Chloride Contributions from Water Softeners and Other Domestic, Commercial, Industrial, and Agricultural Sources to Minnesota Waters

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January 2019

Funding for this project was provided by the Minnesota Environment and Natural Resources Trust Fund as recommended by the Legislative-Citizen Commission on Minnesota Resources (LCCMR)

Executive summary

High chloride levels in surface waters and groundwater are an emerging concern in Minnesota, as they can negatively impact aquatic and plant life. Previous research has shown that road salt is a major source of chloride, particularly in urban areas, but chloride discharge from water softener use, another major source, has not been quantified. Two chloride mass budgets were undertaken: a wastewater treatment plant (WWTP) chloride budget and a statewide chloride budget. The WWTP chloride budget was performed for wastewater facilities with chloride monitoring data to estimate chloride discharged from household and commercial water softeners relative to other household, commercial, and industrial sources. Results of the WWTP chloride budget were applied to WWTPs statewide through a statewide chloride budget undertaken for the State of Minnesota. Secondary objectives of the statewide chloride budget included estimating chloride from other sources for comparison, such as human excreta; household product use; background drinking water concentrations; drinking water and wastewater chlorination; commercial sources; industrial discharge; road salt use; atmospheric deposition; dust suppressant use; fertilizer application; and livestock waste.

The analysis employed multiple types of data, including groundwater monitoring data, spatial data, WWTP monitoring data, purchasing records, and responses from a statewide survey of water conditioning professionals. The results of the WWTP chloride budget accounted for 98% of the chloride discharged from included WWTPs, attributing remaining chloride to hauled septage, and showed that water softener use was the largest chloride point source investigated in the WWTP chloride budget. At the statewide level, household and commercial water softening were estimated to contribute 65% of WWTP chloride discharge. Industries were also major sources, contributing 22% of the estimated chloride load of statewide WWTPs. Human excreta, household product use, background chloride concentrations, chlorination, and other commercial processes contributed relatively small amounts of chloride, less than 5% of the chloride load. However, since the analysis was conducted at a statewide level, the results are not necessarily generalizable to the local level, where sources may have different chloride discharge and importance based on local conditions.

In the statewide chloride budget, road salt use was the largest chloride source, contributing 403,600 metric tons (t) of chloride annually to surface waters. Chloride from fertilizer use was the next largest chloride source (221,300 t), followed by WWTPs (209,900 t), livestock waste (62,600 t) and residential septic systems (33,100 t). Although fertilizer is a major source of chloride at the statewide level, application rates and monitoring data from research literature indicate that it has a lesser impact on surface water and groundwater quality than sources such as road salt. Previous research on chloride in animal waste, septic system effluent, and dust suppressant application rates suggests that these may be important local sources of chloride; future research investigating their effects on chloride levels in groundwater and surface water would better characterize their environmental impacts. Factors affecting importance of chloride sources at the local level can also include timing of chloride application or discharge and sensitivity of receiving waters. The results of the statewide chloride budget show that water softeners are major sources of chloride and indicate that increasing efficiency of water softener salt use could be a viable strategy to manage chloride levels in wastewater and receiving waters.

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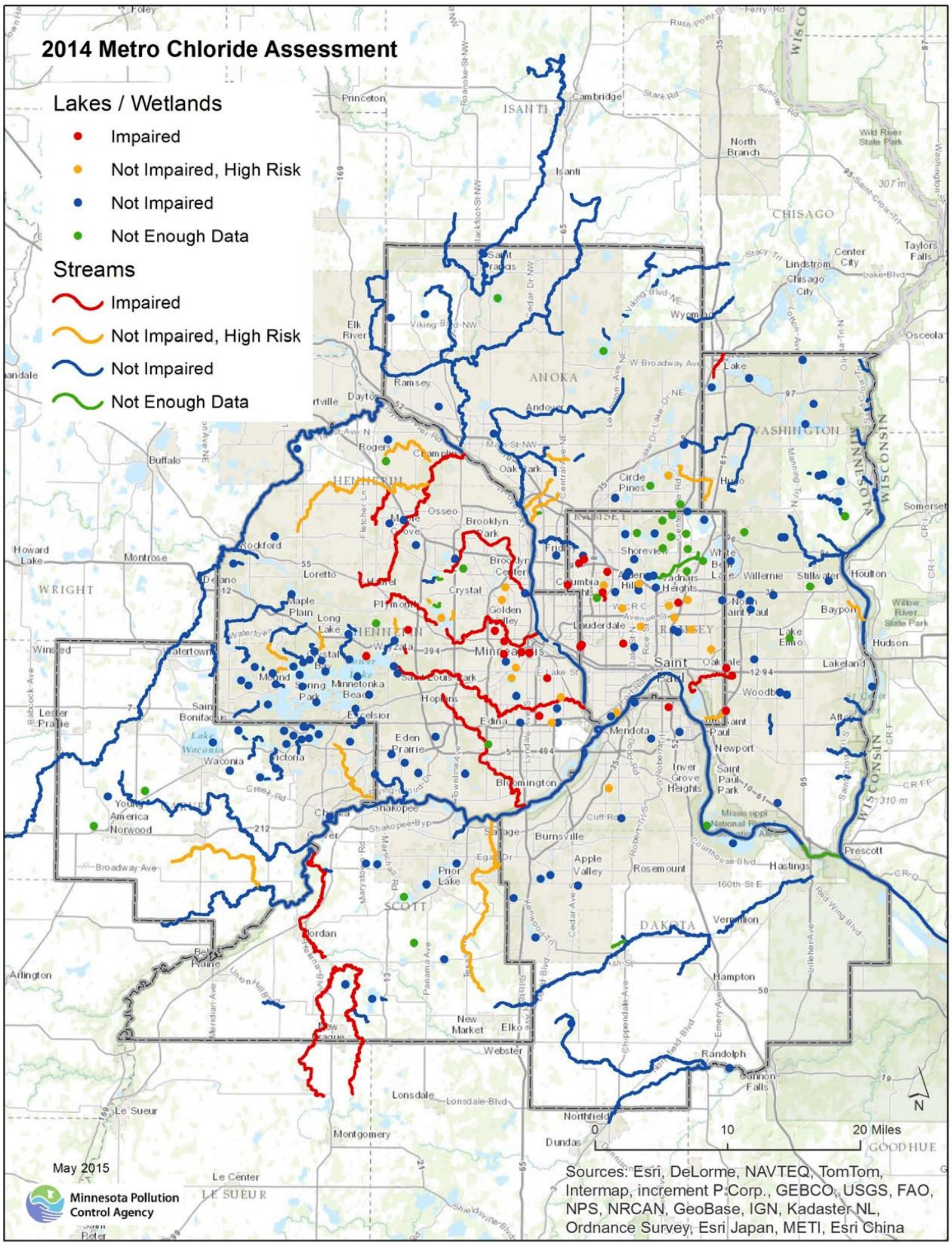
1 Introduction

Chloride levels in Minnesota waters have been increasing over time (Novotny et al., 2008), making chloride an emerging environmental concern across the state (EQB, 2015). Chloride accumulates in water and elevated chloride levels have many negative effects on plant (Miklovic and Galatowitsch, 2005; Richburg et al., 2001; Wilcox, 1986) and aquatic life (Dougherty and Smith, 2006; Karraker et al., 2008).

Due to the harmful impacts of elevated chloride concentrations on aquatic life and ecosystems, the Environmental Protection Agency (EPA) has established water quality standards for chloride. The water quality standard for chronic exposure is a four-day average concentration of 230 mg/L and for acute exposure, a one-hour average concentration of 860 mg/L (EPA, 1986). Water bodies are considered impaired for chloride if they exceed the chronic or acute criteria two times in a three-year period (EPA, 1986). In Minnesota, 39 of its 50 chloride-impaired waters are in the Twin Cities Metropolitan Area (TCMA) and 38 additional water bodies in the TCMA are at high risk of chloride impairment (MPCA, 2016; Figure 1). Furthermore, the number of chloride impairments is dependent on available monitoring data. In the TCMA, monitoring data to assess impairment status is available for less than a third of its water bodies, so there may be a greater number of water bodies in the metropolitan area that exceed chloride standards (MPCA, 2016).



Sources: Esri, DeLorme, NAVTEQ, TomTom, Intermap, increment P Corp., GEBCO, USGS, FAO, NPS, NRCAN, GeoBase, IGN, Kadaster NL, Ordnance Survey, Esri Japan, METI, Esri China

Figure 1. Chloride impairment status of surface waters and wetlands in the Twin Cities Metropolitan Area (from MPCA 2016).

Elevated chloride concentrations have also been found in groundwater below urban regions across the northern United States (Mullaney et al., 2009). Monitoring by the Minnesota Pollution Control Agency (MPCA) found that 27% of groundwater monitoring wells have chloride levels exceeding the EPA secondary drinking water standard of 250 mg/L and that chloride concentrations in shallow groundwater below the TCMA are up to five times higher than in rural wells (MPCA, 2013). In addition, high chloride concentrations can increase corrosivity of drinking water distribution systems and increase the rate of lead release (Stets et al., 2018).

