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Evidence of Road Salt in New Hampshire's Snowpack Hundreds of Meters from Roadways

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Abstract: Salinization of surface and groundwater has been directly linked to the area of road surfaces in a watershed and the subsequent wintertime maintenance used to keep roads free of snow and ice. Most studies that explore road salt in snow along roadways limit the study to within 100 m from a roadway and conclude that there is negligible deposition of de-icing salt at distances greater than 100 m. In this study, we analyze the ion content of the southern New Hampshire snowpack and use Mg^{2+} as a conservative sea-salt tracer to calculate sea salt and non-sea salt fractions of Cl^-. There is a minimum of 60% non-sea salt Cl^-, which we attribute to road salt, in the snowpack at our study sites 115 to 350 m from the nearest maintained roadways. This suggests that larger areas need to be considered when investigating the negative impact of Cl^- loading due to winter-time maintenance.

Keywords: winter-time maintenance; chloride; salinization; ion pulse; sea salt aerosol; impervious surfaces

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1. Introduction

Chloride contamination of surface and groundwater has become an area of increased environmental concern and research focus in New Hampshire (NH) where deicing of roads using road salt is a regular practice during winter months [1]. One study by the NH Department of Environmental Services (DES) that focused on four watersheds in southeastern NH found that road salt was the main contributor to excess Cl^- in streams—accounting for over 90% of total chloride load in some cases [2]. This is likely part of a broader regional trend observed by Kaushal et al., 2005, who note increases in baseline Cl^- concentrations in rural and urban northeastern United States streams over the past 30 years [3]. Many studies attribute increasing Cl^- concentrations in northern freshwater systems to increasing road-surface area and the subsequent increase in winter-time maintenance [3–8]. One study suggests impervious land cover will increase in the United States by at least 19% into the year 2100 [9], which will lead to further increases in Cl^- concentrations. After the year 2100, many surface waters in the northeastern United States could have Cl^- concentrations toxic to freshwater life and unsuitable for human consumption [3].

The state of NH used about 170,000 tons of road salt between November 2014 and April 2015 [10], most of which is applied as coarse sodium chloride (halite) crystals to roads and parking lots. Once applied to such surfaces, salt crystals are either covered up by precipitation and plowed off the surface or left loose on the surface. Salt that is plowed off and mixed with snow is stored in the snowbank until snowmelt. Salt that remains on the surface can be transported through a variety of different mechanisms including airborne transport through splashing and spraying of pooled water or particulate matter, infiltration into the surface, or as runoff [11]. Airborne spreading is perhaps the most prominent mechanism to transport road salt from roads, with resuspended de-icing salt aerosols capable of being transported over 1 km from their source [12]. In any case, conductive species such as Na^+ and Cl^- that enter an adjacent environment during winter are ultimately destined to end up in streams at the onset of melt during the spring time pulse [13,14], or infiltrate through the soil into groundwater where lasting effects are more pronounced [1,15]. In fact, one study notes high road salt contamination year-round in springs up to 800 m from maintained roadways [15].

Prior investigations into the accumulation of de-icing salt in snowpacks near roads often restrict their study areas to within 30 m from the shoulder of the roadside [16,17], and attribute most road side salt accumulation dispersion from the roadside to plowing, splashing, or spraying of fine droplets [18]. Restricted study areas could lead to underestimated road salt loading in snowpacks if, as previously suggested [12], de-icing salts are transported much further than 30 m. Underestimation of total road salt contained in a snowpack, inaccurate representation of its spatial distribution, and uncertainty in transport mechanisms will lead to uncertainty when trying to quantify the impact of salting roads on nearby environments.

Previously, Lazarcik et al. observed that the snowpack near Hanover, NH had about double the Cl^- load compared to snowpacks near Durham, NH, in two consecutive seasons [13]. They note that this was unexpected, because Hanover is much further than Durham from the Atlantic coast and deposition of sea-salt aerosol (SSA) decreases rapidly with distance from the coast [19]. This indicates that there is a source of chloride near the Hanover snowpack in addition to SSA. This study explores the potential for road salt to be a substantial source of chloride in the snowpack at several NH field sites over the course of two winter field campaigns using daily or near-daily measurements of snowpack ions. We hypothesize that wind transport of fine dry road salt particles is the primary process delivering excess Cl^- to the snowpack hundreds of meters from roadways, though this can only be inferred from this study.

