

## Salty Water Trend and Sources of Salt in the Delaware River

Hongbing Sun<sup>1\*</sup>, Taylor Grieshaber<sup>1</sup>, Fatima Sulaman<sup>1</sup>,  
Lauren Margel<sup>1</sup>, Elaine Panuccio<sup>2</sup> and Nancy Lawler<sup>3</sup>

<sup>1</sup>Dept. of Geological, Environmental and Marine Sciences, Rider University, Lawrenceville NJ;

<sup>2</sup>Delaware River Basin Commission, West Trenton, NJ;

<sup>3</sup>Musconetcong Watershed Association, Asbury, NJ; \*Corresponding Author

### Abstract

There were apparent upward trends of sodium and chloride concentrations between 1944 and 2018 in the Delaware River. The increase of chloride concentration shows more significance than that of the increase of sodium concentration and the higher adsorption affinity of sodium to soil particles caused a more sustained sodium concentration than that of chloride. There were 13 recorded periods when sodium concentrations were above the 20 mg/l in drinking water recommended by US EPA and American Heart Association between 2009 and 2018 for the Delaware River at USGS Trenton gage station. If the current trend continues, the projection here is that by approximately year 2050 (or sooner), annual average sodium concentration in the Delaware River at Trenton station will reach this benchmark of 20 mg/l level. Philadelphia Water Department Plants downstream of Trenton station might reach this level sooner than water plants above Trenton station. Among the five sources of sodium chloride (winter deicing road salt, weathering of rocks, agricultural fertilizer, sewage treatment plants and precipitation) deicing road salt contributes to about 2/3 of the total salt loading and the continuing increase in the Delaware River. Annual retention of sodium from the deicing salt is about 30 to 40% (or more depending on the annual precipitation) in the Delaware River based on past studies. Though using calcium chloride as an alternative winter deicing salt might be more beneficial ecologically, short-term impact on the accelerated release of sodium stored in the soil and long-term impact of calcium chloride still need to be studied.

### Introduction - Sodium Chloride Trend

Between 1945 and 2018, sodium concentration in the Delaware River (DR) at Trenton increased about 4 times and chloride concentration increased about 6.3 times. There were 13 recorded periods in the Delaware River at Trenton showing sodium concentrations being above the 20 mg/l limit in drinking water recommended by the US Environmental Protection Agency and America Heart Association between 2009 and 2018. Regular road salt applications are the likely underlying reason for the observed increases in sodium concentrations in the Delaware River Watershed (DRW). Both sodium and chloride

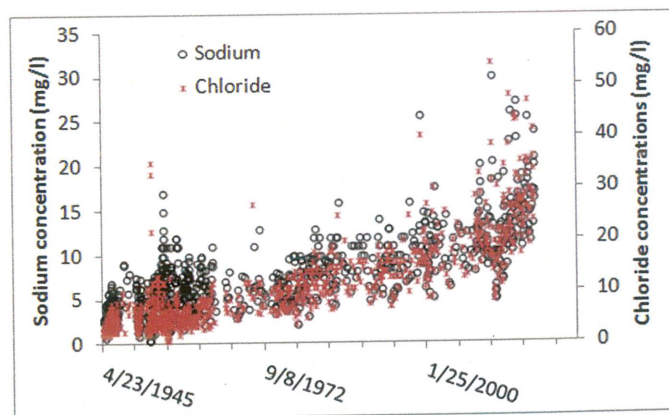
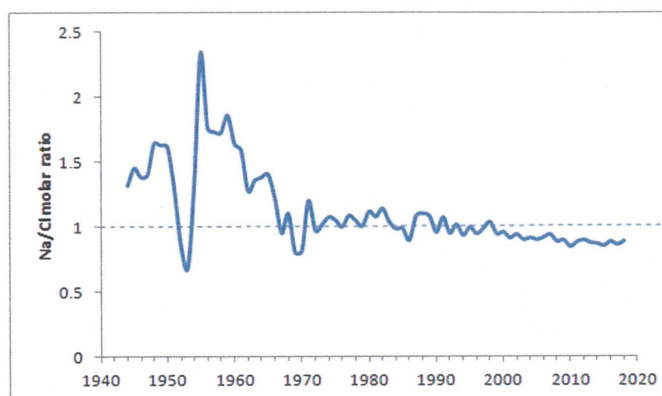


