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Monitoring radionuclides in subsurface drinking water sources near unconventional drilling operations: a pilot study

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Abstract

Unconventional drilling (the combination of hydraulic fracturing and horizontal drilling) to extract oil and natural gas is expanding rapidly around the world. The rate of expansion challenges scientists and regulators to assess the risks of the new technologies on drinking water resources. One concern is the potential for subsurface drinking water resource contamination by naturally occurring radioactive materials co-extracted during unconventional drilling activities. Given the rate of expansion, opportunities to test drinking water resources in the pre- and post-fracturing setting are rare. This pilot study investigated the levels of natural uranium, lead-210, and polonium-210 in private drinking wells within 2000 m of a large-volume hydraulic fracturing operation before and approximately one-year following the fracturing activities. Observed radionuclide concentrations in well waters tested did not exceed maximum contaminant levels recommended by state and federal agencies. No statistically-significant differences in radionuclide concentrations were observed in well-water samples collected before and after the hydraulic fracturing activities. Expanded monitoring of private drinking wells before and after hydraulic fracturing activities is needed to develop understanding of the potential for drinking water resource contamination from unconventional drilling and gas extraction activities.

Keywords

Hydraulic fracturing; Unconventional drilling; NORM; Uranium; Lead-210; Polonium-210

1. Introduction

The application of unconventional drilling (i.e., the combination of hydraulic fracturing and horizontal drilling) techniques to extract natural gas and oil is rapidly expanding throughout the world (Kerr, 2010; USEIA 2014). Although the current political and economic landscape

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is favorable for expanded natural gas exploration, many have voiced concerns for potential environmental and public health impacts (Howarth et al., 2011a, 2011b; Goldstein et al., 2012). One of the most important issues to be considered is the impact of unconventional drilling on local and regional water resources (Gregory et al., 2011; Vidic et al., 2013). Within this context, concern about potential contamination of drinking water resources by naturally occurring radioactive materials (NORM) liberated by unconventional drilling activities is on the rise (Brown, 2014).

Emerging reports indicate that unconventional drilling activities may contaminate ground water, as evidenced by increased levels of methane gas and other geologic signatures apparently originating from the natural gas-bearing formations (Osborn et al., 2011; Warner et al., 2012). Similarly, NORM have the potential to contaminate ground water by numerous mechanisms, including, but not limited to: flowback water spills on the surface, leakage of flowback from containment ponds, well casing failure, and introduction of new/larger fractures to the subsurface (USEPA 2012a). Through any of these processes, NORM contained either within the formation or in wastes removed from the formation (bit cuttings, brine, gas streams), have the potential to partition into ground-water. The potential for migration through the formation depends on the physical-geochemical (redox state, pH sensitivity, gas/solid) and radiochemical properties (half-life, decay products) (Nelson et al., in review).

Studying the impacts of unconventional drilling on NORM levels in private drinking is challenging for several reasons. In the United States (US), landowners are not required to monitor private drinking water wells for radionuclides (USEPA 2012b). Consequently, there are no large publically-accessible databases of NORM levels in private drinking wells in regions with drilling activities. Although the need to monitor private drinking water wells is recognized (DeSimone 2009), access to wells can be difficult and requires explicit permission from landowners. Another challenge is that scientific studies often lack critical temporal perspective (i.e. pre- and post-drilling), as drilling activities often outpace the ability for scientists to commence a well-controlled study. Thus, given that NORM is present in most groundwater, source-term attribution of NORM found in drinking water wells can be challenging. Taken together, these observations and the paucity of well-controlled studies complicates a scientific assessment of the relationship between unconventional drilling activities and NORM mobilization into ground water.

An opportunity for a small pilot study arose to investigate the potential for mobilization of NORM into groundwater due to unconventional drilling activities. Based on our experience with produced fluids from the Marcellus Shale region of the US, we focused our pilot study on levels of natural uranium (^{nat}U : ^{238}U , $t_{1/2} = 4.5 \times 10^9$ years; ^{235}U , $t_{1/2} = 7.0 \times 10^8$ years; ^{234}U , $t_{1/2} = 2.5 \times 10^5$ years), lead-210 (^{210}Pb , $t_{1/2} = 22.2$ years), and polonium-210 (^{210}Po , $t_{1/2} = 138$ days) in private drinking wells (with permission) in the north San Juan Basin of southwestern Colorado, USA (Nelson et al., in review). This area was chosen because we were able to obtain a sample of water that was collected weeks before a hydraulic fracture that occurred in January, 2012. Additionally, the San Juan Basin is a unique location to study the connection between drilling and NORM, as the region is renowned for fossil fuel resources and natural uranium (^{nat}U) deposits (Finch et al., 1989). In 2000 over 80% of the

