

## Investigation of Bedrock Well Contamination by Uranium, Radium and Radon Resulting from Deicing Salt Exchange

<b>Title:</b>	Investigation of Bedrock Well Contamination by Uranium, Radium and Radon Resulting from Deicing Salt Exchange
<b>Project Number:</b>	2017CT304B
<b>Start date:</b>	3/1/2017
<b>End Date:</b>	2/28/2018
<b>Funding Source</b>	104B
<b>Congressional District:</b>	CT-002
<b>Research Category:</b>	Ground-water Flow and Transport
<b>Focus Category:</b>	Groundwater, Radioactive Substances, Water Quality
<b>Descriptors:</b>	None
<b>Principal Investigators:</b>	Michael Dietz, Meredith J. Metcalf, Gary A. Robbins

**Investigation of Bedrock Well Contamination by Uranium, Radium and Radon Resulting  
from Deicing Salt Exchange**

**FINAL REPORT**

**to**

**Connecticut Institute of Water Resources**

**United States Geological Survey**

**Award# G16AP00043**

**May 20, 2019**

**Michael Dietz, UConn Extension (PI)**

**Gary Robbins, UConn NRE (Co-PI)**

**Meredith Metcalf, ECSU (Co-PI)**

**Lukas McNaboe, UConn NRE (Ph.D. student)**

## **Introduction/Research Objective**

Each year, an increasing amount of salt is being used in the U.S. as a deicing agent (Mullaney et al. 2009, Fay and Shi 2012). Researchers have observed a gradual salinization of both surface water (Kaushal et al. 2005; Kaushal 2016) and groundwater systems due to continued deicing practices (Kelly et al. 2008; Novotny et al. 2009; Perera et al. 2013; Trowbridge et al. 2010). In Connecticut, chloride concentrations in groundwater have risen on average over 10 times background levels in the last 100 years (Cassanelli and Robbins, 2013). In some areas of the state, chloride concentrations have exceeded 100 times the background levels. Individual wells have been found to have salinity levels greater than that of sea water.

Radium concentrations in groundwater have been highly correlated with sodium chloride 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 sodium ions (Krishnaswami et al. 1982; Sanders et al. 2013; Tamamura et al. 2014). Radium solubility can also be enhanced by the formation of  $\text{RaCl}^+$  complexes in saline waters (Langmuir and Riese, 1985). Although there are many factors that influence the solubility of radon in aqueous solutions (e.g., temperature, pressure), the observed naturally occurring positive correlation between radium and salt in saline aquifers suggests that radium, and its progeny radon, could be mobilized by deicing salt contamination of groundwater. This represents a public health concern due to the carcinogenic nature of these elements: ingestion of radium can produce harmful health effects such as cataracts and osteosarcoma (CDC, 2015) and radon exposure has been identified as the second-leading cause of lung cancer in the US (Darby et al. 2001).

The metamorphic and igneous bedrocks of Connecticut are known to contain significant levels of uranium and radium which can cause high fluxes of radon under natural conditions (Thomas and McHone 1997). These geologic conditions, along with our findings to date (see related research below), suggest that there is a significant potential for bedrock wells contaminated with high levels of deicing salt in Connecticut to also be contaminated with uranium, radium, and radon at concentrations of concern. In addition, a salinity increase in the shallow groundwater could result in a larger flux of radon to overlying buildings; this region is particularly susceptible due to the shallow nature of its groundwater (Krewski et al., 2005; Thomas and McHone 1997).

This project was a field experiment designed to evaluate whether salt contamination of fractured bedrock has caused elevated levels of uranium, radium and radon in wells. The broader goal of the project was to further the understanding of the risks associated with de-icing salt contamination in the state's supply of subsurface drinking water.