Figure 1. Sodium and chloride concentrations in the Delaware River at Trenton station between 1944 and 2018.

concentrations also increase downstream because of the accumulated salt loading from upstream. Most of the population in the DRW obtain their drinking water from the Delaware River. Sodium is not removed during the water treatment process, and it has been known that high sodium intake is an issue for people with high blood pressure (particularly seniors) and multiple sclerosis (Farez et al., 2015; Sun and Sun, 2018). Therefore, understanding the trend and sources of the sodium chloride in the Delaware River Basin can have a significant public health implication and is the first step for possible future measures in salt reduction in the DRW.

### Sources of sodium in the Delaware River Basin

Before large applications of road deicing salt (around 1960s), weathering of albite and other minerals were the source of sodium in the Delaware River. Average Na/Cl molar ratios were well above 1 (Figure 2). However, continued disturbance of the natural balance of sodium and chloride by the anthropogenic salt inputs has lowered the Na/Cl molar ratios steadily during the last 50 years. A lower Na/Cl molar ratio in the DR water is a reflection of the increasing sodium retention in the basin because of the higher adsorption affinity of sodium to soil particles than that of chloride (Weil and Brady, 2017; Drever, 1997). Retention of sodium will likely contribute to the sustained increases of sodium chloride concentration in the DR water in the near future even if the anthropogenic salt input is suspended. There are five major sources of sodium chloride that contribute to the total sodium chloride concentration and related aqueous geochemical change in the DRW.



**Figure 2. Decline of Na/Cl molar ratios because of the increased application of deicing salt. (Modified from Sun et al., 2012).**

#### 1) Deicing Salt Application

The largest salt source in the DRW is the winter deicing salt. The contribution of its proportion has increased with time. Currently, deicing salt is estimated to contribute to about 2/3 of the total sodium input in the Delaware River (Sun et al., 2012, and Sun et al., 2014). The sodium and chloride retention rates are 30-40% which are comparable to retention rates reported in other previous studies (Kelly et al., 2008; Howard and Haynes, 1993). There are many similar situations in other areas of the Northern United States, Canada and Europe where road salt application is the main source of sodium chloride in their water bodies (Corsi et al., 2010; Dailey et al., 2014; Jackson and Jobbagy, 2005). Therefore, understanding of the road salt input and sodium chloride retention can have implication for studies in regions around the world (Lofgren et al., 2001).

#### 2) Weathering Supply from Nature

Our estimation is that sodium supply from natural weathering is less than 15% of the total sodium in the Delaware River (Sun et al., 2014). Because surficial geology of the DRW is mainly sedimentary rock, little natural salts remain. The main sources of natural sodium are albite, a type of feldspar, various clay particles and organic matters that have adsorbed sodium (Weil and Brady, 2017).

**Table 1. Normalized 10-year average annual concentrations and regression trends of major ions and pH in precipitation at Milford, PA and Washington Crossing, NJ and Delaware River water at Trenton, NJ USGS station\*. Units: Kg/hectare/year except for pH (Modified from Sun et al., 2014).**

	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Na <sup>+</sup>	K <sup>+</sup>	Cl <sup>-</sup>	pH
Milford, PA (precipitation)						
1981-1990	0.88	0.27	1.12	0.26	2.33	4.29
1991-2000	0.84	0.22	1.27	0.20	2.43	4.35
2001-2010	0.94	0.23	1.24	0.31	2.42	4.54
Washington Crossing, NJ (precipitation)						
1981-1990	1.25	0.47	2.09	0.29	4.12	4.33
1991-2000	0.83	0.37	2.26	0.24	4.28	4.38
2001-2010	1.01	0.34	2.13	0.24	4.12	4.50
Delaware River at Trenton (n=695, basin area, 17560 square kilometers, downstream)						
1944-1950	90.16	30.39	31.46	8.60	31.68	7.03
1951-1960	91.61	31.10	32.68	9.54	36.16	7.10
1961-1970	63.82	22.08	25.73	6.26	35.79	7.12
1971-1980	107.85	36.55	46.27	11.78	67.06	7.92
1981-1990	94.26	32.89	50.83	7.91	75.11	8.00
1991-2000	89.38	29.87	56.82	7.96	88.10	7.88
2001-2011	115.07	38.11	84.84	9.91	143.10	7.86
Regression Trends of Ion Concentrations in the Delaware River at Trenton between 1944 and 2012. For SO <sub>4</sub> <sup>2-</sup> , between 1980-2012						
Regression t-test	4.09	3.73	25.61	-2.92	34.30	18.34
Number of datum	694	694	693	466	694	693