US coalbed methane was extracted from the San Juan Basin, mostly from the Fruitland formation (late Cretaceous) (Ayers, 2002). Most (90%) of this gas is extracted from an area termed the “Fairway,” which is hydrostatically over-pressured (US EPA, 2004). In addition to large volumes of natural gas, wells in this area produce large volumes of water with 1000–4000 mg/L total dissolved solids (TDS) (US EPA 2004). Like many oil and gas rich regions in the world, the San Juan Basin is experiencing a boom in unconventional drilling, as operators seek to extract oil and natural gas from deeper shale formations (Sakelaris, 2014). Many landowners are concerned that these expanded unconventional drilling operations may introduce contaminants into drinking water resources. Thus, the goal of this pilot project was to determine if levels of ^{nat}U , ^{210}Pb , and/or ^{210}Po significantly changed in the year following a hydraulic fracture in a relatively small sub-geographical area within an area of intensive unconventional drilling activity.

2. Materials and methods

2.1. General

All reagents were ACS reagent grade or higher. NIST-traceable certified reference materials (CRM's, Eckert Ziegler Radioisotopes, Atlanta, GA USA) were used to prepare ^{232}U tracer (92403) and ^{209}Po tracer (92565), as previously described (Knight et al., 2014). All half-lives and alpha-emission energies, with the exception of ^{209}Po , were extracted from the United States National Nuclear Data Center (NNDC Brookhaven National Laboratory, US Department of Energy). Emission energy of ^{209}Po was extracted from NNDC; however, we chose to use a half-life of 128.3 years for ^{209}Po based on reports from the literature (Collé et al., 2007). All uncertainties are “standard uncertainties” with a coverage factor $k = 1$, unless stated otherwise (Currie, 1968). For all laboratory experiments, the analyst was blinded from the source of samples. Unless otherwise explicitly stated, all reported radioactivity concentrations were decay corrected to 2 January, 2013, 8:00 AM CST using standard differential equations (Bateman, 1910).

2.2. Sampling description

Duplicate groundwater samples from three private residences in the “Fairway” of the San Juan Basin in southwestern Colorado, USA; one surface water sample; and one municipal water sample were collected on 2 January, 2013 – based on standard collection methods (US Geologic Survey 2006). For all water samples, 1 L was collected and acidified (pH 2 using concentrated HNO_3), at least one week prior to analysis. An additional groundwater sample from residence A was collected by the owner on 22 January, 2012. Private drinking water wells ranged from approximately 40–100 m in depth according to information obtained from Colorado Division of Water Resources (<http://www.dwr.state.co.us/WellPermitSearch/>). Wells were approximately 2000 m from an unconventional well that was hydraulically fractured in January 2012 with ~750,000 L of hydraulic fracturing fluid at a depth of ~2400 m, according to information from FracFocus Chemical Disclosure Registry (<http://fracfocus.org/>). Additionally, within a 3 km radius of the homes, there are approximately 131 natural gas wells (12 of which were horizontally drilled) and 28 natural-gas/produced-water storage pits according to publically available information from the Colorado Oil and Gas Conservation Commission (COGCC, <http://cogcc.state.co.us/Home/gismain.cfm>).

Municipal water (purified from surface water) was collected from a city recreation center. Surface water was collected downstream of the site of a former U smelter. Note that surface and municipal water samples were collected upstream of any drilling activity in the region.

2.3. Determination radionuclide activities

250 to 500 mL aliquots of acidified samples were analyzed for activities of ^{nat}U , ^{226}Ra , ^{210}Po , and ^{210}Pb using established methods (Knight et al., 2014; Eichrom 2009, Nelson et al., 2014). Briefly, ^{238}U , ^{235}U , and ^{234}U were determined by preconcentration with $FeOH_3$, separation on TRU resin (Eichrom Technologies, USA) and TEVA resin (Eichrom), and prepared by CeF_3 microprecipitation, as previously described (Knight et al., 2014). 250 mL glass vials were hermetically sealed and stored for 30 days prior to quantitation of ^{226}Ra by ^{222}Rn emanation using a RAD7 as recommended by the manufacturer (DurrIDGE Company, Inc., USA). ^{210}Po activities were determined by preconcentration with $FeOH_3$, separation on Eichrom SR-Resin, and autodeposition on Ag disks (Eichrom, 2009). ^{210}Pb was determined by holding samples for 467 days and measuring ^{210}Po as previously described (Vesterbacka and Ikäheimonen, 2005). Approximately 3.5 half-lives of ^{210}Po had elapsed, suggesting that if modeled by secular equilibrium, the ^{210}Po had grown in to ~90% of the supporting ^{210}Pb activity. Alpha spectra of U and Po isotopes were collected on vacuum-controlled alpha spectrometers (Alpha Analyst, Canberra Industries, USA) employing passivated ion-implanted silicon detectors (PIPS, Canberra) with a source to detector distance of approximately 10 mm as described by us previously (Knight et al., 2014). Contamination of detectors was prevented by use of thin-films (Inn et al., 2008). Activities were determined using standard isotope dilution techniques (Makarova et al., 1983).

