

# Impact of Road Salt on Drinking Water Quality and Infrastructure Corrosion in Private Wells

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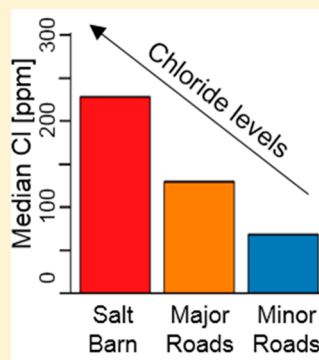
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## S Supporting Information

**ABSTRACT:** Increased road salt use and resulting source water contamination has widespread implications for corrosion of drinking water infrastructure, including chloride acceleration of galvanic corrosion and other premature plumbing failures. In this study, we utilized citizen science sampling, bench-scale corrosion studies, and state-level spatial modeling to examine the potential extent of chloride concentrations in groundwater and the resulting impact on private wells in New York. Across the sampled community, chloride levels varied spatially, with the highest levels in private wells downgradient of a road salt storage facility followed by wells within 30 m of a major roadway. Most well users surveyed (70%) had stopped drinking their well water for aesthetic and safety reasons. In the bench-scale experiment, increasing chloride concentration in water increased galvanic corrosion and dezincification of plumbing materials, resulting in increased metal leaching and pipe wall thinning. Our simple spatial analysis suggests that 2% of private well users in New York could potentially be impacted by road salt storage facilities and 24% could potentially be impacted by road salt application. Our research underscores the need to include the damage to public and privately owned drinking water infrastructure in future discussion of road salt management.



## 1. INTRODUCTION

Rising chloride levels nationally in freshwater sources are receiving increased scrutiny.<sup>1–3</sup> Road salt is applied to de-ice roads in the winter for highway safety, with more than 18 million metric tons applied annually and most used in northeastern and midwestern states.<sup>4</sup> This translates to 62.1 kg (137 lbs) of road salt per person each year.<sup>5</sup> Nearly all of this road salt eventually enters adjacent rivers, streams, and aquifers,<sup>6–9</sup> with detrimental impacts to both ecosystem function<sup>10</sup> and drinking water supplies.<sup>11–13</sup> Herein we argue that road salt's impact on drinking water infrastructure—in terms of lifetime, leaks, and water contamination—is an emerging problem for both private well owners and municipal water suppliers.

For more than 50 years, researchers have reported increased corrosion rates of metals in the presence of chloride.<sup>11,13,14</sup> The Larson Index was the first corrosion index to document the relationship between chloride and sulfate in drinking water and its impact on steel and cast iron water main corrosion.<sup>13</sup> However, chloride acceleration of corrosion became highly publicized in the early 2000s when water utilities switched

from sulfate-based to chloride-based coagulants, as it affected lead release from lead-solder joints.<sup>14</sup> Researchers determined that by increasing the chloride-to-sulfate mass ratio (CSMR) above the critical threshold of 0.5, galvanic corrosion (i.e., corrosion between dissimilar metals) was promoted.<sup>14–16</sup> While sulfates inhibit corrosion by forming passive protective film layers and reducing galvanic currents between dissimilar metals, chlorides prevent the formation of such passive layers and stimulate galvanic current.<sup>14,16</sup> Increases in chloride (and corrosion indices) may not be immediately apparent, as water utilities are not required to monitor chloride in water. However, water utilities have experienced increased infrastructure corrosion rates, higher levels of lead in water, and even Lead and Copper Rule (LCR) action level exceedances as a result of higher chlorides in water.<sup>11,14,17,18</sup> Moreover, using water quality data sets and the U.S. Environmental Protection

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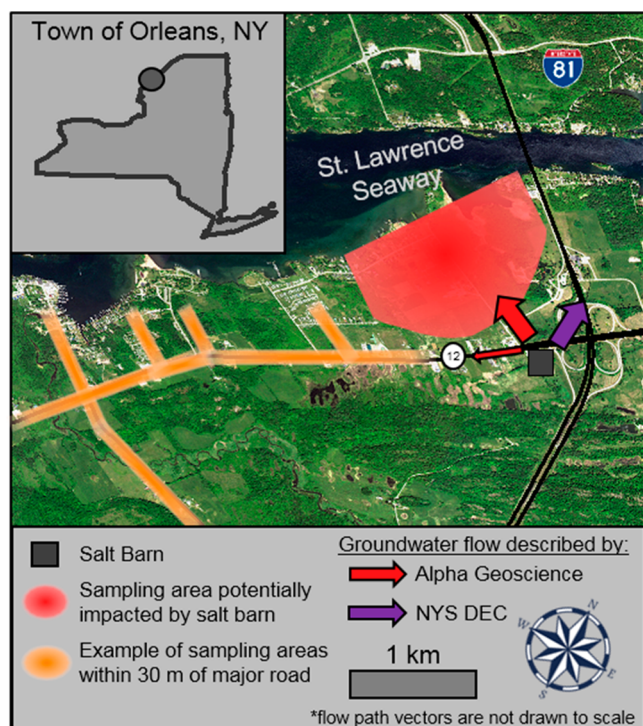
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Agency (USEPA) Safe Drinking Water Information System database, Stets et al. (2018) illustrated the statistical relationship between surface water chemistry and resulting corrosion in drinking water utilities.<sup>11</sup>

In New York State, there are concerns that road salt storage facilities (i.e., point sources of chloride) and roadway applications (i.e., nonpoint or diffuse sources of chloride) are increasing chloride levels in private wells.<sup>12,19,20</sup> In Jefferson County, there has been a multiyear controversy regarding the source(s) of high chloride found in private wells, which has been attributed to a road salt storage facility (Figure 1), general



