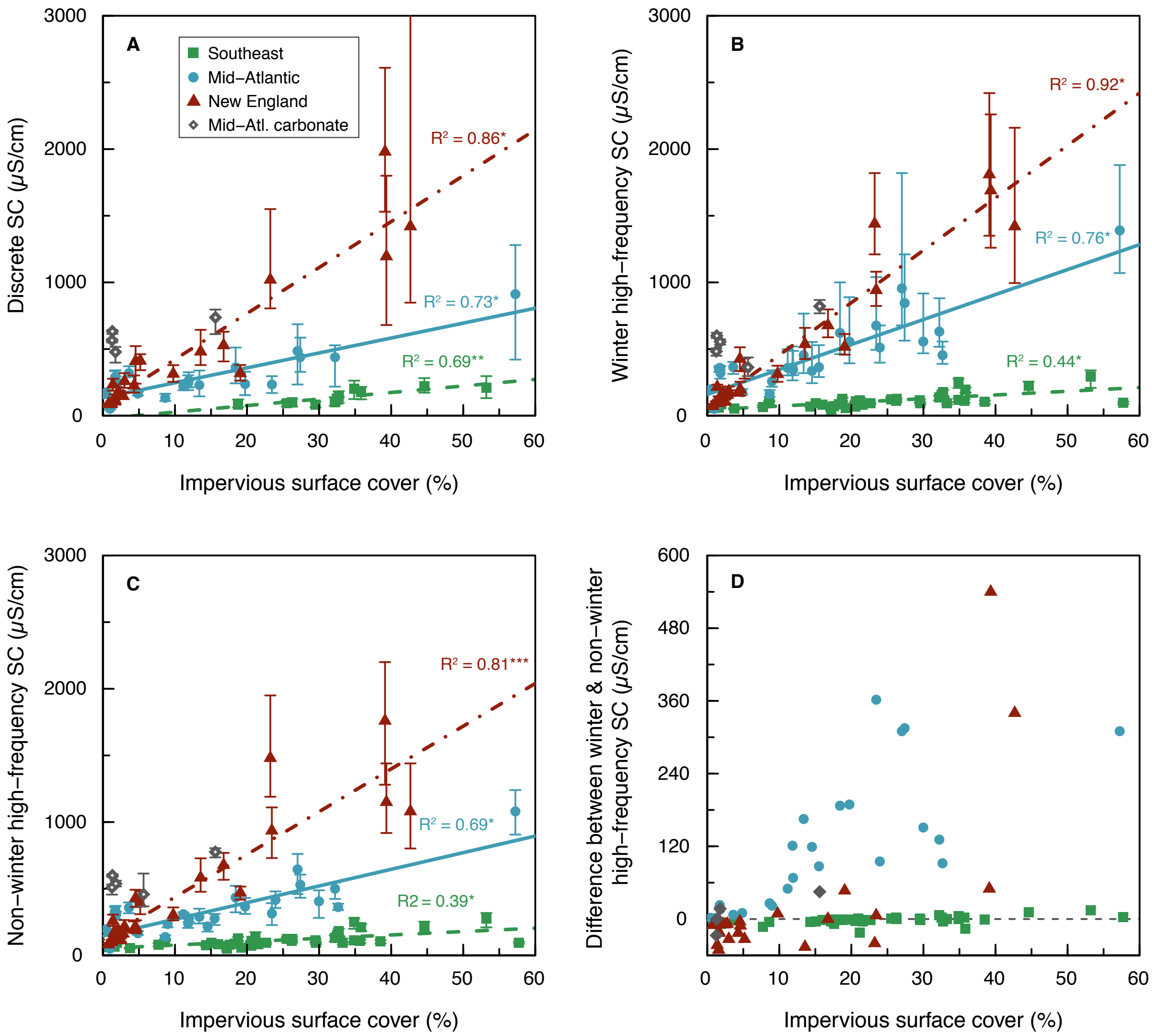

Figure S1. (A) Discrete SC (for 1998–2018) versus impervious surface cover (ISC) for each of the three regions and for four Mid-Atlantic carbonate watersheds. The 75th percentile for discrete SC for the highest ISC New England site is 8010 µS/cm. (B) Winter and (C) non-winter high-frequency specific conductance (SC) versus ISC. The symbols represent median values with uncertainties given as the interquartile range (25th to 75th percentile). (D) The difference between median winter and non-winter SC versus ISC. The median number of high-frequency SC observations per watershed was ~305,000, which at a collection interval of 15 minutes represents 8.7 years of data; for a site with 305,000 observations, the 5th percentile represents 15,250 observations. R^2 values (adjusted R^2 as reported in R) for regressions are shown along with statistical significance indicated as * $p < 0.001$, ** $p < 0.01$, *** $p < 0.05$, ^ $p < 0.1$. Full equations for regressions in plots are reported in Table S7.

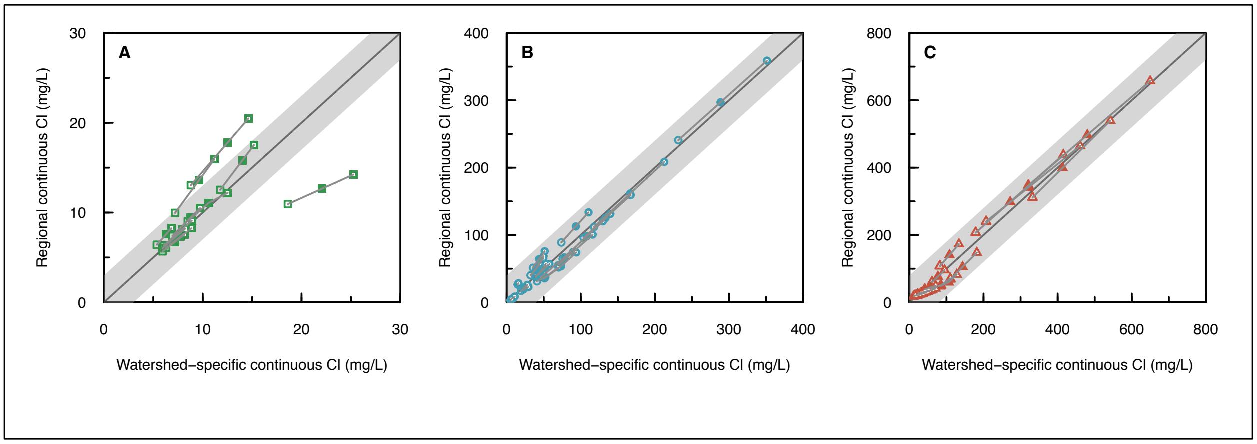

Figure S2. Predicted high-frequency [Cl] for the 25% quantile (open square, thin line), median (filled square), and 75% quartile (open square, thick line) calculated from the regional piecewise regression versus the watershed-specific regression relationships for [Cl] versus SC for the (A) Southeast, (B) Mid-Atlantic, and (C) New England regions (n = 10, 20, 21, respectively). The grey shaded area represents approximately 10% uncertainty from the 1:1 line. The points for individual sites are connected with lines. Predicted [Cl] is similar between regional versus watershed-specific relationships with greater similarity at concentrations higher than 150–200 mg/L.

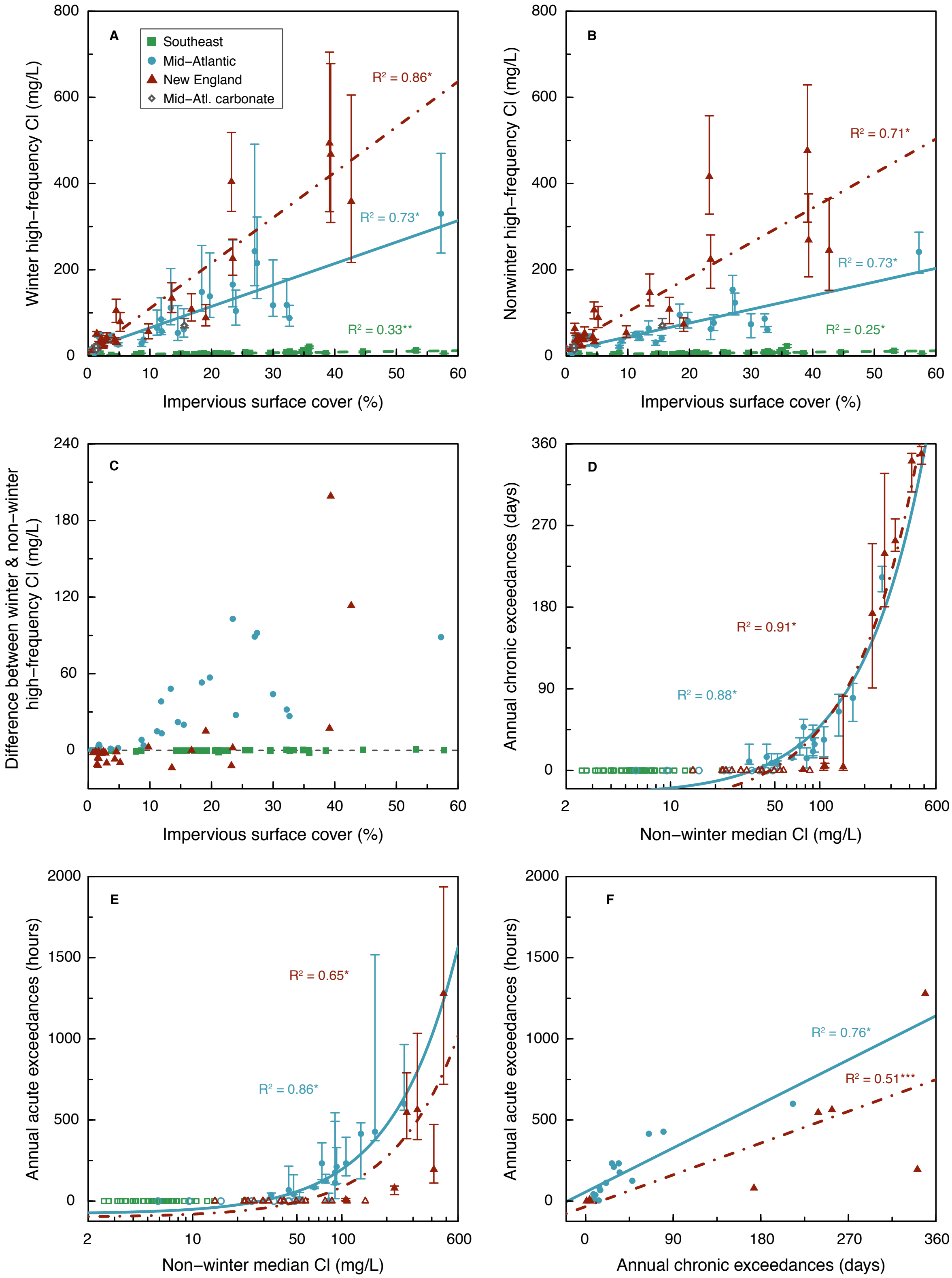

Figure S3. (A) Winter [Cl] and (B) non-winter [Cl] versus impervious surface. The symbols represent median values with uncertainties given as the interquartile range (25th to 75th percentile). (C) Difference between median winter and median non-winter [Cl] versus impervious surface cover. (D) Chronic exceedances versus median non-winter [Cl]. (E) Acute exceedances versus median non-winter [Cl]. (F) Median acute versus median chronic exceedances

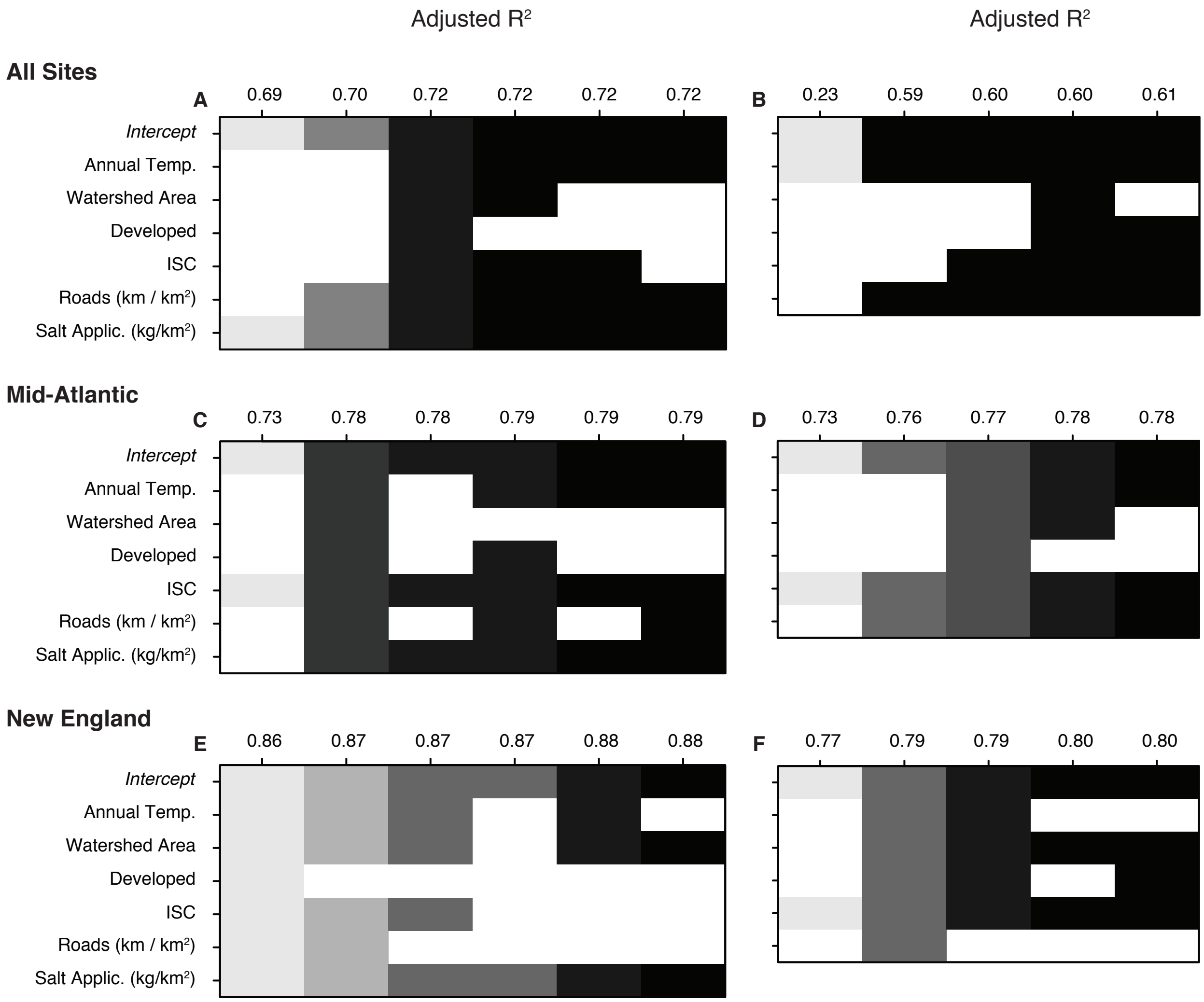

Figure S4. (A) Best subset regression for land cover characteristics and average deicing salt application rates across all three regions. (B) Best subset regression for land cover characteristics only (salt application rates excluded) across all three regions. (C) Best subset regression for land cover characteristics and average deicing salt application rates for the Mid-Atlantic. (D) Best subset regression for land cover characteristics only (salt application rates excluded) for the Mid-Atlantic. (E) Best subset regression for land cover characteristics and average deicing salt application rates for New England. (F) Best subset regression for land cover characteristics only (salt application rates excluded) for New England. Shaded boxes within each column are included in the single or multiple linear regression for which the R^2 value displayed at the top of the column. Boxes are shaded with different intensities to help distinguish between different groups of models.