## **Procedures/Progress**

The project began with a statewide search for wells impacted by salt contamination. First, the Connecticut Department of Public Health (DPH) was contacted for a list of wells with known salt contamination. This list contained sites impacted by de-icing salts, and possibly by saltwater intrusion in some coastal locations. Sites with concentrations of sodium ( $\text{Na}^+$ ) and chloride ( $\text{Cl}^-$ ) below 250 mg/L were excluded from the investigation as they were well below 1000 mg/L. This value is the salinity at which detectable radionuclide mobilization is expected to occur (McNaboe et al. 2017, Tamamura et al. 2014). A total of 26 potential sites were obtained from the DPH database, 9 of which were within a 1-mile radius in Brookfield, CT. These sites are enumerated in table 1. As the database did not include coordinates, addresses, nor phone numbers, these sites needed to be reverse-georeferenced based on the water system name. Of these sites, all but one declined to participate in the study for reasons that include: there is no access to well, they already have undergone rigorous testing, or a presumed lack of interest due to a lack of correspondence.

Additional sites were identified by distributing a flyer seeking volunteers for well testing; the flyer is attached to this report. The flyer was sent to town sanitarians and municipalities, with the intent that they inform their citizens of the program. There were 2 responses to this flyer, which are also listed in table 1; one of these was amenable to sampling. The other respondent referred us to a neighborhood with saline groundwater, but the property owners were not interested in participating. The CT Department of Transportation (DOT) was also contacted to sample any saline wells they were aware of, as salt storage facilities are particularly susceptible to being sources of groundwater salt contamination (Dennis 1973). They informed of us at least two sites (table 1), but did not permit any sampling activities. The CT Department of Energy and Environmental Protection (DEEP) was contacted to sample any salt-impacted wells they were aware of; DEEP officials informed us that they weren't aware of any wells that met our investigation's criteria of Na<sup>+</sup> or Cl<sup>-</sup> concentrations above 1000 mg/L.

We travelled to the locations to collect well samples by low flow sampling, according to the procedure attached in this document. For each site, we delivered 3 bottles to the CT DPH laboratory for radionuclide analysis (Rn, Ra, U), and delivered 1 bottle to UConn Center for Environmental Science and Engineering (CESE) laboratory for cation analysis (Na<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>). An additional bottle was delivered to the UConn Natural Resources (NRE) water quality laboratory for Cl<sup>-</sup> analysis.

Due to the low response rate of our statewide inquiry, the scope of sampling was expanded to include shallow monitoring wells that penetrate only to the top of the water table. Five such wells were identified around the UConn campus in Storrs, CT (Figure 3); samples were collected from these wells by low-flow sampling during February 2018, as this period is historically the annual maximum for groundwater salinity in this area (Cassanelli 2011, McNaboe 2017).

Table 1: List of prospective sampling locations. Locations in bold were sampled during this study.

Map index	Location	Type	Wells	Recent Na <sup>+</sup> (mg/L)	Recent Cl <sup>-</sup> (mg/L)	Bedrock	Surficial
0	Old Lyme, DEEP HQ	Municipal	1	568	1580	Granitic gneiss	Till
1	70 Merrow Road, Tolland CT	Commercial	1	110	601	Gneiss	Till
2	Greenwich, Fairview Country Club	Commercial	1	347	656	Gneiss	Till
3	Brookfield commons area	Comm, res, sch.	15	399	1005	Marble	Fines
4	East Hampton town hall	Municipal	2	168	651	Gneiss	Till
5	North Stonington Congregational Church	Church	1	316	--	Gneiss	Sand/Gravel
6	Franklin Elementary School	School (public)	1	280	--	Schist	Gravel
7	Dayville, Crystal Water Co.	Commercial	?	750	--	Granitic gneiss	Sand/Gravel
8	Brookfield, Down the Hatch Restaurant	Commercial	1	261	231	Granitic gneiss	Till
9	Knollbrook Rd. Bethany, CT	Residential	4	370	550	Gneiss	Till
10	Tony's Drilling, Montville CT	Industrial	1	--	3247	Quartzite	Till
<b>11</b>	<b>Private well, South Windsor, CT</b>	Residential	1	182	440	Arkose	Sand/Gravel
<b>12</b>	<b>New Canaan CT, St. Luke's School</b>	School (private)	1	312	908	Schist	Till
13	Colebrook school	School (public)	1	211	836	Gneiss/Schist	Till
14	Eastford; Whitcraft corp.	Industrial	2	58	626	Schist	Till
15	DOT Garage, New Milford, CT	Government	10	148	340	Marble	Alluvial/Fines
<b>16</b>	<b>Storrs, CT**</b>	School	5	--	1627	Gneiss	Till
17	DOT Garage, Ashford, CT	Government	1	--	--	Granofels	Till