Research has shown that use of deicing salt for winter road maintenance is a major contributor of chloride to surface waters and groundwater (Kelly et al., 2008; Novotny et al., 2009; Perera et al., 2013). In the TCMA, an estimated 365,000 metric tons of deicing salt are applied to surfaces each year (Sander et al., 2007). Analysis of chloride to bromide ratios in groundwater indicated that deicing salt is the dominant source of chloride in TCMA groundwater resources (MPCA, 2013), although water softening salt could also be a source, since sodium chloride salt is commonly used for both applications. While research has shown deicing salt application to be a major chloride source, brine discharged from household water softeners is an important chloride source that has not been closely examined in research. Households commonly use ion exchange water softeners to remove ions that cause water hardness, typically calcium and magnesium. Sodium chloride is used in the ion exchange process and eventually discharged to either wastewater treatment plants (WWTPs) or septic systems. In Minnesota, residential water softener use is prevalent (MPCA, 2016) due to high hardness across groundwater resources (Briggs and Ficke, 1977).

The chloride contributions from water softener use to the environment have not been well characterized in research. Additionally, chloride discharge from water softeners is a concern of many wastewater facility operators; in Minnesota, approximately 100 communities have high chloride levels in their wastewater facility effluent and reasonable potential to exceed their chloride water quality standards (MPCA, 2017a). A WWTP chloride mass budget was conducted for wastewater facilities with chloride monitoring data to estimate chloride discharge from water softening and other household, commercial, and industrial sources. Using the results of the WWTP chloride budget, a chloride budget for the State of Minnesota was undertaken to estimate the annual chloride contributions from water softening and other major point and nonpoint sources for comparison.

2 Wastewater treatment plant chloride budget

2.1 Methods

Monitoring data were available for 135 wastewater facilities in 2016 through the Minnesota Pollution Control Agency (MPCA) Wastewater Browser (MPCA, 2017b). Data were available for wastewater facilities' total monthly discharge and 24-hour composite chloride concentrations. Municipal facilities with continuous discharge and at least 11 months of chloride and discharge monitoring data were included in the WWTP chloride budget. Monitoring data were downloaded from the MPCA Wastewater Browser. For wastewater facilities with continuous discharge that lacked data for certain months, monthly chloride and flow values were estimated using their annual average values for 2016.

Multiple data sources were used to estimate contributions of domestic, commercial, and industrial chloride sources in cities discharging to WWTPs. Chloride contributions were estimated for the following sources discharging to wastewater treatment facilities: household water softeners, household cleaning agents, human excreta, background drinking water chloride concentration, commercial water softening, other commercial discharge, and industrial discharge.

2.1.1 Household water softening

Chloride from water softening was calculated with a conversion equation adapted from Thompson et al. to estimate softening based on per capita water use, rather than household water use (Thompson et al., 2006):

$$\left(\frac{Hardness\ removed\ (gpg) * Per\ capita\ water\ use\ (gal) * Population\ using\ softeners}{Average\ softener\ efficiency\ \left(\frac{grains}{lb\ NaCl}\right)}\right) = lb\ NaCl \quad \textbf{(EQ. 1)}$$

For all cities discharging to WWTPs included in the chloride budget, cities' annual residential water use was estimated using 2016 Census populations (U. S. Census Bureau, 2017) and a rate of 58.6 gallons per capita per day (gcpd), estimated in a national study of residential water use (WRF, 2016). Efficiencies of 2000 and 4000 grains/lb salt were used for timer and demand-based softeners, respectively (Baker, N.D.). It was assumed that all indoor residential water was softened and that water was softened to zero grains per gallon (gpg), based on softener factory settings to remove all hardness and industry definitions of soft water as less than 1 gpg (ASABE, 1999).

2.1.1.1 Water hardness

Groundwater provinces developed by the Minnesota Department of Natural Resources (MDNR) were used to characterize regions across the state with similar groundwater sources for drinking water (MDNR, 2018). Groundwater monitoring data from wells serving as drinking water sources were obtained from the MPCA, which included well location and calcium carbonate concentrations, used to calculate water hardness. Monitoring data were imported into ArcGIS (Esri, 2015) and groundwater hardness was interpolated across the state using kriging methods; a Gaussian model was used in kriging to fit the hardness data (Nas and Berktay, 2009). Kriging was conducted within groundwater provinces to interpolate hardness values in areas with similar groundwater characteristics and drinking water sources (Figure 2). Kriging results from all groundwater provinces were compiled for a statewide map of drinking water hardness.

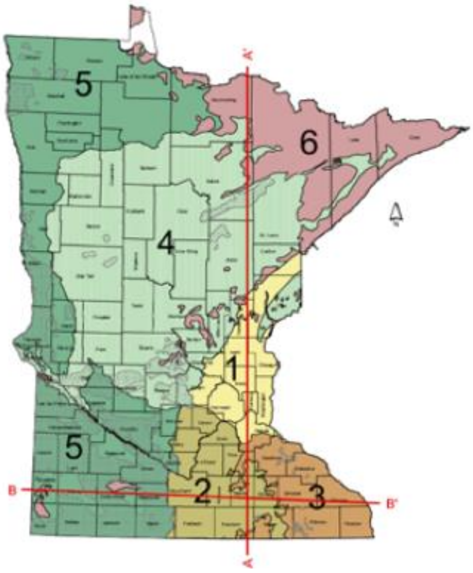


Figure 2. Groundwater provinces in Minnesota. Adapted from (MDNR, 2018).

Average hardness values were calculated for cities and townships. For cities that had well monitoring data within their city boundaries, monitoring data were used instead of interpolated data. For cities with surface water sources for drinking water, hardness data was collected from water utility reports or by contacting drinking water utilities. Average surface water hardness values were calculated and used for surface-water sourced cities where hardness values could not be obtained.

2.1.1.2 Survey of water conditioning professionals and plumbers

A survey of water conditioning professionals and plumbers was conducted to characterize household water softening practices across the state. The electronic survey included questions on the prevalence of water softener use within their primary service area. The survey protocol and questions were reviewed by the University of Minnesota Institutional Review Board and granted exemption from full review. The survey was disseminated via Qualtrics (Qualtrics, 2017) to listservs of water conditioning professionals and plumbers from the Minnesota Water Quality Association (MWQA) and Minnesota Department of Health (MDH). The survey was sent to 1,747 individuals. All survey response data were used, including responses from incomplete surveys.

Survey results were used to estimate the following: prevalence of demand-based softener use; prevalence of timer-based softener use; and prevalence of water softening in communities with centrally softened water. Survey participants' responses were used for the primary cities they serviced. Prevalence of softening was expected to vary based on water hardness; therefore, for cities without survey response data, averaged survey estimates from their groundwater provinces were used. Survey results on the percentage of population using demand-based and timer softeners were used for Equation 1. For cities with centrally softened water, identified from MDH data, the percentages of the population using softeners were adjusted in Equation 1 based on survey results.

2.1.2 Drinking water chloride concentrations

Chloride concentrations in wells supplying public or private drinking water were obtained from MDH and MPCA. Kriging was used to interpolate groundwater chloride concentrations within MDNR groundwater regions (MDNR, 2018) and a Gaussian model was used in kriging. Average background chloride concentrations were calculated for cities. For cities that had well monitoring data for chloride, these data were used instead of interpolated data. Drinking water chloride concentrations for cities with surface water sources was collected from water utility reports or by contacting drinking water utilities; chloride concentrations were not estimated for 11 cities having a surface water drinking water source without available data, due to variability in chloride concentrations from anthropogenic influences. Due to low chloride concentrations in most drinking water sources, this was expected to have little impact on results. Chloride loading from background concentrations in drinking water use was estimated by multiplying the estimated background concentration with the total WWTP discharge for 2016 (WWTP inflow and discharge rates were assumed to be equivalent).

2.1.3 Other household sources

Humans ingest and excrete salt from their diets. Chloride from excretion of feces and urine was estimated using the value of 4,818 mg per capita per day (Thompson et al., 2006). Additionally, household cleaning agents such as soaps, detergents and toilet cleansers frequently contain chloride. A value of 10.8 g chloride per capita per week was used to estimate chloride from use of common households and personal care products, which was estimated in a study of household contributions of inorganic elements and heavy metals to wastewater (Tjandraatmadja et al., 2010).

2.1.4 Commercial sources

Commercial organizations such as laundromats, hotels, and restaurants may soften water for aesthetic benefits, to reduce detergent use, and to reduce build-up of mineral scaling in pipes, fixtures, and appliances. Commercial water softening within each city was estimated using Equation 1 and the city's hardness and estimated softening prevalence. Commercial water use was estimated using city groundwater pumping data from the MDNR, which include estimated commercial water use for each city. A linear regression was created between city population and commercial water use, and the equation of the fitted line was used to estimate commercial water use in cities without pumping data.

Commercial organizations may discharge chloride from use of cleaning agents and other products. Chloride from commercial cleaning agents and other commercial processes was estimated using a concentration of 33 mg/L, a flow-weighted concentration calculated by the city of Santa Clarita, CA from monitoring of commercial wastewater (LASCD, 2012).

