Supplemental information for:

## Increasing chloride in US rivers and its link to potential corrosivity of source waters

Stets, E.G.<sup>1</sup>, C. J. Lee<sup>2</sup>, D.A. Lytle<sup>3</sup>, M.D. Schock<sup>3</sup>

Contents:

Figure S1. Map of U.S. displaying regions used in status assessment.

Table S1. Numbers of stations in the trend and status analyses by land use categories and regions.

Figure S2. Map of U.S. displaying trend results for chloride concentration.

Figure S3. Map of U.S. displaying trend results for chloride-sulfate mass ratio (CSMR).

Figure S4. Map of U.S. displaying trend results for Larson Ratio (LR).

Figure S5. Plots showing change in sulfate and alkalinity plotted against changes (1992-2012) in chloride-sulfate mass ratio (CSMR) and Larson ratio (LR).

Table S2. Results of Wilcoxon rank sum test for differences in chloride concentration (mg / L), CSMR, and LR between 1992 and 2012 for sites based on land use categories.

Table S3. Results of Kruskal-Wallis non-parametric ANOVA and Dunn's post-hoc pairwise multiple comparison test among sites included in the status assessment.

Figure S6. Plot of the number of reported Pb ALEs per 10 upstream surface drinking water intakes on the y-axis versus (A) chloride concentration (mg/L); and, (B) Larson ratio on the x-axis.

Figure S1. Map of U.S. displaying regions used in status assessment.



Table S1. Numbers of stations in the trend and status analyses by land use categories and regions.

## UPDATED 08/15/2017

Land use category	Status assessment			Trend analysis		
	Chloride	CSMR	LR	Chloride	CSMR	LR
All sites	323	248	104	152	74	76
Undeveloped	156	141	27	36	25	26
Agricultural	52	33	20	27	8	8
Mixed	84	57	39	66	31	32
Urban	28	17	17	12	10	10
Region	Chloride	CSMR	LR	Chloride	CSMR	LR
Northern tier	148	107	47	NA	NA	NA
Southern tier	43	37	30	NA	NA	NA
Interior West	92	87	14	NA	NA	NA
Pacific	40	17	12	NA	NA	NA

NA – Not analyzed.

Figure S2. Map of U.S. displaying trend results for chloride concentration.



Figure S3. Map of U.S. displaying trend results for chloride-sulfate mass ratio (CSMR).



Figure S4. Map of U.S. displaying trend results for Larson Ratio (LR).



**Table S2.** Results of Wilcoxon rank sum test on raw chloride concentration, CSMR, and LR from 1991-1993 versus 2011-2012 by land-use categories. The median values from each time period are displayed along with the Wilcoxon test statistics (*W*) and the associated *P*-value. Test results with P < 0.01 are shown in bold. **Red numbers have not been updated 08/15/2017** 

Parameter	Land-use	Season	1991-1993	2011-2012	W	P-value
Chloride	Undeveloped	Cold	40	46	3339	0.18
		Warm	79	99	5842	0.90
	Mixed	Cold	25	21	3679	0.36
		Warm	23	22	39587	0.79
	Agricultural	Cold	39	41	1400	0.66
		Warm	27	28	2137	0.21
	Urban	Cold	87	144	48	0.005
		Warm	49	56	1251	0.17
CSMR	Undeveloped	Cold	0.17	0.15	2421	0.56
		Warm	0.52	0.49	5721	0.02
	Mixed	Cold	0.88	1.81	639	0.001
		Warm	0.64	0.71	16593	0.004
	Agricultural	Cold	0.53	0.82	313	0.07
		Warm	0.47	0.84	267	0.01
	Urban	Cold	2.9	6.0	28	<0.001
		Warm	1.6	2.2	689	<0.001
LR	Undeveloped	Cold	1.9	2.5	2039	0.006
		Warm	1.9	3.0	4309	0.01
	Mixed	Cold	1.00	1.03	1126	0.44
		Warm	0.82	0.86	16902	0.08
	Agricultural	Cold	0.48	0.50	356	0.26
		Warm	0.48	0.55	324	0.11
	Urban	Cold	2.19	4.96	50	0.005
		Warm	1.58	1.92	844	<0.001

Table S3. Results of Kruskal-Wallis non-parametric ANOVA and Dunn's post-hoc pairwise multiple comparison test among sites included in the status assessment. Comparisons were performed on site-averaged chloride concentration, chloride-sulfate mass ratio (CSMR), and Larson ratio (LR). The  $\chi^2$ , degrees of freedom (df) and *p*-values are displayed for the Kruskal-Wallis test along with the *Z* scores and *p* values of the pairwise Dunn's test.

	Chloride	CSMR	LR
Kruskal-Wallis	X <sup>2</sup> = 73, df = 3, <i>p</i> < 0.01	X <sup>2</sup> = 75, df = 3, <i>p</i> < 0.01	X <sup>2</sup> = 28, df = 3, <i>p</i> < 0.01
Agricultural-Mixed	0.04	-1.89**	-4.62***
Agricultural-	5.28***	3.03***	-3.71***
Undeveloped			
Agricultural-Urban	-1.96**	-3.90***	-4.70***
Mixed-Undeveloped	6.20***	6.37***	0.71
Mixed-Urban	-2.13**	-2.71***	-0.95
Undeveloped-Urban	-6.36***	-6.82***	-1.47*

\* - p < 0.1 \*\* - p < 0.05 \*\*\* - p < 0.01 Figure S5. Comparisons of the changes (1992-2012) in flow-normalized sulfate and alkalinity concentration compared to changes (1992-2012) in the chloride-sulfate mass ratio (CSMR) and the Larson ratio. Grey dots show results for individual sites and boxes show median changes by land use category. A) Change in sulfate versus change in Larson ratio. B) Change in CSMR versus change in sulfate. C) Change in Larson ratio versus change in alkalinity.



2.0



∆ Larson Ratio

Figure S6. Plot of the number of reported Pb ALEs per 10 upstream surface drinking water intakes on the y-axis versus (A) chloride concentration (mg/L); and, (B) Larson ratio on the x-axis. Open circles show results for individual stations and the red line shows a locally-weighted regression (LOESS) smooth through the data points.