\*Cells with a hyphen (--) represent that no sample was analyzed for that respective constituent.

\*\* Shallow, overburden wells were sampled at this location

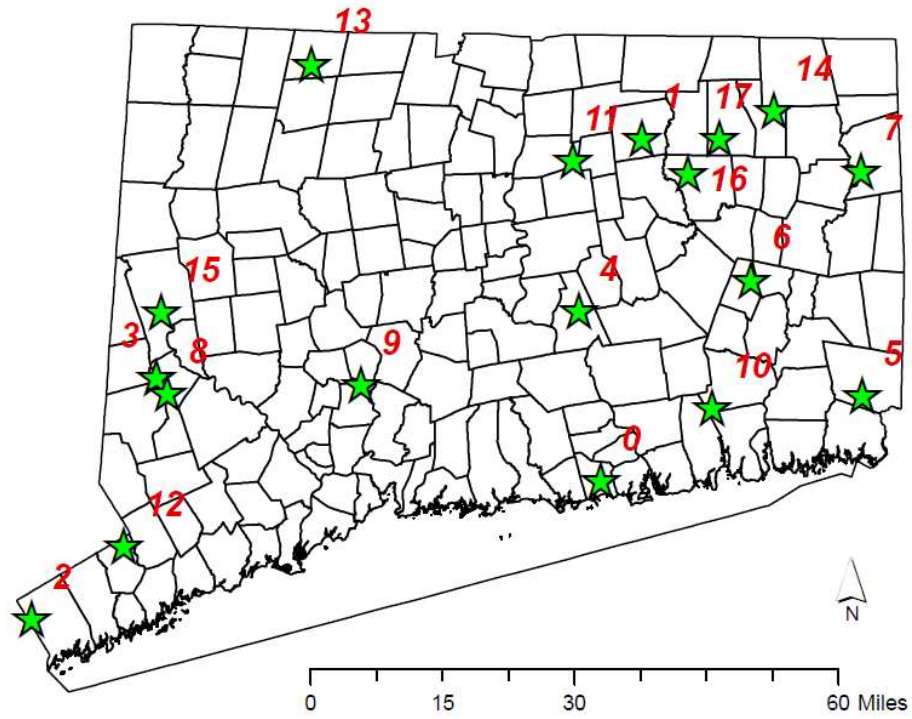


Figure 1: Prospective sites that were identified for groundwater sampling. Indices on this figure correspond to those on table 1.