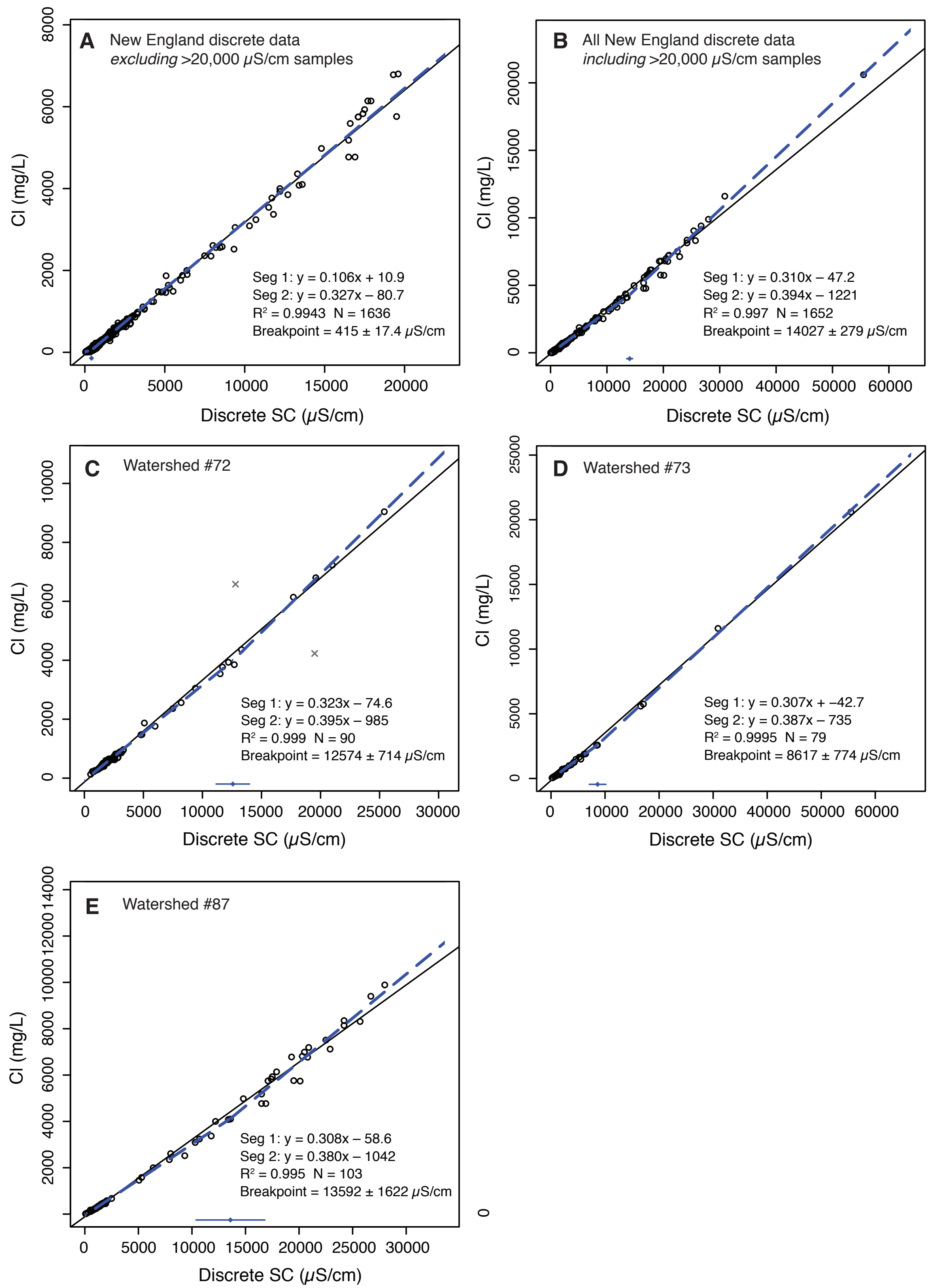

Figure S5. Discrete [Cl] versus SC with piecewise regression models for (A) all discrete data from the New England region, including samples >20,000 µS/cm, (B) watershed #72, (C) watershed #73, and (D) watershed #87 from New England. Circles represent discrete data, "x" symbols represent discrete data excluded from the regression model, and the blue diamond and associated line represent the breakpoint and uncertainty. The blue dashed line is the piecewise regression model, and the solid black line is the single linear regression model. In all of these models, the slope of [Cl] versus SC increases from ~0.30 to ~0.39 at higher [Cl] and SC.

Table S1. Watershed characteristics 1 – location, area, relief, geology

USGS Gage #	Short name for USGS site	Site #	Latitude	Longitude	Watershed area	Relief	Felsic igneous & meta-morphic[a]	Mafic	Ultra mafic	Carbonate[b]	Silicate sedimentary	Unconsolidated sediments	Water[c]
					km^2	m	%	%	%	%	%	%	%
Southeast													
02203700	IntrenCr	1	33.68900	-84.33048	27.68	85.5	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02336360	NancyCr1	2	33.86917	-84.37889	68.03	104.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02336410	NancyCr2	3	33.83844	-84.43937	96.46	120.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02336120	NFPeachC	4	33.83149	-84.34270	91.23	94.3	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02336300	PeachCrk	5	33.82031	-84.40764	222.90	110.4	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02336526	ProctorC	6	33.79427	-84.47437	35.13	135.8	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02336240	SFPeachC	7	33.80288	-84.34076	71.60	88.1	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02203603	SouthRi2	8	33.68389	-84.41528	6.26	63.4	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02203655	SouthRiv	9	33.67900	-84.35798	59.60	90.8	93.8	0.0	6.2	0.0	0.0	0.0	0.0
02336728	UtoyCrek	10	33.74344	-84.56827	90.02	99.3	99.8	0.0	0.0	0.0	0.0	0.0	0.2
02336313	WoodCrk	11	33.82177	-84.43882	7.20	75.2	100.0	0.0	0.0	0.0	0.0	0.0	0.0
Southeast high-frequency only													
02208150	AlcovRiv	12	33.91761	-83.88796	79.76	125.0	80.4	19.6	0.0	0.0	0.0	0.0	0.0
02218565	ApalaRiv	13	34.01038	-83.89407	14.82	79.8	79.3	20.7	0.0	0.0	0.0	0.0	0.0
02207385	BigHaCrk	14	33.81511	-83.99019	44.62	81.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02207400	BruFkCrk	15	33.82150	-83.94241	21.26	61.1	100.0	0.0	0.0	0.0	0.0	0.0	0.0
023362095	BurnFCrk	16	33.82139	-84.27472	8.23	53.0	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02203873	CobbsCrk	17	33.70955	-84.23937	20.88	89.1	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02208493	CorniCrk	18	33.62850	-83.79886	70.05	94.2	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02335350	CrookCrk	19	33.96510	-84.26492	22.92	74.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02337410	DogRiver	20	33.65381	-84.82103	171.97	157.6	99.1	0.9	0.0	0.0	0.0	0.0	0.0
02203831	DooliCrk	21	33.70566	-84.29242	10.76	79.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02334578	LevelCrk	22	34.09649	-84.07963	13.12	63.1	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02344630	LineCrek	23	33.35762	-84.58243	180.80	92.9	89.2	10.6	0.0	0.0	0.0	0.0	0.1
02207135	LittStCk	24	33.83066	-84.13936	5.67	65.8	99.8	0.0	0.2	0.0	0.0	0.0	0.0
02336340	NancyCr3	25	33.89788	-84.34548	45.26	94.2	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02336030	NFPeach1	26	33.90566	-84.22492	3.69	44.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02207185	NoBusCrk	27	33.77816	-84.03797	26.11	136.0	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02217643	ParksCrk	28	34.16206	-83.52509	7.89	68.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02204037	PoleBCrk	29	33.66844	-84.15103	41.65	107.8	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02334480	RichlCrk	30	34.13260	-84.06991	24.33	93.0	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02101726	RockyRiv	31	35.73514	-79.42308	178.55	104.4	92.4	0.0	7.6	0.0	0.0	0.0	0.0
02336152	SFPeach2	32	33.81044	-84.24797	14.77	55.5	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02203863	ShoalCrk	33	33.69319	-84.25389	22.50	103.1	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02203950	Snapfin1	34	33.76344	-84.22020	34.21	75.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02203960	Snapfin2	35	33.69667	-84.19861	84.00	105.9	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02207160	StoneMCk	36	33.77344	-84.07714	72.48	286.8	96.7	0.0	3.3	0.0	0.0	0.0	0.0
02334885	SuwanCrk	37	34.03232	-84.08936	121.89	116.3	100.0	0.0	0.0	0.0	0.0	0.0	0.0

USGS Gage #	Short name for USGS site	Site #	Latitude	Longitude	Watershed area	Relief	Felsic igneous & meta-morphic[a]	Mafic	Ultra mafic	Carbonate[b]	Silicate sedimentary	Unconsolidated sediments	Water[c]
					km^2	m	%	%	%	%	%	%	%
02205865	SweetwCk	38	33.94111	-84.10778	54.53	98.8	100.0	0.0	0.0	0.0	0.0	0.0	0.0
02217274	WheelCrk	39	34.08233	-83.85462	4.30	63.4	98.3	1.7	0.0	0.0	0.0	0.0	0.0
Mid-Atlantic													
01654000	Accotink	40	38.81289	-77.22832	61.68	90.2	97.5	0.0	2.4	0.0	0.0	0.1	0.0
01493112	ChesteBr	41	39.25706	-75.94014	17.12	18.9	0.0	0.0	0.0	0.0	0.0	100.0	0.4
01573695	ConewCr1	42	40.19528	-76.56778	52.83	220.3	0.0	21.7	0.0	0.0	78.3	0.0	0.0
01573710	ConewCr2	43	40.15111	-76.68944	122.52	255.0	0.0	21.1	0.0	0.0	78.9	0.0	0.0
01646000	DifficuR	44	38.97594	-77.24581	149.81	112.6	92.3	0.0	4.3	0.0	0.4	2.6	0.0
01585075	FosterBr	45	39.40853	-76.34264	4.73	70.7	0.0	0.0	0.0	0.0	0.0	100.0	0.0
01651770	HickeyRu	46	38.91681	-76.96922	2.58	62.7	0.0	0.0	0.0	0.0	0.0	100.0	0.0
01636845	LitCato1	47	39.34117	-77.63397	10.86	236.5	82.7	17.3	0.0	0.0	0.0	0.0	0.0
01636846	LitCato2	48	39.33761	-77.62647	11.86	243.1	84.2	15.8	0.0	0.0	0.0	0.0	0.0
01408000	ManRivSq	49	40.16139	-74.15472	113.49	75.9	0.0	0.0	0.0	0.0	0.0	100.0	0.0
01658000	MattawCr	50	38.59614	-77.05603	143.74	60.9	0.0	0.0	0.0	0.0	0.0	100.0	0.0
0166818623	MillCrk1	51	38.12961	-77.23217	36.60	68.1	0.0	0.0	0.0	0.0	0.0	100.0	0.0
0166818985	MillCrk2	52	38.12894	-77.22350	73.10	69.7	0.0	0.0	0.0	0.0	0.0	100.0	0.0
01649500	NeBrAnac	53	38.96025	-76.92597	188.06	165.0	21.4	0.0	0.0	0.0	0.0	78.6	0.0
01649190	PaintBra	54	39.03314	-76.96428	33.96	123.5	91.9	0.0	0.0	0.0	0.0	8.1	0.0
01581752	Plumtree	55	39.49628	-76.34744	6.46	74.7	80.4	19.6	0.0	0.0	0.0	0.0	0.0
01648010	RockCree	56	38.96017	-77.04206	169.01	143.3	97.5	0.0	1.4	0.0	0.0	1.1	0.0
01408029	SFLiDifR	57	38.90889	-77.33826	7.15	68.8	95.6	0.0	0.0	0.0	4.4	0.0	0.0
01650800	SligoCre	58	38.98622	-77.00486	16.71	93.0	99.0	0.0	0.0	0.0	0.0	1.0	0.0
01651800	WattsBra	59	38.90128	-76.94328	8.71	85.5	0.0	0.0	0.0	0.0	0.0	100.0	0.0
Mid-Atlantic high-frequency only													
01585219	Accotin2	60	38.86624	-77.28638	10.32	54.7	85.0	0.0	15.0	0.0	0.0	0.0	0.0
01646305	DeadRunW	61	38.95978	-77.17567	6.25	59.2	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01656903	FlatlicB	62	38.88239	-77.43191	10.29	68.4	0.2	0.0	0.0	0.0	99.8	0.0	0.0
01585219	HerringR	63	39.31796	-76.55513	42.29	142.9	47.1	3.9	0.0	0.0	0.0	49.0	0.0
01654500	LongBran	64	38.81095	-77.23498	9.68	78.0	100.0	0.0	0.0	0.0	0.0	0.0	0.0
0165389205	ManRivAl	65	40.14667	-74.12222	164.30	84.1	0.0	0.0	0.0	0.0	0.0	100.0	0.0
Mid-Atlantic carbonate													
015765195	BigSprgR	66	39.99594	-76.26404	4.49	59.0	1.6	0.0	0.0	98.4	0.0	0.0	0.0
01616100	DryMarsR	67	39.19259	-78.06863	28.43	53.4	0.0	0.0	0.0	95.3	4.7	0.0	0.0
01632900	SmithCrk	68	38.69345	-78.64279	241.74	731.5	0.0	0.0	0.0	61.3	38.7	0.0	0.0
01636316	SpoutRun	69	39.06703	-78.00375	57.28	75.9	0.0	0.0	0.0	96.3	3.7	0.0	0.0
New England													
01115275	BearTBrk	70	41.78260	-71.67479	1.62	105.5	94.9	0.0	0.0	0.0	0.0	0.0	5.1

USGS Gage #	Short name for USGS site	Site #	Latitude	Longitude	Watershed area	Relief	Felsic igneous & meta-morphic[a]	Mafic	Ultra mafic	Carbonate[b]	Silicate sedimentary	Unconsolidated sediments	Water[c]
					km^2	m	%	%	%	%	%	%	%
01104410	CamTrib1	71	42.43648	-71.26422	0.99	36.0	99.9	0.1	0.0	0.0	0.0	0.0	0.0
01104415	CamTrib2	72	42.43593	-71.26006	1.15	50.8	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01104420	CamTrib3	73	42.41982	-71.25756	1.87	49.5	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01115280	CorkBrok	74	41.80399	-71.64979	4.68	86.7	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01115190	DollyBrk	75	41.82232	-71.70035	12.88	114.7	99.6	0.0	0.0	0.0	0.0	0.0	0.4
01115265	HemloBrk	76	41.79065	-71.69868	21.67	102.4	99.9	0.0	0.0	0.0	0.0	0.0	0.1
01104405	HobbsBrk	77	42.43648	-71.26950	5.03	57.0	80.9	19.1	0.0	0.0	0.0	0.0	0.0
01115110	HuntiBrk	78	41.84677	-71.61173	15.68	119.2	99.7	0.0	0.0	0.0	0.0	0.0	0.3
01115098	PeeptBrk	79	41.85260	-71.60618	13.18	120.1	99.2	0.0	0.0	0.0	0.0	0.0	0.8
01115187	PonagRiv	80	41.81871	-71.70507	38.83	149.1	97.5	0.0	0.0	0.0	0.0	0.0	2.5
01196500	QuinnRiv	81	41.45026	-72.84128	285.28	309.0	16.2	0.8	0.0	0.0	82.9	0.0	0.0
01115183	QuonaBrk	82	41.79760	-71.58479	5.07	72.3	99.3	0.7	0.0	0.0	0.0	0.0	0.0
01115114	RushBrok	83	41.83760	-71.61201	12.39	130.8	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01201487	StillRiv	84	41.46582	-73.40320	155.09	248.6	73.7	9.0	0.0	17.3	0.0	0.0	0.0
01095220	StillwaR	85	42.41093	-71.79118	78.80	486.7	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01104460	StonBR20	86	42.36899	-71.27061	57.11	140.1	97.0	3.0	0.0	0.0	0.0	0.0	0.0
01104455	StonBWal	87	42.37260	-71.27033	1.30	114.6	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01104475	StonBWes	88	42.35454	-71.26811	2.04	83.5	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01104370	StonyBrk	89	42.38556	-71.28944	27.00	87.2	97.3	2.7	0.0	0.0	0.0	0.0	0.0
01115276	WestcStr	90	41.78538	-71.66729	14.18	57.6	70.5	0.0	0.0	0.0	0.0	0.0	0.0
New England high-frequency only													
01095434	GatesBrk	91	42.36454	-71.77535	8.22	137.4	97.8	0.0	0.0	2.2	0.0	0.0	0.0
011277905	LatimBrk	92	41.36861	-72.20222	45.81	179.3	100.0	0.0	0.0	0.0	0.0	0.0	0.0
01095375	QuinaRiv	93	42.37287	-71.82813	115.38	310.8	99.1	0.9	0.0	0.0	0.0	0.0	0.0

[a] In six cases, intermediate bedrock (named diorite or intermediate) was mapped as present with >50% diorite mapped for sites #72, #72, #87.

