Oil and gas wastewater as road treatment: radioactive material exposure implications at the residential lot and block scale

Daniel J Bain¹*, Tetiana Cantlay²,³, Brittany Garman² and John F Stolz²,³

¹ Department of Geology and Environmental Science, University of Pittsburgh, Pittsburgh, PA, United States of America
² Center for Environmental Research and Education, Duquesne University, Pittsburgh, PA, United States of America
³ Department of Biological Sciences, Duquesne University, Pittsburgh, PA, United States of America
* Author to whom any correspondence should be addressed.

E-mail: dbain@pitt.edu

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Abstract

The resurgence of oil and gas extraction in the Appalachian Basin has resulted in an excess of oil and gas brines in Pennsylvania, West Virginia, and Ohio. Primarily driven by unconventional development, this expansion has also impacted conventional wells and consequently, created economic pressure to develop effective and cheap disposal options. Using brine as a road treatment, directly or as a processed deicer, however, creates substantial concern that naturally occurring radioactive material in the brines can contaminate roads and road-side areas. Current decision making is based on risk exposure scenarios developed by regulatory agencies based on recreational users in rural areas and exposures to drivers during a typical commute. These scenarios are not appropriate for evaluating exposures to residential deicer users or people living near treated streets. More appropriate exposure scenarios were developed in this work and exposures predicted with these models based on laboratory measurements and literature data. Exposure scenarios currently used for regulatory assessment of brine road treatment result in predicted exposures of 0.4–0.6 mrem/year. Residential exposures predicted by the scenarios developed in this work are 4.6 mrem/year. If the maximum range of near-road soil radium concentrations observed in the region is used in this residential scenario (60 pCi/g ²²⁶Ra, 50 pCi/g ²²⁸Ra), residents living near these roads would be exposed to an estimated 296 mrems/year, above regulatory exposure thresholds used in nuclear facility siting assessments. These results underline the urgent need to clarify exposure risks from the use of oil and gas brines as a road treatment, particularly given the existing disparities in the distribution of road impacts across socioeconomic status.

Introduction

The rapid expansion of unconventional oil and gas (O&G) extraction in the Marcellus and Utica/Point Pleasant plays have generated substantial volumes of contaminated wastewater (i.e., produced water) (Lutz et al 2013). Traditional disposal methods for this wastewater were overwhelmed by the increased volumes and toxicity (i.e., high salt and heavy metal content) and this quickly led to fundamental changes in the management of this wastewater (Ferrar et al 2013, Rahm et al 2013). Capacity at wastewater treatment plants that previously diluted brine from coal and conventional gas operations was saturated by the unconventional brine volumes and resulted in discharge of waters with elevated total dissolved solids (TDS) and bromide (States et al 2013). The nationwide prohibition of brine dilution by the US EPA in 2016 (US Environmental Protection Agency 2016) has curtailed the practice. However, the problems created by disposal of unconventional wastewaters have cascaded to other disposal method options for conventional O&G brines, and, in some cases, resulted in reassessment of traditional methods such as road brining. Specifically, the evaluation of naturally occurring radioactive material (NORM) loadings from the use of conventional brines for dust suppression in Pennsylvania (Tasker et al 2018) resulted in a moratorium on this practice.
This reevaluation of traditional, grandparented disposal methods for conventional wastewaters occurs as demand for acceptable disposal methods (i.e., underground injection) increases. With disposal of unconventional production waste waters being increasingly limited to recycling and underground injection, supply and demand dictates that underground injection is less available and more expensive for conventional extractors. Therefore, the incentive to utilize traditional disposal methods available to conventional extractors has likely increased.

Increased brine production (Wilson and VanBriesen 2012) and the potential shift of conventional brines to traditional disposal methods may increase risks above the levels posed by historical use of these disposal practices. Even if these risks posed by historical conditions and practice were tacitly accepted, changes in risk associated with increased utilization of these practices should be evaluated. In isolation, use of O&G brines as a deicing or dust suppression product is a potential win-win scenario, saving both oil and gas operators and road maintainers money while lowering salt loadings to near road systems (Haake and Knouft 2019). However, the NORM in O&G brines poses potential exposure risks that are poorly defined, particularly through unintended accumulation of contaminants with repeated application. This study measured radioactivity in a sample of a brine product intended for individual property owner use to confirm the copious measurements of the products available in the grey literature. Then, exposure modeling based on these values and typical parcel resident behavior was developed. The consequences of potential O&G brine use on major commuter routes was examined in a neighborhood, equity, and stormwater context. In general, these risks warrant additional scrutiny of traditional brine disposal practices.