2.1.5 Industrial sources

Monitoring data on industrial discharge and chloride concentrations were obtained from Metropolitan Council and from previous chloride monitoring reports conducted for Alexandria, Marshall, and Long Prairie (Bolton & Menk, 2017; Bolton & Menk, 2018; Wenck, 2014). Data were obtained for 39 industries discharging to wastewater treatment facilities. Using discharge and concentrations from all facilities, an average annual discharge rate and flow-weighted chloride concentration were calculated. The discharge rate and flow-weighted concentration were used to estimate industrial discharge to wastewater treatment plants based on their number of significant industrial user permits, retrieved from MPCA.

2.1.6 Chlorination of drinking water and wastewater

Chloride from chlorination of wastewater and surface drinking water sources was estimated using literature values. Chlorination of wastewater has been estimated to contribute 5 mg/L chloride per capita (Peters, 1981). MPCA provided a list of WWTPs that chlorinate wastewater; this chlorination rate was used with Census data to estimate chloride loading from wastewater disinfection for communities discharging to WWTPs using chlorine for disinfection from April through October. Chlorination of surface drinking water sources has been estimated to add between 8-12 mg/L chloride (Fairfax County Water Authority, 2014); a value of 10 mg/L was used with WWTP discharge rates to estimate chloride loading for chlorination in cities with surface water drinking water sources.

2.1.7 Road salt inflow and infiltration

Seasonal chloride loading from inflow and infiltration (I&I) of road salt was estimated using WWTP monitoring data. Facilities with average chloride loads from December to April exceeding average loads from May to November were assumed to have road salt influence during winter and snowmelt months; the average May-November chloride loads were subtracted from December-April loads to estimate chloride from road salt infiltration and inflow. An analysis of variance (ANOVA) test was conducted in RStudio (R. Studio Team, 2015) on log-transformed chloride data from these facilities to test for significant differences in monthly chloride loads.

2.2 Results

2.2.1 Household water softening

2.2.1.1 Drinking water hardness

From 1176 drinking water wells with groundwater monitoring data, 91% had hard or very hard water, and 84% had very hard water. The average water hardness across all wells was 350 mg/L $CaCO_3$ equivalents, (20 gpg) with a median of 289 mg/L $CaCO_3$ equivalents (17 gpg). The highest hardness values were observed in the western and southwestern areas of the state; the hardest water was observed in the Sioux Quartzite aquifer in southwestern Minnesota. Figure 3 shows hardness in groundwater drinking water sources from monitoring data and interpolation.

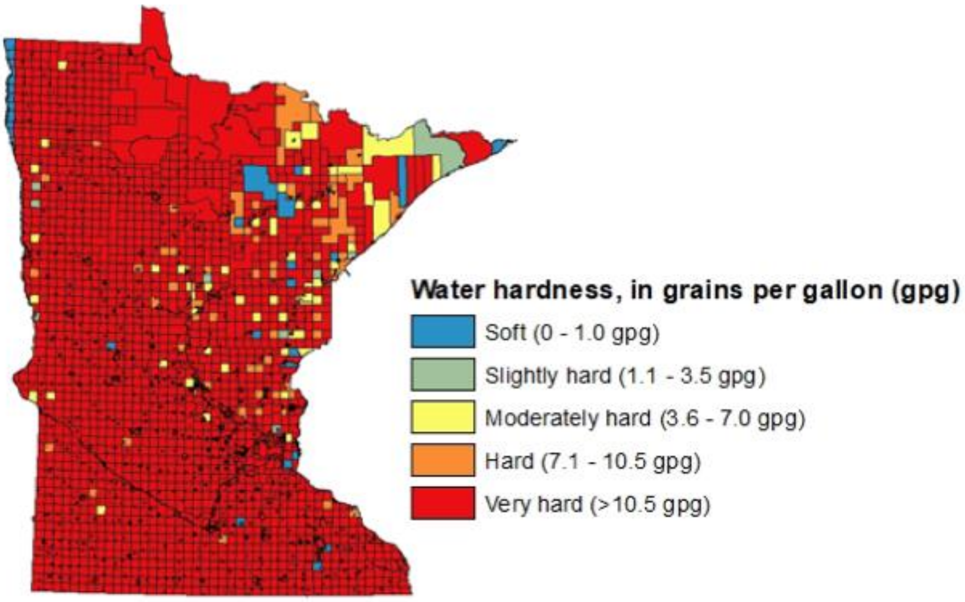


Figure 3. Level of water hardness by city in grains per gallon (gpg) from groundwater monitoring data and spatial interpolation.

2.2.1.2 Water softening prevalence

The electronic survey on water softening practices had 191 participants from the water conditioning and plumbing industries (a 11% response rate). Survey participants were distributed geographically across the state (Figure 4).

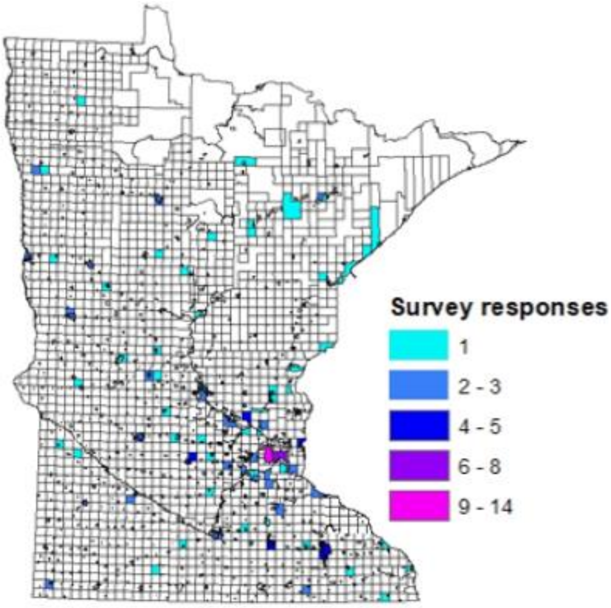


Figure 4. Geographical distribution of survey participants by city.

Among households softening water, the survey results indicated that the majority of households used demand-based softeners; the average survey response for use of demand-based softeners was 72% compared to 28% for timer-based softeners. In communities with centrally softened water, households may soften water in addition to the centralized treatment. The average survey estimate of household softening prevalence in communities with centrally softened water was 35%. Analyzing survey results by groundwater provinces showed substantially lower water softening in northeastern Minnesota, with an average of 52% of households softening (Figure 5); this area has softer water than the remainder of the state (Figure 3).

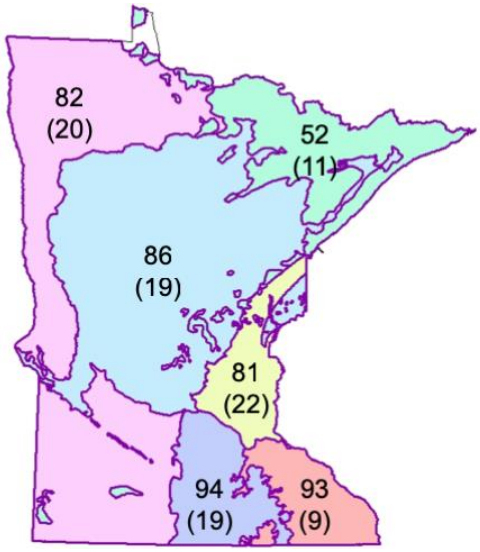


Figure 5. Estimated prevalence of household water softening by groundwater province. Number is percent of households softening, followed by number of survey responses in parentheses.

2.2.2 Drinking water chloride concentrations

From 2,326 drinking water wells with monitoring data for chloride, 14 wells had samples above the drinking water limit of 250 mg/L, four of which were supplying public drinking water. These were generally in the western part of the state, where chloride concentrations are naturally high from weathering of geological formations (MPCA, 2013). Chloride concentrations in drinking water wells from monitoring data and interpolation are shown in Figure 5. Most cities (95%) were found to have drinking water chloride concentrations under 20 mg/L (Figure 6).

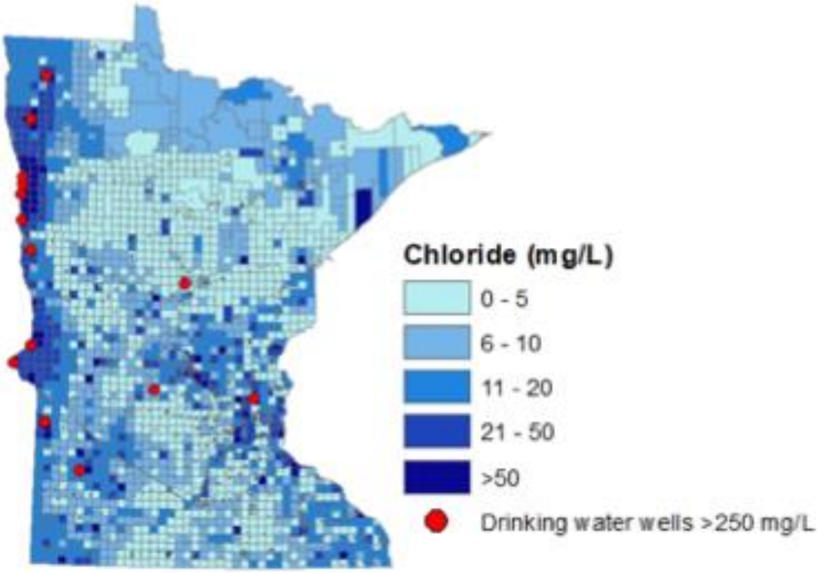


Figure 6. Drinking water background chloride concentration by city, from groundwater monitoring data and spatial interpolation.