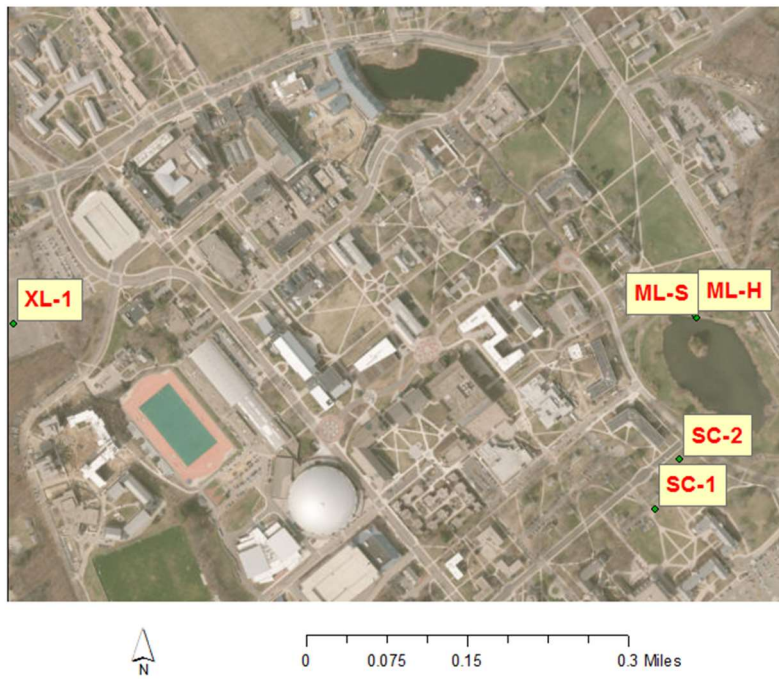


Figure 2: UConn sites that were sampled during this study.

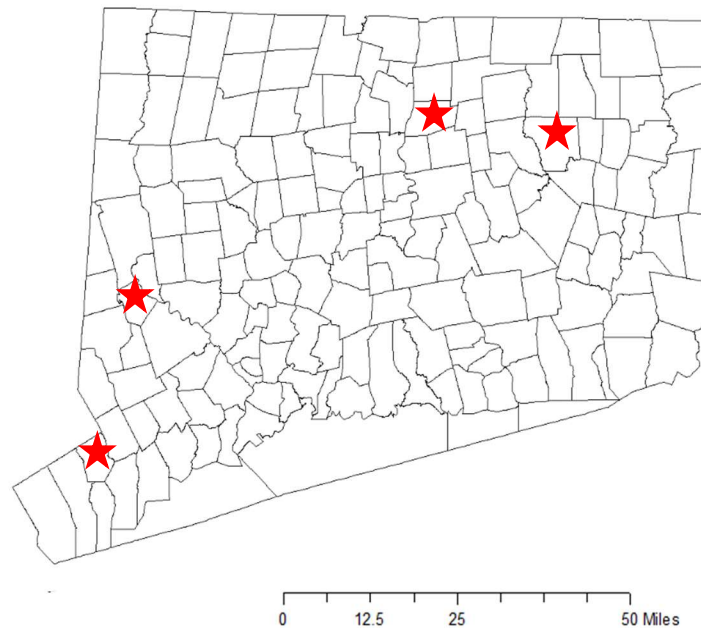


Figure 3. Locations of actual sampling sites.