**Methods**

An O&G brine deicer product, Aquasalina® (Nature’s Own Source LLC, Brecksville, OH), was acquired from a Lowes in Ohio on June 13, 2018. Radioactivity of the brine product sample was measured on a broad energy germanium detector (Canberra BE3825) in Marinelli beakers after an equilibration period of >1 month. 226Ra activities were determined from the 214Bi (609 keV) and 214Pb (259 keV, 351 keV) energies to avoid uranium interferences. 228Ra activities were measured using the 228Ac daughter activity (911 keV). This measurement was not intended to be a rigorous assessment of residential de-icing products, a product was measured to evaluate the copious measurements of residential products available in the grey literature (McCracken 2017). Such an assessment is beyond the scope of this work.

Exposures were estimated for individual lots using RESRAD-ONSITE 7.2 (Kamboj et al 2018). Model defaults were adopted unless otherwise specified. In addition, RESRAD-ONSITE was used to reproduce the exposure estimates developed to assess use of O&G brine as a deicing or dust control agent (Pennsylvania Department of Environmental Protection 2016, Ohio Department of Health Bureau of Environmental Health and Radiation Protection 2018).

RESRAD simulations require the specification of the concentration and ‘geometry’ of the radioactivity. To generate reasonable estimates of these values, we used the recommended residential O&G brine product application (8 l per 92.9 m²) and an average area of driveway and sidewalk to determine an average activity of radium per application. This radium was assumed to impact a 2 m wide, 2 cm deep layer of soil adjacent to the driveway/sidewalk (the brine impacted area figure 1). To retain simplicity, radium transfer was assumed to be quantitative and be uniformly distributed across the width and depth. Soil bulk density is assumed to be ~1855 kg m⁻³ (a compacted soil, resulting in a conservative Ra soil concentration). These data were used to estimate the radioactivity for the RESRAD simulation. In road scenarios, these estimates were based Ohio Department of Transportation (ODOT) deicer application rates (table 1) to an area defined by a 1 m length of road and half of a typical road width. This activity was assumed to impact a soil volume similar to the residential case at the roadside (2 m width, 2 cm depth, by the 1 m length). To evaluate focusing of storm flows into storm water infrastructure, we used this same ODOT application rate to a 100 m length of a 4 lane highway (again, assuming a half width of the entire right way) and impacting a single soil volume as defined above. These scenarios are summarized in table 1.

The median new residential lot for a detached home in the United States is ~791 m² (US Census Bureau 2017). Based on literature values, a 791 m² lot has ~22–79 m² of sidewalk and ~42–142 m² of driveway (Akhbari et al 2003, Stone 2004, Bochis et al 2008).

RESRAD simulations require the specification of a variety of parameters (table 2). One of the first choices is the selection of exposures. For example, the PADEP scenario (Pennsylvania Department of Environmental Protection 2016) assess exposure for people using the roads recreationally, a reasonable choice for the dirt roads being considered. This choice dictates exposures. In the recreational scenario, given the potential for hunting, ingestion of meat is an exposure considered in the simulation. These exposures can be selected by the user independent of scenarios provided in RESRAD (e.g., the ODH exposure scenario was selected independently of
an exposure scenario, table 2). These exposure scenarios also dictate the exposure time and divide it into indoor and outdoor exposures (this influences the magnitude of radon and external gamma exposure). Brine impacted area and depth used for this study are described above or taken from the information in the studies cited. Likewise, activities are taken from the study cited, or in the case of this study, road concentrations from the PADEP recreationist scenario is used. All other parameters used were RESRAD defaults.

Cleveland is used as a case study to map spatial patterns of potential residential exposure and patterns in socio-economic status. Cleveland has an annual average of 46 winter storms and is the location of an O&G brine deicer production facility. Road, parcel, and census data were collected (Cuyahoga County Department of Information Technology - GIS Department 2019) and the distance of each parcel from a major road (NOACA codes 1 and 3) was measured (nearest function, ArcMap 10.x). Socioeconomic indicators for each of the lots located within 40 m of a major road were determined from 2010 US Census data (US Census Bureau 2010).