2.2.3 Commercial water use

For cities without MDNR pumping data, commercial water use was extrapolated. A relationship was developed between community population and commercial pumping rates, which had a high R^2 value (0.73; Figure 7). Cities with high per capita commercial water use that were outliers included Grand Forks, Rochester, and Bloomington (Figure 7). The high per capita commercial water use can be explained in part by their commercial activity; Grand Forks and Rochester have large medical centers, and the Mall of America is located in Bloomington.

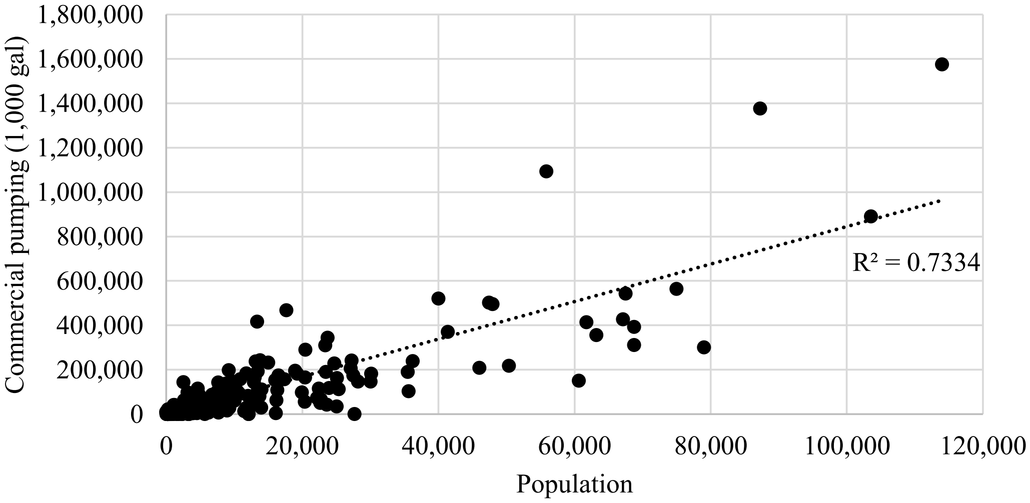


Figure 7. Correlation between commercial pumping rates and city population.

2.2.4 Industrial sources

Across 39 industrial facilities discharging to WWTPs with monitoring data for chloride concentrations and annual discharge rates, a flow-weighted average chloride concentration of 1,746 mg/L was estimated and an average discharge rate of 108 million L per year (Table 1). The average industrial annual load was estimated at approximately 189,200 kg chloride. Among industries discharging to wastewater treatment plants, industries with the highest chloride loads included waste management and food processing facilities (Table 1).

Table 1. Discharge characteristics of industries with available monitoring data.

ID	Industry type	Flow-weighted chloride concentration (mg/L)	Average discharge (million gal/yr)	Average chloride load (kg/yr)
I-1	Manufacturing facility	1,202	8.40	38,211
I-2	Metals manufacturing facility	277	4.60	4,828
I-3	Metals manufacturing facility	926	4.70	16,483
I-4	Dairy food manufacturing facility	77	1.30	378
I-5	Grain product manufacturing facility	781	60.60	179,082
I-6	Food and beverage manufacturing facility	1,038	54.40	213,723
I-7	Metal finishing industry facility	774	4.20	12,309
I-8	Dairy food manufacturing facility	23,251	0.94	82,913
I-9	Dairy food manufacturing facility	50,112	0.70	132,271
I-10	Food manufacturing facility	97,724	0.52	191,338
I-11	Food manufacturing facility	571	0.08	182
I-12	Waste disposal facility	1,004	0.34	1,293
I-13	Commodity processing and transport facility	5,580	0.24	4,967
I-14	Recycling center	664	0.04	109
I-15	Food and beverage manufacturing facility	11,373	0.02	757
I-16	Waste disposal facility	1,703	2.66	17,145
I-17	Waste disposal facility	1,583	8.73	52,313
I-18	Membrane manufacturing facility	1,707	52.58	339,740
I-19	Chemical manufacturing facility	66	1.69	422
I-20	Waste to energy plant	34,611	0.11	14,008
I-21	Waste to energy plant	34,300	0.03	3,895
I-22	Dairy food manufacturing facility	398	54.32	81,907
I-23	Dairy food manufacturing facility	52,150	0.14	26,844
I-24	Waste disposal facility	612	0.58	1,335
I-25	Food manufacturing facility	500	12.59	23,838
I-26	Waste disposal facility	17,962	61.60	4,188,490
I-27	Metal manufacturing facility	67	11.96	3,037
I-28	Food manufacturing facility	242	129.63	118,830
I-29	Medical center	128	10.73	5,199

ID	Industry type	Flow-weighted chloride concentration (mg/L)	Average discharge (million gal/yr)	Average chloride load (kg/yr)
I-30	Brewery	548	1.28	2,645
I-31	Food manufacturing facility	185	39.32	27,560
I-32	Educational institution	791	12.44	37,251
I-33	Food and beverage manufacturing facility	5,679	4.46	95,899
I-34	Food manufacturing facility	727	130.94	360,108
I-35	Cargo and freight facility	921	5.98	20,852
I-36	Food manufacturing facility	1,100	68.26	284,210
I-37	Food manufacturing facility	1,600	54.75	331,602
I-38	Food manufacturing facility	200	10.95	8,290
I-39	Food manufacturing facility	400	299.30	453,189

Industrial chloride loading estimates for Alexandria, Long Prairie, and Marshall WWTPs based on the number of significant industrial user (SIU) permits were comparable to industrial chloride loading estimates from previous studies (Table 2).

Table 2. Comparison of WWTP industrial chloride loads based on number of significant industrial user permits (SIUs) and from previous studies.

WWTP	Number of SIUs	Estimated chloride load per SIU (kg/yr)	Estimated WWTP industrial chloride load by SIUs (kg/yr)	Estimated WWTP industrial chloride load from previous studies (kg/yr)
Alexandria Lakes Area Sanitary District WWTP	2	189,165	378,330	464,638[a]
Long Prairie WWTP and Pretreatment Facility	6	189,165	1,134,989	1,076,811[b]
Marshall WWTP	3	189,165	567,495	453,257[c]

[a](Wenck, 2014) [b](Bolton & Menk, 2018) [c](Bolton & Menk, 2017)

2.2.5 Road salt infiltration and inflow

Twenty-six of 98 wastewater facilities had above-average chloride loads in winter and snowmelt months. Across these facilities, chloride loads in winter and snowmelt months were 20% higher than the remainder of the year, on average (Table 3). Results from the ANOVA on monthly chloride loading from the 26 WWTPs showed that the differences in chloride loads across months were statistically significant ($p<0.05$).

Table 3. Average monthly chloride loads by season for WWTP facilities exhibiting road salt infiltration and inflow.

WWTP	Average monthly chloride load (kg) Dec-Apr	Average monthly chloride load (kg) May-Nov	Per cent difference
Hector WWTP	16,878	5,013	337%
New Richland WWTP	4,810	3,585	134%
Trimont WWTP	6,538	4,968	132%
Eveleth WWTP	5,224	3,980	131%
Aurora WWTP	3,161	2,574	123%
Dawson WWTP	9,154	7,603	120%
Preston WWTP	6,232	5,203	120%
Rogers WWTP	59,963	50,490	119%
Hoyt Lakes WWTP	2,094	1,791	117%
Hibbing WWTP South Plant	40,153	34,848	115%
Worthington WWTP	78,888	71,352	111%
Wykoff WWTP	478	437	109%
Braham WWTP	3,447	3,177	109%
Central Iron Range Sanitary Sewer District WWTP	7,570	6,983	108%
Windom WWTP	27,434	25,354	108%
Rochester WWTP/Water Reclamation Plant	443,998	414,334	107%
Fairmont WWTP	42,110	39,715	106%
Zimmerman WWTP	5,081	4,831	105%
Virginia WWTP	29,141	27,762	105%
Holdingford WWTP	3,213	3,138	102%
Jordan WWTP	22,958	22,422	102%
Lonsdale WWTP	14,205	13,881	102%
Starbuck WWTP	6,657	6,517	102%
Adams WWTP	2,284	2,242	102%
Watertown WWTP	19,712	19,526	101%
Austin WWTP	297,561	294,810	101%

2.2.6 WWTP chloride budget

Ninety-six municipal WWTPs had at least 11 months of chloride and discharge monitoring and were included in the WWTP chloride budget and all facilities discharged to surface waters. In 2016, the 96 WWTPs discharged 114,600 metric tons (t) of chloride. The fraction of chloride from sources discharging to wastewater facilities are shown in Figure 8. These point sources are estimated to be 98% of the total chloride mass discharged from the WWTPs; the remaining chloride mass could be from hauled septage, as 24 facilities accepted hauled waste.