3. Results and discussion

3.1. Uranium

Alpha spectra indicated that ground water samples collected from private drinking water wells had varying activities of U isotopes (Fig. 1A, Table 1). The activities of ^{238}U in groundwater samples were 2.8–47 mBq/L and the activities of ^{234}U were 27–84 mBq/L. These levels of U do not exceed maximum contaminant levels (MCL; 30 mg/L or ~750 mBq/L) established by the United States Environmental Protection Agency (US EPA), but $^{234}U/^{238}U$ ratios ranged from 1.7 to as large as 10, suggesting alpha-daughter recoil has greatly enriched ^{234}U in some samples (Osmond et al., 1983). Notably, levels of U isotopes did not significantly change from pre-hydraulic fracture to post-hydraulic fracture at Home A. This observation suggests that U enrichment attributed to drilling activities has either not occurred or that any ^{nat}U plumes have not yet entered drinking water supplies. Importantly, the MCL of ^{nat}U in drinking water is often determined by the mass of ^{nat}U . The majority (99%) of ^{nat}U is comprised of ^{238}U , meaning the mass of ^{238}U is used to calculate total U activity, under the assumption that the activity of ^{238}U and ^{234}U are equal (US EPA 2014). Our results suggest that the groundwater may not be in equilibrium, thus by measuring ^{nat}U by mass, the true activity of ^{nat}U may be greatly underestimated.

3.2. Lead and polonium

^{210}Pb and ^{210}Po are the longest-lived radioactive decay products of ^{222}Rn , and are thus indicators of the accumulation of ^{222}Rn . The US EPA does not publish specific guidelines for evaluating ^{210}Pb or ^{210}Po concentrations in drinking waters. Consequently, little monitoring is performed for these radionuclides and little is known about their levels in drinking water sources, particularly from unregulated groundwater wells (Ruberu et al., 2007). When we tested for ^{210}Pb (average recovery 44%) and ^{210}Po (average recovery 30%), we found relatively low levels, each below recommended gross-beta and gross-alpha levels, respectively (Fig. 1B, Table 1). Levels of ^{210}Po ranged from 1.8 ± 0.7 mBq/L to 15.4 ± 1.6 mBq/L in groundwater. Note, because of sample holding time (>one year) and the number of elapsed ^{210}Po half-lives (>3), we were unable to quantitate ^{210}Po levels in the pre-fracture sample from Home A. After holding samples for over one year, we remeasured ^{210}Po to estimate ^{210}Pb , based on the principles of secular equilibrium. Levels of ^{210}Pb ranged from 7.6 ± 0.5 to 25.0 ± 1.2 mBq/L, post-fracture. The higher levels of ^{210}Pb relative to ^{210}Po in ground water samples is likely due to differences in particle reactivity and mobility of Pb and Po in the aquifer. We observed a potential increase in levels of ^{210}Pb at Home A, one year after the hydraulic fracturing operation. However, large uncertainties and small sample size of this pilot study limit the scope of any conclusions based on these data. Some stakeholders are concerned that contamination from hydraulic fracturing may be introduced to drinking water aquifers through: (1) ^{222}Rn migration from geologic formations due to introduced fractures, (2) ^{226}Ra contamination arising from spills, or (3) ^{226}Ra and/or ^{222}Rn migration through cracked well-casing. Thus, although we noted a slight increase in ^{210}Pb , our data does not suggest that increases of NORM in groundwater have occurred outside the range of natural variations (Fig. 2).

3.3. Limitations

Opportunities to sample private drinking water resources in the pre- and post-drilling setting are rare and this study represents a pilot investigation of important natural radionuclides of interest within this context. Apparent increases in ^{210}Pb observed at Home A in this study are not statistically significant and are limited by the small number of samples collected and analyzed. The levels of ^{226}Ra were below detection limits for our instrumentation, thus we were unable to determine its equilibrium status with ^{210}Pb . Further studies are needed to expand the scope and range of future work to enable statistically significant conclusions. Future studies are expected to include assessment of the levels of ^{222}Rn in ground water. More sensitive methods to determine the activities of ^{226}Ra , such as MnO_2 coprecipitations traced with ^{133}Ba (Maxwell, 2006), can also be employed to improve detection limits for ^{226}Ra .