**Figure 1.** Location of the Town of Orleans in New York State and grouping of private wells based on proximity to road salt sources. Specifically, the red area denotes where private wells may potentially be impacted by the salt barn as described by Alpha Geoscience,<sup>21</sup> orange area denotes where private wells may potentially be impacted by road salt application as described by Kelly et al. (2018),<sup>12</sup> and areas with no color denote where private wells are not likely impacted by the salt barn or road salt application. Groundwater flow paths described by Alpha Geoscience are shown by red arrows while flow paths described by New York State Department of Environmental Conservation (NYS DEC) are shown by a purple arrow.

road salt application, discharge from septic and water softener systems, and natural sources.<sup>21–23</sup> In this study, we utilize citizen science sampling, bench-scale corrosion studies, and spatial modeling to (1) characterize drinking water quality in a private well community potentially impacted by road salt storage and application; (2) determine the effect of increasing road salt concentrations on the corrosion of plumbing materials; and (3) estimate the number of private well users in New York potentially impacted by road salt.

## 2. MATERIALS AND METHODS

**Salt Storage Facility in Town of Orleans Area.** The Collins Landing Salt Storage Facility (hereafter termed the salt barn; Figure 1) is located at the intersection of Interstate 81

and New York State Route 12. Built in the 1980s, the facility initially stored road salt on an open concrete slab but was eventually covered following a New York State Department of Environmental Conservation (DEC) mandate in 1990.<sup>24–27,52</sup> The DEC stated that groundwater quality concerns may result from improper road salt storage and strongly encouraged proper road salt storage practices, especially when private wells are in the immediate vicinity.<sup>27</sup> Today, the Collins Landing Salt Storage Facility stores road salt in a covered barn structure.<sup>28</sup>

In the study area, the subsurface structure is a heterogeneous mixture of fractured bedrock and overlying unconsolidated deposits.<sup>21</sup> Alpha Geoscience observed that wells located directly downgradient from the salt barn experienced elevated chloride concentrations and suggested that groundwater flowed from southeast to northwest (perpendicular to the St. Lawrence River; Figure 1).<sup>21</sup> In addition, Alpha Geoscience noted a secondary flow path along Route 12 (parallel to the St. Lawrence River). These flow paths described by Alpha Geoscience were also observed in a study by Fourth Coast Inc.<sup>23</sup> These studies were paid for by a resident and the Town of Orleans. In contrast, an assessment by DEC stated that regional groundwater gradients flow north and away from the private wells.<sup>22,25</sup> Further, the DEC suggested that other sources of chloride are responsible for the observed chloride concentrations (e.g., road salt applied to adjacent roadways, discharge from local septic and water softener systems, and geologic sources).

**Citizen Science Testing of Well Water.** Between April and June 2016, 125 well water sampling kits were made available to residents in the Town of Orleans area. Local media outlets (e.g., newspaper, television) were contacted to notify well users of this testing opportunity. The well water sampling protocol used in this study was developed by Pieper et al. (2015) to characterize premise plumbing and groundwater quality.<sup>29</sup> In brief, participants were instructed to collect two water samples from the cold-water kitchen tap at normal flow: (1) 250 mL first draw of water collected after 6+ hours of stagnation and (2) 250 mL flush sample collected after 1 min of flushing. Water samples were shipped to Virginia Tech for metal analysis via inductively coupled plasma-mass spectrometry (ICP-MS) per Standard Method 3125-B.<sup>30</sup> Water samples were acidified with 2% v/v concentrated nitric acid and digested for a minimum of 16 h before analysis. For data quality assurance and quality control, blanks and spikes of known concentrations were measured every 10–15 samples. Participation in this water sampling campaign was free and voluntary, and all protocols were approved by VT IRB (16–302). In total, 95 of the 125 sampling kits (76%) were returned for analysis. Water quality results from five wells were removed from this analysis, as these homes were not located within the targeted study area (i.e., located on neighboring islands).

In addition to water sampling, residents were asked to complete a questionnaire about household and system characteristics, water use patterns, and perception of water quality (Supporting Information Section SI-1). Eighty nine of the 90 accompanying questionnaires were completed (99%). An overview of reported system characteristics is included in Section SI-2. Based on addresses provided on the questionnaire, private wells were divided into three groups (Figure 1). The first group included homes downgradient of the salt barn as defined by the Alpha Geoscience report,<sup>21</sup> and were also downgradient of the intersection of Interstate 81 and State

Route 12. The second group was located within 30 m of major roadways (not including neighborhood roads), as Kelly et al. (2018)<sup>12</sup> concluded that such wells are impacted by road salting practices. The third group of private wells were not in proximity to the salt barn or major roadways (only impacted by neighborhood roads).

**Bench-Scale Corrosion Testing.** As we were not able to adequately characterize the effect of road salt on drinking water infrastructure during the citizen science testing, we conducted a follow-up bench-scale experiment. Specifically, using chloride levels observed in the Town of Orleans area, we isolated the impact of increasing chloride on corrosion. A synthetic water was developed based on water quality from a well in this study, which had a pH of  $7.4 \pm 0.1$ , alkalinity of  $122 \pm 5$  mg/L as  $\text{CaCO}_3$ , sulfate concentration of  $32.9 \pm 1.0$  mg/L, and no disinfectant residual (Table SI-1). To isolate the effect of increasing road salt, sodium chloride ( $\text{NaCl}$ ; 99.5% pure) was added to the synthetic well water at varying levels spanning the range observed in field sampling: 10 mg/L, 62.5 mg/L, 250 mg/L (USEPA secondary standard for chloride), and 1,000 mg/L. Since the background sulfate concentration was held constant, the CSMR values in the synthetic water ranged from 0.3 to 30.4.