Results

Measurement of radium concentrations revealed substantial concentrations of radium in the O&G brine deicer product. $^{226}$Ra activity was $625 \pm 86$ pCi/L and $^{228}$Ra $516 \pm 44.9$ pCi/L. These measurements fall within the range of measurements of Aquasalina (McCracken 2017, ToxStrategies, Inc. 2019) and dust suppression brines (Tasker et al 2018). In the Ohio Department of Natural Resources samples (McCracken 2017) (i.e., ‘post’ values), $^{226}$Ra averaged $994 \pm 245$ pCi/L and $^{228}$Ra averaged $737 \pm 112$ pCi/L. It is difficult to place these in a larger context as a general radium water standard has not been set (radium discharge standards are developed on a permit by permit basis).

Application rates of the residential O&G brine product for ice control are $\sim 0.1$ times the application rates for dust suppression in Pennsylvania, but 10 times higher than application rates recommended for the use of brine as a road deicing agent in Ohio (table 1). The residential transfer scenarios result in $^{226}$Ra + $^{228}$Ra increases in the accumulation zone of 0.005–0.01 pCi/g soil per application, assuming an average Ra activity ($^{226}$Ra and $^{228}$Ra combined) of 1730 pCi/L, i.e., the sum of average ODNR values.

In both the PADEP and ODH simulations, RESRAD results indicate relatively minor risk increases (table 2). However, neither scenario considers a residential exposure scenario, where individuals spend relatively large proportions of time in or around their home. The RESRAD manual suggests an 18 h/day, 7 days/week exposure for residents on a contaminated site (12 h indoors, 6 h outdoors) (Kamboj et al 2018). A scenario predicting exposure for individuals living on a lot where brine products are used as primary deicing agent for sidewalks and driveways was developed (figure 2). Note, only external gamma exposure, dust and radon inhalation, and soil and plant ingestion exposure pathways were considered. Otherwise, default RESRAD values were used. The precise geometry of radium contamination along the edges of sidewalks and driveways cannot be specified in the RESRAD system, so the area of sidewalk and driveway was based on literature for a typical lot (Akbari et al 2003,.

Figure 1. Illustration of the assumed transfer of residential deicing applications to soils near driveways and sidewalks. For example, drainage from one side of a 4 m wide driveway would accumulate in a 2 m wide strip of soil next to the driveway. Likewise, a 1 m wide sidewalk would accumulate at ~ one half this rate. The sensitivity of the model to this geometry is documented in table 3.

Figure 2. Diagram illustrating the geometry of radium contamination along the edges of sidewalks and driveways. Drainage from one side of a 4 m wide driveway would accumulate in a 2 m wide strip of soil next to the driveway. Likewise, a 1 m wide sidewalk would accumulate at ~ one half this rate. The sensitivity of the model to this geometry is documented in table 3.
Table 1. O&G brine application rates and per application accumulation in roadside soils. This assumes a deicer activity of 1730 pCi/L radium.

| Application type                                      | Application rate (L/m²) | Road width (m) | Road length (m) | Activity per application (pCi/g soil/application) |
|-------------------------------------------------------|-------------------------|----------------|-----------------|--------------------------------------------------|
| Residential O&G deicer                                | 0.008 (Anonymous)       | 2 (half of 4 m wide driveway) | 1.0             | 0.008                                            |
| ODOT Anti-icing Road Application (low)                | 0.013 (Ohio Department of Transportation 2019) | 3.7            | 1.0             | 0.002                                            |
| ODOT Anti-icing Road Application (high)               | 0.026 (Ohio Department of Transportation 2019) | 3.7            | 1.0             | 0.004                                            |
| PA Dust Suppression (max initial application during dust suppression season) | 2.3 (Pennsylvania Department of Environmental Protection 2013) | 3.7            | 1.0             | 0.4                                              |
| PA Dust Suppression (max for subsequent applications during dust suppression season) | 1.5 (Pennsylvania Department of Environmental Protection 2013) | 3.7            | 1.0             | 0.3                                              |
| ODOT Road Application (low deicing rate, 4 lane highway, 100 m outfall spacing) | 0.013 (Ohio Department of Transportation 2019) | 7.4            | 100             | 17                                               |
Table 2. RESRAD parameters and results.