### Results/Significance

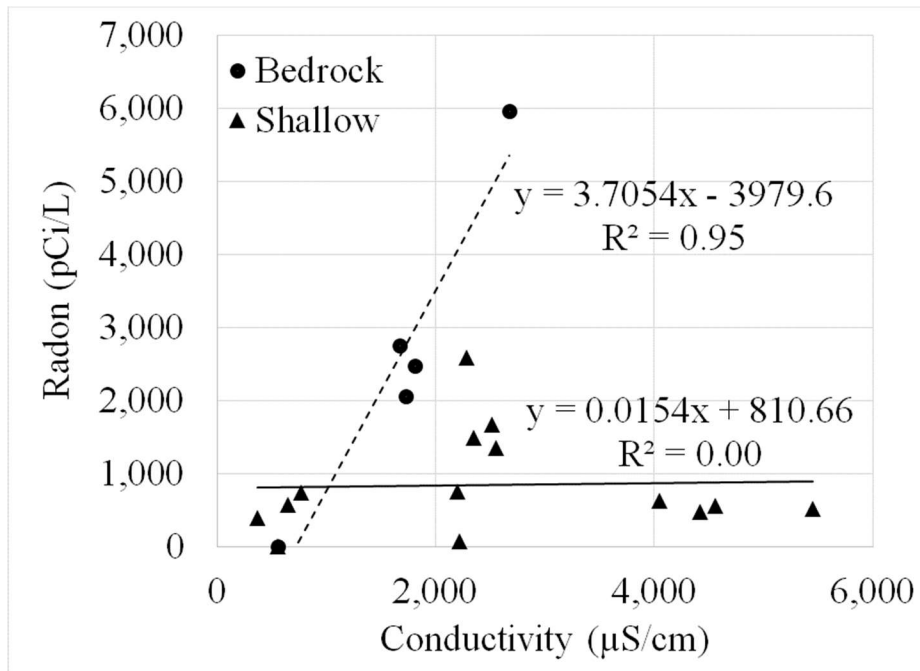
Due to a laboratory error, no analysis of Ca, Na, Mg or Ca was performed for samples from the Brookfield sites and several of the UConn sites (-SOB, -GB5, -BHK, -BHF). However, all samples were analyzed for EC and Rn (Table 2). Regression analysis indicated a significant positive relationship between EC and Rn ( $R^2=0.95$ ) for the bedrock wells (Figure 4), however the regression only contained five data points. Only one sample had a concentration of Rn above the 4,000 pCi/L action level. Recent analysis of shallow wells in Storrs, CT indicated a weak but statistically significant *negative* relationship between conductivity and Rn (McNaboe et al., 2017). Shallow groundwater has more potential to lose Rn in gaseous form to the atmosphere. Water drawn from deep bedrock wells would likely have higher dissolved radon, because it has cannot readily escape in gaseous form. No discernible relationship was evident for the samples from shallow groundwater sites in the current study (Figure 4).

Radium concentrations in the bedrock wells sampled were generally low, with only one site having a detectable concentration (Table 2). Uranium was detected in all of the bedrock wells sampled, but all samples were below the 30  $\mu\text{g/L}$  drinking water standard. A significant positive relationship was found between conductivity and U (Figure 5). Uranium concentrations were generally low or not detected (61%) in shallow groundwater samples (Table 2).

The concentration of radium in all of the shallow groundwater samples taken from the UConn campus was above the drinking water standard. This does not pose a direct health concern, because the shallow groundwater in this area is not currently being used as a drinking water source. A weak negative relationship between Na and Ra was found (Figure 6).

**Table 2. Concentrations of salts and radionuclides for samples taken in this study. Samples above the dashed line were collected from bedrock wells; samples below the dashed line were collected from shallow overburden wells or curtain drains (Brookfield-1). ND=non-detect, dashes=no result available.**

Well location	EC ( $\mu\text{S}/\text{cm}$ )	Cl (mg/L)	Na (mg/L)	Mg (mg/L)	Ca (mg/L)	Rn (pCi/L)	Ra (pCi/L)	U ( $\mu\text{g}/\text{L}$ )
New Canaan	1730	397	42.7	57.6	189.8	2070	ND	17.0
South Windsor	558	164	65.0	4.2	31.7	5	<3	0.5
Brookfield-2	1670	--	--	--	--	2760	--	22.0
Brookfield-3	2670	--	--	--	--	5970	--	20.0
Brookfield-4	1810	--	--	--	--	2480	--	4.7
Brookfield-1	2280	--	--	--	--	2590	--	3.6
Storrs -- SC-1	4050	1520	720.6	18.0	115.8	628	63	<1
Storrs -- SC-2	371	52	17.9	6.4	38.0	390	52	<1
Storrs -- XL-1	4420	1630	983.5	11.1	28.7	486	33	<1
Storrs -- ML-H	2200	795	312.2	14.3	79.0	764	80	<1
Storrs-- ML-S	2350	767	238.3	20.6	129.1	1490	117	<1
Storrs -- ML-O	2220	788	357	11.8	65.8	83	26	1.8
Storrs -- SOB	768	--	--	--	--	748	--	1.1
Storrs -- GB5	4560	--	--	--	--	565	--	<1
Storrs -- BHK	646	--	--	--	--	573	--	<1
Storrs -- BHF	5450	--	--	--	--	524	--	<1
BRK-5a	2520	--	--	--	--	1680	--	2.3
BRK-5b	2550	--	--	--	--	1360	--	1.7