**Metal Leaching Jar Tests.** Iron wires (99.9% pure; 2 mm (0.08 in.) diameter), zinc wires (99.5% pure; 2 mm (0.08 in.)), stainless steel 304 wires (3.18 mm (0.125 in.)), and brass alloy 360 wires (3.18 mm (0.125 in.)) of 5.7 cm (2.25 in.) length were epoxied to the center of 125 mL glass jar lids (Figure SI-1). Each wire material was tested in triplicate with weights within  $\pm 3\%$ . To create simulated lead solder joints, 50/50 lead–tin solder (Alpha 13505) was applied to the inner surface of 30 copper coupling (2.54 cm (1 in.) by 0.95 cm (0.375 in.)). After conditioning the samples for 3 days using the NSF 61 testing water (pH  $8.5 \pm 0.5$ , alkalinity  $500 \pm 25$  mg/L as  $\text{CaCO}_3$ ),<sup>31</sup> 3-day composite water samples (375 mL) were analyzed for lead and copper. The 12 lead solder joints with the lowest achievable relative standard deviation (RSD) for copper (RSD = 5.1%) and lead (RSD = 16.1%) were selected for testing. The selected 12 solder joints had an average copper level of  $853 \pm 49$   $\mu\text{g/L}$  and average lead of  $59.1 \pm 9.5$   $\mu\text{g/L}$ . To examine deposition corrosion (plating of dissolved copper on metal surfaces and resulting corrosion of the microgalvanic cells),<sup>32</sup> iron and zinc wires were also exposed to the four chloride water conditions with 3 mg/L cuprous chloride ( $\text{CuCl}$ ) as copper ( $\text{Cu}$ ) added. For these conditions, the amounts of  $\text{NaCl}$  added to the synthetic waters were adjusted to maintain the target chloride concentrations. In total, 84 test apparatuses were exposed to four water conditions with and without  $\text{CuCl}$ .

Each jar was filled completely with the corresponding water to exclude air and was mixed on a MaxQ3000 shaker table to simulate water flow within home and well plumbing. A dump-and-fill protocol was conducted 3 times per week (Monday, Wednesday, and Friday), and water samples were collected during each water change. Water samples were processed as previously described via ICP-MS,<sup>30</sup> except samples from iron wire apparatuses were also digested with 2% v/v hydroxylamine and heated at 50 °C for a minimum of 24 h.<sup>33</sup> For the final sampling round (Day #47), water samples from iron wires were lost during incubation.

**Weight Loss Analysis.** To determine the weight loss of each wire apparatus (lead solder joints were not included), the dried weight (60 °C for 24 h) of each apparatus was measured

before the experiment and at the end of the experiment after cleaning based on revised ASTM G1 standard practice.<sup>34</sup> Specifically, after the experiment, the surface scales on zinc, iron, and stainless steel wires were mechanically removed with a Dremel tool equipped with cloth polishing bits to ensure minimal removal of metallic surface. Scales on brass wires were chemically removed using commercial Brasso. The total weight loss was calculated as the difference between dried weight before and after the experiment.

**Exploring Well Communities Potentially Impacted by Road Salt.** A simple spatial model was developed to examine the potential impact of road salt on private wells across New York. The model estimated the relative number of private well users potentially impacted by both point sources (i.e., salt storage facilities) and nonpoint sources (i.e., salt runoff from major roads). Data sources included: private well locations from the U.S. Geological Survey (USGS),<sup>35</sup> catchment and flow accumulation data sets from the National Hydrography Plus Database (NHD Plus Version 2.1, <http://nhd.usgs.gov>), salt barn locations provided by the New York State Department of Transportation (NYSDOT), and roadways derived from USGS National Transportation Data set.<sup>36</sup> It is important to note that there is uncertainty associated with USGS well location data set, which were derived from 1990 US Census data, proximity to roadways, and municipal boundaries.<sup>35</sup> Although the DEC is developing a database of private wells (NYS Water Wells Database; <https://gis.ny.gov/gisdata/inventories/details.cfm?DSID=1203>), it is largely incomplete at this time. Thus, the USGS data set represents the most complete data set currently available. In addition to uncertainty associated with well locations, site specific characteristics such as geology, well depth, and salt storage and applications practices will greatly affect road salt concentrations and such data were not available during this effort. Thus, this effort represents a first order, back-of-the-envelope, estimate of private well users potentially impacted by road salt, which aims to provide insights into the potential extent of the problem.

To estimate the number of private well users potentially impacted by salt storage facilities, we combined well locations, NHD catchments, NHD flow accumulation raster layers, and NYSDOT salt barn locations. First, NHD catchments with road salt facilities were identified, which ranged in size from roughly 1 to 880 ha (median of 130 ha). We then determined areas downgradient of the storage facility within the identified NHD catchments using the flow accumulation raster. Essentially, this area represents the potential flow path for a contaminant plume if it followed topographic gradients. While this is often the case in shallow aquifers, flow patterns in bedrock aquifers can be more complicated both spatially and temporally.<sup>37</sup> To estimate the number of well users impacted by road salt application, we combined well locations with major roadway locations. Researchers observed that private wells in East Fishkill, NY within 30 m of a major roadway had significantly higher chloride levels.<sup>12</sup> Thus, we tabulated the number of well users within a 30 m buffer of major roadways. The R code for this model is included in Section SI-3.

**Statistical Analysis.** All statistics were conducted in R (version 3.3.2) using an alpha value ( $\alpha$ ) of 0.05. Non-parametric statistics including Wilcoxon test and Kruskal–Wallis test were used to compare the median concentrations among chloride groupings and levels, CSMR values, and resident use of water. Spearman's rho ( $\rho$ ) was used to evaluate

the association between water quality materials, distance to the salt barn, and CSMR values. Chi-squared test was used to measure association between reported corrosion issues and chloride groupings.