| RESRAD model | Exposure Scenario (and exposure pathways) | Exposure times (per year) | Brine impacted area & depth | Radionuclide activity in soil | Dose (at time zero/year 1) |
|--------------|------------------------------------------|--------------------------|----------------------------|----------------------------|--------------------------|
| ODH          | /doesn’t correspond to named scenario/ (external gamma, dust inhalation, ingestion of plants, ingestion of meat, ingestion of fish, ingestion of soil) | 2 h/day, 7 days/week, all outdoors | 93 m², 5 cm depth into soil | 0.965 pCi/g ²²⁶Ra 1.111 pCi/g ²²⁸Ra | 0.6 mrem/year |
| PADEP        | 'Recreationist' (external gamma, dust inhalation, radon inhalation, ingestion of plant, ingestion of meat, ingestion of milk ingestion of soil) | 2 h/day, 3 days/week, all outdoors | Area not specified, 15 cm depth into soil | 1 pCi/g ²²⁶Ra 0.5 pCi/g ²²⁸Ra (Above background) | 0.441 mrem/year |
| This Study   | 'Suburban Resident' (external gamma, dust inhalation, radon inhalation, ingestion of plants, ingestion of soil) | 12 h/day indoors, 6 h/day outdoors, 7 days/week | 221 m², 2 cm depth into soil | 1 pCi/g ²²⁶Ra 0.5 pCi/g ²²⁸Ra (Above background) | 4.6 mrem/year |
That is an area of 221 m² using the concentration observed by the PA DEP (1 pCi $^{226}$Ra and 0.5 pCi $^{228}$Ra) (Pennsylvania Department of Environmental Protection 2013) to a depth of 2 cm. This scenario produces an exposure of 4.6 mrem/year primarily from ground and radon exposure.

Discussion

RESRAD scenarios

The Pennsylvania Department of Environmental Protection (figure 2) modeled exposure risk based on radium concentrations elevated above background in sediments collected from roads historically treated with brines (i.e., 1 pCi $^{226}$Ra and 0.5 pCi $^{228}$Ra, 15 cm depth) (Pennsylvania Department of Environmental Protection 2016). Exposure was estimated for a recreationist (2 h/day, 3 day/week, includes fish, game, and milk consumption exposure pathways). Consumption of game accounted for the majority of estimated exposure in year one of this scenario (0.35 of the total 0.44 mrem/year). The Ohio Department of Health modeled the average risk for an application of O&G brine on a road (Ohio Department of Health Bureau of Environmental Health and Radiation Protection 2018). While ODH modeled both adult and child risk, given the similar results in these two cases, the adult case is considered here to facilitate comparison with other scenarios. The highest concentrations of Ra measured in brine product samples (1.1 pCi/g $^{226}$Ra and 1.3 pCi/g $^{228}$Ra) were distributed across a soil surface (93 m² area, 5 cm depth). The concentrations used in the model seemed to be based on the liquid concentration, not the soil concentration. Occupancy is assumed to be 2 h/day, 7 days/week and all exposure paths in RESRAD were modeled. The majority of exposure in this scenario is external exposure (0.46 of the total 0.6 mrem/year).

The observed activities of radium on or near roads can range much higher. The PADEP sampled 33 roads recently treated with O&G brines and 18 roads assumed to be not treated (Pennsylvania Department of Environmental Protection 2016). The ‘background’ roads actually had higher concentrations of Ra. This was attributed to an inability to rule out historical dust suppression with O&G brines (Pennsylvania Department of Environmental Protection 2016). In these background samples, concentrations ranged up to $\sim$60 pCi/g $^{226}$Ra and $\sim$50 pCi/g $^{228}$Ra. If this much Ra accumulated in a residential lot, RESRAD estimates using our residential scenario and these concentrations translate to exposures ($\sim$290 mrem, table 3) well above Nuclear Regulatory Commission general public exposure limits of 100 mrem/year (Zelac et al 2001).

Given the substantial uncertainties in these parameters, a series of sensitivity tests based on the scenario described above were conducted (table 3). Concentrations, area widths, area depths, and activities were varied and exposures assessed using the updated parameters. In general, exposure increases with activity in the brine impacted zone. Assuming a deeper brine impacted zone results in minimal changes in exposure estimates. In contrast, smaller ground surface areas resulted in higher exposure estimates. Unexpected interactions between parameters (e.g., change both depth and concentration) were not apparent. The use of the highest observed road concentrations (from the PADEP study) in a residential scenario resulted in substantial exposures to the residents.

