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# Mobilization of Radium and Radon by Deicing Salt Contamination of Groundwater

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**Abstract** Elevated radium (Ra) concentrations have been observed in aquifers with high naturally occurring salinity. The flux of radon (Rn) gas from the decay of Ra out of saline aquifers can be enhanced owing to salting-out effects. This raises the issue as to whether increased salinization of groundwater from road deicing practices can enhance Ra and Rn mobility to the extent that they become a human health concern. Continued use of salt (NaCl) as a road deicing agent has resulted in a gradual salinization of groundwater systems in snow-affected regions. This study presents groundwater data from a monitoring well field installed around a permeable pavement parking lot at the University of Connecticut, Storrs campus. The data suggest a connection between road salting and (a) the mobilization of dissolved Ra as well as (b) enhanced Rn gas flux from the water table. A positive correlation ( $R^2 = 0.92$ ) was identified between dissolved  $\text{Na}^+$  and isotopes of Ra; a negative relationship was observed between specific conductance and

dissolved Rn. In two monitoring locations, concentrations of Ra were detected that exceeded the EPA MCL of 5 pCi/L. Concentrations of Rn in the groundwater were found to be at a level that theoretically could generate gas concentrations in the vadose zone that exceed the indoor Rn standard by orders of magnitude. Given these findings, it appears that salt contamination of groundwater could increase the potential for human exposure to these radioactive and carcinogenic elements.

**Keywords** Road salt · Radium · Radon · Groundwater · Urban aquifers

## 1 Introduction

Radium (Ra) concentrations in groundwater have been highly correlated with sodium ( $\text{Na}^+$ ) chloride ( $\text{Cl}^-$ ) levels in saline aquifers (Sturchio et al. 2001; Vinson et al. 2009). This phenomenon has been attributed to increased competition for adsorption sites due to the abundance of  $\text{Na}^+$  ions (Krishnaswami et al. 1982; Sanders et al. 2013; Tamamura et al. 2014). Also, the solubility of Ra can be enhanced by the formation of  $\text{RaCl}^+$  complexes in saline waters (Langmuir and Riese 1985). The observed naturally occurring correlation between Ra and salt in saline aquifers suggests that Ra, and its progeny Rn, could be mobilized by deicing salt contamination of groundwater.

Each year, an increasing amount of deicing salt is used in the USA (Mullaney et al. 2009; Fay and Shi 2012).

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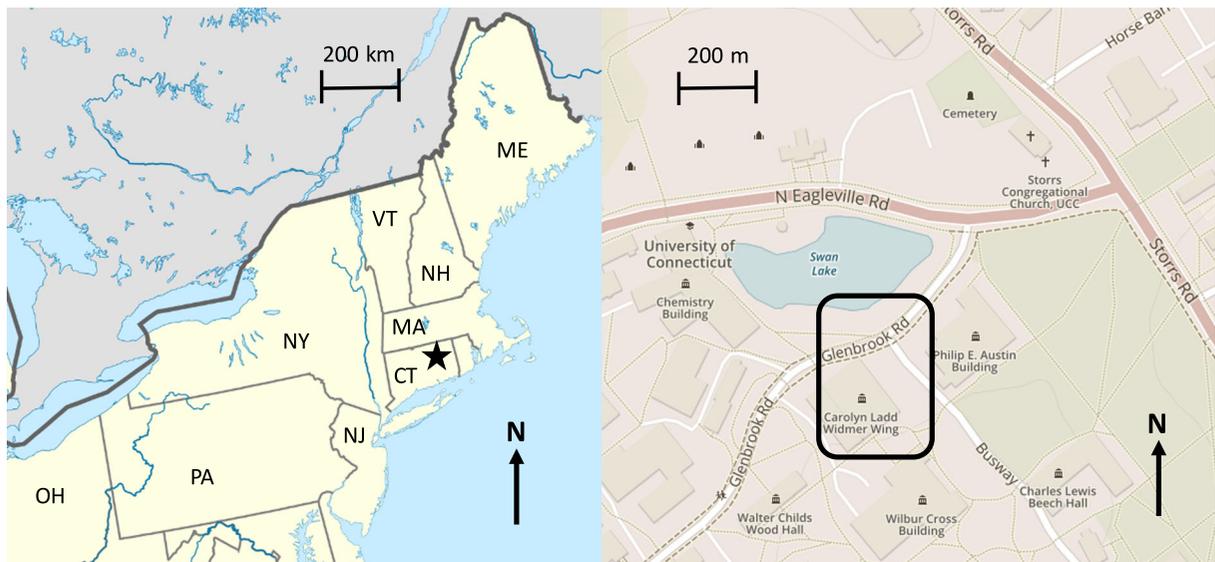
While the immediate transportation and safety benefits of this practice cannot be overlooked, further research is necessary to fully understand the human health impacts of deicing salt once it has left the road. The majority of the annual deicing salt mass will either run off directly into surface water bodies or percolate down to groundwater systems. A gradual salinization of both surface water and groundwater systems due to the continued prevalence of deicing has been observed (Kaushal et al. 2005; Kelly et al. 2008; Novotny et al. 2009; Trowbridge et al. 2010; Perera et al. 2013). These trends are documented across large timescales (Cassanelli and Robbins 2013) and on different continents (Kaushal 2016). The increased salinity of freshwater systems has been observed to persist throughout the year due to input from reservoirs of saline groundwater (Kincaid and Findlay 2009; Cooper et al. 2014). Deicing salt can be present in groundwater at concentrations sufficient to mobilize metal cations such as Cu, Pb, and Zn (Norrström and Jacks 1998; Bäckström et al. 2004).