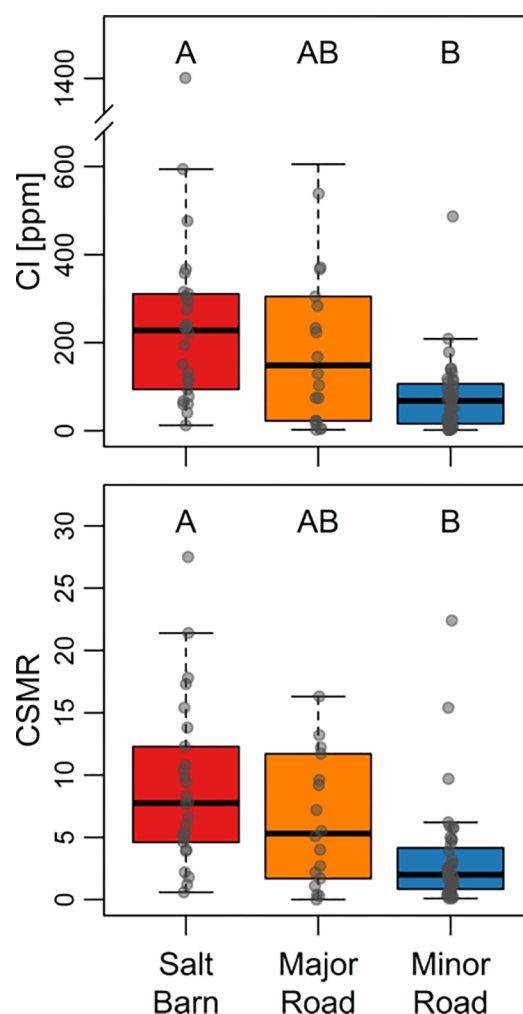
### 3. RESULTS

#### Citizen Science Testing: Private Well Water Quality.

Citizen science sampling was used to explore water quality in the Town of Orleans area. Specifically, this campaign focused on documenting spatial patterns in chloride concentrations and corrosion and water quality perception concerns. Results from this effort documented that (1) chloride levels varied spatially and trends were consistent with prior work;<sup>12,21,23</sup> (2) some wells had elevated lead and copper concentrations and CSMR levels were above 0.5 in most wells; (3) most well users had stopped drinking their well water for aesthetic and safety concerns; and (4) more detailed information on plumbing characteristics was necessary to evaluate the impact of increasing chloride on corrosion.

**Chloride Concentrations Varied Based on Distance to the Salt Barn.** During this study, chloride levels were elevated above the USEPA standard of 250 mg/L in 21% of the wells sampled, with concentrations as high as 1,401 mg/L (Table SI-2 and Figure SI-2). Sodium levels were also problematic, as 82% of wells had concentrations above the recommended standard of 20 mg/L with a high of 786 mg/L. There was a strong negative correlation between a well's distance to the salt barn and the chloride in water concentration ( $\rho = -0.40$ ;  $n = 90$ ), implying a connection between the salt barn and chloride in groundwater. To evaluate the potential impact of road salt from both the salt barn and application, we classified the private wells into three groups (Figure 1).<sup>12,21</sup> There were significant differences in chloride concentrations based on this grouping (Kruskal Wallis,  $p < 0.05$ ), as wells downgradient of the salt barn had the highest median concentrations (228 mg/L;  $n = 26$ ), followed by those within 30 m of a major roadway (116 mg/L;  $n = 20$ ), and those not in proximity to either source (70 mg/L;  $n = 44$ ) (Figure 2). These findings suggest that chloride levels are highest in proximity to the salt barn but are also elevated near major roadways, consistent with expectations based on other research.<sup>12,21,23</sup>

**Lead and Copper Were Concerns for Well Users Throughout the Study Area.** One in five first draw samples (20%) in this study exceeded either the LCR lead and/or copper action levels (Table SI-2 and Figure SI-3). Specifically, 12% of wells sampled had lead above 15  $\mu\text{g/L}$  and 13% had copper above 1.3 mg/L. Detectable lead ( $\geq 1 \mu\text{g/L}$ ) was observed in 74% of first draw samples, with concentrations as high as 40.6  $\mu\text{g/L}$ . Copper concentrations were observed as high as 8.2 mg/L. There was no relationship between lead and copper levels in well water based on distances to the salt barn (Spearman's  $\rho$ ,  $p = 0.20$ ,  $p = 0.87$ ), or based on well groupings (Kruskal–Wallis,  $p = 0.96$  and  $p = 0.33$ , respectively). Increases in lead and copper concentrations are heavily dependent on the plumbing infrastructure, which varied from home to home. The highest proportion of homeowner-reported copper plumbing use was in wells downgradient of the salt barn (67%;  $n = 24$ ), compared to wells within 30 m of a major roadway (47%;  $n = 17$ ) and not in proximity to the salt barn or major roadways (64%;  $n = 42$ ). Interestingly, there were no differences in reported corrosion issues (e.g., pinhole leaks, plumbing failures) based on groupings (Chi-squared,  $p = 0.35$ ), as 50% of wells downgradient of the salt barn reported