[b] For watersheds #67–#69, the carbonate bedrock is mapped as a mixture of limestone and dolostone. For watersheds #84 and #91, the carbonate bedrock is mapped as marble.

[c] The bedrock geology data are mapped at a coarser scale than the land cover data in Table S2. While the open water data in Table S2 are more accurate than the water column in this table, this column is included for the sake of completeness, *i.e.*, so that the bedrock geology categories add up to 100%.

Table S2. Watershed characteristics 2 – climate and land cover

Site #	Mean annual temp.	Mean Dec – Mar temp.	Mean annual precip.	Annual precip. as snow	Developed -low	Developed -high	Forest	Grass -land & shrub	Pasture	Cultiv- ated crops	Wet- lands	Open water	Imperv- ious surface cover	Road length	Road crossings
	°C	°C	mm	%	%	%	%	%	%	%	%	%	%	km / km²	crossings / km²
Southeast															
1	16.21	7.86	1294	2.00	52.0	31.4	12.5	1.1	2.7	0.0	0.0	0.1	35.84	13.26	0.397
2	16.00	7.26	1329	3.07	55.1	25.7	17.7	0.1	0.1	0.0	0.4	0.9	29.49	9.40	1.323
3	16.06	7.38	1325	2.99	53.2	21.5	23.8	0.2	0.2	0.0	0.3	0.7	25.51	8.98	1.213
4	16.09	7.40	1330	3.01	54.3	28.5	15.7	0.1	0.1	0.0	0.9	0.4	32.17	10.26	1.217
5	16.12	7.56	1322	2.64	53.9	29.4	15.7	0.1	0.1	0.0	0.5	0.3	32.55	11.34	1.144
6	16.22	7.84	1307	2.00	53.0	30.6	13.1	1.7	1.0	0.0	0.1	0.1	34.87	12.57	0.712
7	16.09	7.57	1319	2.66	58.3	21.1	19.6	0.2	0.2	0.0	0.4	0.2	26.25	10.61	1.187
8	16.31	7.94	1277	2.00	52.8	40.3	6.9	0.0	0.0	0.0	0.0	0.0	44.60	12.06	0.799
9	16.29	8.03	1278	2.00	51.0	28.2	19.0	0.7	0.2	0.0	0.8	0.1	32.73	11.22	0.956
10	16.30	7.98	1297	2.00	51.6	12.8	33.4	1.1	0.2	0.0	0.4	0.5	18.76	8.04	0.700
11	16.28	7.87	1310	2.00	31.0	56.9	9.8	0.5	0.3	0.0	0.0	1.5	53.21	6.11	1.111
Southeast high-frequency only															
12	15.95	7.37	1314	2.96	46.5	16.0	25.4	5.1	5.3	0.0	0.8	0.5	21.45	6.57	1.016
13	15.87	7.17	1324	3.37	55.5	13.7	24.4	2.0	1.6	0.0	2.2	0.2	20.54	6.96	0.405
14	16.05	7.47	1302	2.65	54.0	8.1	25.2	2.6	7.6	0.0	1.2	1.1	17.16	7.04	0.762
15	16.02	7.48	1297	2.65	48.4	10.0	19.7	5.9	12.4	0.0	1.8	1.2	17.20	6.82	1.035
16	16.01	7.40	1326	3.00	53.1	32.5	13.9	0.0	0.1	0.0	0.2	0.3	35.71	9.90	1.215
17	16.21	7.84	1297	2.00	68.4	13.2	17.4	0.5	0.2	0.0	0.2	0.1	22.63	11.22	1.389
18	16.42	8.17	1261	2.00	6.2	0.2	40.2	14.9	28.7	0.2	3.4	5.3	0.83	2.07	0.343
19	15.96	7.11	1341	3.90	47.0	36.4	12.7	1.0	1.3	0.0	0.7	0.8	38.53	8.37	0.872
20	15.93	7.45	1325	2.49	16.3	1.6	56.9	8.3	13.9	0.0	1.0	1.3	3.73	3.45	0.384
21	16.22	7.80	1296	2.00	71.7	8.7	17.1	1.3	0.5	0.0	0.0	0.7	18.25	10.02	0.836
22	15.72	7.11	1349	4.00	55.4	7.5	29.1	2.9	3.7	0.0	0.8	0.1	16.00	6.94	1.601
23	16.17	8.33	1281	2.00	23.3	4.8	44.2	6.1	12.7	0.0	6.5	1.5	7.72	3.83	0.658
24	16.06	7.47	1320	3.00	55.0	0.4	43.4	0.3	0.2	0.0	0.4	0.2	8.61	6.54	0.529
25	15.94	7.16	1332	3.11	51.9	32.3	14.1	0.2	0.1	0.0	0.4	1.0	34.85	9.70	1.348
26	15.96	7.19	1341	3.00	36.2	58.2	5.0	0.3	0.0	0.0	0.4	0.0	57.74	10.49	0.813
27	16.16	7.62	1296	2.53	51.9	10.9	31.2	1.1	3.6	0.0	0.4	0.8	17.58	7.79	0.192
28	16.15	7.40	1295	3.00	7.3	0.7	52.1	11.6	25.7	0.0	1.7	0.3	1.61	1.39	0.127
29	16.26	7.98	1283	2.00	50.3	15.9	27.5	2.7	1.0	0.0	1.7	0.7	21.57	7.70	0.720
30	15.67	7.05	1357	4.00	49.7	7.7	33.8	5.7	1.8	0.0	0.4	0.3	15.02	6.36	0.986
31	15.10	6.23	1170	5.00	8.3	1.0	42.4	7.3	39.0	0.5	0.6	0.9	1.96	2.99	0.622
32	15.99	7.39	1325	3.00	55.0	24.4	19.8	0.1	0.1	0.0	0.4	0.3	29.56	10.65	1.015
33	16.20	7.79	1299	2.00	66.4	12.7	19.5	0.2	0.4	0.0	0.6	0.2	20.65	9.79	0.844
34	16.12	7.60	1314	2.65	60.0	20.7	18.3	0.3	0.1	0.0	0.2	0.3	26.23	9.98	1.462
35	16.17	7.74	1305	2.37	61.7	13.8	21.9	1.0	0.2	0.0	0.9	0.5	20.96	9.07	0.774
36	16.15	7.70	1307	2.58	49.0	8.0	34.3	1.6	1.2	0.0	0.9	3.8	14.33	6.70	0.593

Site #	Mean annual temp. °C	Mean Dec – Mar temp. °C	Mean annual precip. mm	Annual precip. as snow %	Developed -low %	Developed -high %	Forest %	Grass -land & shrub %	Pasture %	Cultiv-ated crops %	Wet-lands %	Open water %	Imperv-ious surface cover %	Road length km / km^2	Road crossings crossings / km^2
37	15.69	7.06	1345	3.92	44.3	16.7	29.1	2.7	4.4	0.0	2.3	0.3	21.17	7.84	1.034
38	15.92	7.25	1334	3.15	49.4	30.5	14.9	1.4	2.1	0.0	0.6	1.0	33.29	8.63	1.339
39	15.95	7.45	1322	3.00	42.5	15.8	26.5	3.6	10.2	0.0	1.1	0.1	19.22	7.48	0.232
Mid-Atlantic															
40	13.13	3.03	1071	11.00	55.6	18.7	22.7	0.2	0.1	0.1	2.7	0.0	23.45	12.97	2.189
41	12.99	2.84	1100	12.00	4.4	0.0	1.6	0.3	11.0	77.9	4.6	0.2	0.47	3.61	0.351
42	11.20	0.62	1122	17.90	13.8	0.6	50.4	5.8	10.0	16.8	2.4	0.2	1.82	4.73	0.700
43	11.33	0.85	1098	17.74	15.7	1.8	37.5	7.1	16.1	19.4	2.1	0.1	3.61	4.23	0.759
44	12.97	2.85	1072	11.39	45.2	8.4	38.9	1.0	1.0	0.8	4.3	0.4	11.83	8.79	1.068
45	13.51	3.01	1166	12.00	31.5	8.9	54.8	0.3	1.5	0.0	2.9	0.1	11.91	5.32	0.211
46	13.89	3.44	1086	10.03	35.8	62.7	1.5	0.0	0.0	0.0	0.0	0.0	57.24	16.60	0.000
47	12.00	2.17	1017	13.00	6.6	0.6	21.3	0.8	64.0	4.0	2.1	0.6	1.66	3.57	1.289
48	12.02	2.18	1017	13.00	7.3	0.6	19.8	0.8	65.3	3.7	2.1	0.5	1.74	3.73	1.180
49	11.92	1.50	1181	16.01	34.8	7.6	23.3	2.4	4.2	7.7	16.8	2.6	11.16	5.20	1.005
50	13.74	3.87	1079	9.98	26.4	5.8	40.9	3.0	1.5	6.9	12.5	0.4	8.65	5.03	0.557
51	14.04	3.92	1076	9.00	7.3	0.6	77.5	5.5	0.3	1.7	6.9	0.1	1.20	2.45	0.410
52	14.09	3.96	1078	9.00	7.3	0.4	71.8	11.4	0.3	1.8	6.6	0.2	0.97	2.56	0.492
53	13.08	2.88	1095	11.76	47.3	14.5	23.4	2.4	5.4	2.0	4.6	0.2	19.73	9.50	1.547
54	13.13	2.70	1100	11.93	54.5	6.7	29.1	1.1	3.7	2.2	2.7	0.0	13.38	7.34	0.795
55	12.93	2.56	1222	12.96	60.0	22.6	13.0	1.3	1.5	1.5	0.0	0.0	27.39	11.30	1.703
56	12.88	2.70	1083	11.63	57.0	12.3	20.9	1.0	5.3	1.2	1.9	0.3	18.42	9.80	0.757
57	12.81	2.87	1064	12.00	43.3	0.7	50.9	0.7	0.3	0.0	3.8	0.0	4.85	7.36	0.560
58	13.04	2.80	1091	10.75	70.7	17.1	11.8	0.3	0.0	0.0	0.1	0.0	27.01	15.52	0.599
59	13.78	3.38	1081	11.00	59.8	26.6	13.5	0.0	0.0	0.0	0.1	0.0	32.21	14.10	4.247
Mid-Atlantic high-frequency only															
60	12.87	2.93	1061	11.00	59.9	25.6	14.3	0.2	0.0	0.0	0.1	0.0	29.98	15.79	2.615
61	13.47	3.09	1095	10.28	60.6	9.0	29.0	0.3	1.0	0.0	0.2	0.0	15.50	11.89	1.120
62	12.89	2.79	1062	11.97	71.4	14.6	11.6	0.9	0.2	0.2	0.9	0.1	23.95	10.61	1.653
63	13.32	2.87	1176	11.08	69.6	24.0	5.6	0.0	0.0	0.0	0.1	0.6	32.65	17.96	1.442
64	13.17	3.11	1060	11.00	65.8	4.5	27.3	0.1	0.0	0.0	2.3	0.0	14.53	10.82	0.723
65	11.90	1.56	1182	16.08	29.5	6.0	26.9	2.4	3.2	7.0	22.1	1.8	8.98	4.53	0.962
Mid-Atlantic carbonate															
66	11.63	1.00	1105	16.11	33.6	12.8	3.0	0.7	22.8	26.8	0.1	0.0	15.62	4.86	0.223
67	12.06	2.20	974	13.00	7.0	0.4	21.1	0.0	68.3	2.3	0.8	0.2	1.31	5.28	0.985
68	11.67	2.27	993	13.57	7.4	0.7	45.7	0.0	67.8	2.0	0.0	0.1	1.74	3.27	0.443
69	12.13	2.25	977	13.10	9.2	0.4	20.4	0.1	43.1	3.0	0.1	0.0	1.27	5.18	1.170

Site #	Mean annual temp.	Mean Dec – Mar temp.	Mean annual precip.	Annual precip. as snow	Developed -low	Developed -high	Forest	Grass -land & shrub	Pasture	Cultiv- ated crops	Wet- lands	Open water	Imperv- ious surface cover	Road length	Road crossings
	°C	°C	mm	%	%	%	%	%	%	%	%	%	%	km / km^2	crossings / km^2
New England															
70	9.44	-0.88	1281	26.36	9.0	0.4	71.8	0.6	3.0	2.2	8.0	4.7	1.39	2.78	0.705
71	9.50	-1.10	1215	26.00	31.6	22.2	7.7	5.6	0.5	0.0	0.0	0.0	23.23	7.40	0.000
72	9.50	-1.07	1217	26.00	47.0	40.8	0.2	0.5	0.8	0.0	0.0	0.0	39.15	18.84	0.000
73	9.52	-1.05	1216	26.00	35.1	44.7	10.2	0.0	0.7	0.0	9.3	0.0	39.34	13.91	0.000
74	9.39	-0.89	1291	25.00	10.6	1.5	64.3	1.1	5.4	0.0	17.2	0.0	2.80	2.87	0.214
75	9.37	-0.93	1310	26.92	7.7	0.9	75.1	1.6	0.5	0.0	13.2	1.0	1.69	2.78	0.543
76	9.35	-1.04	1314	27.81	8.0	0.8	64.0	0.7	2.8	0.0	23.6	0.0	1.67	2.49	1.015
77	9.50	-1.13	1210	26.00	15.1	3.4	49.0	1.0	0.5	1.1	29.8	0.0	4.53	4.65	0.596
78	9.48	-0.84	1291	25.18	5.0	0.2	79.0	1.7	3.6	0.0	10.0	0.4	0.77	1.71	0.638
79	9.52	-0.82	1281	24.34	8.0	1.3	71.5	1.7	6.6	0.2	10.2	0.6	2.14	2.21	0.759
80	9.20	-1.12	1325	27.97	6.0	0.5	74.2	1.2	2.6	0.3	12.5	2.7	1.20	2.32	0.824
81	10.06	-0.84	1293	22.66	38.1	16.3	33.8	3.1	1.2	0.0	5.7	1.3	19.07	6.37	0.887
82	9.74	-0.69	1264	25.00	15.1	2.6	16.9	20.3	0.0	0.0	5.2	0.0	4.31	2.96	0.592
83	9.45	-0.88	1290	25.05	10.8	2.0	64.8	1.7	4.5	0.0	16.3	0.0	2.99	3.71	0.323
84	9.88	-0.78	1277	23.17	30.3	15.2	42.6	1.0	2.4	0.1	5.7	2.6	16.75	6.70	1.612
85	8.61	-2.82	1221	28.83	7.0	0.7	70.8	1.5	6.8	0.7	10.8	1.1	1.59	2.56	0.977
86	9.52	-1.05	1209	25.71	24.8	12.8	36.5	0.7	3.4	0.2	15.5	6.1	13.56	6.55	0.648
87	9.58	-0.94	1211	26.00	14.5	51.8	10.2	2.5	0.0	0.2	1.5	0.0	42.67	16.81	0.000
88	9.57	-0.87	1213	26.00	51.3	4.6	32.9	0.1	3.0	0.0	7.4	0.6	9.76	4.85	2.940
89	9.51	-1.08	1207	25.43	21.8	2.6	44.9	0.7	6.2	0.1	20.4	3.2	5.18	4.28	0.704
90	9.59	-0.78	1288	26.25	6.7	0.6	7.7	17.0	0.3	2.1	25.1	0.0	1.56	2.01	1.237
New England high-frequency only															
91	8.97	-2.45	1205	28.00	37.0	21.7	26.3	1.7	5.4	0.0	7.9	0.0	23.44	7.84	2.433
92	10.05	-0.67	1276	22.96	9.0	1.5	68.7	2.5	5.1	0.0	8.5	2.7	2.49	3.10	1.157
93	8.47	-2.91	1255	28.93	13.0	3.3	58.2	1.3	6.8	0.2	12.9	3.8	4.60	3.07	0.763