**Figure 4. Electrical conductivity vs. radon in bedrock (circles) and shallow (triangles) wells.**

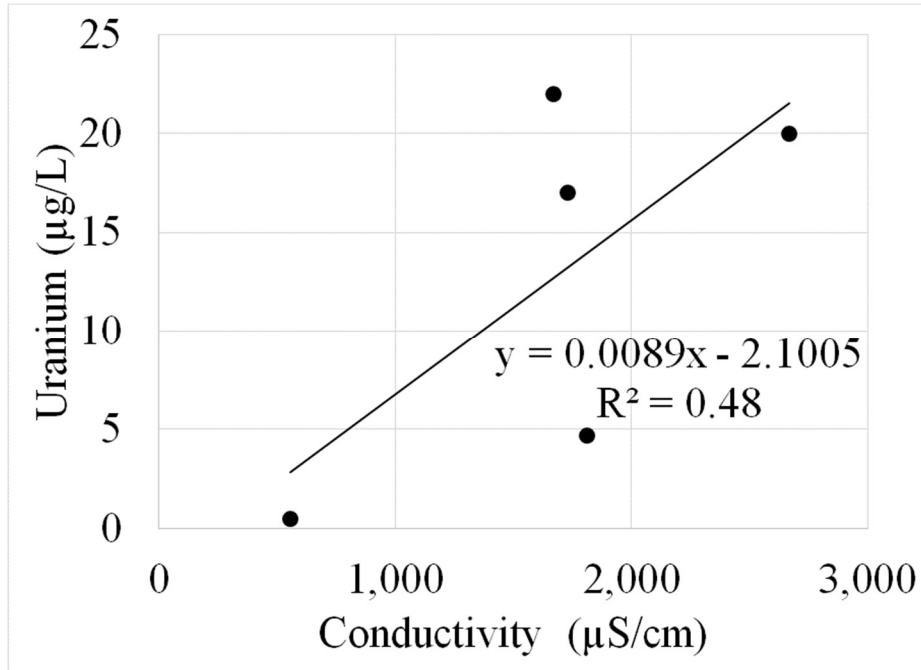


Figure 5. Electrical conductivity vs. uranium in bedrock wells, statewide in CT.