*\*Precipitation data are from National Atmospheric Depositional Program and stream data are from the US Geological Survey (USGS). Regression t-test is for the regression slope of concentration vs. sample date. Any t value >1.97 or <-1.97 indicates a significant trend with 95% confidence. A positive t value indicates an increasing trend while a negative t value indicates a decreasing trend. The higher the t value is, the stronger the trend*

### 3) Agricultural supply

Contribution of salt from agricultural sources to the total salt in the DRW might be significant before the application of deicing road salt becomes dominant. However, between 1950 and 2004, the farmland in the DRW was reduced by about 47.6%, while the national farmland was reduced only by 22.1% during the same period (Sun et al., 2006). Reduction of the farmland in DRW was almost twice as fast as the national average. Therefore, it is unlikely that agricultural supply is a significant contributor to the sodium and chloride concentration increases in the DRW.

### 4) Precipitation

Precipitation accounts for less than 4% of the sodium and chloride concentrations in the Delaware River (see the underneath Table 1). Its contribution have not changed significantly based upon the available data between 1983 and 2013. There are no significant trends for sodium and chloride concentrations in the precipitation at the two stations (shown in Table 1) in the DRW.

### 5) Discharge from Water Treatment Plants

Sodium level in recycled water can be twice the sodium level in potable water (PWD, 2007). The salt here mainly comes from the salt in food, water softener, disinfectants (sodium hypochlorite), etc. Increased salt proportion in this category over the years is mainly because of the population increase. In 1950, the human population of the DRW was about 5.1 million. By 2010, the population increased to about 8.7 million people. This source can account for 3-4 % of the total sodium in the DRB.

### **Projection of the Salt Trend in the Delaware River**

Projection based upon the regression trend of Figure 1 is that by about year 2050 or sooner, average annual sodium concentration in the Delaware River at Trenton will reach the 20 mg/l EPA and AHA recommended limit. By the end of the century (or sooner), the average annual sodium concentration will be about 29 mg/l, well above the 20mg/l benchmark. Sodium concentration at the intake points of the Philadelphia Water Department will reach this 20 mg/l benchmark sooner than at the Trenton gauge station. Between now and 2050, there will be more periods in January and February where sodium concentrations will exceed the EPA and AHA recommended limit of 20 mg/l.

### **Impact on Water Quality by Increased Salt Application**

#### 1) Water is getting saltier and harder.

Table 1 shows the normalized 10-year average annual concentrations and regression trends of major ions, pH in precipitation at Milford station, PA and Washington Crossing station, NJ, and in the Delaware River at Trenton, NJ station (Sun et al., 2014). Statistically significant upward trends can be identified for calcium and magnesium concentrations between 1944 and 2013 in the Delaware River. Trends for concentrations of other elements can be identified as well. However, not all the trends are due to the cation exchange of sodium with other cations or the anion exchange/complexation of chloride with other ions.

#### 2) Concentrations of heavy metals in water may be affected.

Complexation of chloride with lead and mercury can lead to in-situ mobilization of these metals in soil solution. Dispersion from hydrated sodium can also lead to the increased concentration of arsenic in soil solution. There are positive concentration correlations between Na, Hg, Cl and Pb from the Centennial Lake Watershed in the DRW (Sun et al., 2015; Sun et al., 2016).