Table S3. Watershed characteristics 3 – deicing salt application rates normalized to watershed area

Site #	Deicing salt application rates		
	1999	2011	2015
	kg/km²	kg/km²	kg/km²
Southeast			
1	4816	4163	6008
2	3663	3196	4612
3	3457	3014	4350
4	3960	3432	4953
5	4185	3620	5224
6	4172	3601	5197
7	3754	3254	4695
8	4777	4101	5917
9	4267	3682	5313
10	2949	2606	3760
11	2787	2410	3477
Southeast continuous-only			
12	718	653	942
13	1544	1860	2684
14	1423	1955	2822
15	1668	1966	2837
16	3691	3220	4647
17	3854	3334	4810
18	1023	2110	3044
19	4238	3700	5339
20	1186	1293	1866
21	3913	3375	4870
22	1993	2221	3204
23	3979	3425	4943
24	2372	2151	3104
25	998	1127	1626
26	3788	3292	4750
27	1933	2144	3094
28	1669	2138	3085
29	3041	2973	4290
30	2214	2136	3082
31	3979	3425	4943
32	3844	3323	4795
33	4426	3821	5514

Site #	Deicing salt application rates		
	1999	2011	2015
	kg/km²	kg/km²	kg/km²
Mid-Atlantic			
40	55,137	28,354	111,160
41	24,805	14,206	93,934
42	50,947	69,243	98,235
43	51,651	72,024	102,180
44	36,572	19,377	75,969
45	39,543	23,867	157,813
46	608,947	36,804	1,127,556
47	41,894	25,346	167,590
48	44,774	27,336	180,753
49	13,729	29,847	250,901
50	33,092	21,683	143,373
51	11,589	5,918	23,202
52	12,114	6,191	24,271
53	69,658	41,373	273,567
54	58,270	34,155	225,838
55	74,218	44,568	294,692
56	86,520	39,775	293,369
57	27,785	15,438	60,524
58	114,822	65,802	435,092
59	253,598	47,480	591,761
Mid-Atlantic continuous-only			
60	64,043	32,933	129,112
61	54,946	28,045	109,949
62	46,788	24,833	97,358
63	120,623	69,210	457,628
64	44,441	22,697	88,982
65	11,709	25,715	216,170
Mid-Atlantic carbonate			
66	56,719	86,947	123,350
67	28,345	14,577	57,149
68	21,942	11,365	44,558
69	26,097	13,672	53,601

Site #	Deicing salt application rates		
	1999	2011	2015
	kg/km²	kg/km²	kg/km²
New England			
70	50,951	1,125	191,667
71	163,702	226,698	489,978
72	186,571	256,347	554,060
73	168,162	226,125	488,739
74	61,832	1,276	217,331
75	60,375	1,257	214,069
76	74,597	1,561	265,976
77	67,275	95,454	206,312
78	48,935	1,015	172,889
79	50,361	1,052	179,224
80	72,190	1,511	257,412
81	59,338	87,646	344,743
82	60,986	1,308	222,855
83	73,664	1,530	260,657
84	63,059	93,802	368,897
85	34,556	46,748	101,040
86	90,501	125,693	271,667
87	185,828	249,083	538,360
88	83,696	113,688	245,720
89	61,225	84,951	183,610
90	49,513	1,093	186,270
New England continuous-only			
91	110,984	154,248	333,386
92	17,465	27,373	107,670
93	41,771	62,112	134,247

Site #	Deicing salt application rates		
	1999	2011	2015

	kg/km^2	kg/km^2	kg/km^2
34	3530	3062	4418
35	2698	2545	3672
36	2386	2326	3357
37	2984	2660	3839
38	2485	2170	3131
39	570	531	766

Table S4. Summary of years with discrete and high-frequency data for each site

Site #	Discrete			High-frequency		
	Years with discrete samples	Specific conductance	Chloride	Years included	Years excluded for incomplete (<85%) winter record	Years of data included
		N	N			N
Southeast						
1	1999 – 2018	357	237	2008 – 2018	none	11
2	1976 – 2018	360	221	2008 – 2018	none	11
3	1976 – 2018	275	236	2008 – 2018	none	11
4	1976 – 2018	392	242	2008 – 2018	2008, 2013, 2016	8
5	1959 – 2018	752	296	2008 – 2018	2013, 2014	9
6	1976 – 2018	399	239	2008 – 2018	none	11
7	2003 – 2018	336	226	2008 – 2018	none	11
8	2003 – 2016	98	99	2008 – 2018	2010, 2014, 2015	8
9	2001 – 2018	375	235	2008 – 2018	none	11
10	1993 – 2010	232	197	2008 – 2018	none	11
11	1976 – 2018	289	144	2008 – 2018	none	11
Southeast high-frequency only						
12	1997 – 2018	164	0	2008 – 2018	2013	10
13	2001 – 2018	147	0	2008 – 2018	2010	10
14	1996 – 2018	234	0	2008 – 2018	2011, 2012, 2015	8
15	1996 – 2018	180	0	2008 – 2018	2015, 2018	9
16	2013 – 2018	116	0	2013 – 2018	none	6
17	1999 – 2018	113	0	2014 – 2018	2016	4
18	None	0	0	2015 – 2018	none	4
19	1976 – 2018	164	0	2008 – 2018	none	11
20	2007 – 2018	255	11	2008 – 2018	2010	10
21	1999 – 2018	136	10	2014 – 2018	none	5
22	2001 – 2018	151	0	2008 – 2018	none	11
23	None	0	0	2009 – 2018	2016	9
24	1999 – 2018	111	0	2014 – 2018	none	5
25	1976 – 2018	120	0	2013 – 2018	2013	5
26	1976 – 2018	151	0	2008 – 2018	2016	10
27	1999 – 2018	170	0	2008 – 2018	none	11
28	None	0	0	2013 – 2018	none	6
29	2013 – 2018	122	10	2013 – 2018	2016, 2018	4
30	1976 – 2018	154	0	2008 – 2018	none	11
31	None	0	0	2009 – 2018	2009	9
32	2012 – 2018	139	10	2012 – 2018	2012	6
33	2012 – 2018	145	10	2012 – 2018	2012	6
34	2012 – 2018	134	0	2012 – 2018	2012	6
35	2013 – 2018	119	0	2013 – 2018	2016	5
36	2013 – 3018	95	0	2006 – 2018	none	3

Site #	Discrete			High-frequency		
	Years with discrete samples	Specific conductance N	Chloride N	Years included	Years excluded for incomplete (<85%) winter record	Years of data included N
37	1976 – 2018	228	8	2008 – 2018	none	11
38	2010 – 2018	19	0	2010 – 2018	2010	8
39	2001 – 2018	138	0	2008 – 2018	none	11
Mid-Atlantic						
40	1968 – 2018	87	92	2015 – 2018	2015	3
41	1991 – 2018	186	188	2013 – 2018	2016	5
42	2012 – 2018	159	136	2014 – 2018	none	4
43	2011 – 2018	171	143	2014 – 2018	none	4
44	1968 – 2018	181	156	2008, 2013 – 2018	none	7
45	2016 – 2018	67	70	2016 – 2018	none	3
46	2013 – 2018	131	90	2013 – 2018	2013	5
47	2017 – 2018	79	79	2017 – 2018	none	2
48	2017 – 2018	71	71	2017 – 2018	none	2
49	1959 – 2018	201	164	2015 – 2018	none	4
50	1961 – 2018	421	47	2008 – 2011	none	4
51	2014 – 2016	33	32	2014 – 2016	none	2[a]
52	2014 – 2016	35	34	2014 – 2016	none	2[a]
53	1959 – 2018	450	158	2008 – 2018	2008	10
54	2007 – 2018	429	217	2008 – 2018	none	11
55	2005 – 2018	197	166	2014 – 2018	none	5
56	1999 – 2018	427	318	2013 – 2018	none	6
57	2008 – 2018	150	20	2008 – 2018	2008	10
58	2012 – 2018	202	204	2013 – 2018	2013	5
59	2012 – 2018	122	88	2013 – 2018	2013, 2017	4
Mid-Atlantic high-frequency only						
60	None	0	0	2012 – 2015	2015	3
61	2008 – 2018	127	0	2008 – 2018	none	11
62	2008 – 2018	130	0	2008 – 2018	2018	10
63	None	0	0	2016 – 2018	none	3
64	1969 – 2018	74	2	2013 – 2018	2013	5
65	None	0	0	2008 – 2018	none	11
Mid-Atlantic carbonate						
66	2009 – 2018	141	130	2016 – 2018	2016	2
67	1969 – 2017	52	28	2008 – 2016	2008, 2009, 2011, 2012, 2014	4
68	1969 – 2018	192	140	2008 – 2016	2009, 2010	9
69	2006 – 2017	44	21	2008 – 2016	2010	8

Site #	Discrete			High-frequency		
	Years with discrete samples	Specific conductance	Chloride	Years included	Years excluded for incomplete (<85%) winter record	Years of data included
		N	N			N
New England						
70	2000 – 2018	30	28	2010 – 2018	none	9
71	1997 – 2018	65	59	2012 – 2018	none	7
72	1997 – 2018	101	90	2008 – 2018	none	11
73	1997 – 2018	86	79	2012 – 2018	2012	6
74	2000 – 2018	31	29	2010 – 2018	2010	8
75	2000 – 2018	30	28	2010 – 2018	2010	8
76	2000 – 2018	31	29	2009 – 2018	2009	9
77	1997 – 2018	77	69	2012 – 2018	none	7
78	2000 – 2018	64	36	2009 – 2018	2009	9
79	1994 – 2018	77	30	2009 – 2018	2009, 2015	8
80	1994 – 2018	41	29	2009 – 2018	2009	9
81	1953 – 2018	660	490	2014 – 2018	none	5
82	2000 – 2018	66	39	2010 – 2018	none	9
83	2009 – 2018	63	38	2010 – 2018	none	9
84	1993 – 2018	337	206	2014 – 2018	none	5
85	1973 – 2016	81	74	2008 – 2018	2009	10
86	1997 – 2018	77	72	2008 – 2018	none	11
87	1998 – 2018	117	106	2008 – 2018	none	11
88	1998 – 2018	77	71	2008 – 2018	none	11
89	2011 – 2018	36	35	2010 – 2018	2010	8
90	2009 – 2018	26	26	2010 – 2018	none	9
New England high-frequency only						
91	None	0	0	2013 – 2018	2017	5
92	2005 – 2012	59	3	2010 – 2012	none	3
93	None	0	0	2008 – 2018	none	11

[a] Sites #51 & 52 – ~2 complete years of data across 3 water years

Table S5. Statistical summaries of discrete data from 1998 – 2018 for sites with ≥20 [Cl] and SC discrete samples and for entire record for sites categorized as high-frequency only

Site #	Specific Conductance (µS/cm)								Chloride (mg/L)							
	Min	5%	25%	Median	75%	95%	Max	N	Min	5%	25%	Median	75%	95%	Max	N
Southeast																
1	31	67.8	123	179	216	345	1180	357	1.34	4.35	7.85	14.0	21.9	86.8	288	237
2	34	47	64	81	106	137	242	359	0.59	1.85	2.80	3.81	5.30	7.86	10.8	221
3	42	54.7	70	94.5	116	127	161	274	1.61	2.49	3.56	5.30	7.26	9.16	12.1	236
4	31	51.1	76	102	140	161	186	383	1.00	1.84	3.07	4.98	7.10	9.40	14.3	242
5	37	59	83.5	116	156	186	253	399	1.38	2.59	4.00	6.13	8.73	11.6	14.7	233
6	48	82	124	204	261	314	935	394	3.02	4.02	7.12	11.2	14.3	18.2	28.6	239
7	29	38	67.5	99.5	128	145	209	336	0.80	1.02	2.57	4.74	7.15	8.98	11.7	226
8	30	92.1	173	222	282	343	421	98	0.62	2.84	6.82	10.6	12.4	16.3	20.7	99
9	37	61.7	97	144	184	214	439	375	0.97	1.99	4.04	6.51	9.76	16.2	110	235
10	38	49	62	81	122	143	155	228	1.08	2.06	2.85	3.75	5.81	7.16	8.63	194
11	46	75	132	211	297	377	420	279	1.22	1.81	3.89	6.50	14.2	19.5	22.4	143
Southeast high-frequency only																
12	32	48	70	76	84	94.9	202	164	*nd*							
13	20	38.3	56	60	65	74	80	147	*nd*							
14	27	42.3	53	60	65	84.4	137	234	*nd*							
15	28	34	40	44	48	53.1	89	180	*nd*							
16	40	59.8	82	111	119	125	158	116	*nd*							
17	28	63.6	86	94	98	104	145	113	*nd*							
18	*nd*								*nd*							
19	20	50.2	91	103	109	121	238	164	*nd*							
20	21	38	46	50	55	63.3	69	255	2.42	2.46	2.82	3.26	3.30	3.41	3.46	11
21	31	59.8	83	87	90	97	101	136	4.75	5.68	6.82	6.94	7.07	7.21	7.29	10
22	26	38.5	58	73	76	87	92	151	*nd*							
23	*nd*								*nd*							
24	43	81	101	111	117	123	127	111	*nd*							
25	42	65.7	85	114.5	128	145	202	120	*nd*							
26	20	25	47.5	90	100	107	131	151	*nd*							
27	34	52.3	79	100	117	141	222	170	*nd*							
28	*nd*								*nd*							
29	34	48.1	71	82.5	90	104	136	122	3.65	3.72	3.99	4.84	6.07	6.32	6.45	10
30	25	34	60	78	83	99.7	110	154	*nd*							
31	*nd*[a]								*nd*							
32	35	56.9	81	96	99.5	112	159	139	2.25	3.57	5.35	5.91	6.27	10.8	14.10	10
33	39	52.4	95	109	114	121	159	145	2.58	4.41	6.65	7.02	7.52	7.87	7.97	10
34	29	65.7	91	112	118	128	182	134	*nd*							
35	31	51.8	82.5	100	105	118	211	119	*nd*							
36	56	59.7	68	72	77.5	84	93	95	*nd*							
37	40	47	79.5	100	110.5	175	326	228	3.73	3.75	3.95	5.30	5.57	6.07	6.33	8
38	55	87.4	101	103	107	113	138	19	*nd*							
39	20	28	46	51	57	65	69	138	*nd*							