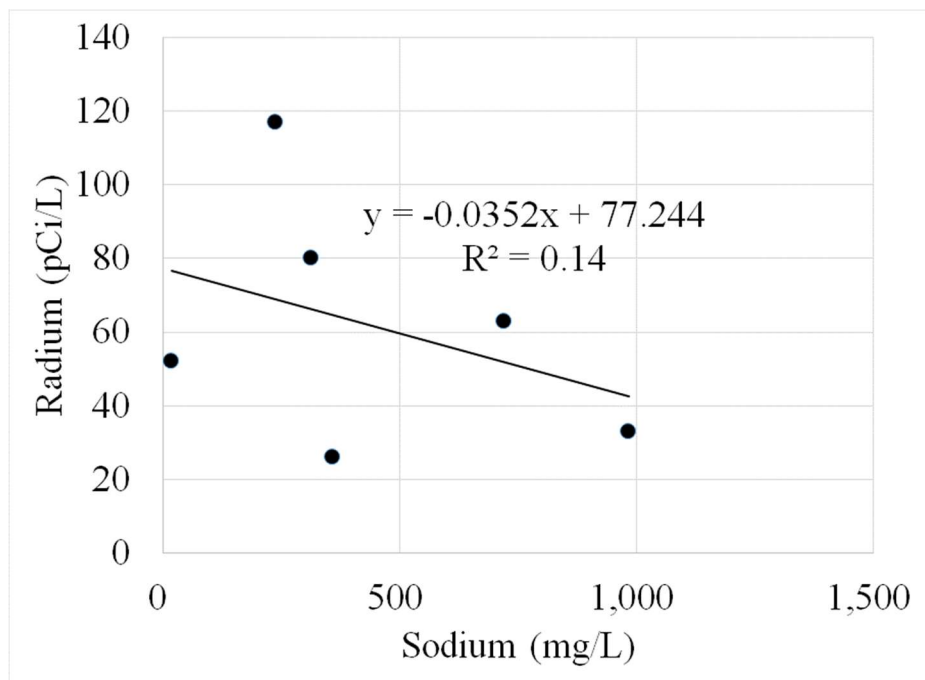


Figure 6. Sodium vs. radium in shallow groundwater wells on the UConn campus.



## Upcoming work

Unfortunately we were only able to test a small number of bedrock wells in this study; only five wells had a sodium concentration high enough to release radium and/or radon. We are routinely learning about new sites with groundwater that has been contaminated by road salt, and we intend to sample these wells for radionuclides as resources become available. There will likely be opportunities to further investigate the relationship between high salt in groundwater and radionuclides.

## Literature Cited

Cassanelli, J.P. (2011) Salt contamination of ground and surface water in Connecticut: a compilation and synthesis of historic data and local scale testing. Master's Theses, Univ. of Connecticut. 69.

Cassanelli, J., and Robbins, G.A. (2013). Road Salt Impact to Connecticut's Groundwater: A Statewide Centennial Perspective, *Journal of Environmental Quality*, 42, 737-748.

Cooper, C.A., Mayer, P.M., Faulkner, B.R. (2014) Effects of road salts on groundwater and surface water dynamics of sodium and chloride in an urban restored stream. *Biogeochemistry*, 121, 149-166.

Corsi, S.R., De Cicco, L.A., Lutz, M.A., Hirsch, R.M. (2015) River chloride trends in snow-affected urban watersheds: increasing concentrations outpace urban growth rate and are common among all seasons. *Science of the Total Environment*, 508, 488-497.

Darby, S., Hill, D., Doll, R. (2001) Rn: a likely carcinogen at all exposures. *Annals of oncology*, 12(10), 1341-1351.

Fay, L., Shi, X. (2012) Environmental impacts of chemicals for snow and ice control: state of the knowledge. *Water Air Soil Pollution*, 223, 2751-2770.

Kaushal, S.S., Groffman, P.M., Likens, G.E., Belt, K.T., Stack, W.P., Kelly, V.R., Band, L.E., Fisher, G.T. (2005). Increased salinization of fresh water in the northeastern United States. *Proceedings of the National Academy of Sciences*, 102(38), 13517-13520.

Kaushal, S.S. (2016). Increased salinization decreases safe drinking water. *Environ. Sci. Technol.*, 50, 2765-2766.

Kelly, V.R., Lovett, G.M., Weathers, K.C., Findlay, S.E.G., Strayer, D.L., Burns, D.J., Likens, G.E. (2008). Long-term sodium chloride retention in a rural watershed: Legacy effects of road salt on streamwater concentration. *Environ. Sci. Technol.*, 42, 410-415.

Krewski, D., Lubin, J.H., Zielinski, M., Alavanja, V.S., Field, R.W. (2005). Residential Radon and Risk of Lung Cancer: A Combined Analysis of 7 North-American Case-control Studies. *Epidemiology*, 16(2), 137-145.

Krishnaswami, S., Graustein, W.C., Turekian, K.K. (1982). Radium, thorium and radioactive lead isotopes in groundwaters: application to the in situ determination of adsorption-desorption rate constants and retardation factors. *Wat. Res. Research*, 18(6), 1663-1675.

Langmuir, D., Riese, A.C. (1985). The thermodynamic properties of radium. *Geochim. et Cosmochim. Acta*, 49(7), 1593-1601.