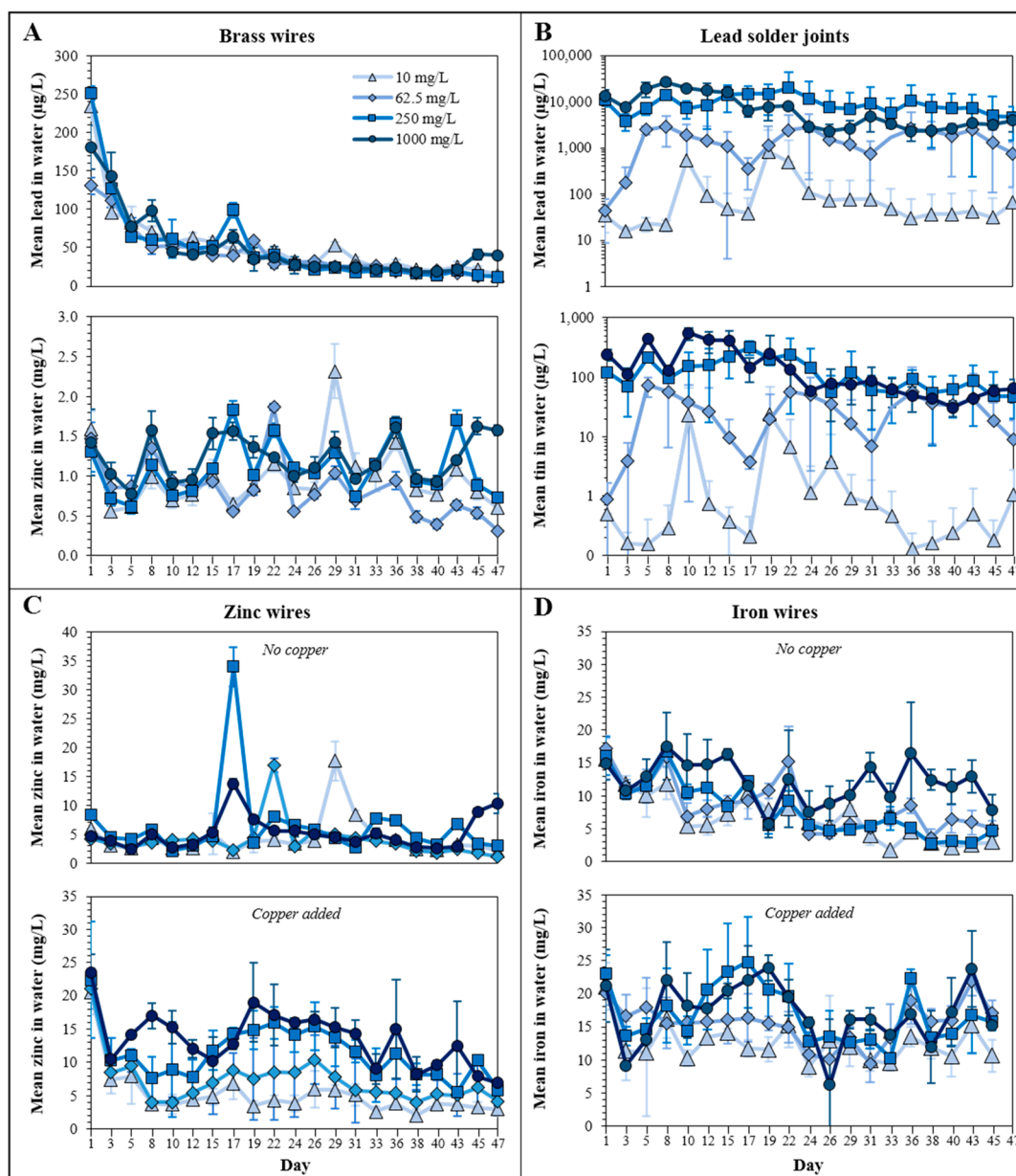


**Figure 2.** Chloride and CSMR values based on groups: (1) downgradient of the salt barn and intersection of Interstate 81 and State Route 12; (2) within 30 m of a major roadway; and (3) not in proximity to the salt barn or major roadways. Cl: chloride concentrations. CSMR: chloride-to-sulfate mass ratio. A and B are used to denote significant differences between groups (Kruskal–Wallis,  $p < 0.05$ ).

issues, 50% of wells within 30 m of a major roadway, and 35% of wells not proximity to either source. Correlating corrosion damage to chloride levels in a random sampling campaign was not possible given the variabilities in plumbing infrastructure from home to home.

Lead was highly correlated with copper ( $\rho = 0.52$ ) and zinc ( $\rho = 0.59$ ), suggesting the corrosion of leaded brass fittings.<sup>38,39</sup> Only one home had measurable tin in water (1.2  $\mu\text{g/L}$ ), indicating possible corrosion of lead solder joints. After 1 min of flushing, 4% of flushed samples ( $n = 4$ ) exceeded LCR action levels—two homes had elevated lead (17.4 and 19.1  $\mu\text{g/L}$ ) and two homes had elevated copper (1.5 and 2.1 mg/L). Despite the reduction in water lead associated with flushing, 26% of homes ( $n = 23$ ) had detectable lead after 1 min of water use. Lead was still correlated with copper ( $\rho = 0.47$ ) and zinc ( $\rho = 0.39$ ), suggesting corrosion of brass fittings in the well plumbing and/or sloughing of leaded scales.<sup>29</sup>

A primary drinking water concern related to road salt is the increase in CSMR values, but there is no database of typical CSMR values and corresponding metal concentrations in



**Figure 3.** Mean metal concentrations that leached into water during 47-day exposure to 10–1000 mg/L chloride from (A) brass wires, (B) lead solder joints, (C) zinc wires, and (D) iron wires. Error bars denote the 95th confidence interval.

private wells. A data set from Virginia<sup>40</sup> reports that 59% of wells in the Ridge & Valley region ( $n = 654$ ) and 78% of wells in the Piedmont region ( $n = 508$ ) have CSMR values above 0.5. During this study, 89% of wells had CSMR values above the critical threshold of 0.5 (Figure SI-4), with median CSMR values of 7.7 in wells adjacent to the salt barn, 4.5 in wells in proximity to major roadways, and 1.9 in wells not in proximity to either source (Figure 1). Both median lead and copper levels were significantly higher when CSMR values were greater than 0.5 (Wilcoxon Test,  $p < 0.05$ ), with median lead of  $<1 \mu\text{g/L}$  vs  $2.7 \mu\text{g/L}$  and median copper  $44 \text{ mg/L}$  vs  $179 \text{ mg/L}$ . However, there were weak correlations between CSMR and both lead ( $\rho = 0.31$ ) and copper concentrations ( $\rho = 0.22$ ).