A review of the occurrence of Ra in principal US aquifer systems found that dissolved Ra commonly occurs in the groundwater of fractured crystalline bedrock and the overlying glacial till of New England (Szabo et al. 2012). Ra is known to have a high solid/liquid distribution coefficient ( $K_D$ ) and as such would have minimal mobility in groundwater when the total dissolved solids are low (Sturchio et al. 2001; Vinson et al. 2009; Szabo et al. 2012). In more saline aquifers,

greater quantities of  $\text{Na}^+$  compete with Ra for adsorption sites; this enhances the mobility of Ra as reflected by lower  $K_D$  values (Tamamura et al. 2014). Elevated levels of Ra in groundwater can produce harmful health effects such as cataracts and osteosarcoma (CDC 2015).

Salt contamination of groundwater could also impact the rate of Rn (the progeny of Ra) exhalation. With a dimensionless Henry's Law coefficient ( $K_H$ ) of 0.35 at 10 °C, dissolved Rn readily partitions to the gaseous phase. This volatility increases with temperature. Furthermore, the solubility of a gas dissolved in solution decreases as the electrolyte concentration (i.e., salinity) increases (Weiss 1970). Schubert et al. (2012) developed a model to predict the  $K_H$  of Rn as a function of temperature and salinity. The model demonstrated that Rn will more readily partition to the gas phase (i.e., lower  $K_H$ ) under warmer and increasingly saline conditions (Schubert et al. 2012). Given the shallow nature of groundwater in the northeast, a decrease in  $K_H$  due to increased salinity could result in an increase in flux of Rn to overlying buildings (Krewski et al. 2005; Thomas and McHone 1997). This represents a public health concern due to the carcinogenic nature of the element; Rn exposure has been identified as the second leading cause of lung cancer in the USA (Darby et al. 2001).

The extent to which contamination of the shallow groundwater in the northeast by deicing salt can mobilize Ra and Rn is not known. The objective of this research is to evaluate the degree to which deicing salt



**Fig. 1** (Left) Black star denotes location of Storrs, CT within the Northeastern USA. (Right) Circled at center of the right map is the approximate location of the study area within Storrs, CT

contamination of shallow groundwater can enhance Ra and Rn mobility.