### **Alternative to Sodium Chloride for Deicing: Calcium Chloride?**

Since calcium is a macronutrient element in soil and water, it can be taken up by organisms in soil easily and moderate amount of calcium can be beneficial. Calcium salt might also help neutralize acidity in soil and water from acid rain. Therefore, calcium chloride ( $\text{CaCl}_2$ ) can be used as an alternative salt in place of sodium chloride for deicing. However, a few drawbacks of calcium chloride application need to be recognized as well. Because calcium has a higher cation exchange capacity than sodium, there will be an initial accelerated release of sodium stored in soil from previous application of sodium chloride salt in the past few decades. We will not expect a decrease in the concentration of sodium in the Delaware

River for many years to come, even if all the deicing salt were switched to calcium chloride now. Strong cation exchange capacity of the calcium might accelerate the release of other unwanted metals from soils locally. Also increased calcium and magnesium concentrations increase the hardness of water. Studies on the long-term ecological impact of calcium chloride salt are needed as well.

### References

- Corsi SR, Graczyk, DJ, Geis SW, Booth NL, Richards KD. 2010. A fresh look at road salt: aquatic toxicity and water-quality impacts on local, regional, and national scales. *Environmental Science & Technology*, 44:7376-7382.
- Dailey KR, Welch KA, Lyons WB. 2014. Evaluating the influence of road salt on water quality of Ohio rivers over time. *Applied Geochemistry*, 47:25-35
- Drever JI. 1997. *The Geochemistry of Natural Waters*, 3rd Ed., Prentice Hall, New Jersey.
- Jackson RB, Jobbagy EG. 2005. From icy roads to salty streams. *Proceedings of the National Academy of Sciences of USA*, 102, 11487-11488.
- Kelly VR, Lovett GM, Weathers KC, Findlay SEG, Strayer DL, Burns DJ, Likens GE. 2008. Long-term sodium chloride retention in a rural watershed, Legacy effects of road salt on stream water concentration. *Environmental Science & Technology*, 42:410-415.
- Lofgren S. 2001. The chemical effect of deicing salt on soil and stream water of five catchments in southeast Sweden. *Water, Air and Soil Pollution*, 130:863-868.
- Philadelphia Water Department (PWD), Baxter Water Treatment Plant Surface Water Intake, 2007. *The Delaware River Watershed Source Water Protection Plan*, Philadelphia, PA.
- Farez MF, Fiol MP, Gaitán MI et al. 2015. Sodium intake is associated with increased disease activity in multiple sclerosis. *Journal of Neurology, Neurosurgery and Psychiatry*, 86:26-31.
- Sun H, Sun M. 2018. Age and gender dependent associations of blood pressure and serum sodium and potassium-renal and extrarenal regulations. *Journal of the American Society of Hypertension*, 12: 392-401.
- Sun H, Alexander J, Gove B, Koch. 2015. Mobilization of arsenic, lead, and mercury under conditions of sea water intrusion and road deicing salt application. *Journal of Contaminant Hydrology*, 180:12-24
- Sun H, Alexander J, Gove B, Pezzi E, Chakowski N, Husch J. 2014. Mineralogical and Anthropogenic Controls of Stream Water Chemistry in Salted Watersheds. *Applied Geochemistry*, 48:141-154.
- Sun H, Hewins D, Latini D, Husch J. 2006. Changes in Impervious Surface Area, Flood Frequency, and Water Chemistry within the Delaware River Basin during the Past 50 Years: Initial Results. *Proceedings of the 7th Int. Conf. on Hydrosience and Engineering (ICHE-2006)*, Sep 10 -Sep 13, Philadelphia, USA ISBN: 0977447405.15pp.
- Sun H, Sinpatanasakul L, Husch JM, Huffine M. 2012. Na/Cl molar ratio changes during a salting cycle and its application to the estimation of sodium retention in salted watersheds. *Journal of Contaminant Hydrology*, 136-137:96-105
- Sun H, Sulaman F, Dell'oro A. 2016. Changes of mercury concentration in response to chloride complexation under deicing salt condition. *GSA Abstracts with Programs*. 48, 2.
- Weil RR, Brady NC. 2017. *The Nature and Properties of Soils*. 15<sup>th</sup> Edition. Prentice Hall, New Jersey.