- McNaboe, L.A. (2017). Impacts of de-icing salt contamination of groundwater in a shallow urban aquifer. Master's Theses, Univ. of Connecticut. 1138.
- McNaboe, L.A., Robbins, G.A., and Dietz, M. E., (2017). Mobilization of Radium and Radon by Deicing Salt Contamination of Groundwater. *Water, Air & Soil Pollution*, 228(94).
- Mullaney, J.R., Lorenz, D.L, Arnston, A.D. Chloride in groundwater and surface water in areas underlain by the glacial aquifer system, northern United States. U.S. Geological Survey Scientific Investigations Report 2009-5086. U.S. Geological Survey: Reston, VA, 2009.
- Novotny, E.V., Sander, A.R., Mohseni, O., Stefan, H.G. (2009). Chloride ion transport and mass balance in a metropolitan area using road salt. *Water Resour. Res.*, 45, W12410.
- Perera, N., Gharabaghi, B., Howard, K. (2013). Groundwater chloride response in the Highland Creek watershed due to road salt application: A re-assessment after 20 years. *Journal of Hydrology*, 479, 159-168.
- CDC. (2015). *Public health statement for Ra*. Agency for Toxic Substances and Disease Registry. Center for Disease Control. Atlanta, GA, 2015.
- Sanders, L.M., Luiz-Silva, W., Machado, W. Sanders, C.J., Marotta, H., Enrich-Prast, A., Bosco-Santos, A., Boden, A., Silva-Filho, E.V., Santos, I.R., Patchineclam, S.R. (2013). Rare earth element and radionuclide distribution in surface sediments along an estuarine system affected by fertilizer industry contamination. *Water Air Soil Pollution*, 224, 1742.
- Schubert, M., Paschke, A., Lieberman, E., Burnett, W.C. (2012) Air-water partitioning of  $^{222}\text{Rn}$  and its dependence on water temperature and salinity. *Environ. Sci. Technol.*, 46, 3905-3911.
- Sturchio, N.C., Banner, J.L., Binz, C.M., Heraty, L.B., Musgrove, M. (2001). Radium geochemistry of ground waters in Paleozoic carbonate aquifers, midcontinent, USA. *Appl. Geochem.*, 16(1), 109-122.
- Tamamura, S., Takahiro, T., Tomita, J., Nagao, S., Fukushi, K., Yamamoto, M. (2014). Salinity dependence of  $^{226}\text{Ra}$  adsorption on montmorillonite and kaolinite. *J. Radioanal. Nucl. Chem.*, 299(1), 569-575.
- Thomas, M.A., McHone, N.W. (1997). Indoor radon potential map of Connecticut. Connecticut Geological and Natural History Survey, Connecticut Department of Environmental Protection.
- Trowbridge, S.T., Kahl, J.S., Sassan, D.A., Heath, D.L, Walsh, E.M. (2010). Relating road salt exceedances of the water quality standard for chloride in New Hampshire streams. *Environ. Sci. Technol.*, 44, 4903-4909.
- US EPA. (1996). *Test Methods for Evaluating Solid Wastes Physical/Chemical Methods*. EPA SW-846 Third Edition, United States Environmental Protection Agency Office of Solid Waste: Economic, Methods, and Risk Analysis Division. Washington, D.C.
- Vinson, D.S., Vengosh, A., Hirschfield, D., Dwyer, G.S. (2009). Relationships between radium and radon occurrence and hydrochemistry in fresh groundwater from fractured crystalline rocks, North Carolina (USA). *Chemical Geology*, 260, 159-171.
- Weiss, R.F. (1970) The solubility of nitrogen, oxygen and argon in water and seawater. *Deep-Sea Research*, 17(4), 721-735.

*Graduate students involved in this project*

Name: Lukas McNaboe

Department: UConn department of Natural Resources and the Environment

Degree: Ph.D.

Expected graduation date:

Name: Luke Davis

Department: ECSU department of Environmental and Earth Sciences

Degree: B.S.

Expected graduation date: May 2018