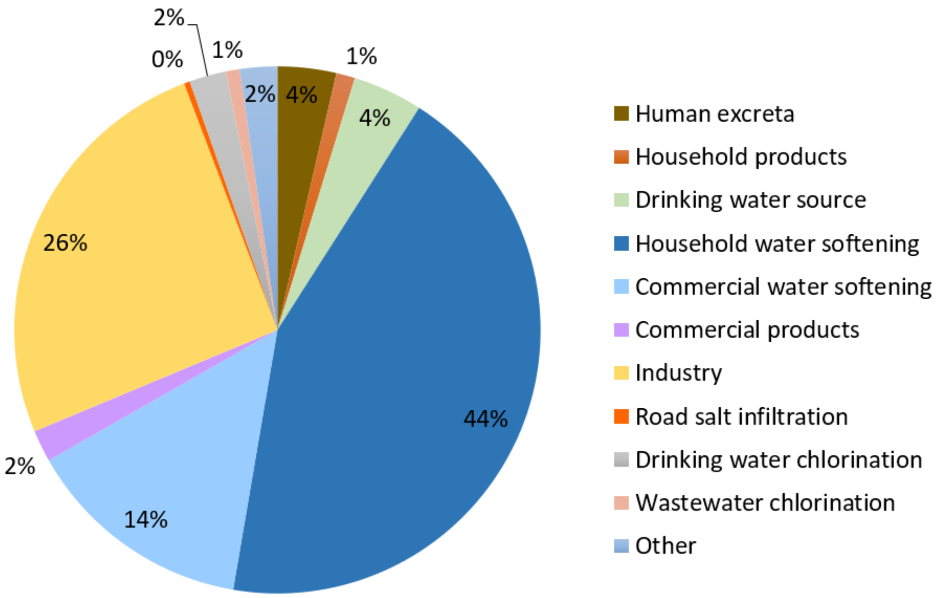


Figure 8. Fraction of wastewater treatment plant chloride contributed from domestic, commercial, and industrial sources among 96 wastewater facilities with monitoring data.

Water softeners were the largest source of chloride discharging to WWTPs, with household and commercial water softening comprising 58% of the chloride load. High chloride loads were estimated for industry, contributing 26% of the chloride mass discharged. Human excreta, background chloride concentrations in drinking water, commercial non-softening processes, household product use, chlorination of drinking water, chlorination of wastewater, and deicing salt infiltration each contributed less than 5% of the total chloride mass discharged from these facilities.

3 Statewide chloride budget

3.1 Methods

Multiple data sources were used to estimate annual chloride contributions of point and nonpoint sources in order to compare WWTP discharges to those in the State of Minnesota. Point sources included in the analysis were WWTPs and industries permitted to discharge to the environment. Chloride contributions were estimated from the following nonpoint sources: de-icing activities (road salt), livestock waste, fertilizer application, atmospheric deposition, and dust suppressant.

3.1.1 WWTPs

Chloride loads from domestic, commercial, and industrial sources discharging to wastewater treatment plants were estimated as described in sections 2.1.1-2.1.6 for all WWTPs in the state. For cities without wastewater treatment plant discharge data, their water use was estimated using residential, commercial, and industrial water use rates. Road salt infiltration into sewage pipes was not estimated for WWTPs without monitoring data due to unknown seasonal variation in chloride loading and unknown vulnerability to chloride infiltration. Road salt infiltration can be an important chloride source for individual WWTPs (Table 3), but Figure 8 indicates that road salt infiltration is not a substantial source of chloride to WWTPs in aggregate.

3.1.2 Permitted industries

Chloride from industries with National Pollutant Discharge Elimination System (NPDES) permits to discharge to the environment was estimated using monthly monitoring data from the MPCA Wastewater Browser (MPCA, 2017b). Discharge and chloride concentration data from the same discharge stations were multiplied to calculate monthly chloride loading and then summed to calculate annual chloride loading. For facilities with continuous discharge that lacked chloride concentrations for certain months, monthly chloride concentrations were estimated using their average chloride concentrations for 2016.

3.1.3 Septic systems

MPCA provided data on the number of residential and non-residential septic treatment systems in each county in 2016. The number of residential septic systems in each county was multiplied by the county average number of household members (SDC, 2017) to estimate the statewide population using septic systems.

The percent of households using demand-based and timer softeners was estimated using averaged survey results for the groundwater region where the county was located, and mean values for hardness and drinking water chloride concentrations were calculated for each county. Domestic water use and chloride loading was estimated as described in Sections 2.1.1-2.1.3.

3.1.4 Road salt

Salt contract records were obtained from the Minnesota Department of Administration. Government agencies in Minnesota can contract road salt purchases through the Department of Administration to obtain road salt at a rate set through the state's bidding process. The contract records include all salt purchases by MnDOT and for cities, counties, and organizations that participate through the Cooperative Purchasing Venture (CPV). Agencies are required to purchase at least 80% of the contracted amounts, and suppliers must be able to deliver up to 120% of the contracted amounts. Contract amounts are set at the beginning of each winter and can vary across years.

For MnDOT, contract amounts were obtained from 2011 to 2018. For cities, counties, and other organizations participating in the CPV, data were available for 2014 to 2018. To account for use of surplus road salt across different years, the contract amounts of road salt were averaged across years with available data. The sodium chloride content of regular and treated road salt was established from purchasing contract specifications (95% and 91.2%, respectively). It was estimated that 93% of contract amounts were used, based on previous research (Sander et al., 2007).

For cities that did not participate in the CPV, chloride from road salt use was estimated using results from linear regressions between population and estimated road salt use for cities in the CPV, following the methods of Sander et al. (2007). Separate linear regressions were conducted for cities located within and outside the Twin Cities Metropolitan Area (TCMA). For counties that did not participate in the CPV, chloride from road salt use was estimated using results from linear regressions between county statistics on average vehicle miles traveled and road salt use for counties participating in the CPV.

The chloride from road salt application from public agencies was estimated by summing estimated chloride use from MnDOT, counties, cities, and other participating agencies. Statistics on salt sales from the Salt Institute were used to estimate sales of bulk road salt by nongovernment entities as well as packaged road salt for private application on parking lots and sidewalks. The most recent available data is from 2006, when 80% of bulk road salt was used by governments and 20% was used by private users ((The Salt Institute, 2006) as cited in (Sander et al., 2007)). Bulk road salt sales constituted 93-95% of road salt sales, with packaged road salt for home or commercial use constituting the remaining 5-7% of road salt sales ((The Salt Institute, 2006) as cited in (Sander et al., 2007)). Given the estimated bulk road salt use from public agencies, these statistics were used to estimate use of bulk road salt by private entities and use of packaged road salt (Sander et al., 2007).

3.1.5 Atmospheric deposition

Data on chloride deposition from precipitation was obtained from the National Atmospheric Deposition Program (NADP). Data from 2016 were not available, so data from 2013 were used, the most recent data available at the time of the analysis. Using a spatial join function in ArcGIS, overlaying the Minnesota city and township shapefile with the atmospheric deposition shapefile, average deposition values were calculated in ArcGIS for each city.

3.1.6 Fertilizer

Fertilizer sales data were retrieved from the Minnesota Department of Agriculture (MDA, 2015; MDA, 2016a; MDA, 2017) to estimate use of potash (KCl). Muriate of potash (0-0-60) sales were retrieved from available sales reports from 2014-2016 and the three-year average fertilizer sales figure was used to account for fertilizer surplus, storage, and use across growing seasons.

3.1.7 Livestock

Data on livestock inventory was taken from the Minnesota Department of Agriculture (MDA, 2016b). The report detailed inventories of the following: milk cows; hogs; beef cattle; chickens, turkeys, sheep; and milk goats. Daily chloride excretion rates from the American Society of Agricultural Engineers (ASAE) were used for milk cows, hogs, chickens, and sheep (ASAE, 2003). Chloride excretion rates from previous studies were used for turkeys (Sherwood, 1989) and a rate of 4.34 lb chloride per ton manure were used for beef (Wilson, 2018). Chloride from milk goat excretions was not estimated due to the comparatively small milk goat inventory and lack of data on chloride excretion from milk goats.

3.1.8 Dust suppressant

Chloride compounds are commonly used in dust suppressant for gravel roads. Results from a MnDOT survey on dust control practices were used to estimate dust suppressant use among counties that participated in the survey (Marti and Kuehl, 2013). Additionally, 23 counties were contacted for dust control application totals; counties were purposively selected to represent urban and rural regions across the state. Information on dust control programs was retrieved for 36 of 87 counties. Twelve counties did not have dust control programs; dust application rates from the 24 counties with dust control application rates were used to estimate dust suppressant application statewide. Most county programs reported total product applied instead of application rates, since dust suppressant is often applied to small residential areas.