Site #	Specific Conductance (µS/cm)								Chloride (mg/L)							
	Min	5%	25%	Median	75%	95%	Max	N	Min	5%	25%	Median	75%	95%	Max	N
Mid-Atlantic																
40	44	110	172	255	407	713	2220	47	5.10	12.9	22.5	44.5	82.8	164	626	48
41	46	99.8	153	175	194	203	213	177	1.95	6.17	11.7	13.3	15.2	16.3	26.7	179
42	98	151	222	265	297	336	601	159	4.80	10.5	22.6	28.5	34.6	47.8	143	136
43	110	177	293	319	371	402	874	171	2.40	9.82	25.1	32.6	40.9	54.4	198	143
44	52	112	203	246	304	545	2740	177	7.70	17.8	36.0	46.9	62.7	166	899	152
45	81	146	213	265	326	436	850	67	5.88	17.3	26.5	38.8	51.9	80.7	359	70
46	52	140	421	912	1280	2555	6540	131	8.04	16.5	78.2	184	281	570	1940	90
47	117	151	227	278	336	418	462	79	3.78	10.1	19.8	33.1	45.4	58.9	66.9	79
48	74	160	234	293	341	399	449	71	6.65	11.5	23.4	36.9	47.5	61.1	68.4	71
49	117	154	228	260	298	454	2600	86	11.2	16.3	23.6	33.2	45.8	108	737	85
50	38	79	110	134	165	218	479	421	12.0	13.3	18.8	25.4	30.8	47.6	69.8	47
51	46	54	64	71	84	117	150	33	4.49	6.27	8.67	9.9	11.4	17.8	20.5	32
52	30	31.7	44.5	52	59.5	67	68	35	3.40	3.52	4.52	5.82	6.87	8.76	9.59	34
53	43	80.4	145	243	332	775	3280	389	3.62	8.06	21.8	39.9	65.5	97.6	632	144
54	16	84.8	145	230	339	781	3530	429	8.87	13.9	33.0	54.9	88.2	301	1010	217
55	83	131	329	440	586	1252	3150	197	5.09	17.1	60.6	95.4	142	401	888	166
56	73	120	243	363	512	1334	8390	427	6.54	16.0	35.9	63.5	104	343	1430	318
57	51	106	151	166	181	202	418	150	13.7	14.2	23.4	25.8	28.1	29.0	29.8	20
58	10	93.4	234	485	686	1855	9720	202	4.29	15.1	43.0	100	155	486	2810	204
59	61	127	218	439	528	938	1900	122	3.88	11.3	26.3	63.4	86.0	186	474	88
Mid-Atlantic high-frequency only																
60	*nd*								*nd*							
61	76	122	208	285	342	533	2730	127	*nd*							
62	119	197	334	418	506	817	1170	130	*nd*							
63	*nd*								*nd*							
64	31	81.3	168	222	270	437	545	74	*nd*							
65	*nd*								*nd*							
Mid-Atlantic carbonate																
66	183	287	612	737	796	863	2080	141	21.2	27.0	44.6	59.7	79.6	120	600	130
67	386	568	614	633	643	653	656	51	11.2	12.0	13.2	13.9	14.8	17.7	21.6	27
68	165	260	400	478	516	545	567	191	5.23	11.5	17.7	21.3	25.0	31.8	44.2	139
69	275	507	549	566	575	586	589	44	11.7	12.0	12.3	12.9	13.7	14.4	15.8	21
New England																
70	146	156	188	234	277	313	341	30	34.3	35.8	48.9	56.5	68.2	80.5	85.4	28
71	388	592	816	1170	1660	2798	3730	55	148	165	244	332	504	820	1110	49
72	189	599	1560	2080	2840	13000	25400	91	219	296	472	621	862	4449	9040	80
73	188	315	784	1370	2070	10192	55500	77	44.9	82.8	215	395	731	4235	20600	70
74	76	86	127	147	173	200	262	31	19.8	21.9	29.9	36.4	42	49.4	66.6	29
75	63	71	100	114	137	150	155	30	15.1	19.0	24.5	27.2	33.7	37.6	38.6	28
76	60	67	90	106	121	151	168	31	14.7	15.2	21.2	24.6	29.4	36.9	42	29

Site #	Specific Conductance (µS/cm)								Chloride (mg/L)							
	Min	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**	**N**	**Min**	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**	**N**
77	134	198	290	428	607	2529	3270	67	36.7	40.0	69.1	109	148	712	968	59
78	37	58	72	87	94	103	125	64	7.86	9.04	11.5	13.7	15.8	18.3	19.4	36
79	109	115	165	202	219	246	293	59	21.2	23.6	31.7	37.6	48.1	55.4	75	30
80	47	57	77	97	110	131	137	31	11.5	13.8	17.4	22.2	25.6	31.1	34.2	29
81	146	195	296	367	434	562	707	300	15.6	25.6	38.0	48.6	62.4	105	150	167
82	78	135	173	226	302	615	1270	66	14.1	26.8	34.5	45.4	57.2	266	352	39
83	89	133	209	259	320	362	634	63	20.7	28.1	42.0	54.0	75.1	113	183	38
84	134	231	419	540	652	793	1490	297	16.8	33.2	55.9	73.8	97.9	140	241	166
85	56	74	107	126	162	284	325	79	9.67	12.9	18.3	25.1	30.6	63.0	71.5	72
86	163	237	418	531	669	984	1440	67	36.7	47.1	96.1	132	178	276	396	62
87	61	159	848	1420	8010	22580	28000	117	12.2	83.4	253	418	4080	8340	11500	106
88	144	175	254	314	379	970	1540	77	20.7	23.0	41.1	54.7	72.7	254	425	71
89	244	247	342	413	461	574	1190	36	44.9	48.4	77.4	96.1	115	138	323	35
90	70	72	88	107	118	149	156	26	14.1	15.2	19.3	22.9	26	33.9	37.8	26
New England high-frequency only																
91	*nd*								*nd*							
92	47	66.2	91.5	102	120	138	155	59								
93	*nd*								*nd*							

[a] *nd* = no data

Table S6. Statistical summaries of high-frequency data

Site #	**Specific Conductance** (observed) µS/cm								**Chloride** (modeled) mg/L						
	Min	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**	**N**	**Min**	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**
Southeast															
1	30	113	178	204	228	270	2040	375044	0.70	10.0	18.6	22.1	25.3	30.8	532
2	28	69	98	114	126	143	434	375759	0.46	3.67	5.95	7.20	8.14	9.47	32.3
3	25	70	104	123	137	158	1100	371059	0.80	3.65	6.29	7.77	8.86	10.5	83.8
4	20	64	108	135	150	168	791	258955	0.50	3.13	6.08	7.89	8.90	10.1	51.8
5	21	76	124	155	171	195	871	288398	0.77	4.06	6.93	8.78	9.74	11.2	51.6
6	33	115	202	252	278	317	1290	373445	4.14	7.82	11.7	14.0	15.1	16.9	60.6
7	25	62	103	125	136	152	587	376013	0.55	3.10	5.93	7.44	8.20	9.30	39.3
8	21	107	163	219	254	294	1050	265077	1.00	4.76	7.21	9.65	11.2	12.9	46.0
9	27	88	149	180	197	219	1310	374538	0.50	4.34	8.49	10.6	12.5	25.5	668
10	15	68	109	127	137	148	699	371343	0.50	3.19	5.33	6.27	6.79	7.36	36.1
11	29	104	210	282	323	372	1080	377985	0.60	3.35	8.80	12.5	14.6	17.1	53.6
Southeast high-frequency only															
12	16	64	81	89	94	101	167	336229	0.32	3.46	4.57	5.09	5.42	5.88	10.2
13	19	54	68	72	75	80	293	340845	0.52	2.81	3.72	3.98	4.18	4.51	18.4
14	17	50	62	66	69	73	124	254796	0.39	2.54	3.33	3.59	3.79	4.05	7.38
15	9	37	46	50	52	57	89	292426	0.30	1.69	2.28	2.54	2.67	3.00	5.09
16	15	61	94	110	118	128	1010	192929	0.30	3.26	5.42	6.47	6.99	7.65	244
17	18	54	84	93	99	107	1860	130303	0.45	2.81	4.77	5.36	5.75	6.27	543
18	29	45	55	61	68	79	111	137784	1.17	2.22	2.87	3.26	3.72	4.44	6.53
19	21	60	88	103	114	127	285	370122	0.65	3.20	5.03	6.01	6.73	7.58	17.9
20	20	44	49	53	58	65	94	337116	0.58	2.15	2.48	2.74	3.07	3.53	5.42
21	14	63	83	88	92	98	1860	172780	0.30	3.39	4.70	5.03	5.29	5.68	543
22	18	57	77	82	86	90	421	380640	0.45	3.00	4.31	4.64	4.90	5.16	37.1
23	31	57	64	74	87	107	169	305956	1.30	3.00	3.46	4.11	4.96	6.27	10.3
24	13	85	107	113	117	123	4250	167057	0.30	4.83	6.27	6.66	6.93	7.32	1383
25	28	67	98	115	127	147	438	168372	1.11	3.66	5.68	6.80	7.58	8.89	43.1
26	10	52	87	96	103	111	1590	336338	0.30	2.67	4.96	5.55	6.01	6.53	448
27	18	53	69	75	83	97	134	374816	0.45	2.74	3.79	4.18	4.70	5.62	8.04
28	16	54	61	65	68	71	212	185970	0.32	2.81	3.26	3.53	3.72	3.92	13.1
29	21	53	74	83	89	101	220	121484	0.65	2.74	4.11	4.70	5.09	5.88	13.7
30	17	70	89	94	100	108	190	371672	0.39	3.85	5.09	5.42	5.81	6.34	11.7
31	22	72	84	92	100	114	178	305342	0.71	3.98	4.77	5.29	5.81	6.73	10.9
32	18	55	84	95	100	111	955	208247	0.45	2.87	4.77	5.49	5.81	6.53	225
33	23	65	98	108	113	121	898	196872	0.78	3.53	5.68	6.34	6.66	7.19	205
34	21	61	98	112	118	132	2000	202224	0.65	3.26	5.68	6.60	6.99	7.91	592
35	15	58	87	99	105	114	502	164983	0.30	3.07	4.96	5.75	6.14	6.73	65.6
36	36	58	67	72	78	89	114	103338	1.63	3.07	3.66	3.98	4.38	5.09	6.73
37	27	64	102	126	158	241	400	377089	1.04	3.46	5.94	7.51	9.61	15.0	29.7
38	21	61	82	94	103	115	501	276627	0.65	3.26	4.64	5.42	6.01	6.80	65.2
39	15	42	55	59	63	68	1240	379145	0.30	2.02	2.87	3.13	3.39	3.72	325

Site #	Specific Conductance (observed) µS/cm								Chloride (modeled) mg/L						
	Min	5%	25%	Median	75%	95%	Max	N	Min	5%	25%	Median	75%	95%	Max
Mid-Atlantic															
40	37	145	256	368	550	1340	12600	102002	1.80	14.5	46.1	77.9	130	354	3558
41	33	155	189	196	201	205	219	167247	1.62	12.0	14.9	15.5	15.9	16.2	17.4
42	79	221	273	309	336	405	1330	130159	4.58	23.3	30.1	34.9	45.1	71.1	420
43	94	251	315	359	399	462	1220	132391	4.19	25.7	34.5	40.5	46.0	62.6	312
44	42	157	219	257	317	663	8710	235198	2.00	20.3	40.0	52.0	71.0	181	2728
45	32	170	259	294	336	614	5500	306958	2.90	12.5	41.4	52.8	66.5	157	1745
46	7	487	959	1150	1360	2530	57900	1290763	4.00	71.9	207	261	321	656	16486
47	54	273	314	337	363	404	2470	178564	1.90	32.6	40.0	44.2	48.9	56.4	432
48	47	268	322	348	374	413	2070	173358	3.30	32.9	43.4	48.5	53.5	61.1	383
49	28	217	289	314	334	693	4420	130251	4.80	22.9	44.4	51.8	57.8	165	1275
50	33	101	131	145	166	286	628	100959	6.00	14.7	19.4	22.9	29.5	67.4	176
51	35	55	63	69	79	136	185	72590	4.68	7.50	8.62	9.47	10.9	18.9	25.8
52	23	30	45	51	55	64	121	69477	2.45	3.30	5.13	5.86	6.35	7.44	14.4
53	28	222	328	395	483	1190	9020	793322	1.80	37.8	69.8	90.0	117	330	2692
54	15	187	247	323	393	1190	13000	805886	4.40	33.7	51.3	73.5	93.9	327	3778
55	39	230	466	566	682	1610	12100	495214	2.50	36.1	105	134	168	439	3501
56	10	206	360	474	587	1400	9020	341763	3.30	30.5	74.2	107	139	370	2535
57	23	127	155	168	186	227	1620	332289	6.80	17.7	22.6	24.9	28.1	35.3	280
58	33	212	521	690	849	2680	17200	491793	2.10	29.3	118	167	212	738	4910
59	6	267	443	524	595	1250	11800	987090	1.90	35.4	63.5	78.4	98.4	283	3253
Mid-Atlantic high-frequency only															
60	29	148	335	462	556	1530	10600	89224	0.70	19.5	53.1	90.1	118	401	3043
61	34	145	246	292	345	789	13400	369151	0.70	19.0	36.2	44.1	56.0	185	3859
62	36	251	366	433	517	881	7260	341634	0.70	37.1	62.2	81.7	106	212	2070
63	48	268	350	377	431	736	7140	296873	2.40	40.0	57.5	65.4	81.1	170	2035
64	16	110	192	231	285	817	8860	168839	0.70	13.0	27.0	33.7	42.9	194	2536
65	53	162	213	239	270	446	3100	351809	3.26	21.9	30.6	35.0	40.3	85.5	859
Mid-Atlantic carbonate															
66	110	599	748	785	824	905	2500	67181	22.9	55.3	65.2	70.7	86.6	120	772
67	139	551	587	600	613	633	741	139813	2.30	10.1	11.9	12.5	13.1	14.1	19.3
68	137	369	453	500	525	549	642	302524	10.1	18.3	21.3	23.0	23.8	24.7	28.0
69	78	473	522	543	566	589	873	272925	9.84	12.5	12.8	13.0	13.1	13.3	15.2
New England															
70	52	163	208	235	287	342	411	468136	17.1	38.2	50.0	57.1	70.8	85.2	103
71	24	858	1200	1470	1900	2690	13900	300323	61.0	229	332	413	542	779	4145
72	26	460	1310	1770	2260	3120	21500	427363	65.0	65.0	321	480	650	947	7307
73	28	470	982	1290	1670	2700	30700	221390	22.4	22.4	207	321	461	840	11164
74	36	110	138	161	181	252	422	399274	9.90	25.0	32.6	38.8	44.2	63.5	110
75	59	85	105	123	143	163	637	397497	12.0	19.1	24.6	29.5	35.0	40.5	171
76	28	70	92	110	133	165	309	469973	7.30	14.4	20.7	25.8	32.3	41.4	82.4
77	65	261	356	429	500	695	4460	347210	18.4	55.1	84.0	106	128	187	1331

Site #	Specific Conductance (observed) µS/cm								Chloride (modeled) mg/L						
	Min	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**	**N**	**Min**	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**
78	16	52	67	79	89	103	324	447469	3.90	9.05	11.9	14.1	16.0	18.6	60.1
79	11	123	149	179	204	232	288	329739	10.6	24.8	32.3	41.1	48.4	56.6	72.9
80	29	68	85	98	112	137	193	307912	5.80	13.8	18.5	22.1	26.0	33.0	48.5
81	145	322	432	482	540	673	1410	167656	13.6	34.0	61.2	77.3	95.9	139	375
82	47	127	161	186	228	357	3980	443493	7.00	23.3	33.4	40.8	53.3	91.7	1169
83	32	116	164	205	246	323	614	429578	10.3	22.8	37.2	49.5	61.8	84.9	172
84	132	407	577	678	777	919	3210	166535	8.4	49.7	77.8	108	138	181	872
85	48	88	115	148	195	283	587	336593	6.37	15.7	22.0	29.7	40.7	61.2	132
86	61	349	458	570	700	1010	4280	573149	18.4	78.2	110	144	182	274	1240
87	0	183	881	1160	1590	3580	98500	521049	6.10	6.10	179	272	415	1079	32735
88	1	205	260	305	364	435	3530	406052	10.3	25.7	41.4	54.2	71.1	91.4	976
89	119	236	295	378	464	578	1600	419242	22.4	44.7	61.7	85.7	110	143	438
90	42	75	91	106	133	154	388	471515	7.61	15.8	19.8	23.5	30.3	35.5	93.7
New England high-frequency only															
91	74	565	756	939	1100	1390	6430	236978	18.7	103	165	225	277	372	2013
92	48	82	97	109	124	150	1020	99704	16.0	19.6	21.2	22.4	24.0	26.8	251
93	81	126	161	203	259	359	807	373950	19.5	24.2	27.9	32.4	38.3	48.8	182