**Perception of Water Quality Was Influenced by Distance to Salt Barn.** Based on questionnaire results ( $n = 89$ ), 70% of residents stated that they were not drinking their tap water.

Most residents living downstream of the salt barn were not drinking their well water (85%) compared to those in proximity to major roads (75%) and only 58% for those not in proximity to either source. Residents that stopped drinking their water had significantly higher ( $p < 0.05$ ) median lead levels ( $2.8$  vs  $1.1 \mu\text{g/L}$ ) and chloride ( $124.4$  vs  $27.2 \text{ mg/L}$ ), but this did not hold true for copper ( $155$  vs  $178 \text{ mg/L}$ ;  $p = 0.42$ ). Residents who answered survey questions about why they stopped drinking the water ( $n = 50$ ) described four general reasons: 22% indicated that they were advised by others to stop (e.g., advice from neighbors, state provided bottled water); 46% noted aesthetic issues with the water (e.g., taste, color, odor problems); 32% were concerned about the safety of the water (e.g., tested positive for lead and/or bacteria, contamination concerns); and 6% reported other reasons. All but one (95%) of these residents switched to bottled water as their primary drinking water source. While the state is

providing some residents with bottled water service, others are personally bearing the costs of this alternative supply.

**Bench-Scale Testing: Impact of Increasing Chloride on Plumbing Corrosion.** Follow-up bench-scale experiments were conducted to isolate the impact of road salt on corrosion of four key building plumbing materials. Results from this effort confirmed that (1) increasing chloride concentrations increased galvanic corrosion and dezincification; (2) chloride levels did not impact stainless steel corrosion; and (3) metal leaching, thinning of plumbing materials, and degradation of aesthetic water quality must be considered when evaluating the impact of road salt on drinking water.

**Lead Solder Corrosion Increased with Road Salt Loading.** CSMR is key factor controlling leaching from lead solder joints,<sup>14</sup> and previous studies have examined the impact of CSMR on lead leaching mostly with low alkalinity waters (<50 mg/L as CaCO<sub>3</sub>).<sup>16,41</sup> Even though synthetic water in this study had an alkalinity of 122 mg/L as CaCO<sub>3</sub>, increasing chloride levels up to 250 mg/L resulted in a significant median increase in lead leaching from lead solder joints (Kruskal–Wallis Test,  $p < 0.05$ ; Figure 3B), with no difference observed between 250 and 1000 mg/L chloride ( $p = 0.10$ ). The increased corrosion rate was also reflected by more white lead suspended particles at higher (250 and 1000 mg/L) versus lower (10–62.5 mg/L) chloride levels (Figure SI-5). The lack of difference in median lead among the 250 and 1000 mg/L chloride water conditions was attributed to the high variability, which has been observed in prior studies.<sup>14,42</sup> Specifically, median lead values ( $\pm$ standard deviation) were  $47 \pm 61$   $\mu$ g/L at 10 mg/L chloride,  $1,466 \pm 1232$   $\mu$ g/L at 62.5 mg/L chloride,  $7710 \pm 6296$   $\mu$ g/L at 250 mg/L chloride, and  $4850 \pm 1322$   $\mu$ g/L at 1000 mg/L chloride. After 14 days, lead leaching from the 250 mg/L chloride conditions became more corrosive than the 1000 mg/L chloride condition, but again there was high variability in the 250 mg/L triplicates (standard deviation of 8423  $\mu$ g/L for days 15–47).

Similar trends were observed with tin leaching, as 250 and 1000 mg/L chloride waters had significantly higher median levels compared to 10 and 62.5 mg/L waters ( $88\text{--}97$  vs  $<1\text{--}30$   $\mu$ g/L;  $p < 0.05$ ). In contrast, copper leaching levels were higher in lower chloride waters ( $p < 0.05$ ). Median copper levels in 10 and 62.5 mg/L chloride conditions (630 and 592  $\mu$ g/L, respectively) were higher than 1000 mg/L chloride (98  $\mu$ g/L), which was higher than 250 mg/L chloride water (57  $\mu$ g/L). This might be due to protective effects of chloride on copper release or due to sacrificial protection of the copper by galvanic connection to the lead.<sup>43</sup> Overall, increasing chloride concentrations increased the galvanic corrosion between lead solder and copper pipes, resulting in increased lead and tin leaching. However, corrosion rates between 250 and 1000 mg/L should be further explored, as 250 mg/L chloride waters released higher tin and lead levels but there was high variability among the triplicates.