## 2 Methods

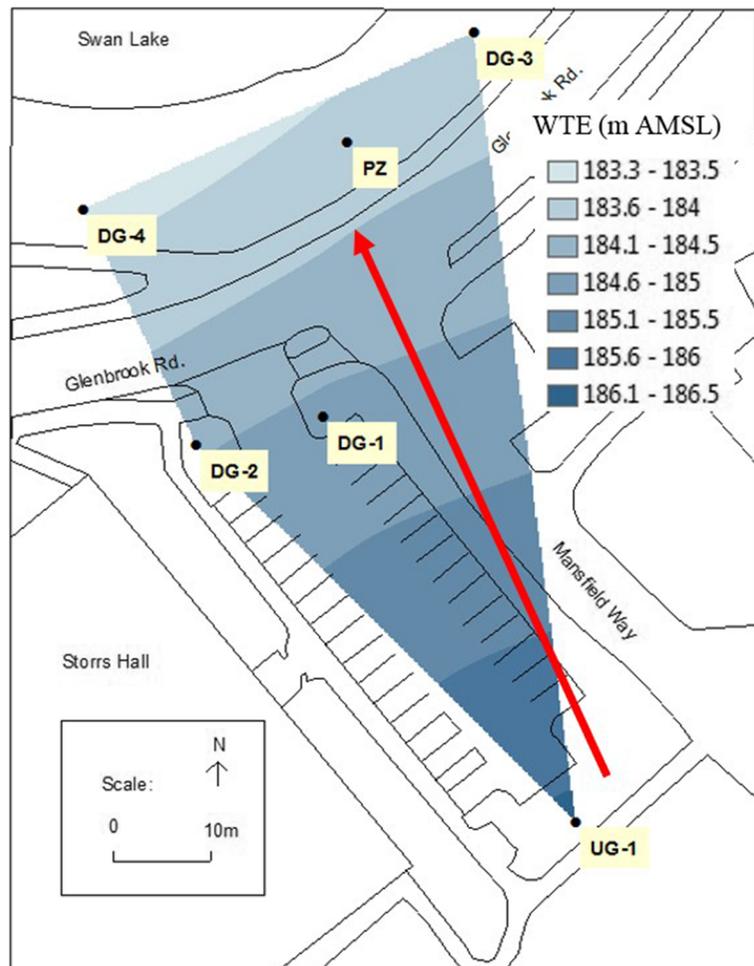
### 2.1 Study Site

The magnitude of salt contamination of groundwater depends largely on the soil permeability. As a worst-case analysis, this study focused on an area of very high permeability: an area of permeable pavement which is heavily salted in winter months. The site investigated is on the University of Connecticut campus in Storrs, CT, USA (Fig. 1). Within the study site is an 860-m<sup>2</sup> permeable asphalt parking lot. The overburden soil surrounding the permeable asphalt consists of highly disturbed glacial till and native sandy silt. The overburden

soil in the study area is underlain by the Hebron Gneiss at a depth ranging from about 2.1 to 5.2 m. Due to the prevalence of till overburden formations in Connecticut, this site was assumed to be representative of subsurface conditions in this state.

Six overburden monitoring wells were installed around the study area to investigate the shallow groundwater (Fig. 2). Depth to the water table is approximately 3 m under the permeable pavement and 1 m near the piezometer cluster. Pertinent specifications for each monitoring well are enumerated in Table 1. Wells UG-1, DG-1, and DG-2 were drilled in August 2014 (Dietz et al. 2016), and wells DG-3 and DG-4 and the nested piezometer (PZ) were drilled in October 2015. As shown in Fig. 2, the shallow groundwater tends to flow in a northwesterly direction: traveling below the permeable asphalt (PA) then following the topography downhill to Swan Lake, a small stormwater detention pond. Well WB-1

**Fig. 2** Study site in the University of Connecticut campus in Storrs, CT. The red arrow shows direction of groundwater flow during dry periods. Contours show the elevation of the water table. Black labeled and numbered dots are shallow wells.



**Table 1** Monitoring well specifications (distances are approximate)

Well ID	Surface elevation (m AMSL)	Depth (m)	Intake length (m)	Location/description
UG-1	189.91	5.18	1.52	3 m southeast of the PA
DG-1	188.12	5.18	1.52	1 m north of the PA
DG-2	188.18	5.18	1.52	3 m northeast of PA
DG-3	184.51	2.44	1.52	North of Glenbrook Rd, easternmost location
DG-4	186.96	3.96	0.91	North of Glenbrook Rd, westernmost location
PZ-S	184.95	2.13	0.91	North of Glenbrook Rd, directly downgradient from
PZ-M		3.05	0.30	the PA, cluster of 3 piezometers installed to discrete depths
PZ-D		3.96	0.30	
WB-1	188.88	6.71	3.05	100 m south of PA, not pictured in Fig. 2.

PA: permeable asphalt

(not shown in Fig. 2) is a shallow monitoring well located about 100 m south of the permeable pavement. It was sampled to obtain a background water quality measure, distant from areas being salted on campus.