MnDOT-specified concentrations were used for calcium chloride ($CaCl_2$; 38%) and magnesium chloride ($MgCl_2$; 28%) to calculate chloride mass from dust suppressant (MnDOT, 2015). For each county, dust control use was divided by the total length of gravel roads to estimate an application rate, and the average application rate across all counties was applied to length of gravel roads for counties without data.

3.2 Results

3.2.1 Road salt

To estimate chloride from road salt application, data from 88 cities participating in the CPV were used in the linear regression for cities in the TCMA, and data from 97 cities were used in the regression for non-TCMA cities; the linear regressions had high R^2 values (0.89 and 0.91, respectively; Figures 9-10). Data from 58 counties participating in the CPV were included in the linear regression against average daily vehicle miles traveled on highways and city streets. Results from the linear regression had an R^2 value of 0.70 (Figure 11). Results from the regression were used to estimate chloride from road salt use for the remaining 29 counties in Minnesota.

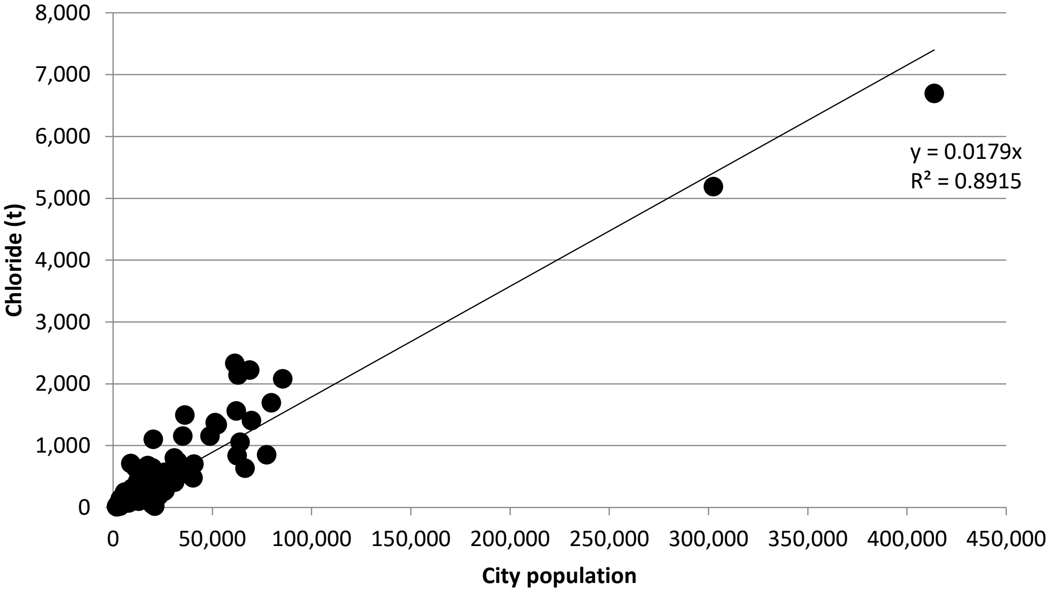


Figure 9. Per capita chloride use (metric tons) for road salt among TCMA cities participating in CPV.

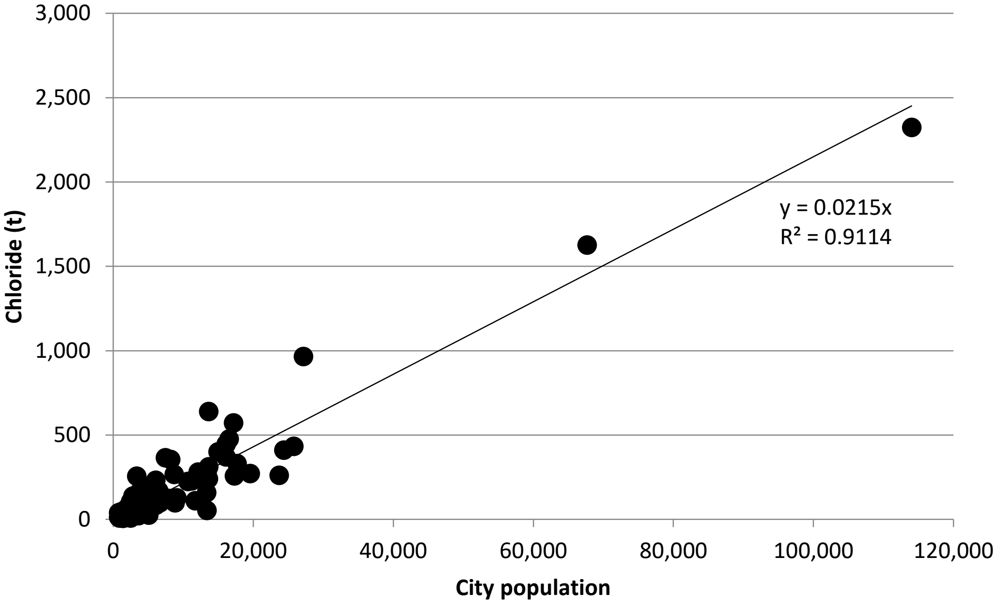


Figure 10. Per capita chloride use (metric tons) for road salt among non-TCMA cities participating in CPV.

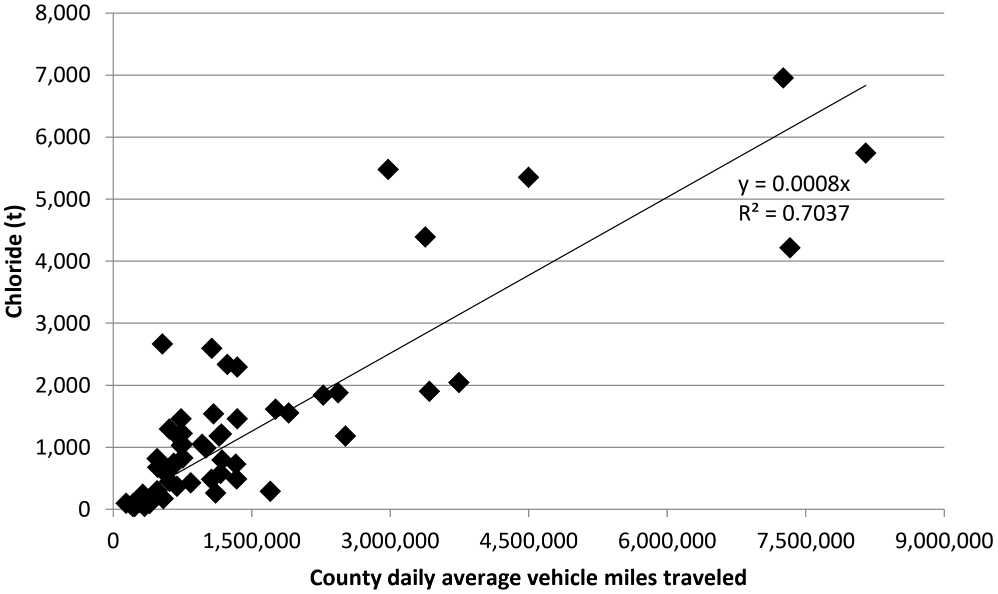


Figure 11. Chloride (metric tons) from county road salt use against average daily vehicle miles traveled.

Following estimation for cities and counties lacking road salt purchase records, the amount of chloride from private application of bulk road salt and packaged road salt were estimated using market share values reported in Sander *et al.* (2007). The estimated chloride use from bulk and packaged road salt is shown in Table 4. Of the estimated seasonal road salt use, 249,100 t of NaCl road salt are estimated to be used in the TCMA, contributing 151,200 t chloride.

Table 4. Estimated road salt use and chloride loads per season.

Bulk road salt	**NaCl salt (t)**	**Chloride (t)**
MnDOT	186,700	113,300
Counties	162,600	98,700
Cities	151,800	92,200
Other agencies in CPV	4,100	2,500
Private users	126,300	76,700
Packaged road salt	33,200	20,200
Total	664,900	403,600

3.2.2 Dust suppressant

The most frequently reported dust suppressant application rate was 0.3 gal $CaCl_2$ per square yard. Summing the reported dust suppressant use and estimated dust suppressant for counties without data or county programs, an estimated 9,400 t of chloride are applied annually statewide for dust control.

3.2.3 Permitted industries

Industries permitted to discharge to the environment were estimated to contribute 14,200 t of chloride to the environment in 2016. The industries with the highest concentrations (exceeding 230 mg/L) included corn processing, food processing, industrial manufacturing, concentrated animal feeding operations (CAFOs), ethanol manufacturing, soybean processing, rendering, waste to energy, and milk-based powder production. Three industry types contributed 72% of the chloride load discharged by industrial permit holders: iron ore mining, corn processing, and egg product food processing.