2. Materials and Methods

Snow samples were collected at daily or near daily intervals at several sites spanning the southern half of NH for two winter seasons in 2013–2014 to 2014–2015 [13,20], hereby denoted as Winter 2 and Winter 3, respectively (Table 1, Figure 1). Winters 2 and 3 are used to be consistent with a previous study; study sites, sampling methods, data analysis methods, and quality control procedures are identical to those previously described [13]. During the first year of the study in winter 2012–2013, sampling was conducted at six sites, creating regular gaps of 2–3 days between repeat visits of all sites which made time series difficult to interpret [13]. As an additional quality measure, five consecutive days (10–14 February) of Winter 3 data at the two sites near Durham—Thompson Farm (TFO) and Burley-Demeritt (BDO)—were rejected due to analytical errors caused by progressive failure of the pump on the autosampler making injections into the cation channel of the ion chromatograph.

Table 1. Site location overview.

Site (Abbr.)	Distance to Nearest Maintained Road (m)	Distance to Atlantic Ocean (km)	Elevation (m)	Latitude and Longitude
CRREL Yard Open (CYO) [a]	290	150	143	N 43°43′ W 72°16′
Dartmouth Farm Open (DFO) [a]	114	150	119	N 43°44′ W 72°15′
Burley-Demeritt Open (BDO)	330	23	35	N 43°05′ W 70°59′
Thompson Farm Open (TFO)	348	22	19	N 43°06′ W 70°56′

[a] Less than 1 km from I 91 in Vermont.