Table S7. Regression relationships for figures

	Region[a]	Slope	1 σ uncertainty	R^2 [b]	p value	intercept	1 σ uncertainty	p value
Figure 2A High-frequency SC vs ISC[c]	SE	2.62	0.50	0.41	*<0.001*	49.2	13.6	*<0.001*
	MA	13.7	1.72	0.71	*<0.001*	147	35.1	*<0.001*
	NE	34.0	2.95	0.85	*<0.001*	99.2	50.6	<0.1
Figure 2B High-frequency SC variability vs ISC	SE	2.81	0.55	0.67	*<0.001*	6.39	14.9	0.70
	MA	37.8	5.64	0.64	*<0.001*	101	115	0.39
	NE	64.7	5.06	0.88	*<0.001*	–82.9	86.7	0.35
Figure 3A Discrete [Cl] vs ISC	SE	0.165	0.105	0.13	0.15	1.56	3.64	0.68
	MA	2.59	0.26	0.84	*<0.001*	12.7	5.11	<0.05
	NE	10.1	2.14	0.51	*<0.001*	28.4	32.7	0.40
Figure 3B High-frequency [Cl] vs ISC	SE	0.150	0.038	0.28	*<0.001*	2.94	1.02	<0.01
	MA	3.53	0.41	0.75	*<0.001*	12.8	8.34	0.14
	NE	8.67	1.00	0.77	*<0.001*	17.0	17.1	0.33
Figure 5A Chronic [Cl] exceedances vs ISC	MA	2.67	0.38	0.66	*<0.001*	–17.7	7.83	<0.05
	NE	7.52	0.94	0.73	*<0.001*	–26.2	16.1	0.12
Figure 5B Acute [Cl] exceedances vs ISC	MA	9.43	1.47	0.62	*<0.001*	–39.3	30.0	0.20
	NE	17.7	2.83	0.62	*<0.001*	–84.4	48.4	<0.1
Figure 5C Chronic [Cl] exceedances vs median [Cl]	MA	0.75	0.06	0.88	*<0.001*	–27.3	4.92	*<0.001*
	NE	0.86	0.05	0.93	*<0.001*	–39.7	8.14	*<0.001*
Figure 5D Acute [Cl] exceedances vs median [Cl]	MA	2.74	0.21	0.87	*<0.001*	–77.9	18.2	*<0.001*
	NE	1.86	0.27	0.68	*<0.001*	–99.3	45.6	<0.05
Figure 6C Annual [Cl] difference	MA	0.060	0.013	0.66	<0.01	0.808	1.18	0.51
Figure 6D Annual [Cl] difference	NE	0.079	0.006	0.90	*<0.001*	–0.381	1.00	0.71
Figure S1A Discrete SC vs ISC	SE	4.90	1.03	0.69	<0.01	–22.0	35.3	0.55
	MA	11.2	1.53	0.73	*<0.001*	135	30.1	*<0.001*
	NE	34.4	3.06	0.86	*<0.001*	78.3	53.7	0.16
Figure S1B Winter SC vs ISC	SE	2.78	0.50	0.44	*<0.001*	44.3	13.6	<0.001
	MA	18.7	2.11	0.76	*<0.001*	159	43.1	<0.001
	NE	39.3	2.46	0.92	*<0.001*	61.9	42.1	0.16
Figure S1C Non-winter SC vs ISC	SE	2.53	0.50	0.39	*<0.001*	52.1	13.6	*<0.001*
	MA	12.5	1.67	0.69	*<0.001*	146	34.2	*<0.001*
	NE	32.1	3.19	0.81	*<0.001*	116	54.6	<0.05
Figure S3A Winter [Cl] vs ISC	SE	0.16	0.04	0.33	*<0.001*	2.65	0.97	<0.01
	MA	4.98	0.59	0.73	*<0.001*	15.3	12.1	0.22
	NE	10.5	0.88	0.86	*<0.001*	4.84	15.0	0.75
Figure S3B Non-winter [Cl] vs ISC	SE	0.15	0.04	0.25	*<0.001*	3.11	1.06	<0.01
	MA	3.17	0.39	0.73	*<0.001*	12.8	7.88	0.12
	NE	8.02	1.07	0.71	*<0.001*	22.2	18.3	0.24

	Region[a]	Slope	1 σ uncertainty	R^2 [b]	p value	intercept	1 σ uncertainty	p value
Figure S3D Chronic exceedances vs non-winter median [Cl]	MA	0.83	0.62	0.88	*<0.001*	–27.2	4.91	*<0.001*
	NE	0.88	0.06	0.91	*<0.001*	–41.0	9.39	*<0.001*
Figure S3E Acute exceedances vs non-winter median [Cl]	MA	2.99	0.24	0.86	*<0.001*	–78.6	18.8	*<0.001*
	NE	1.91	0.29	0.65	*<0.001*	–100	47.8	<0.05
Figure S3F Acute vs chronic exceedances	MA	3.02	0.43	0.76	*<0.001*	56.1	27.2	<0.1
	NE	2.17	0.71	0.51	<0.05	–34.0	148	0.83

[a] SE = Southeast, MA = Mid-Atlantic (silicate only), NE (New England)
[b] Reported R^2 values are the adjusted R^2 as calculated by R.
[c] SC = specific conductance; ISC = impervious surface cover (% of watershed)

Table S8. Statistical summaries of *winter* high-frequency data

Site #	Specific Conductance (observed) µS/cm								Chloride (modeled) mg/L						
	Min	5%	25%	Median	75%	95%	Max	N	Min	5%	25%	Median	75%	95%	Max
Southeast															
1	34	112	173	195	211	257	2040	125006	0.7	9.88	18.0	20.9	23.0	29.1	532
2	31	66	98	115	126	155	434	126040	0.69	3.44	5.95	7.28	8.14	10.4	32.3
3	25	70	105	124	137	176	1100	124886	0.80	3.65	6.37	7.85	8.86	11.9	83.8
4	20	69	115	138	149	175	791	87884	0.50	3.47	6.55	8.09	8.83	10.6	51.8
5	24	76	127	156	168	191	871	96265	0.95	4.06	7.11	8.84	9.56	10.9	51.6
6	37	117	208	252	277	337	1290	121832	4.32	7.91	12.0	14.0	15.1	17.8	60.6
7	27	60	104	125	133	148	587	125973	0.69	2.96	5.99	7.44	7.99	9.03	39.3
8	22	100	165	226	256	290	1050	88075	1.04	4.45	7.29	9.96	11.3	12.8	46.0
9	31	85	149	178	193	220	1310	124056	0.5	4.13	8.49	10.5	11.5	26.1	668
10	15	66	108	126	135	149	699	123371	0.50	3.08	5.28	6.22	6.69	7.42	36.1
11	29	91	209	292	340	389	1080	124311	0.60	2.68	8.75	13.0	15.5	18.0	53.6
Southeast high-frequency only															
12	16	61	79	88	94	101	167	113719	0.32	3.26	4.44	5.03	5.42	5.88	10.2
13	23	52	66	70	73	77	99	112036	0.78	2.67	3.59	3.85	4.05	4.31	5.75
14	17	47	60	65	68	71	81	86801	0.39	2.35	3.20	3.53	3.72	3.92	4.57
15	9	33	42	47	50	54	89	100765	0.30	1.43	2.02	2.35	2.54	2.81	5.09
16	21	62	97	112	121	149	1010	65019	0.65	3.33	5.62	6.60	7.19	9.02	244
17	22	61	84	92	98	115	1860	43749	0.71	3.26	4.77	5.29	5.68	6.80	543
18	33	43	49	59	65	78	94	46487	1.43	2.09	2.48	3.13	3.53	4.38	5.42
19	25	59	86	103	113	125	285	124442	0.91	3.13	4.90	6.01	6.66	7.45	17.9
20	20	42	48	52	57	63	91	112499	0.58	2.02	2.41	2.67	3.00	3.39	5.23
21	23	63	82	87	90	96	1860	57159	0.78	3.39	4.64	4.96	5.16	5.55	543
22	23	56	75	81	84	88	175	127156	0.78	2.94	4.18	4.57	4.77	5.03	10.7
23	31	54	61	66	78	91	140	102344	1.30	2.81	3.26	3.59	4.38	5.23	8.43
24	28	83	105	110	114	120	4250	56032	1.11	4.70	6.14	6.47	6.73	7.12	1383
25	33	69	104	118	128	169	438	55676	1.43	3.79	6.08	6.99	7.65	10.3	43.1
26	14	52	87	98	105	114	1590	112091	0.30	2.67	4.96	5.68	6.14	6.73	448
27	18	50	65	71	75	81	126	126454	0.45	2.54	3.53	3.92	4.18	4.57	7.51
28	28	52	58	61	65	70	212	67026	1.11	2.67	3.07	3.26	3.53	3.85	13.1
29	29	56	74	81	85	91	220	44405	1.17	2.94	4.11	4.57	4.83	5.23	13.7
30	17	67	86	92	97	106	190	121524	0.39	3.66	4.90	5.29	5.62	6.21	11.7
31	30	66	81	87	94	106	178	102244	1.24	3.59	4.57	4.96	5.42	6.21	10.9
32	18	54	83	94	99	109	955	69558	0.45	2.81	4.70	5.42	5.75	6.40	225
33	26	66	100	107	114	124	898	66346	0.97	3.59	5.81	6.27	6.73	7.38	205
34	25	62	101	113	119	161	2000	67336	0.91	3.33	5.88	6.66	7.06	9.80	592
35	25	61	90	99	105	130	502	55308	0.91	3.26	5.16	5.75	6.14	7.78	65.6
36	37	54	64	70	73	78	105	33555	1.69	2.81	3.46	3.85	4.05	4.38	6.14
37	32	61	95	114	134	164	242	124908	1.37	3.26	5.49	6.73	8.04	10.0	15.1
38	25	62	83	94	101	117	501	91902	0.91	3.33	4.70	5.42	5.88	6.93	65.2
39	15	42	55	59	62	67	1240	125337	0.30	2.02	2.87	3.13	3.33	3.66	325

Site #	Specific Conductance (observed) µS/cm								Chloride (modeled) mg/L						
	Min	5%	25%	Median	75%	95%	Max	N	Min	5%	25%	Median	75%	95%	Max
Mid-Atlantic															
40	76	205	494	676	1040	2090	12600	34078	1.80	31.6	114	166	269	568	3558
41	46	158	188	198	202	207	219	56419	2.73	12.2	14.8	15.6	16.0	16.4	17.4
42	104	231	278	321	370	436	1330	45076	7.87	24.6	30.7	39.4	57.9	82.8	420
43	142	256	313	364	404	498	1220	45353	10.8	26.4	34.2	41.2	46.7	74.4	312
44	97	212	265	361	519	1150	8710	76800	2.00	37.7	54.5	84.9	135	335	2728
45	49	199	304	344	486	745	5500	100149	2.90	21.9	56.1	69.1	115	199	1745
46	44	697	1070	1390	1880	5470	57900	434537	4.00	131	239	330	470	1497	16486
47	152	307	329	348	362	421	2470	64548	10.5	38.7	42.8	46.2	48.8	59.5	432
48	51	323	347	359	375	430	2070	55211	3.30	43.6	48.3	50.6	53.7	64.4	383
49	94	234	302	359	498	1180	4420	43874	4.80	28.0	48.2	65.2	107	310	1275
50	50	108	142	164	192	366	628	45238	6.62	15.8	21.9	28.9	37.7	92.7	176
51	42	55	61	65	68	75	155	22996	5.66	7.50	8.34	8.90	9.33	10.3	21.6
52	34	46	49	51	55	60	81	19945	3.79	5.25	5.62	5.86	6.35	6.96	9.51
53	61	279	393	555	889	2500	9020	253879	1.80	55.0	89.4	138	239	725	2692
54	34	221	319	454	766	2800	13000	261890	4.40	43.7	72.3	112	203	797	3778
55	58	254	563	845	1210	2860	12100	173967	2.50	43.1	133	216	322	804	3501
56	100	317	462	621	1000	3180	9020	112901	3.30	62.0	103	148	256	875	2535
57	58	138	159	176	200	340	1620	114673	6.80	19.6	23.3	26.3	30.5	55.2	280
58	92	321	677	955	1820	5290	17200	164854	2.10	60.6	163	243	491	1488	4910
59	74	321	517	631	881	2760	11800	341657	4.47	44.0	76.4	108	179	708	3253
Mid-Atlantic high-frequency only															
60	46	262	468	556	917	3190	10600	32833	2.06	39.0	91.9	118	223	885	3043
61	46	198	290	364	529	1750	13400	123574	2.06	28.0	43.7	61.6	110	465	3859
62	99	297	398	511	677	1320	7260	114241	11.1	44.9	71.5	104	153	340	2070
63	65	341	389	454	556	1500	7140	101967	5.31	54.9	68.9	87.8	118	392	2035
64	43	143	244	336	553	1730	8860	56648	1.55	18.6	35.9	53.4	117	459	2536
65	62	166	217	257	340	726	3100	123212	4.79	22.6	31.3	38.1	54.6	167	859
Mid-Atlantic carbonate															
66	319	652	768	821	868	960	2500	22643	36.8	58.8	66.5	85.4	105	142	772
67	224	532	583	600	622	640	692	46451	2.30	9.21	11.7	12.5	13.6	14.5	17.0
68	206	387	450	483	505	547	642	98303	12.5	18.9	21.2	22.4	23.1	24.6	28.0
69	211	495	533	555	573	590	686	91057	10.7	12.6	12.9	13.0	13.2	13.3	13.9
New England															
70	56	149	193	216	233	256	305	156607	17.1	34.5	46.1	52.1	56.6	62.6	75.5
71	339	891	1210	1440	1820	2500	13900	110265	73.7	239	335	404	518	722	4145
72	79	704	1350	1810	2420	4830	21500	145442	65.0	111	335	494	705	1539	7307
73	73	820	1260	1690	2260	3590	30700	77251	22.4	147	310	468	678	1169	11164
74	61	108	132	156	172	196	375	139236	11.7	24.4	30.9	37.5	41.8	48.3	96.9
75	62	82	100	122	138	157	637	139009	12.8	18.3	23.2	29.2	33.6	38.8	171
76	28	64	84	97	112	134	309	155665	7.30	12.7	18.4	22.1	26.4	32.6	82.4
77	153	246	335	426	513	764	4460	123886	22.3	50.6	77.6	105	132	208	1331

Site #	Specific Conductance (observed) µS/cm								Chloride (modeled) mg/L						
	Min	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**	**N**	**Min**	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**
78	16	48	62	72	85	98	324	152605	3.90	8.30	10.9	12.8	15.2	17.7	60.1
79	46	115	138	174	199	234	288	109923	10.6	22.4	29.1	39.6	46.9	57.1	72.9
80	29	67	80	96	108	120	149	103173	5.80	13.5	17.1	21.6	24.9	28.2	36.3
81	195	361	458	517	613	812	1410	58013	19.4	38.5	69.6	88.5	119	183	375
82	47	120	149	175	191	247	3980	150018	7.00	21.2	29.8	37.5	42.3	58.9	1169
83	32	107	147	183	219	268	614	149314	10.3	20.1	32.1	42.9	53.7	68.4	172
84	226	468.3	588	678	797.5	1190	3210	56647	21.7	59.3	81.1	108	144	263	872
85	48	82	106	123	146	169	228	114664	6.37	14.3	19.9	23.9	29.2	34.6	48.4
86	158	323	425	537	659	860	4280	191116	21.8	70.5	101	134	170	229	1240
87	31	452	995	1420	2160	7680	98500	178699	6.10	35.7	217	359	605	2446	32735
88	42	212	254	312	375	460	3530	134375	10.3	27.7	39.7	56.2	74.2	98.5	976
89	150	230	278	356	430	524	1600	138874	22.4	43.0	56.8	79.3	101	128	438
90	42	71	84	94	104	126	388	156645	7.61	14.8	18.1	20.6	23.0	28.5	93.7
New England high-frequency only															
91	221	616	823	942	1080	1830	6430	85934	34.3	120	187	226	271	515	2013
92	48	77	93	104	120	151	1020	33687	16.0	19.1	20.8	21.9	23.6	26.9	251
93	85	117	150	195	255	364	807	126717	19.9	23.3	26.8	31.5	37.8	49.4	182