## 2.2 Monitoring

Long-term monitoring for salt contamination of the site began after well installation and development. Each well was equipped with an Instrumentation Northwest, Inc. (INW) Aquistar® CT2X pressure transducer/specific conductance sensor and data logger. Before installation, the conductance probes were calibrated to a reference solution of 2000  $\mu\text{S}/\text{cm}$ . Pressure transducers were calibrated in the well to manual water level measurements taken with a Solinst® water level sounder. The sensors were programmed to record relative water level, conductance, and temperature on a 10-min interval. Data records were downloaded from each sensor on a monthly to bi-weekly basis.

## 2.3 Sampling and Analysis

Water samples were collected by low-flow sampling on four dates between November 16 and December 18 2015. Stagnant water was removed from the well

casing using a peristaltic pump at rates under 120 mL/min. Samples were analyzed for chloride ( $\text{Cl}^-$ ) with an ion-specific electrode (ISE) using EPA method 9212 (USEPA, 1991). Additionally, the specific conductance of each sample was measured using the INW sensor in the well from which it was obtained.

Between the 1st and 3rd of March 2016, a groundwater sample was collected from each well by low flow sampling for analyses of major dissolved cations ( $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ),  $\text{Cl}^-$ , and radionuclides ( $^{222}\text{Rn}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$ ). As some of the target analytes are redox-sensitive or volatile, care was taken to monitor water quality parameters such as dissolved  $\text{O}_2$ , redox potential, and specific conductance while purging. After these parameters stabilized, 3.79 L of water was collected for Ra analysis, 500 mL was collected for cation analysis, 250 mL was collected for  $\text{Cl}^-$  analysis, and a 40-mL volatile organic analysis (VOA) vial was filled for Rn analysis. Metals were analyzed at the University Center for Environmental Science and Engineering (CESE) laboratory by ICP-OES using EPA method 200.7 (EPA 2001). Rn and Ra were analyzed at the Connecticut Department of Public Health Laboratory using EPA methods 913.0 and 903.0/904.0, respectively.

## 2.4 Equilibrium Vapor-Phase Rn Calculations

The equilibrium vapor-phase Rn concentrations were estimated using the method described in

**Table 2** Empirical parameters for Rn application of Weiss equation, derived in Schubert et al. (2012)

$a_1$	-76.14	$b_1$	-0.2631
$a_2$	120.36	$b_2$	0.1673
$a_3$	31.26	$b_3$	-0.0270

**Table 3** Groundwater data from samples collected 1–3 March 2016 (units of depth to water (DTW) and water table elevation (WTE) are in meters, specific conductance (SC) values are microseimens per centimeter, units of common ions are milligrams per liter, and units of radionuclides are picocuries per liter)

Well ID	DTW	WTE	SC	Na <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Cl <sup>-</sup>	<sup>222</sup> Rn	<sup>226</sup> Ra	<sup>228</sup> Ra	Ra <sub>tot</sub>
UG-1	3.52	186.39	6520	1424.0	372.3	55.7	3944.5	417.0	5.30	3.48	8.78
DG-1	2.91	185.21	18,300	4936.0	183.8	34.9	10899.0	242.0	8.66	10.60	19.26
DG-2	2.93	185.25	719	65.3	78.4	20.5	212.1	610.0	<1.00	<1.00	<2.00
DG-3	0.73	183.78	3787	450.6	397.0	117.7	2422.0	907.0	1.13	<1.00	1.13
DG-4	3.51	183.45	3040	402.4	65.2	11.9	1825.3	243.0 <sup>a</sup>	2.03	<1.00	2.03
PZ-S	1.16	183.79	5520	1302.0	289.7	64.5	3417.0	470.0	1.66	<1.00	1.66
PZ-D	1.14	183.81	3400	425.9	397.5	105.6	1946.0	979.0	1.72	1.42	3.14
WB-1	2.91	185.99	390	36.8	29.3	6.0	80.0	498.0	<1.00	<1.00	<2.00

<sup>a</sup> Dubious result due to gas loss by aeration during sampling

Schubert et al. (2012). This process uses the Weiss equation (Eq. 1)

$$\ln\beta = a_1 + a_2\left(\frac{100}{T}\right) + a_3\ln\left(\frac{T}{100}\right) + S\left\{b_1 + b_2\left(\frac{T}{100}\right) + b_3\left(\frac{T}{100}\right)^2\right\} \quad (1)$$

where *S* is the salinity of the aqueous solution (g/L), *T* is the temperature of the solution (K),