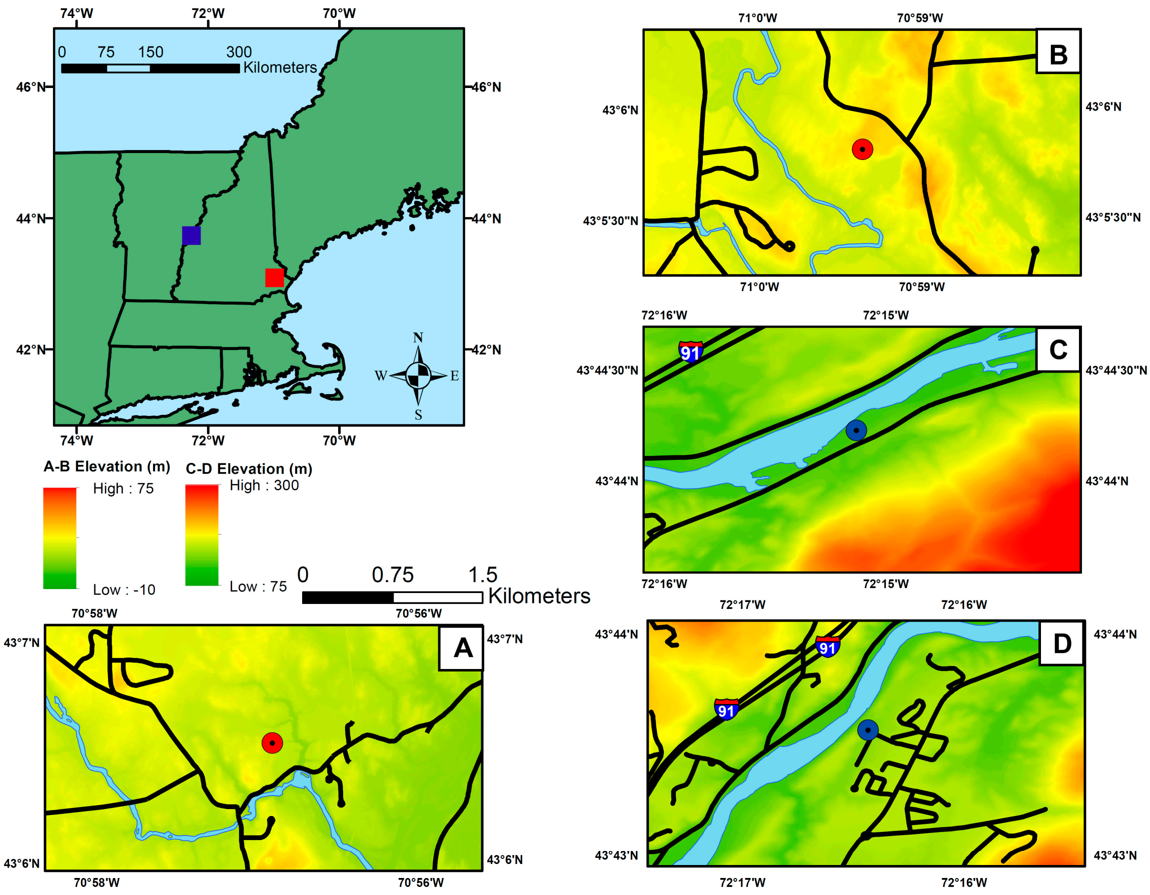

Figure 1. A map of TFO (panel **A**), BDO (panel **B**), DFO (panel **C**) and CYO (panel **D**) in relation to the nearest roads. Each site location in panels **A**–**D** is color coded to its location within New Hampshire in the broader region shown (upper left). CYO was located near the CRREL yard drive, but the nearest public road is 290 m away.

Sea salt chloride fractions in precipitation are often calculated from a measured reference species within the sample and a known bulk seawater composition [21]. Magnesium and sodium are typical reference species for samples of liquid precipitation and aerosol; however, sodium may not be suitable as a reference species due to prolific road salt application in NH. Road salt aerosol resuspended from the road and deposited on the snowpack will increase sodium load above what is expected from bulk seawater compositions.

Sea salt fractions of Cl^- and Na^+ are therefore calculated using the amount of Mg^{2+} present in each snow sample and multiplying by the expected sea salt ratio for each with respect to Mg^{2+} (the molar ratios of these ions in sea salt are $Cl^-/Mg^{2+} = 10.33$; $Na^+/Mg^{2+} = 8.88$ [22]). Non-sea salt, or excess, Cl^- (Na^+) is simply total measured Cl^- (Na^+) minus the calculated sea salt Cl^- (Na^+) fraction. Dust deposition may increase magnesium inventories above the amount due to sea salt; however, dust deposition containing Mg^{2+} is at a yearly low during the winter in the eastern United States [23,24]. It should be noted that any addition of Mg^{2+} to the snowpack not derived from sea-salt aerosol, which includes small amounts of Mg^{2+} found in road salt, will cause calculated non-sea salt Na^+ and Cl^- fractions in snow to be underestimated. Similarly, if Cl^- is lost to the gas phase due to reaction of NaCl with sulfuric or nitric acid derived from fossil fuel emissions [25], non-sea salt Cl^- in NH snow could also be underestimated.

3. Results and Discussion

On average, non-sea salt Cl^- accounts for over 60% of the measured Cl^- at all sites in both winters, with excess Cl^- averaging 81% of total Cl^- at the Dartmouth Organic Farm site (DFO) in

Winter 3 (Table 2; Figure 2). Additionally, linear regression of excess Cl^- versus excess Na^+ in all samples from the two seasons shows very strong correlation and slopes slightly below the value of 1.0 expected for halite, which is much lower than the Cl^-/Na^+ molar ratio of 1.16 in sea salt (Figure 3). At the highest concentrations, Cl^- decreases with respect to Na^+. This is likely caused by acid reactions displacing Cl^- to the gas phase. Therefore, it appears that most of the Cl^- in southern NH snowpacks is not sourced from sea salt, but is actually from road salt.

Table 2. Average sample Cl^-/Na^+ ratio, average sample % non-sea salt Cl^-, and cumulative non-sea salt Cl^-.