3.2.4 WWTPs

The estimated contributions of chloride from domestic, commercial, and industrial sources to all WWTPs statewide, including those without chloride monitoring data, is shown in Figure 12. Water softeners were the largest point source of chloride investigated in the analysis, exceeding chloride contributions from drinking water background concentrations and other domestic sources by over an order of magnitude. The chloride contributions are similar to the results from the WWTP chloride budget (Figure 8), although industry contributed a larger fraction in the WWTP chloride budget because included WWTPs had many industrial wastewater permits, particularly the Metropolitan Council Metro plant.

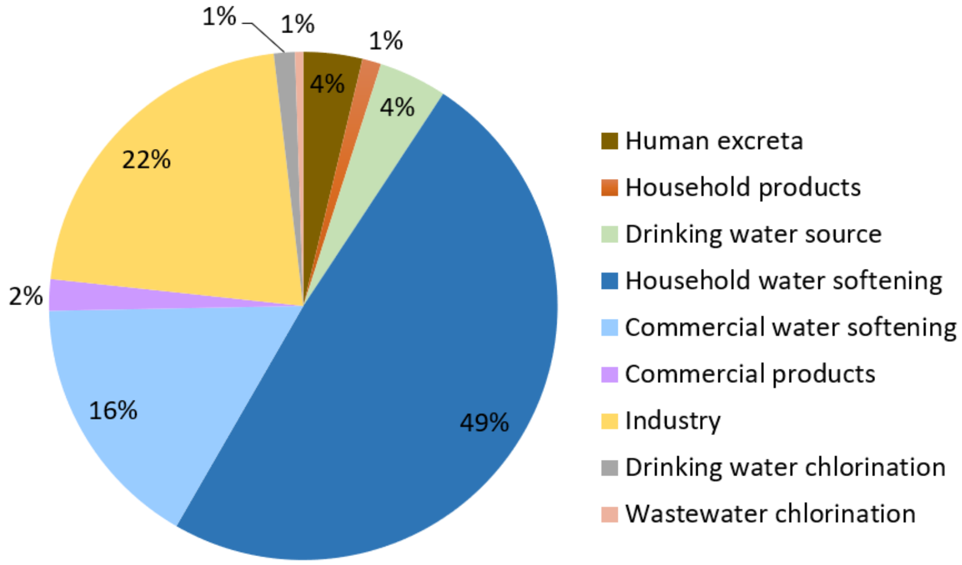


Figure 12. Fraction of chloride contributed from domestic, commercial, and industrial sources to all WWTPs in state of Minnesota.

Based on the survey results, it was estimated that 72% of the state population softens water. Household water softeners contributed 132,500 t of chloride annually, and divided by the population softening water, household salt used averaged 25 lb salt per household per month. Although only 26% of water softeners were estimated to be timer-based, timer-based water softeners contributed 42% of the chloride load from softening due to their lower efficiency. Most of the chloride loading from water softeners was from softening in areas with hard (7-10.5 gpg) to very hard (above 10.5 gpg) water; less than 5% of the chloride load was from softening in communities with slightly to moderately hard water.

3.2.5 Statewide chloride budget

The chloride contributions from non-point and point sources are shown in Table 5 and Figure 13. An estimated 403,600 t of chloride from road salt was applied in 2016, making it the largest source of chloride to the environment statewide. Approximately 221,300 t of chloride from fertilizer was found to be applied statewide and chloride from livestock waste totaled 62,600 t. WWTPs statewide contributed 209,900 t to the environment; only 2% of the chloride discharged to WWTPs was estimated to be land-applied. Of the 33,100 t of chloride discharged annually from residential septic systems, 29,600 t is estimated to be from water softening; 1,800 t were from human waste, 1,100 t were from drinking water background chloride concentrations, and the remaining 600 t were from household product use.

Table 5. Statewide annual chloride contributions from major point and nonpoint sources.

Source	Chloride mass (t)	Per cent of total
WWTPs	209,900	22%
Permitted industries	14,200	1%
Residential septic systems	33,100	3%
Fertilizer use	221,300	23%
Livestock waste	62,600	6%
Atmospheric deposition	14,200	1%
Dust suppressant use	9,400	1%
Road salt use	403,600	42%
Total	968,300	100%

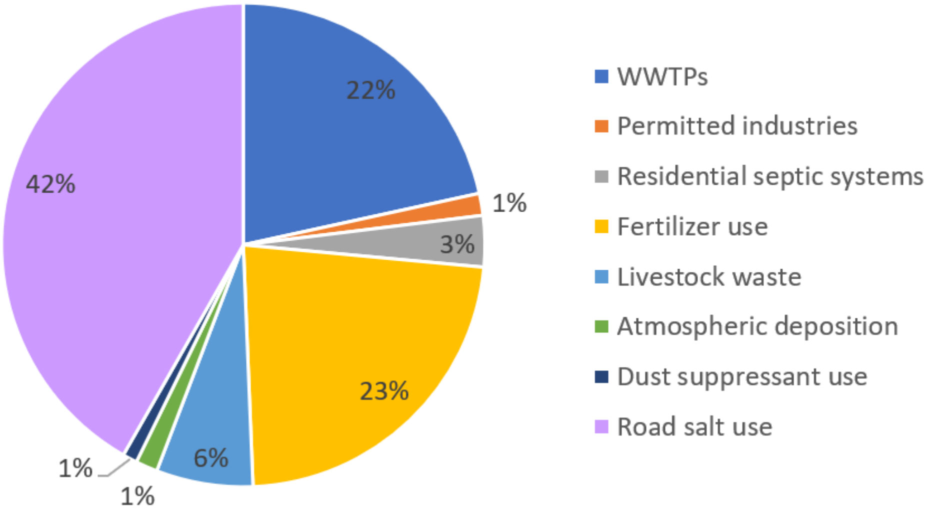


Figure 13. Fraction of annual chloride contributions from major point and nonpoint sources for State of Minnesota.

4 Discussion

WWTPs were found to the largest point sources of chloride in the analysis and water softeners were found to be the highest chloride sources discharging to WWTPs. Residential and commercial water softeners were estimated to contribute 58% of the chloride loading in the WWTP chloride budget and 65% of chloride to WWTPs statewide. Additionally, industries were found to be major chloride sources and water softening likely contributes a substantial portion of industrial chloride loads. Chloride loads from the following sources were relatively minor by comparison, comprising less than 5% of WWTP chloride loads; drinking water chloride concentrations, human excreta, household products, drinking water chlorination, and wastewater chlorination. Deicing salt I&I was a very small contributor of chloride in the WWTP chloride budget and was not estimated in the statewide chloride budget due to unknown infrastructure vulnerability to I&I in non-monitored WWTPs.

The amount of chloride discharged from water softening statewide is comparable with other literature estimates. In the statewide chloride budget, residential water softening yielded 132,500 t of chloride per year, compared to an estimated 135,000 t of chloride from softening in Illinois (Kelly et al., 2012). While the statewide chloride budget results in higher per capita softening salt use than in Illinois, the estimates for Illinois did not take into account regional variation in water softening due to differences in water hardness or treatment. Other researchers have found that commercial and industrial water softening are major chloride sources. In Phoenix, commercial organizations softening water were estimated to discharge between 1,000 to 500,000 pounds of softening salt per year, depending on the size and type of commercial organization (Daugherty et al., 2010). Additionally, a survey of Phoenix industries found that 77% of surveyed industries softened water, and softening salt contributed 17% of the total industrial chloride load (HDR Engineering, 2009).

Road salt use was found to be the dominant chloride source statewide, contributing 403,600 t per season, over 40% of the total chloride estimated in the statewide chloride budget. Although the data were based on purchasing records and market share statistics, the estimates used methods and data sources adopted in previous research (Novotny et al., 2009; Sander et al., 2007). An estimated 249,100 t of road salt are used in the TCMA compared to 349,000 t by Sander *et al.* in 2007, indicating decreases in road salt use over the last decade. This is consistent with MnDOT reports of declining salt use since 2012 due to warmer winters (MnDOT, 2017) and could also be attributed to increasing efficiency in road salt application. Additionally, the statewide estimate is comparable to chloride from deicing salt use in Illinois, estimated at 471,000 t (Kelly et al., 2012). The WWTP chloride budget indicated that infiltration of road salt into sanitary sewer pipes was a small fraction of total chloride discharged, but contributions of road salt I&I may be more important to individual WWTPs.

Agricultural sources were found to be among the highest chloride sources in the statewide chloride budget. The chloride budget estimated that 221,300 t of chloride from KCl fertilizer were used statewide. While agriculture contributes high amounts of chloride at the statewide scale, fertilizer is applied over large areas and may have a lesser impact on surface water and groundwater quality compared to road salt and WWTPs. Fertilizer application rates of 49.9 kg Cl/ha have been used in previous studies (Thunqvist, 2004) and chloride levels in drain tile from row crop fields have been reported between 5.7-36.5 mg/L (Panno et al., 2006), which is well below the established cronic toxity level of 230 mg/L. Road salt application rates can range from 23-176 kg Cl/ha (Fortin and Dindorf, 2012) and runoff from roads can be highly concentrated; Kelly et al. reported concentrations of 1570-8930 in drainage from bridges in the Chicago area (Kelly et al., 2010). Fertilizer was found to be the primary chloride source in a study of two agricultural watersheds in Illinois (David et al., 2016) and is more important in areas with less wastewater discharge and higher deicing salt use (Kelly et al., 2010).