Figure 2. Illustration of the brine exposure scenarios. Grey zones are road surfaces and brown-grey areas roadside brine impacted areas. Left panel) RESRAD exposure scenario used in the PA DEP evaluation of O&G brine application for dust suppression. This scenario is for ‘recreation’ dictating limited time exposures but including fish, game, and milk consumption exposure pathways. Middle) Ohio Department of Health exposure scenario for application of O&G brine as a deicing agent. This scenario seems to translate to road commuting exposures. Right) Residential scenario developed to model exposure from use of O&G brine product as residential deicing agent. Exposure times are longer due to residency.
Rates of contamination from O&G brine deicer
The accumulation of Ra near O&G brine road treatments can be substantial (Pennsylvania Department of Environmental Protection 2016). That said, the rate of Ra accumulation from O&G brine deicers in a residential lot is not clear. Using the mean Ra concentrations at recommended application rates, if all Ra remains in this brine impacted zone, a brine product application adds 0.005 to 0.01 pCi/g total Ra to adjacent brine impacted zones as defined above. In areas with frequent winter storms (Cleveland experiences an average of 46 distinct winter storms in a year), 1.0 pCi/g of Ra could accumulate in 2–5 ‘average’ years. Higher concentrations of Ra or application of additional product as ‘insurance’ (i.e., application of the product before a predicted storm that doesn’t arrive or extra product application to ensure the snow and ice dissolves completely) would accelerate this process. Likewise, uneven drainage or preferential flow could create zones of more concentrated Ra or deeper contamination depths.

The fate of the Ra once deposited in the soil is not clear. Previous studies of brine application have focused on the road materials themselves and observed minimal accumulation in laboratory experiments (Tasker et al. 2018). However, the mineralogy and surface area of a typical soil would increase the potential for Ra accumulation relative to road materials. The default distribution coefficient used in RESRAD for both Ra isotopes (i.e., the amount of Ra on the soil relative to solution) is 0.7. Therefore, a substantial portion remains in solution and is transported further than this brine impacted zone. Regardless, we know most contamination associated with road emissions is deposited relatively close to the road (Cape et al. 2004, Mielke et al. 2010) and expect a similar general fate for Ra. Given the uncertainties in loading and accumulation, this approach provides a reasonable approximation of potential contamination and therefore reasonable exposure estimates.

Fundamentally, contamination would have to move in relatively unexpected ways to get a substantially smaller concentration for a given brine product treatment.

Contamination impacts beyond brine product users
All RESRAD exposure estimates are used in the context of risk. Often the exposure estimates are compared to recommended dose limits for the average individual. However, specific individuals (people working in facilities with radioactive materials, pregnant humans, or those with a history of intense radiological treatments) may have more stringent dose limits. In a residential scenario, it is easy to assume one can avoid this risk by not using brine products for deicing. However, it is a challenge to prevent a neighbor from using the product, assuming that use of a brine product could even be reliably detected. Moreover, as with decaying lead paint in an old house, estimated risk changes for future owners. While RESRAD estimates generate risk levels that are deemed safe for the general population, risk assessments for specific portions of the current and future populations are not so simple.

Further, the concept of risk assumes that individuals can make choices to adjust their exposure. It is not clear this is the case for a resident that purchases a property with historical O&G brine deicer use or lives next to a neighbor who uses a brine product. What is even less clear is the ability of residents living near major transportation routes to control their risk. For example, in Cleveland, there are 2156 residential lots within 40 m of a major transportation route. If Ra accumulates in these parcels due to road application of O&G brines, the risk estimates described above would likely be on the low end of potential exposure. However, the focusing of road runoff may result in larger amounts of contamination (the 2 m contamination zone has the potential to receive brine product applied to multiple lanes of roadway). While storm water infrastructure has the potential to mitigate accumulation in the soil by rerouting the contamination to sewers, leaking infrastructure has the potential to introduce the contamination at depth (Rossi et al. 2017), a process that increases radon exposure risk. The use of O&G brine product on public roads has the potential to increase exposure to a substantial number of individuals who have limited control over that risk.