**Table 4** Predicted gaseous Rn concentrations based on aqueous Rn concentrations, temperature, and salinity

Location	Rn <sub>AQ</sub> (pCi/L)	Salinity (g/L) <sup>a</sup>	Temp (°C)	<i>K<sub>H</sub></i> (dimensionless)	Rn <sub>G</sub> (pCi/L)
UG-1	417	5.37	11.96	0.313	1330
DG-1	242	15.84	21.60	0.221	1100
DG-2	610	0.28	22.14	0.233	2610
DG-3	907	2.87	10.57	0.333	2720
DG-4	243 <sup>b</sup>	2.23	13.38	0.304	800
PZ-S	470	4.72	10.48	0.331	1420
PZ-D	979	2.37	10.48	0.336	2920
WB-1	498	0.12	14.90	0.292	1706

*K<sub>H</sub>* = dimensionless Henry’s Law constant values, calculated from Schubert et al. 2012, Eq. 3a

<sup>a</sup> Grams Na<sup>+</sup> grams Cl<sup>-</sup>

<sup>b</sup> Radon gas loss likely during sampling due to limited amount of water in the well

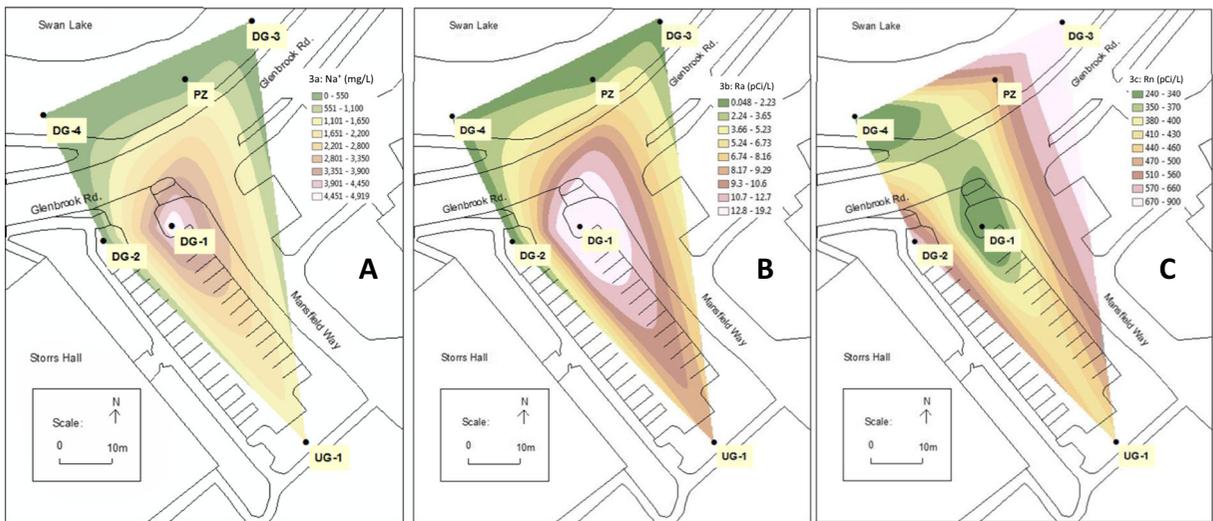
*a*<sub>1</sub>, *a*<sub>2</sub>, *a*<sub>3</sub>, *b*<sub>1</sub>, *b*<sub>2</sub>, and *b*<sub>3</sub> are experimentally determined coefficients derived in Schubert et al. (2012) and listed in Table 2 for reference. The β term is related to the Henry’s law coefficient *K<sub>H</sub>* as

$$K_H = \beta \frac{T}{273.15} \quad (2)$$

These two equations can be used to calculate the *K<sub>H</sub>* of a gas based on the temperature and salinity of the liquid solution.