Table S9. Statistical summaries of *non-winter* high-frequency data

Site #	Specific Conductance (observed) µS/cm								Chloride (modeled) mg/L						
	Min	5%	25%	Median	75%	95%	Max	N	Min	5%	25%	Median	75%	95%	Max
Southeast															
1	30	113	182	211	235	272	861	250038	0.70	10.0	19.2	23.0	26.2	31.1	188
2	28	70	98	114	126	140	204	249719	0.46	3.75	5.95	7.20	8.14	9.24	14.3
3	34	70	104	123	136	153	296	246173	0.85	3.65	6.29	7.77	8.78	10.1	21.2
4	25	62	104	132	150	166	249	171071	0.52	3.00	5.81	7.69	8.90	9.97	15.5
5	21	75	123	154	173	196	323	192133	0.77	4.00	6.87	8.72	9.86	11.2	18.8
6	33	114	199	252	278	315	968	251613	4.14	7.78	11.6	14.0	15.1	16.8	46.1
7	25	62	103	125	138	153	348	250040	0.55	3.10	5.93	7.44	8.34	9.37	22.8
8	21	110	162	215	253	296	612	177002	1.00	4.89	7.16	9.48	11.1	13.0	26.8
9	27	89	149	182	200	219	1270	250482	0.50	4.41	8.49	10.7	14.3	25.5	645
10	16	68	109	128	138	147	254	247972	0.50	3.19	5.33	6.32	6.84	7.31	12.9
11	32	115	210	278	318	362	452	253674	0.60	3.91	8.80	12.3	14.4	16.6	21.3
Southeast high-frequency only															
12	29	66	82	90	94	101	160	222510	1.17	3.59	4.64	5.16	5.42	5.88	9.74
13	19	55	69	73	75	80	293	228809	0.52	2.87	3.79	4.05	4.18	4.51	18.4
14	18	52	63	66	69	74	124	167995	0.45	2.67	3.39	3.59	3.79	4.11	7.38
15	16	39	48	51	53	58	80	191661	0.32	1.82	2.41	2.61	2.74	3.07	4.51
16	15	60	92	110	117	125	286	127910	0.30	3.20	5.29	6.47	6.93	7.45	18.0
17	18	52	83	94	99	105	233	86554	0.45	2.67	4.70	5.42	5.75	6.14	14.5
18	29	50	57	62	69	79	111	91297	1.17	2.54	3.00	3.33	3.79	4.44	6.53
19	21	60	90	104	115	128	253	245680	0.65	3.20	5.16	6.08	6.80	7.65	15.8
20	26	45	50	54	58	66	94	224617	0.97	2.22	2.54	2.81	3.07	3.59	5.42
21	14	62	84	88	93	99	135	115621	0.30	3.33	4.77	5.03	5.36	5.75	8.10
22	18	58	79	83	86	91	421	253484	0.45	3.07	4.44	4.70	4.90	5.23	37.1
23	42	61	67	79	92	110	169	203612	2.02	3.26	3.66	4.44	5.29	6.47	10.3
24	13	86	110	115	118	125	499	111025	0.30	4.90	6.47	6.80	6.99	7.45	64.5
25	28	66	96	113	126	141	219	112696	1.11	3.59	5.55	6.66	7.51	8.50	13.6
26	10	53	87	95	102	110	184	224247	0.30	2.74	4.96	5.49	5.94	6.47	11.3
27	22	56	71	79	86	99	134	248362	0.71	2.94	3.92	4.44	4.90	5.75	8.04
28	16	58	63	66	68	72	208	118944	0.32	3.07	3.39	3.59	3.72	3.98	12.9
29	21	51	74	85	90	107	147	77079	0.65	2.61	4.11	4.83	5.16	6.27	8.89
30	19	72	90	96	101	108	158	250148	0.52	3.98	5.16	5.55	5.88	6.34	9.61
31	22	75	87	95	104	116	153	203098	0.71	4.18	4.96	5.49	6.08	6.86	9.28
32	18	55	84	96	100	111	450	138689	0.45	2.87	4.77	5.55	5.81	6.53	47.3
33	23	64	97	108	112	120	226	130526	0.78	3.46	5.62	6.34	6.60	7.12	14.1
34	21	60	96	111	118	126	460	134888	0.65	3.20	5.55	6.53	6.99	7.51	50.8
35	15	56	85	98	105	112	393	109675	0.30	2.94	4.83	5.68	6.14	6.60	27.3
36	36	62	68	75	80	90	114	69783	1.63	3.33	3.72	4.18	4.51	5.16	6.73
37	27	66	106	136	174	269	400	252181	1.04	3.59	6.21	8.17	10.7	16.9	29.7
38	21	60	82	94	103	114	213	184725	0.65	3.20	4.64	5.42	6.01	6.73	13.2
39	18	42	56	60	64	68	270	253808	0.45	2.02	2.94	3.20	3.46	3.72	16.9

Site #	Specific Conductance (observed) µS/cm								Chloride (modeled) mg/L						
	Min	5%	25%	Median	75%	95%	Max	N	Min	5%	25%	Median	75%	95%	Max
Mid-Atlantic															
40	37	135	227	314	392	505	1220	67924	1.80	11.7	37.8	62.6	84.8	117	320
41	33	153	189	196	200	204	216	110828	1.62	11.8	14.9	15.5	15.8	16.2	17.2
42	79	218	271	305	328	358	667	85083	4.58	22.9	29.8	34.3	42.1	53.4	170
43	94	250	316	357	397	436	566	87038	4.19	25.6	34.6	40.2	45.7	54.1	96.8
44	42	146	206	240	274	333	1730	158398	2.00	16.9	35.8	46.6	57.4	76.1	518
45	32	163	251	276	308	358	607	206809	2.90	10.2	38.8	47.0	57.4	73.6	155
46	7	423	906	1080	1240	1440	6210	856226	4.00	53.6	192	241	287	344	1708
47	54	257	304	329	363	403	525	114016	1.90	29.6	38.2	42.8	48.9	56.2	78.4
48	47	251	314	336	371	412	475	118147	3.30	29.6	41.8	46.1	52.9	60.9	73.1
49	28	212	283	309	323	338	2180	86377	4.80	21.4	42.6	50.3	54.5	59.0	608
50	33	98	125	138	151	171	291	55721	6.00	14.2	18.5	20.7	24.8	31.1	69.0
51	35	55	64	74	85	155	185	49594	4.68	7.50	8.76	10.2	11.7	21.6	25.8
52	23	28	42	50	56	67	121	49532	2.45	3.06	4.76	5.74	6.47	7.81	14.4
53	28	207	311	366	431	509	3190	539443	1.80	33.3	64.7	81.3	101	124	933
54	15	176	239	289	350	407	738	543996	4.40	30.5	48.9	63.5	81.4	98.0	195
55	39	218	432	530	606	687	1800	321247	2.50	32.6	95.0	124	146	169	494
56	10	184	323	434	522	625	965	228862	3.30	24.2	63.7	95.2	120	150	246
57	23	122	153	166	182	195	381	217616	6.80	16.8	22.3	24.6	27.4	29.7	62.4
58	33	190	460	645	761	863	1340	326939	2.10	23.0	101	154	187	216	353
59	6	238	423	500	546	612	1510	645433	1.90	30.7	60.3	72.7	84.6	103	356
Mid-Atlantic high-frequency only															
60	29	133	281	405	488	601	768	56391	0.70	16.9	42.2	73.5	97.7	131	179
61	34	132	228	277	311	363	1390	245577	0.70	16.7	33.1	41.5	47.3	61.3	360
62	36	236	355	416	480	552	795	227393	0.70	34.5	59.0	76.7	95.4	116	187
63	48	241	340	362	388	450	634	194906	2.40	35.4	54.6	61.0	68.6	86.6	140
64	16	101	179	217	243	293	545	112191	0.70	11.5	24.8	31.3	35.7	44.2	114
65	53	161	210	235	259	289	1520	228597	3.26	21.7	30.1	34.3	38.4	43.6	398
Mid-Atlantic carbonate															
66	110	558	735	776	804	859	1470	44538	22.9	52.6	64.3	67.0	78.5	101	351
67	139	556	588	600	610	624	741	93362	2.30	10.4	11.9	12.5	13.0	13.7	19.3
68	137	360	456	510	531	549	573	204221	10.1	18.0	21.4	23.3	24.1	24.7	25.6
69	78	465	517	538	560	588	873	181868	9.84	12.4	12.8	12.9	13.1	13.3	15.2
New England															
70	52	172	220	259	306	350	411	311529	17.1	40.6	53.2	63.4	75.7	87.3	103
71	24	830	1190	1480	1950	2760	4310	190058	61.0	221	329	416	557	800	1266
72	26	388	1280	1760	2200	2750	11800	281921	65.0	65.0	310	477	629	819	3950
73	28	357	918	1150	1440	1810	9320	144139	22.4	22.4	183	269	376	512	3281
74	36	113	140	164	189	262	422	260038	9.90	25.8	33.1	39.6	46.4	66.2	110
75	59	87	108	125	146	165	459	258488	12.0	19.6	25.4	30.1	35.8	41.0	122
76	36	75	98	119	143	170	215	314308	7.30	15.9	22.4	28.4	35.2	42.9	55.7
77	65	276	363	430	492	659	2870	223324	18.4	59.7	86.1	106	125	176	848

Site #	Specific Conductance (observed) µS/cm								Chloride (modeled) mg/L						
	Min	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**	**N**	**Min**	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**
78	19	55	71	82	91	105	230	294864	3.90	9.61	12.6	14.7	16.4	19.0	42.5
79	11	127	156	183	208	232	255	219816	10.6	25.9	34.4	42.3	49.6	56.6	63.3
80	42	70	87	99	115	142	193	204739	6.61	14.4	19.1	22.4	26.9	34.3	48.5
81	145	306	418	470	516	568	776	109643	13.6	32.2	56.8	73.4	88.2	105	172
82	59	135	169	198	255	392	881	293475	7.00	25.6	35.7	44.4	61.3	102	247
83	46	122	177	216	265	333	491	280264	10.3	24.6	41.1	52.8	67.5	87.9	135
84	132	379	567	678	769	855	1490	109888	8.40	45.4	74.7	108	136	162	353
85	53	92	129	174	222	293	587	221929	7.53	16.6	25.3	35.8	47.0	63.5	132
86	61	364	477	583	728	1040	1920	382033	18.4	82.7	116	147	190	282	543
87	0	137	802	1080	1440	1850	28600	342350	6.10	6.10	152	245	365	502	9423
88	1	203	263	303	360	427	2580	271677	10.3	25.1	42.2	53.7	70.0	89.1	704
89	119	240	309	389	479	585	682	280368	22.4	45.9	65.8	88.8	115	145	173
90	47	79	98	117	142	156	386	314870	8.85	16.8	21.5	26.3	32.5	36.0	93
New England high-frequency only															
91	74	544	730	936	1110	1330	4150	151044	18.7	96.4	157	224	281	352	1271
92	50	84	100	113	126	150	400	66017	16.2	19.8	21.5	22.9	24.2	26.8	53.2
93	81	130	166	206	263	358	551	247233	19.5	24.7	28.5	32.7	38.7	48.7	98.7

Table S10. Chloride concentration versus SC regression relationships

Site #	Linear regression					(S)ingle or (P)iece wise model	Second linear regression (if piecewise)[b]				Observed versus predicted		
	Slope[c]	Intercept	R^2	N	Excluded pts[d]		Slope	Intercept	Break-point	Break-point uncertainty	R^2	Median diff (mg/L)	N
Southeast													
1	0.133	-4.97	0.950	227	0	P	0.292	-62.6	362	23.2	0.843	5.42	4
2	0.078	-1.74	0.785	220	1	S					0.808	0.19	7
3	0.078	-1.80	0.823	236	0	S					0.923	-0.82	19
4	0.067	-1.15	0.850	238	0	S					0.927	-0.12	14
5	0.060	-0.49	0.705	294	0	S					0.936	0.01	12
6	0.045	2.66	0.566	239	1	S					0.907	1.91	20
7	0.069	-1.17	0.847	225	2	S					0.749	-0.29	18
8	0.044	0.082	0.715	98	1	S					0.685	-0.61	9
9	0.068	-1.65	0.879	230	0	P	0.589	-104	195	1.83	0.636	-0.62	15
10	0.052	-0.36	0.829	197	1	S					0.036	0.57	5
11	0.051	-2.01	0.866	144	1	S					0.825	0.48	7
Southeast – piecewise regression	0.065	-0.72	0.852	2348	7	P	0.351	-111	385	4.79	0.682	1.01	130
Southeast – single linear regression	0.114	-6.45	0.582	2348	7	S	–	–	–	–	0.625	0.23	130
Mid-Atlantic													
40	0.285	-26.7	0.979	87	1	S					0.999	-1.58	5
41	0.085	-1.18	0.762	186	1	S					0.845	0.02	73
42	0.131	-5.81	0.776	124	1	P	0.377	-81.7	309	9.29	0.426	2.55	67
43	0.137	-8.70	0.869	129	1	P	0.329	-89.3	421	20.0	0.575	0.71	71
44	0.317	-29.4	0.992	155	0	S					0.986	1.33	129
45	0.325	-42.7	0.781	67	0	S					0.894	3.40	67
46	0.286	-67.4	0.967	90	0	S					0.978	4.47	89
47	0.182	-17.1	0.914	79	1	S					0.915	-0.28	72
48	0.194	-19.1	0.934	71	1	S					0.942	-0.34	67
49	0.298	-41.8	0.982	163	0	S					0.921	-4.66	15
50	0.158	-1.28	0.981	45	0	P	0.316	-22.9	137	5.80	NA	NA	NA
51	0.141	-0.25	0.774	32	1	S					0.802	-0.01	26
52	0.122	-0.35	0.655	34	1	S					0.692	0.09	26
53	0.302	-29.1	0.972	158	2	S					0.980	-3.10	141
54	0.292	-20.9	0.992	209	2	S					0.991	1.68	207
55	0.292	-31.1	0.981	156	0	S					0.930	1.27	127
56	0.284	-28.1	0.987	311	2	S					0.987	1.73	287
57	0.176	-4.65	0.910	20	0	S					0.879	0.27	18
58	0.287	-31.6	0.990	198	2	S					0.991	1.17	153
59	0.160	-7.38	0.979	87	1	S	0.282	-69.1	509	33.8	0.936	-0.64	64

Site #	Linear regression					(S)ingle or (P)iece wise model	Second linear regression (if piecewise)[b]				Observed versus predicted		
	Slope[c]	Intercept	R^2	N	Excluded pts[d]		Slope	Intercept	Break-point	Break-point uncertainty	R^2	Median diff (mg/L)	N
MidAtlantic piecewise regression	0.171	-5.80	0.979	2401	17	P	0.291	-44.5	321	11.2	0.978	-2.85	1704
MidAtlantic – single linear regression	0.283	-32.2	0.976	2401	17	S					0.976	-3.15	1704
Mid-Atlantic carbonate													
66	0.066	15.6	0.900	119	0	S	0.409	-250	776	12.6	0.892	0.86	31
67	0.049***	-16.6	0.199	28	0	S					NA	NA	NA
68	0.035	5.21	0.261	140	0	S					0.248	0.64	121
69	0.007[#]	9.31	0.084	21	0	S					0.529	-0.29	8
New England													
70	0.262	-4.55	0.976	27	0	S					0.977	-1.33	17
71	0.300	-28.1	0.996	59	0	S					0.993	2.39	23
72	0.346	-132	0.996	90	2	S					0.628	2.62	26
73	0.369	-155	0.997	79	0	S					0.346	-23.6	32
74	0.271	-4.89	0.987	28	0	S					0.985	-0.75	15
75	0.274	-4.24	0.976	27	1	S					0.975	0.14	18
76	0.284	-5.47	0.974	28	0	S					0.956	-0.45	20
77	0.304	-24.2	0.998	69	1	S					0.997	0.56	22
78	0.188	-0.71	0.721	35	0	S					0.646	-0.25	29
79	0.292	-11.1	0.977	29	0	S					0.975	-0.89	20
80	0.277	-5.03	0.972	28	0	S					0.928	-0.58	19
81	0.115	-3.04	0.875	490	0	P	0.321	-77.3	361	5.80	0.853	-7.12	39
82	0.297	-14.5	0.997	38	0	S					0.990	-0.90	29
83	0.300	-12.0	0.993	38	0	S					0.780	-3.03	29
84	0.155	-13.4	0.877	206	1	P	0.302	-96.4	566	19.3	0.903	-12.4	37
85	0.233	-4.83	0.980	74	1	S					0.947	-2.27	4
86	0.296	-24.9	0.993	72	1	S					0.988	-2.62	24
87	0.334	-115	0.994	103	0	S					0.985	-18.6	71
88	0.286	-32.9	0.990	71	0	S					0.968	-1.37	34
89	0.288	-23.3	0.990	35	0	S					0.993	1.25	31
90	0.249	-2.85	0.978	26	0	S					0.972	-0.34	20
New England piecewise regression including only samples with SC <20,000 µS/cm	0.106	10.9	0.994	1636	7	P	0.327	-80.7	415	17.4	0.956	-8.75	558
New England piecewise regression - all	0.310	-47.2	0.997	1652	7	P	0.394	-1221	14027	279	0.959	-15.8	559

Site #	Linear regression					(S)ingle or (P)iece wise model	Second linear regression (if piecewise)[b]				Observed versus predicted		
	Slope[c]	Intercept	R^2	N	Excluded pts[d]		Slope	Intercept	Break-point	Break-point uncertainty	R^2	Median diff (mg/L)	N
New England – single linear regression	0.341	-68.3	0.993	1652	7	S					0.956	-16.7	559

[a] S = Single linear regression, P = Piecewise regression. All regression models can be found in the accompanying USGS data release.[10]
[b] Only shown if a piecewise regression was used.
[c] p values for discrete Cl vs SC slopes are <0.001 unless indicated otherwise (** is <0.01, *** is <0.05, ^ <0.1, # >0.1). The only cases in which p values for slopes were not <0.001 were for some of the Mid-Atlantic carbonate streams.
[d] Not included in the reported N values. All excluded discrete samples are reported in Table S15.