4. Conclusions

As national and global demand for oil and natural gas expands, fossil-fuel-rich regions of the world will likely experience more exploratory and commercial unconventional drilling activities. New drilling techniques have been introduced to the San Juan Basin region, allowing operators to extract oil and natural gas from previously unreachable formations, such as the Morrison formation, Mancos Shale, and Gallup Sandstone (Sakelaris, 2014; Zah,

2014). Some of these formations are known to contain significant concentrations of NORM, such as the Morrison formation, from which 150,000,000 kg of U_3O_8 were extracted from between 1948 and 1986 (Turner-Peterson and Fishman, 1986; McLemore and Chenoweth, 2003). Given the presence of NORM in underlying strata, there is the potential for unintended contamination of drinking water sources (aquifers) by radionuclides (e.g., U isotopes, Ra isotopes, ^{210}Pb , and ^{210}Po). These potential contamination events will likely be isolated to areas near the drilling operations, in regions where most drinking water is supplied from aquifers. Yet, many of the private wells used to extract drinking water from such aquifers are unlikely to be monitored by state and local environmental/public health agencies. Thus, there is a need to expand monitoring of these wells, particularly before drilling operations take place. Limited sample size and scope limits any conclusions based on the data presented here. However, this pilot study provides a basis upon which to build future expanded studies to develop a more detailed understanding of the potential impact of co-extracted NORM on drinking water resources.

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Fig. 1.

Activities of NORM in water collected from homes, a river and a municipal building in an area with unconventional drilling operations. (A) natural-uranium activities (^{238}U (blue), ^{235}U (grey), ^{234}U (green)) in Jan 2013. (B) lead-210 (^{210}Pb , (brown)) and polonium-210 (^{210}Po , (red)) activities on Jan 2013 and one home on Jan 2012. Uncertainties are based on duplicate measurements and counting statistics. (*) ^{210}Po reported, but due to sample holding times does not represent unsupported ^{210}Po .

Fig. 2. Schematic of possible NORM contamination pathways of groundwater near hydraulic fracturing operations. Solid lines indicate a radioactive decay or series of radioactive decays. Dashed lines indicate transportation in the environment. Geologic data adapted from a published source (Fassett, 1974). Note, geologic formations are not to scale for illustrative purposes.

Table 1

Activities of radionuclides from selected sites in Southern Colorado.

	^{238}U	^{235}U	^{234}U	$^{234}\text{U}/^{238}\text{U}$	^{226}Ra	$^{210}\text{Pb}^b$	^{210}Po
Municipal	5.0 ± 0.6	0.3 ± 0.1	7.7 ± 0.7	1.5	<LOD ^c	1.0 ± 0.3	1.3 ± 0.4
Stream	36 ± 1.3	1.4 ± 0.3	40 ± 1	1.1	<LOD	3.0 ± 0.4	2.8 ± 0.7
Home B (1)	47 ± 1	1.8 ± 0.3	81 ± 2	1.7	<LOD	7.6 ± 0.5	1.8 ± 0.7
Home B (2)	46 ± 2	3.0 ± 0.5	84 ± 3	1.8	<LOD	10.0 ± 0.6	5.2 ± 1.1
Home C (1)	7.3 ± 0.7	0.4 ± 0.2	73 ± 2	10	<LOD	24.3 ± 1.1	10.0 ± 1.4
Home C (2)	8.7 ± 0.7	0.7 ± 0.2	74 ± 2	8.5	<LOD	20.5 ± 0.9	5.3 ± 1.0
Home A (1)	3.7 ± 0.7	0.3 ± 0.2	32 ± 2	8.4	<LOD	19.9 ± 0.9	13.2 ± 2.0
Home A (2)	3.3 ± 0.8	0.8 ± 0.4	27 ± 2	8.2	<LOD	25.0 ± 1.2	15.4 ± 1.6
Home A (3) ^a	2.8 ± 0.5	0.4 ± 0.2	29 ± 1	10	<LOD	14.3 ± 1.0	15.2 ± 1.6

All activities reported as mBq/L.

All uncertainties correspond to $k = 1$.

All samples collected 2 January 2013, unless otherwise stated.

^aSample collected 22 January 2012.^bInferred from ^{210}Po (assumed in secular equilibrium on 17 April 2014).^cLOD approximately 100 mBq/L.