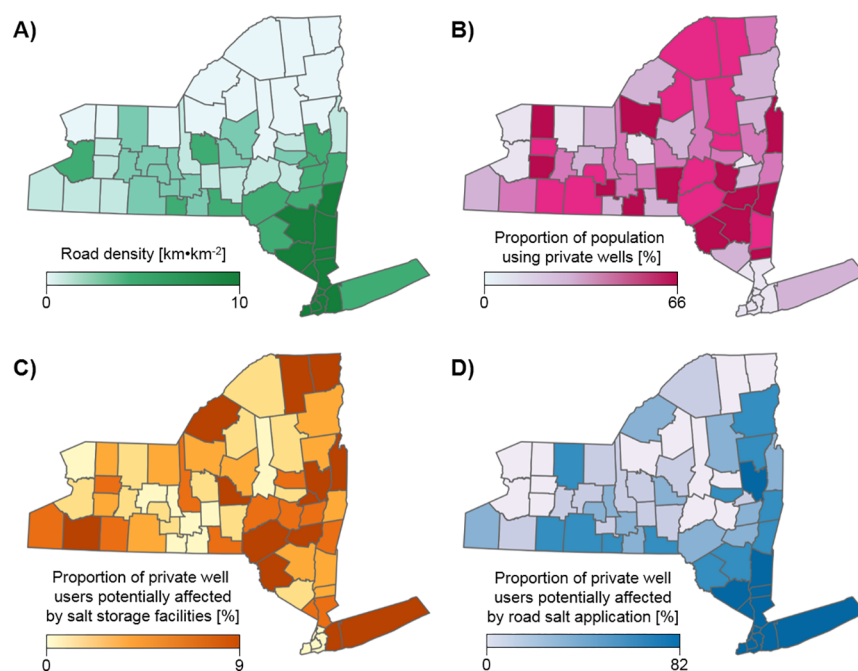
**Road Salt Influenced Galvanized Iron Corrosion When Copper Was Present.** When zinc wires (representing new galvanized iron pipes with 100% zinc surface coating) were exposed to varying chloride concentrations, median zinc values ranged from 3.3 to 4.7 mg/L over the range of chloride levels tested (Figure 3C). However, when 3 mg/L CuCl as Cu was added to the synthetic well water, zinc leaching was strongly influenced by chloride concentrations, as median zinc levels were higher (Kruskal–Wallis,  $p < 0.05$ ) in the presence of CuCl and increased with increasing chloride levels for 62.5–

1000 mg/L chloride waters. Specifically, median zinc values were 3.8 mg/L at 10 mg/L chloride, 4.4 mg/L at 62.5 mg/L chloride, 10.1 mg/L at 250 mg/L chloride, and 14.3 mg/L at 1000 mg/L chloride. Moreover, increased zinc leaching was evident by the white visible particles (Figure SI-6). Wire weight loss was also impacted, as weight loss ranged from 0.5 to 1.3% without copper but increased to 2.1–6.0% when copper was present (Figure SI-7). Thus, new galvanized iron pipes (100% zinc surface coating) would be expected to experience increased galvanic corrosion and weight loss in the presence of road salt.

Iron wires (representing aged galvanized iron pipes with 100% iron surface) were not influenced by chloride concentrations up to 250 mg/L, as median iron leaching ranged 5.8–7.4 mg/L (Figure 3D). However, iron leaching was significantly higher in 1000 mg/L chloride waters (Kruskal–Wallis,  $p < 0.05$ ; median 12.5 mg/L), suggesting that high chloride levels will increase iron corrosion. With the addition of 3 mg/L CuCl as Cu in the synthetic well water, iron leaching was significantly higher ( $p < 0.05$ ) and varied based on chloride concentrations ( $p < 0.05$ ). Specifically, 250 and 1000 mg/L chloride waters had the highest median iron leaching (16.1 mg/L), whereas the 62.5 mg/L chloride waters were not statistically different (14.4 mg/L;  $p = 0.68$  and 0.47, respectively), but the 10 mg/L chloride water (11.0 mg/L) was statistically lower ( $p < 0.05$  for 250 and 1000 mg/L chloride,  $p = 0.49$  for 62.5 mg/L chloride). Weight loss rates varied from 5.6 to 7.2% without copper and was lower at 5.1–5.8% when copper was added. We speculate that the iron wire with copper was oxidized to form iron(III) protective layers, which prevented further corrosion.<sup>32</sup> Overall, aged galvanized iron pipes would be expected to experience increased corrosion for very high chloride waters, but not weight loss in the presence of road salt.

**Brass Corrosion Rates Were Not Impacted by Increased Road Salt.** Lead leaching from C36000 brass (65.2% Cu, 31.9% Zn, and 2.1% Pb) decreased from an average of 181–251  $\mu$ g/L on day 1 to 11.8–40.0  $\mu$ g/L on day 47 (Figure 3A) due to aging of brass.<sup>44</sup> In addition, chloride levels did not influence the lead leaching into water, as there was no significant difference in median lead ( $p = 0.24$ ). However, the increasing chloride levels generally increased zinc leaching into water. Median zinc was 0.85 and 0.84 mg/L at 10 and 62.5 mg/L chloride and leaching was significantly higher at chloride levels of 250 and 1000 mg/L (1.03 and 1.21 mg/L;  $p < 0.05$ ). These results were consistent with prior observations that zinc leaching increases with increasing chloride levels, as dezincification selectively leaches zinc at higher levels of chloride.<sup>45</sup> This was also reflected by darker red color in the wire surface at higher chloride levels (250–1000) compared to lower chloride levels (10–62.5 mg/L; Figure SI-8). Chloride levels did not influence weight loss rates, as rates were less than 0.1% for all wires (Figure SI-7).

To further understand dezincification, prior research has utilized dezincification and lead leaching indices.<sup>45</sup> The dezincification index evaluates the ratio of zinc:copper leaching to water versus that present in the alloy, and values above 1.0 indicate increased dezincification. The index was higher for the 250 mg/L chloride water (median 24.6) compared to the other conditions (11.4–12.3). As for the lead:copper leaching index, there was no difference in the median lead leaching potential index ( $p = 0.054$ ) with indexes ranging from 0.99 to 1.64.



**Figure 4.** Road density (A),<sup>36</sup> portion of population using private wells (B),<sup>35</sup> and estimates of number of potentially impacted private well users by (C) salt storage facilities and (D) roadway runoff for New York State.

Overall, increasing chloride levels did not increase lead leaching but did increase dezincification.