Table S11. Regression relationships between median (high-frequency) [Cl] and watershed characteristics

	Region[a]	Slope	1 σ uncertainty	R^2	p value	intercept	1 σ uncertainty	p value
Salt application rates included								
Median [Cl] vs Average salt application (kg/km^2)	All	7.21e-4	5.20e-5	0.69	*<0.001*	0.327	6.36	0.96
	MA	3.95e-4	5.01e-5	0.70	*<0.001*	22.6	8.18	<0.05
	NE	1.44e-3	1.15e-4	0.87	*<0.001*	–94.2	19.1	*<0.001*
Median [Cl] vs Average salt application (kg/km^2) + Roads per watershed area (km/km^2)	All	6.75e-4 3.04	5.43e-5 1.29	0.70	*<0.001* <0.05	–1.95	1.05	<0.01
	MA	3.16e-4 3.23	5.93e-5 1.46	0.74	*<0.001* <0.05	3.52	11.5	0.76
	NE	1.41e-3 0.722	3.25e-5 5.94	0.87	*<0.001* 0.90	–92.9	22.2	*<0.001*
Median [Cl] vs Average salt application (kg/km^2) + ISC[b]	All	7.00e-4 0.643	5.70e-5 0.367	0.69	*<0.001* <0.1	–9.72	8.50	0.26
	MA	1.96e-4 2.10	7.68e-5 0.664	0.78	<0.05 <0.01	12.4	7.69	0.12
	NE	1.78e-3 –2.19	4.10e-4 2.62	0.87	*<0.001* 0.41	–117	33.5	<0.01
Salt application rates excluded								
Median [Cl] vs Roads per watershed area (km/km^2)	All	8.83	2.01	0.17	*<0.001*	–14.4	17.4	0.41
	MA	7.92	1.70	0.44	*<0.001*	–1.61	16.7	0.92
	NE	24.7	2.90	0.76	*<0.001*	–23.7	20.8	0.27
Median [Cl] vs Roads per watershed area (km/km^2) + Annual Temperature (°C)	All	12.8 –20.1	1.47 2.21	0.59	*<0.001* *<0.001*	235	29.0	*<0.001*
	MA	8.11 –3.61	1.84 11.2	0.43	*<0.001* 0.75	43.3	141	0.76
	NE	25.0 –22.5	2.98 38.0	0.75	*<0.001* 0.56	187	358	0.61
Median [Cl] vs ISC + Annual Temperature (°C)	All	3.61 –23.1	0.485 2.49	0.53	*<0.001* *<0.001*	296	31.3	*<0.001*
	MA	3.76 –12.3	0.437 7.58	0.75	*<0.001* 0.12	167	95.8	<0.1
	NE	8.80 –7.0	1.02 37.3	0.76	*<0.001* 0.48	187	358	0.61

[a] SE = Southeast, MA = Mid-Atlantic (silicate only), NE (New England)
[b] ISC = Impervious surface cover

Table S12. Statistical summaries of chronic exceedance events

Site #	Days with mean [Cl] >230 mg/L	Exceedance days (number of 4-day periods with mean [Cl] >230 mg/L)	Events		Event duration (days)		Maximum event [Cl] (mg/L)		Mean event [Cl] (mg/L)		Time since the last event (days)	Date each year when the last event ends
	Annual median	Annual median	Annual median	Study period total	Median	IQR	Mean	1 σ	Mean	1 σ	Median	Median
Mid-Atlantic												
40	43	48	4	10	12.5	5.75	1316	934	359	85.8	11	March 27
41	0	0	0									
42	0	0	0	1	3	–	420	–	237	–	–	March 9
43	0	0	0	1	1	–	312	–	238	–	–	March 9
44	5	7	2	15	5	4	1093	708	279	57.2	17	March 17
45	1	0	0	2	2.5	1.5	682	322	229	6.21	53	March 26
46	227	213	15	74	6	9	1112	2662	277	91.6	6	September 30[a]
47	0	0	0									
48	0	0	0									
49	9.5	8.5	2	9	4	1	827	253	257	23.8	23	February 26
50	0	0	0									
51	0	0	0									
52	0	0	0									
53	32.5	35	3	29	9	7	1430	588	366	124	9	March 9
54	23	27	3	35	8	5	1659	798	379	122	12	March 10
55	55	65	4	17	12	20	1669	967	360	104	7	March 26
56	31.5	34	3.5	24	6.5	6.5	1063	632	347	121	8.5	March 26
57	0	0	0									
58	72	80	3	16	13	11.25	2151	1535	499	226	10	April 1
59	25	29.5	3	16	7	2.75	1850	845	359	115	6	March 17
Mid-Atlantic high-frequency only												
60	17	21	4	10	8.5	4.25	2133	865	428	149	9	March 30
61	13	15	2	28	5	3	1728	892	336	106	19	February 27
62	13	13.5	2	17	7	8	966	598	287	57.3	14.5	March 14
63	15	14	3	9	4	2	1155	442	282	82.9	16.5	March 20
64	8	10	2	15	5	2.5	1018	692	290	62.9	15.5	March 20
65	3	0	0	4	2.5	1	572	203	222	18.6	98	March 7
New England												
70	0	0	0									
71	344	341	3	20	104.5	144.7	1552	1275	417	113	3	September 30[a]
72	324	349	4	49	52	84.0	2023	2093	433	139	2	September 30[a]
73	254	253	9.5	56	12.5	17.5	1373	2286	325	112	5	September 20[a]
74	0	0	0									
75	0	0	0									
76	0	0	0									
77	7	4	2	9	3	3	810	442	243	28.1	63	February 18

Site #	Days with mean [Cl] >230 mg/L	Exceedance days (number of 4-day periods with mean [Cl] >230 mg/L)	Events		Event duration (days)		Maximum event [Cl] (mg/L)		Mean event [Cl] (mg/L)		Time since the last event (days)	Date each year when the last event ends
	Annual median	**Annual median**	**Annual median**	**Study period total**	**Median**	**IQR**	**Mean**	**1 σ**	**Mean**	**1 σ**	**Median**	**Median**
78	0	0	0									
79	0	0	0									
80	0	0	0									
81	2	1	1	3	1	2	310	16.1	234	2.71	NA	March 9
82	0	0	0									
83	0	0	0									
84	10	5	1	7	3	2.5	596	192	247	28	45.2	February 21
85	0	0	0									
86	3	4	1	25	4	20	515	295	242	19.0	15.5	September 30[a]
87	229	239	13	130	7	15.5	2173	4002	355	237	4	September 30[a]
88	0	0	0									
89	0	0	0									
90	0	0	0									
New England high-frequency only												
91	176	173	7	46	5	18.75	681	479	266.7	54.6	5	September 27[a]
92	0	0	0									
93	0	0	0									

[a] September 30 is the end of the water year meaning that exceedances occur throughout the year. Events also commonly occur in fall months (October, November).
[b] #65 – 0 events from 2008–2014. 2015-2018: 3, 2, 0, 5 (median =2.5)
[c] #85 – 12 events 2008–2014: 2008=4, 2011=1, 2014=7, others=0; 2015–2018 = 60, 113, 104, 126, respectively (median = 108.5)

Table S13. Statistical summaries of acute exceedance events

Site #	Hours with mean [Cl] >860 mg/L	Events		Event duration (hours)		Maximum event [Cl] (mg/L)		Mean event [Cl] (mg/L)		Time since the last event (days)	Date when the last event ends
	Annual median	Annual median	Study period total	Median	IQR	Mean	1 σ	Mean	1 σ	Median	Median
Mid-Atlantic											
40	50	3	8	11.0	30.7	1552	886	1206	520	28.0	March 17
41	0	0	0								
42	0	0	0								
43	0	0	0								
44	4	1	12	3.5	6.7	1445	593	1183	326	5.8	March 16
45	0	0	0								
46	240	24	95	7.0	13.0	3250	2966	1691	919	4.1	March 26
47	0	0	0								
48	0	0	0								
49	5	1.5	7	4.0	3.5	1052	131	976	72.9	23.0	February 25
50	0	0	0								
51	0	0	0								
52	0	0	0								
53	70.5	3	77	5.0	18.0	1292	386	1041	151	5.0	February 21
54	93	5	67	10.0	21.5	1524	613	1122	204	6.9	March 5
55	166	7	41	9.0	16.0	1605	715	1178	269	8.6	March 17
56	93	3	30	22.0	32.8	1310	404	1073	183	8.6	March 18
57	0	0									
58	171	5	41	20.0	45.0	2088	1115	1319	351	5.0	March 20
59	98	6	37	6.0	14.0	1604	715	1224	379	6.0	March 13
Mid-Atlantic high-frequency only											
60	45	3	15	31.0	40.5	1980	785	1413	408	10.4	March 20
61	27	3	44	9.5	14.8	1682	720	1243	304	11.4	March 5
62	1.5	0.5	15	5.0	6.0	1338	451	1091	208	4.2	March 4
63	34	2	6	14.5	4.7	1391	360	1132	188	19.5	March 16
64	15	2	13	8.0	3.0	1376	542	1082	259	4.0	February 26
65	0	0	0								
New England											
70	0	0	0								
71	78	17	121	4.0	5.0	1380	674	1091	265	7.3	April 7
72	511.5	21	306	7.0	18.0	1687	1273	1149	385	4.7	July 15
73	225.2	16	110	4.0	10.0	1966	1847	1217	779	5.6	March 21
74	0	0	0								
75	0	0	0								
76	0	0	0								
77	3.1	1	9	3.0	4.0	1112	171	1008	107	1.8	February 16
78	0	0	0								
79	0	0	0								

Site #	Hours with mean [Cl] >860 mg/L	Events		Event duration (hours)		Maximum event [Cl] (mg/L)		Mean event [Cl] (mg/L)		Time since the last event (days)	Date when the last event ends
	Annual median	Annual median	Study period total	Median	IQR	Mean	1 σ	Mean	1 σ	Median	Median
80	0	0	0								
81	0	0	0								
82	0	0	0								
83	0	0	0								
84	0	0	1	1.0	0.0	872		868			March 4
85	0	0	0								
86	0	0	3	1.0	1.5	1089	138	1000	103		December 14
87	218.3	30	334	4.1	7.1	3131	2831	1651	937	4.8	March 26
88	0	0	0								
89	0	0	0								
90	0	0	0								
New England high-frequency only											
91	32	5	20	6.0	6.7	1237	315	1048	143	10.7	February 26
92	0	0									
93	0	0									

Table S14. Discrete samples excluded from regression analyses [a]

Site #	Sample date and time	Discrete SC	Cl	High-frequency SC
		µS/cm	mg/L	µS/cm
Southeast				
2	2006-07-15 15:00:00	118	15.9	*nd*
4	2004-09-15 08:00:00	121	15.7	*nd*
4	2007-02-01 07:00:00	131	16.5	*nd*
6	2007-02-13 19:30:00	259	32	*nd*
8	2006-11-06 13:15:00	570	9.4	*nd*
10	2005-09-01 11:15:00	247	5.17	*nd*
11	2007-12-11 09:15:00	3	13.8	292
Mid-Atlantic				
40	1999-03-11 08:15:00	3960	1820	*nd*
41	2001-07-03 09:00:00	168	65	*nd*
42	2018-05-29 09:15:00	453	28	266
43	2018-05-29 08:30:00	557	32	310
47	2018-03-23 11:10:00	499	106	494
48	2018-03-23 10:10:00	548	117	546
51	2015-09-30 05:00:00	123	8.21	*nd*
52	2015-09-30 07:30:00	80	4.79	58
53	2016-02-16 09:20:00	1330	508	1330
53	2018-05-14 18:40:00	51	102	51
54	2014-02-03 07:00:00	3830	919	3830
54	2014-02-05 10:30:00	3910	1280	3910
56	2013-12-12 15:45:00	1400	14.7	1400
56	2016-02-16 14:45:00	4690	1510	*nd*
58	2013-12-11 13:15:00	7030	2150	7057
58	2014-02-03 06:35:00	6820	1560	6820
59	2018-02-10 14:00:00	496	208	681
New England				
72	2017-12-23 11:57:00	12,800	6580	6699
72	2017-12-23 14:37:00	19,500	4230	8705
75	2010-06-17 16:10:00	150	20.5	85
77	2017-04-01 03:36:00	1310	261	894
84	2003-11-18 11:30:00	758	50.1	*nd*
85	2000-02-14 13:00:00	150	53.1	*nd*
86	2005-12-15 13:01:00	565	105	*nd*

[a] nd = no data

Table S15. Statistical summaries of sites with ≥3 [Cl] and SC discrete samples up through 1997

Site #	Specific Conductance (µS/cm)								Chloride (mg/L)							
	Min	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**	**N**	**Min**	**5%**	**25%**	**Median**	**75%**	**95%**	**Max**	**N**
Southeast																
5	36	53	108	138	154	185	250	353	1.70	2.10	4.85	7.9	8.8	11	14	63
10	104	106	112	124	137	144	147	4	5.20	5.27	5.55	5.90	6.75	7.43	7.60	3
Mid-Atlantic																
40	50	103	168	213	240	276	296	41	3.50	8.92	22.0	32.3	37.0	48.6	65.0	45
41	37	62	138	143	153	164	169	9	1.40	3.56	10.0	12.0	13.0	15.4	17.0	9
44	74	74	76	81	90	93	94	4	4.00	4.08	4.25	5.95	11.7	14.7	16.0	4
49	109	155	184	200	225	285	545	115	9.70	12.9	14.5	17.0	21.0	31.5	240	79
50	74	74	76	81	90	93	94	4	4.00	4.08	4.25	5.95	11.7	14.7	16.0	4
53	100	155	210	235	310	445	830	61	16.0	18.0	21.0	24.0	44.0	57.7	59.0	14
New England																
71	452	519	725	833	957	1435	1790	10	122	140	200	220	260	390	491	10
72	531	703	1020	1425	1600	1824	1950	10	130	189	269	387	442	503	536	10
73	362	376	460	639	665	704	716	9	75.7	78.1	98.7	163	168	179	185	9
77	183	186	201	215	231	234	234	10	39.9	39.9	40.3	41.0	43.0	47.4	51	10
81	54	190	255	294	336	400	630	360	6.00	17.0	24.0	28.0	33.0	45.9	99	323
84	227	280	397	466	546	634	722	40	17.0	32.9	41.2	52.0	70.5	91.9	140	40
86	266	269	289	304	444	449	450	10	55.0	57.1	63.3	67.5	106	109.8	112	10

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