| Action | Base values (pCi/g) | Changed values (pCi/g) | Exposure estimate (mrems/yr) |
|--------|-------------------|-----------------------|-----------------------------|
| Double Activity | $^{226}$Ra, 0.5 $^{228}$Ra | $^{226}$Ra, 1 $^{228}$Ra | 9.3 |
| Double Depth, same load | 2 cm depth $^{226}$Ra, 0.5 $^{228}$Ra | 4 cm depth 0.5 $^{226}$Ra, 0.25 $^{228}$Ra | 4.5 |
| Half area, same load | 226 m$^2$ $^{226}$Ra, 0.5 $^{228}$Ra | 113 m$^2$ $^{226}$Ra, 0.25 $^{228}$Ra | 8.9 |
| Double area, same load | 226 m$^2$ $^{226}$Ra, 0.5 $^{228}$Ra | 452 m$^2$ 0.5 $^{226}$Ra, 0.25 $^{228}$Ra | 2.4 |
| ODH conc., residential | $^{226}$Ra, 0.5 $^{228}$Ra | $^{226}$Ra, 1.3 $^{228}$Ra | 5.8 |
| Double depth, double conc. | 2 cm depth $^{226}$Ra, 0.5 $^{228}$Ra | 4 cm depth 1 $^{226}$Ra, 0.5 $^{228}$Ra | 8.9 |
| Highest PADEP soil value treated roads | $^{226}$Ra, 0.5 $^{228}$Ra | 4 $^{226}$Ra, 4 $^{228}$Ra | 20 |
| Highest PADEP soil value background roads | $^{226}$Ra, 0.5 $^{228}$Ra | 60 $^{226}$Ra, 50 $^{228}$Ra | 296 |
The socioeconomic characteristics of the parcels in these near road areas suggest these potential exposures will reinforce existing inequities. Major road construction has been used as a tool to reinforce segregation in urban systems (Rose and Mohl 2012). This routing of commuter routes has led to inequitable exposure, where populations are exposed to higher levels of contaminants but derive less benefit from the processes that release the contamination (Tessum et al 2019). Cleveland parcels within 40 m of a major commuter route are, on average, in highly segregated areas (figure 3). Further, the median household income in these parcels (in 2010) was $27,638, well below the 2010 Cuyahoga County median household income of $43,603 (US Census Bureau 2010). This spatial structure indicates the potential risk from brine deicers would reinforce existing socioeconomic inequities and the ability to avoid this risk is not uniform.

**Brine product deicers and stormwater infrastructure**

When applied to roads, the NORM in O&G brine products would accumulate in streams and engineered systems receiving storm water (Ra accumulates in stream sediments downstream of brine disposal facilities (Warner et al 2013, Lauer et al 2018, Van Sice et al 2018)). This accumulation would be much faster than in residential situations as storm water outfalls receive flow and associated NORM from many meters of roadway (not just the length adjacent to the impervious surface used in our residential scenario, table 1). While areas with combined sewer systems would route NORM to the sewage treatment plant, any leaks in these lines or areas with storm sewer systems would introduce NORM to local systems. Municipal managers striving to meet imminent Municipal Separate Storm Sewer (MS4) regulations would face the additional challenge of mitigating Ra accumulations near storm water outfalls or in engineered retention systems. The potential distribution of NORM to waters downstream of stormwater infrastructure following road treatment has the potential to make stormwater management more complicated and expensive.

**Conclusions**

The reuse of wastewater from conventional O&G extraction as a road and residential deicing treatment has the potential to distribute NORM broadly across sometimes densely populated areas. This redistribution likely increases exposures for substantial and unexpecting portions of the public. The actual exposure, while low on an individual deicer application basis, has the potential to accumulate and create higher exposure levels through time. Further, the potential risk may (1) reinforce existing patterns in socioeconomic inequity and (2) complicate efforts to address aging water infrastructure. As this article is being written, the Ohio Legislature is considering legislation to allow sale of products with up to 20 pCi/ml $^{226}$Ra and 2.5 pCi/ml $^{228}$Ra (Hoagland and Rulli 2021), well above levels used in these estimates. The risk profile communicated to the public about O&G brine deicers does not match this level of NORM, nor does it match potential exposure scenarios when these occur on residential lots in areas with dense human populations.
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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Conflict of interest disclosure

The authors declare no competing financial interest.

ORCID iDs

Daniel J Bain \( \text{https://orcid.org/0000-0003-1979-7016} \)

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