Site	Average Cl^-/Na^+	% Non-Sea Salt Cl^-	Cumulative Non-Sea Salt Cl^- (nmol cm^{-2})
		Winter 2 [1]	
CYO	0.94	61 ± 21	510 ± 173
BDO	1.02	63 ± 21	140 ± 48
TFO	0.99	60 ± 20	188 ± 64
		Winter 3 [1]	
DFO	1.02	81 ± 28	1780 ± 605
BDO	0.97	63 ± 21	434 ± 148
TFO	1.02	66 ± 22	667 ± 227

[1] The spatial variability of Cl^- in snow for the study region was previously assessed to be a maximum of 34% [13].

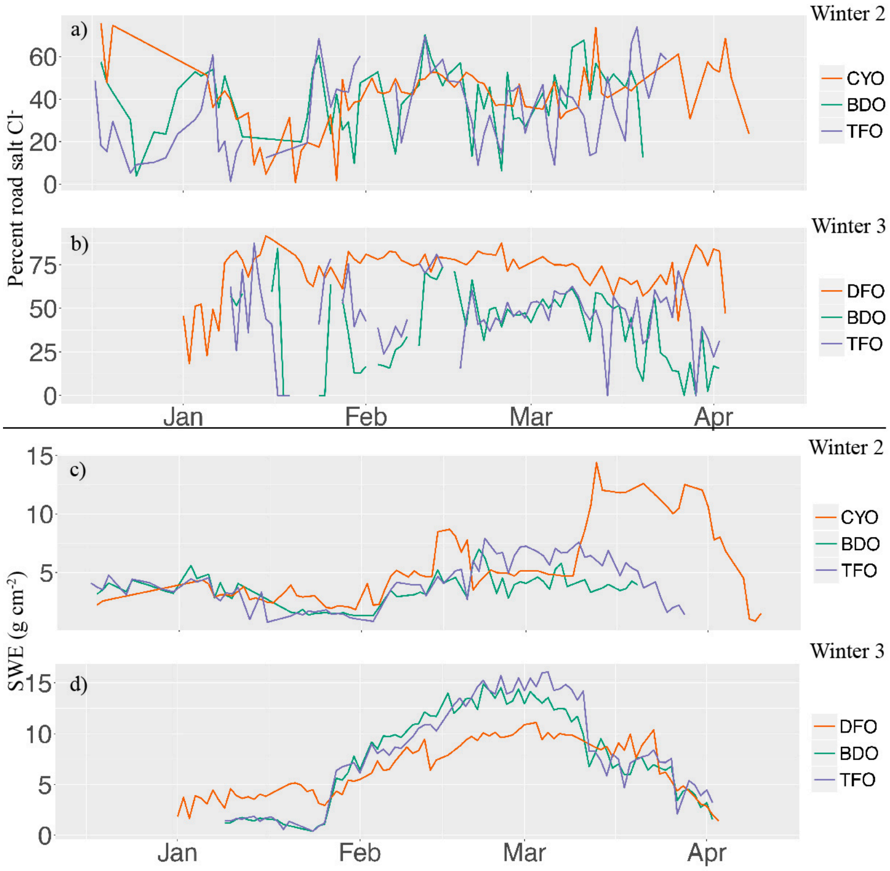

Figure 2. Percent non-sea salt Cl^- inventory time series for each site during Winters 2 (**a**) and 3 (**b**) and total SWE for Winters 2 (**c**) and 3 (**d**). Percent Cl^- are the percent of the total daily pit Cl^- inventory that is calculated to be due to road salt. Percent Cl^- data gaps are days when no snow chemistry data was gathered either due to no snow or severe weather. The SWE data has been interpolated over missing data points. The Hanover site moved to DFO from the CRREL Yard (CYO) between Winters 2 and 3; DFO is more than 150 m closer to a maintained roadway than CYO.