### 3 Results

Water table elevations (WTE) and groundwater quality results from March 1 to March 3, 2016 are summarized in Table 3. The temperatures of the samples are listed in Table 4. Na<sup>+</sup>, Ra, and Rn data were used to generate concentration contour maps of the site (Fig. 3a–c). The highest Na<sup>+</sup> concentrations measured were found directly downgradient of the parking lot (Fig. 3a), indicating that high levels of salt reach the groundwater at the permeable pavement, then travel downgradient along the advective flow path shown in Fig. 2. Correspondingly, high concentrations of Ra were found in locations with high Na<sup>+</sup> concentrations (Fig. 3a, b). The significance of the correlation between the distributions of these elements was tested using ANOVA (IBM 2012). A statistically significant (*p* < 0.001), positive linear correlation (*R*<sup>2</sup> = 0.92) between Ra and Na<sup>+</sup> concentrations was found (Fig. 4). Ra was expressed as a

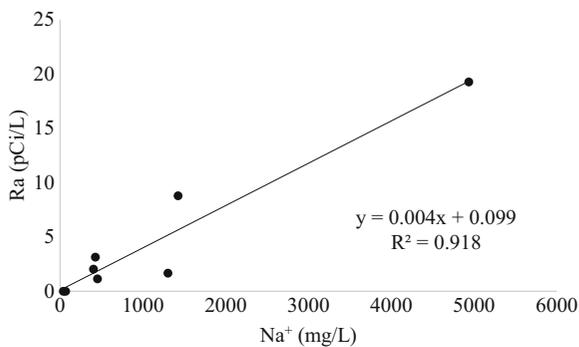


**Fig. 3** Spatial distributions of **a**  $\text{Na}^+$ , **b** combined  $^{226}\text{Ra} + ^{228}\text{Ra}$ , and **c**  $^{222}\text{Rn}$  (Concentration contours were generated in ArcMap using a natural neighbor interpolation algorithm)

combination of its most common isotopes ( $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ) to be consistent with regulatory units. The spatial distribution of Rn at this site (Fig. 3c) is characterized by a low point in the most saline well (DG-1) and relatively higher activities in surrounding, less saline wells. Low concentrations of Rn were also observed in well DG-4; this was likely due to gas loss during sampling. The Rn/specific conductance relationship ( $R^2 = 0.54$ ) was less statistically significant ( $p = 0.059$ ), but negative (Fig. 5), which was different from the highly significant positive relationship found for  $\text{Ra}/\text{Na}^+$ .

**4 Discussion**

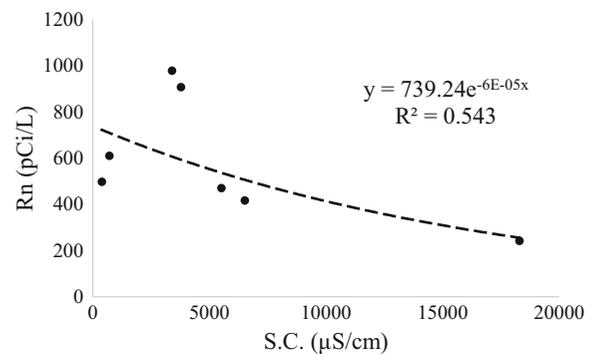
The correlation between Ra and  $\text{Na}^+$  is due to ion exchange at soil particle surfaces. To demonstrate that



**Fig. 4** Positive correlation between Ra activity and  $\text{Na}^+$  concentration for groundwater samples

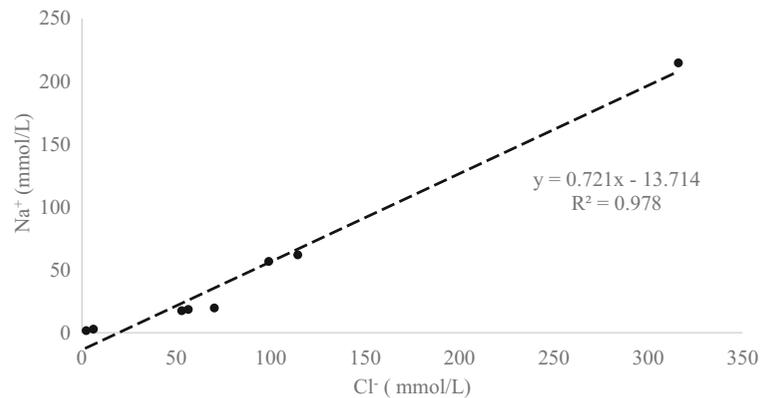
active cation exchange is taking place, Fig. 6 was plotted. While there is a strong correlation between moles of salt-derived  $\text{Na}^+$  and  $\text{Cl}^-$ , the slope is less than one, indicating a deficit of  $\text{Na}^+$ . The occurrence of cation exchange is further supported by relatively high levels of dissolved  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  at the permeable pavement study area compared to those of the background well WB-1 (Table 3).