Livestock waste was estimated to contribute 62,600 t of chloride to the environment. While livestock manure is also applied over large areas of land, livestock manure may have different implications for water quality than fertilizer due to its elevated chloride concentration; chloride levels in horse and hog waste have been reported between 440-1980 mg/L (Panno et al., 2006). Research by USGS and MPCA found elevated chloride levels in seepage from earthen-lined manure storage reached 569 mg/L and high chloride levels in plumes downgradient of manure storage (MPCA, 2001), but there is little research investigating effects of livestock feedlots or manure application practices on chloride levels in groundwater.

While the amount of chloride estimated from statewide dust suppressant use was relatively small at the statewide level compared to sources such as road salt and agriculture, dust suppressant application rates are relatively high. With a typical dust suppressant application rate of 0.3 gal/yd^2 (3,300 kg Cl/ha), the chloride application is nearly 20 times higher than chloride from road salt application rates cited by Fortin and Dindorf (Fortin and Dindorf, 2012). Additionally, higher dust suppressant application rates have been cited in literature, ranging from 2.9 to 19 tons $CaCl_2$ per lane mile (2,860-18,710 kg Cl/ha) (Gesford and Anderson, 2007; Kestler, 2009; Piechota et al., 2004). A study in Colorado found that stream chloride concentrations were significantly higher downstream of dust suppressant application areas (Goodrich et al., 2009), indicating that dust supressant use can have local impacts.

By estimating annual chloride loads from multiple chloride sources, the results provide scale for chloride loads and enable comparisons of different sources; however, the magnitude of chloride loads from nonpoint sources is not necessarily indicative of their impacts on groundwater or surface water quality, or their local importance. Chloride loads from septic systems and livestock waste were also relatively low but monitoring by Panno et al. found concentrations of 21-5620 mg/L in septic system effluent and between 440-1980 mg/L in hog and horse waste (Panno et al., 2006). Additionally, the relative importance of chloride sources can be related to the timing of their use or discharge throughout the year, such as fertilizer application in spring and road salt in snowmelt months.

The methods used in the WWTP chloride budget accounted for 98% of the chloride discharged by monitored wastewater treatment plants. However, the analysis examined chloride sources at an aggregate scale, and not for individual WWTPs. The chloride contributions of various sources and their relative importance may be different at the level of individual WWTPs, limiting the generalizability of the WWTP chloride budget results to the local level. Factors that could affect importance of chloride sources at the local level include: drinking water characteristics; water hardness; prevalence of water softening; wastewater treatment plant chlorination methods; intensity and types of commercial and industrial activities; and infrastructure vulnerability to inflow and infiltration. Additionally, elevated chloride concentrations in WWTP effluent are a greater environmental concern for facilities that discharge to receiving waters that are impaired, at risk of impairment, or have low dilution.

Estimating chloride contributions from multiple sources at a statewide scale has necessary limitations. Reliance on fertilizer sales data and road salt contracts may over-estimate annual chloride contributions from these sources; fertilizer and road salt may be stored across seasons, and road salt through the CPV may purchase between 80-120% of the contracted amount. Fertilizer sales and road salt contracts were averaged over several years of available data to account for annual discrepancies in sales and use. Limited industrial data were available for the analysis, and chloride discharge rates vary by industry, however the predicted industrial chloride discharge using average annual discharge rates and number of wastewater permits was an acceptable approximation to the load estimated from monitoring data for Long Prairie, Alexandria, and Marshall WWTPs.

Estimates of dust suppressant use may also be underestimated; estimates were based on application rates from county programs, whereas dust suppressant may also be applied through private contractors and by townships. However, dust suppressant use was estimated for all counties, including urban counties in the TCMA and counties without dust control programs that may apply less dust suppressant. Without readily available data on dust suppressant application by townships and private companies, it was considered outside the scope of this analysis.

Estimates of water hardness, water softener use, and water softener efficiency are potential sources of error in the calculations. While statistical kriging techniques were used to interpolate hardness for cities without monitoring data, inaccurate water hardness levels could result in errors in water softening salt loading estimates. Two values were chosen to represent efficiencies of demand-based and timer-based softeners, whereas a wide range of efficiency can be expected for residential water softeners. It was also assumed that all water used for indoor residences was softened, whereas some households may soften water for certain appliances. However, the average salt use among households softening water was approximately 25 lb per household per month, which is conservative compared to other estimates from research in Minnesota. Research in Rochester used vendor estimates of 54 lb softening salt per household per month (Wilson, 2007) and a survey in Alexandria estimated that timer-based softeners require 100 lb salt per household per month (Wenck, 2014).

The estimated salt use for water softening statewide is also dependent on the prevalence of water softening, and results from the survey of water softening professionals indicate a high prevalence of softening (72%). While this is a high rate, Minnesota is characterized by very hard water (Briggs and Ficke, 1977; DeSimone and Hamilton, 2009), as evidenced by the groundwater monitoring data and kriging results, and 75% of Minnesotans rely on groundwater for their drinking water supply (MDH, 2017). Additionally, although recent records are not available for national water softening salt statistics, previous research has cited statistics from the Salt Institute of over three million t of water softening salt used annually (Kelly et al., 2010).

Both chloride budgets revealed opportunities for further research. Much research has investigated chloride pollution in urban areas, particularly from road salt, but less evidence is available on loading and impacts of chloride sources in rural and agricultural areas. Limited research has shown that dust suppressant application can significantly impact local surface water quality, and fertilizer has been found to be a major chloride source in agricultural watersheds. Future research on chloride from fertilizer use and septic system effluent, storage and land application of livestock manure, and dust suppressant use could characterize its fate and transport, identify its impacts on local surface water and groundwater quality, and be used to develop best management practices for areas with chloride impairments or with groundwater vulnerability to chloride intrusion. Although a statewide survey was conducted to estimate prevalence of water softening, conducting household surveys on water softening practices would provide valuable data that could be used to identify opportunities for improving softening efficiency, estimate loading reductions, and evaluate strategies that could bring WWTPs into compliance with chloride standards. Additional chloride monitoring data from industries and WWTPs could be used to better quantify chloride loads from sources at a local scale and evaluate potential solutions for chloride pollution across communities in Minnesota.

Water softeners were found to be the largest chloride sources to WWTPs in both chloride budgets and present opportunities for chloride reduction. Chloride reductions can be achieved through ensuring that water softeners are set correctly for water use and water hardness levels. Research in Madison, WI found that optimization of water softeners reduced chloride loading to the WWTP by 27% and replacement of existing water softeners decreased chloride by 48% (Lake et al., 2015). Cities such as Madison and Waukesha have enacted programs offering grants or rebates to optimize softeners and reduce their chloride discharge; others have set ordinances mandating that water softeners installed have demand-based regeneration with minimum softening efficiency levels. Centralized water softening is also an option for some communities but is very capital-intensive and may be less feasible for small, rural communities (MPCA, 2017a). Commercial and industrial softening also presents opportunities for chloride reduction due to the volume of water softened, and efficient softening or treatment alternatives, like brine reuse or reverse osmosis, can also reduce chloride. For cities with elevated chloride levels in WWTP discharge, the feasibility and load reductions of different solutions will depend on many factors, such as: drinking water quality; prevalence of softening; the age and state of drinking water and wastewater infrastructure; community size and economics; and commercial or industrial activity.

5 Conclusions

Findings from the WWTP chloride budget indicate that household and commercial water softening discharge large amounts of chloride to wastewater facilities and septic systems. Industries were also major chloride sources; chloride contributions from other household and commercial sources were relatively small in comparison at the aggregate scale. Road salt use was the largest source in the statewide chloride budget and while fertilizer was also a major source, its impacts on surface water and groundwater quality are not well characterized in research. Chloride from water softeners was the largest chloride source discharging to WWTPs at the statewide scale, indicating that increasing efficiency of water softening practices is a viable solution for chloride management.

Chloride is an environmental concern that affects many communities with chloride impairments or elevated chloride levels in their WWTP discharge. The findings of this research highlight important point and nonpoint sources of chloride in urban and rural areas alike. The results of the analysis show that water softeners are major sources of chloride at the statewide level and suggest that strategies to increase softening efficiency may lower chloride levels in domestic and municipal wastewater. Since chloride is conservative in the environment, strategies to reduce use of salts and other chloride-containing products will have meaningful impacts on surface water quality, groundwater quality, and the environment.

Acknowledgements

We acknowledge and are grateful for funding for this project, provided by the Minnesota Environment and Natural Resources Trust Fund as recommended by the Legislative-Citizen Commission on Minnesota Resources (LCCMR). We also thank the following for providing data and support throughout this research: Sharon Kroening and Jaramie Logelin from the MPCA; members of the MWQA; Doug Heeschen from the Minnesota Department of Administration; Chad Kolstad from MDH; Tina Nelson, Bob Nordquist, and John Clark from Metropolitan Council; Sean Hunt from MDNR; and city and county public works departments.

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