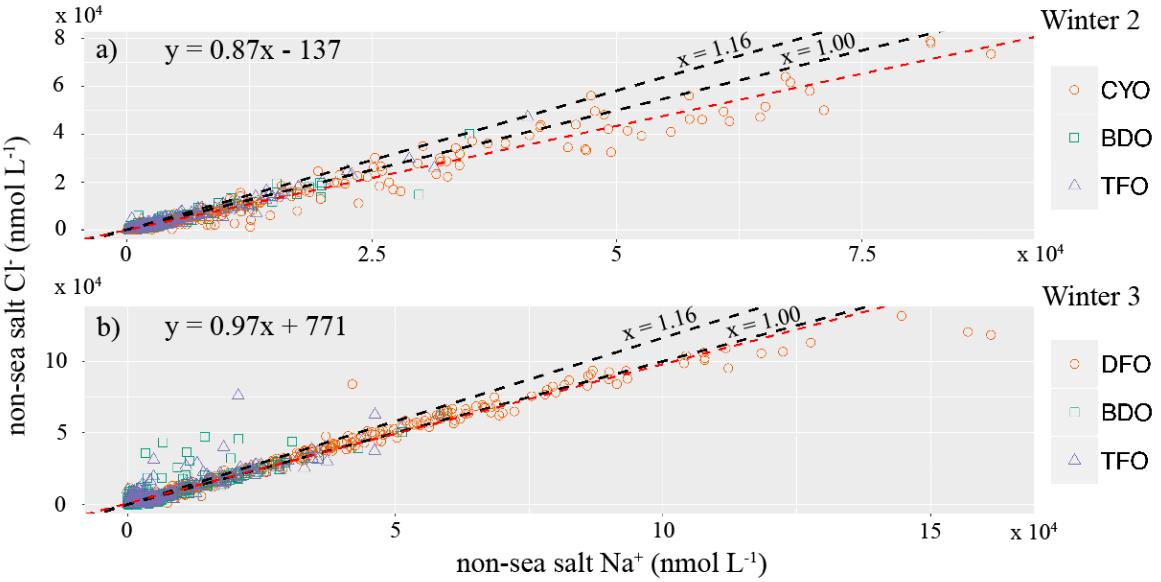

Figure 3. Scatter plot of non-sea salt Na^+ and Cl^- for Winters 2 (**a**) and 3 (**b**) calculated using Mg^{2+} as a conservative SSA tracer. The black lines demonstrate slopes of Cl^-/Na^+ of 1.16 (SSA) or 1.00 (halite). The red dotted lines, and equation in the top left of each panel, show the line of best fit for a yearly data set, and has a slope of 0.87 in Winter 2 and 0.97 in Winter 3.

While the deposition of road salt Cl^- is lower at the seacoast sites BDO and TFO, the fact that road salt is a more important source of Cl^- than sea salt for these sites greater than 100 m from maintained roads is surprising. Many studies either do not sample more than 100 m from the road, or conclude that de-icing salt deposition is negligible at distances greater than 100 m from the road [11,16–18]. Considering that both BDO and TFO are 330 and 350 m away from local secondary roads, respectively (Table 1), it is a matter of concern that more than 400 and nearly 700 nmol cm^{-2} of road salt Cl^- reached BDO and TFO, respectively, in Winter 3 (Table 2). These road salt Cl^- inventories are equivalent to accumulation of 0.25 and 0.39 grams of road salt halite per square meter of land well removed from the nearby roadway. Winter 2 had less frequent snow events compared to Winter 3 [13,20], resulting in less road salt reaching all three sites (Table 2). However, road salt was still the dominant source of Cl^- in the seacoast snowpack with about 140 and 190 nmol cm^{-2} of road salt Cl^- (0.08 and 0.11 g m^{-2} road salt halite) reaching BDO and TFO, respectively (Table 2).

If our findings near Durham are representative of dispersion of road salt to distances of approximately 350 m on both sides of all roads, a minimum of 4000–6000 metric tons of road salt are being widely spread along the nearly 2000 km of roads in New Hampshire in heavy snow years such as Winter 3. Even in a light snow year such as Winter 2 our results suggest at least 1200–1600 tons of road salt is being dispersed hundreds of meters on both sides of salted roadways. This is a small fraction (just 2–3%) of the total amount of applied road salt, all of which is released to the environment, but suggests negative impacts of increased Cl^- loading may need to be considered for a larger portion of the landscape than nearby the streams and lakes that have received close study to date [3–7].