The high concentrations of Ra in some of the water samples were unexpected; two of the wells had Ra activities above the EPA drinking water standard of 5 pCi/L (UG-1 8.78 pCi/L; DG-1 19.26 pCi/L) (EPA 1986). Given the rare occurrence of Ra in groundwater (Hem 1985), the strong relationship found between Ra and  $\text{Na}^+$  at this site indicates that groundwater contaminated with deicing salt can release Ra from soil at levels



**Fig. 5** Inverse relation between Rn activity and specific conductance for groundwater samples as predicted by the Weiss equation (Eq. 1 in this paper)

**Fig. 6** Demonstration of  $\text{Na}^+$  depletion relative to  $\text{Cl}^-$  in groundwater samples



nearly four times the regulatory limit. The high Ra concentration in the upgradient well (UG-1) corresponded to a relatively high  $\text{Na}^+$  concentration. High  $\text{Na}^+$  and  $\text{Cl}^-$  concentrations were found upgradient of the PA lot (Dietz et al. 2016); the suspected cause of this was the cumulative saline runoff from deicing sidewalks in the vicinity of the lot. It should be noted that only during the winter months is the  $\text{Na}^+$  concentration higher in the downgradient wells as compared to the upgradient well; during the rest of the year, precipitation percolating through the pervious lot dilutes high background  $\text{Na}^+$  and  $\text{Cl}^-$  concentrations found in the area (Dietz et al. 2016).

As noted above, a weak inverse correlation was observed between salinity and Rn in the groundwater (Fig. 5). With all other factors held constant, the abundance of Rn in the aqueous phase would be expected to decrease logarithmically as a function of salinity, according to Eq. 1. While this result somewhat supports the “salting out” phenomena quantified by Schubert et al. (2012), other physical factors (e.g., proximity of the well screen to the water table surface, temperature, permeability, and moisture of unsaturated overburden soil) can have significant influence on the observed concentrations of Rn (Ball et al. 1991). Table 4 summarizes calculations of gaseous Rn concentrations that would be in equilibrium (i.e., in the vadose zone directly above the water table) with the observed aqueous Rn concentrations. The  $K_H$  values were adjusted for salinity according to the model developed by Schubert et al. (2012), using groundwater temperatures measured during the collection of aqueous samples. In every case, the equilibrium Rn gas concentration far exceeds the 4 pCi/L EPA indoor air quality standard (EPA 1986). Additionally, wells DG-1 and DG-2 are in very close

proximity to a steam line; this common feature of urban subsurface doubles the groundwater temperature. These higher temperatures substantially decrease the  $K_H$  of Rn, further enhancing Rn volatilization to the vadose zone.

Rn was found in the background water sample from well WB-1. However, Ra, its parent isotope, was not detected. The water also had low  $\text{Na}^+$  concentration. Given that the  $K_D$  of Ra is typically very high in non-saline conditions, the Ra source must be in the solid phases of either the till or the underlying bedrock.

## 5 Conclusion

This study has shown that deicing salt contamination of groundwater can serve to mobilize Ra and Rn in the subsurface. Although our study focused on an area of permeable pavement, the results would be equally applicable to any salted location where there is a high infiltration rate to groundwater, such as an urban riparian floodplain (Ledford et al. 2016). The Henry’s law calculations suggest that buildings adjacent to or downgradient from (a) highly permeable locations that are deiced with salt or (b) locations with elevated subsurface temperature could be periodically subjected to high Rn diffusive fluxes. The Ra data suggest that wells contaminated with high levels of deicing salt could also be contaminated with Ra at levels four times the regulatory standard. The degree of increased radionuclide exposure would depend on a number of key site-specific factors including direction of groundwater flow, location of dwellings, water temperature, depth to groundwater, and soil properties influencing gas migration. The potential public health implications of these findings is strong justification for further study of this phenomenon

to determine (1) if bedrock wells show a similar Ra/Na<sup>+</sup> relationship and (2) the potential geographic extent that might be affected.

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### Compliance with Ethical Standards

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