**Stainless Steel Corrosion Rates Were Not Impacted by Increasing Road Salt.** The grade 304 stainless steel (72.0% Fe, 18.4% Cr, and 8.0% Ni) released minimal levels of iron, chromium, and nickel after 47-day exposure, as most samples contained nondetectable iron of 10  $\mu\text{g/L}$  (95%), chromium of 1  $\mu\text{g/L}$  (98%), and nickel of 0.1  $\mu\text{g/L}$  (93%). Stainless steel has been shown to effectively resist road salt induced corrosion by forming a protective iron–chromium oxide passive layer on the surface.<sup>46–48</sup> The weight loss rate during the 47 days of experiment was <0.04% for all stainless steel wires (Figure SI-7), further suggesting resistance of this material against road salt.

#### 4. IMPLICATIONS

##### Road Salt's Impact on Drinking Water Infrastructure.

Our bench-scale experiments unambiguously demonstrate that increased chloride in drinking water can accelerate galvanic corrosion and dezincification, causing increased water contamination, thinning of plumbing materials, and aesthetic concerns. As observed in our citizen science testing, the impact of increasing chloride will vary at the household level, as it is dependent on the plumbing materials present in the drinking water infrastructure. Both old and new homes are potentially at-risk. Older housing constructed before 1986 may have copper plumbing with lead solder joints, while nearly all housing stock will have brass fittings and fixtures installed.<sup>38</sup> Moreover, galvanized iron is still commonly used in construction of wells.<sup>49</sup> While this research highlighted concerns associated with chloride accelerated corrosion, further research on additional water quality and plumbing materials and configurations should be conducted to fully characterize the extent of this emerging drinking water problem.

The Town of Orleans is not alone in their concerns about road salt. NYSDOT commissioned a study in the Township of

Dannemora to examine the impacts of road salt storage on private wells,<sup>19,50–52</sup> residents in the Town of Pamela have voiced concerns about a local salt storage facility,<sup>52</sup> and a recent study found high chloride levels in wells in East Fishkill attributed to road salt application.<sup>12</sup> Moreover, these road salt challenges are not isolated to New York, as other states have or are starting programs to investigate and/or replace private wells impacted by road salt.<sup>53–55</sup> Due to the road salt concerns in the Town of Orleans area, a new three-mile long municipal water line is being constructed from the Village of Alexandria Bay. This water line will provide municipal water to more than 500 residents and will cost an estimated \$13.2 million, of which \$5 million will be paid through grant funding and \$8.2 million will be the responsibility of the residents.<sup>22,50</sup> Over the 30-year loan period, residents will pay a water bill of \$625 per year (plus \$3.50 per 1000 gallons of water).<sup>52</sup> Although, these costs do not include the replacement of degraded home plumbing and appliances, which have been noted by residents (Figures SI-7 and S8).<sup>20,24</sup> Thus, this new waterline remediating road salt concerns will provide a safer drinking water supply, but may be an expensive solution for some residents.

##### Exploring Well Communities Potentially Impacted by Road Salt.

Combined with recent studies of chloride concentrations in surface water and groundwaters,<sup>1,2,56,57</sup> it is apparent that road salt will be a persistent problem for both municipal water systems and private wells.<sup>11,12</sup> However, without state or federal oversight, private well users are solely responsible for the identification of road salt problems and mitigation of chloride and corrosion concerns. To quantify the potential extent of road salt contamination associated with storage and application, we developed a simple spatial model of private wells across New York State. It is important to note limitations of such an estimate and acknowledge the uncertainty associated with site specific characteristics. Specifically, major sources of uncertainty include: variables that affect the fate and transport of chloride such as underlying

geology,<sup>12,40</sup> watershed characteristics,<sup>7</sup> stormwater management structures,<sup>9</sup> and characteristics of individual well systems (e.g., well depth, treatment<sup>12,40</sup>).

Our model suggests that roughly 35 000 private well users in New York (2% of well population) could be impacted by road salt storage facilities and 460 000 private well users (24% of well population) could be impacted by road salt application (Figure 4; Table SI-3). When scaled to the county level, the number of well users affected by salt barns ranged from 0 to 4100 in a single county (0–9%), with a median of 370 well users per county (1.2%). Note, there are 230 NYSDOT salt storage facilities spread out across 57 counties (out of 62 total counties), and the impact appeared to be evenly distributed across the state (Figure 4a). For well users potentially affected by road salt application, the number ranged from 360 to 60 000 in a single county (<1–82%), with a median of 5100 well users per county (20%). However, some of this impact may likely be overstated, as the model suggests a high occurrence of potential road salt contamination in Bronx, Nassau, and New York counties, areas where municipal water services are the predominate, if not the sole, source of drinking water. Nonetheless, other counties such as Steuben, Ontario, and Wayne are highlighted as counties where a high proportion of private well users are located within 30 m of a road, and therefore, potentially susceptible to the associated nonpoint source chloride contamination.

Taken together, the results from our citizen science sampling, bench-scale testing, and simple spatial model indicate that road salt storage (e.g., point source pollution) and road salt application (e.g., nonpoint source pollution) represent a potential concern for private wells across New York and other states with similar winter management practices. These preliminary estimates can be improved as more data are collected, but serve to understand the potential impact of road salt application and storage on local communities. Moreover, prior research has reported the need to implement watershed management practices to reduce well-known impacts,<sup>9</sup> and our research underscores the need to include drinking water infrastructure (e.g., product lifetimes, leaks, and water contamination) in future discussions.

## ■ ASSOCIATED CONTENT

### ■ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b04709.

Household questionnaire, additional water quality measurements and figures from the citizen science sampling and bench-scale experiments, and R code and results from the spatial analysis (PDF)

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### Notes

The authors declare no competing financial interest.

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