We acknowledge that most salt applied to roads is likely to remain relatively near the road until it dissolves and infiltrates the soil and enters the groundwater, or is dispersed in surface runoff produced by major snow melt events. Coarse halite crystals may bounce to the road edge and salty slush on the road way is frequently pushed into and some short distance beyond roadside snowbanks by plows. The brine on wet salted roadways is clearly lifted by passing traffic but these droplets apparently tend to be deposited within 30–100 m of the roadway, based on the previous studies mentioned above. We propose that remobilization of fine dry road salt that covers de-iced roadways a few days after each snowfall that required de-icing may contribute to dispersion over distances of 100 s of m. A comparison of non-sea salt Cl inventory and the overall snowpack snow-water equivalence (SWE) shows one case where road salt Cl inventory increases in the snowpack days after new snow accumulates (Figure 4). This case was recorded in Winter 3, 7–9 February, where snow was deposited at a rate of 3 to 9 cm per hour for most of the period, which halted travel and required many de-icing operations. Road salt Cl^- inventory takes several days to increase in turn. This example may support the remobilization of dry salt as a more important source of distant dispersion than spray of salty droplets during and immediately after a snowfall event. In contrast, the very beginning of a spring melt is accompanied by an immediate rapid decrease in road salt Cl^- inventory as impurities are flushed from the snowpack in what is commonly called an 'ion pulse' [13]. However, the evidence presented in favor of fine dry road salt dispersion in this work is insufficient to draw firm conclusions and further work must be conducted to better understand the various mechanisms of road salt transport.

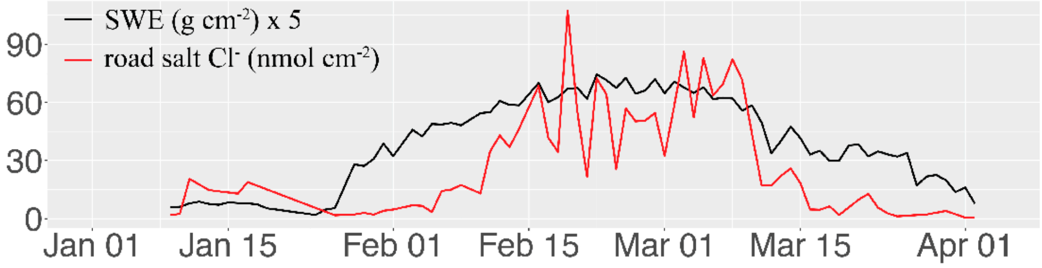

Figure 4. A comparison of total snowpack road salt Cl^- and SWE during Winter 3 at BDO. Cl^- data has been interpolated over missing data points in the time series.

4. Conclusions

Calculating road salt Cl^- using Mg^{2+} as a conservative seawater tracer likely leads to an underestimation of total road salt Cl^-, which makes the high amounts of road salt Cl^- calculated for CYO and DFO especially concerning considering nearly all snowpack Cl^- must eventually enter the nearby watershed. At BDO and TFO, a significant amount of road salt Cl^- appears to have travelled at least 300 m. This indicates that studies focusing on only the first 30 m of the snowpack nearest to roads are inadequately estimating total road salt Cl^- in snowpacks. On average, total snowpack Cl^- was calculated to have at least a 60% fraction originating from road salt at all sites in both study years, which means more than twice as much Cl^- than naturally comes from sea salt is being deposited onto the snowpack over an area far greater than the first initial 30 m alongside roadways. Road salt widely spread over the landscape as opposed to culverts and ditches, which are designed to deliver snowmelt and rain quickly to streams, may result in a larger fraction of road salt infiltrating into the groundwater system. Therefore, larger land areas and the presence of road salt in groundwater and surface water need to be considered when studying the negative effects of Cl^- loading due to wintertime maintenance.

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Author Contributions: Jack Dibb conceived and designed the experiments; James Lazarcik made the field observations and collected samples; Jack Dibb supported and oversaw the laboratory that did the ion chemistry analyses; James Lazarcik and Jack Dibb analyzed the data; James Lazarcik wrote and Jack Dibb edited the paper.

Conflicts of Interest: The authors declare no conflict of interest.

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