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Features

ANALYSIS OF RESERVE PIT SLUDGE FROM UNCONVENTIONAL NATURAL GAS HYDRAULIC FRACTURING AND DRILLING OPERATIONS FOR THE PRESENCE OF TECHNOLOGICALLY ENCHANCED NATURALLY OCCURRING RADIOACTIVE MATERIAL (TENORM)

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ABSTRACT

Soil and water (sludge) obtained from reserve pits used in unconventional natural gas mining was analyzed for the presence of technologically enhanced naturally occurring radioactive material (TENORM). Samples were analyzed for total gamma, alpha, and beta radiation, and specific radionuclides: beryllium, potassium, scandium, cobalt, cesium, thallium, lead-210 and -214, bismuth-212 and -214, radium-226 and -228, thorium, uranium, and strontium-89 and -90. Laboratory analysis confirmed elevated beta readings recorded at 1329 ± 311 pCi/g. Specific radionuclides present in an active reserve pit and the soil of a leveled, vacated reserve pit included ^{232}Thorium decay series (^{228}Ra, ^{228}Th, ^{208}Tl), and ^{226}Radium decay series (^{214}Pb, ^{214}Bi, ^{210}Pb) radionuclides. The potential for impact of TENORM to the environment, occupational workers, and the general public is presented with potential health effects of individual radionuclides. Current oversight, exemption of TENORM in federal and state regulations, and complexity in reporting are discussed.

Keywords: reserve pit, radiation, Technologically-Enhanced Naturally Occurring Radioactive Materials (TENORM), Naturally Occurring Radioactive Materials (NORM), Barnett Shale, natural gas mining, fracking

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Reserve pits are commonly seen throughout areas of unconventional natural gas extraction. The purpose of the reserve pits (commonly referred to as pits, ponds, cellars, tanks, impoundments, etc.) is to hold the large quantities of water and drilling mud required for hydraulic fracturing ("fracking") operations. These pits also provide a depository for brine water that occurs naturally in natural gas deposits, drilling mud, drilling cuttings and hydraulic fracturing fluids. Hydraulic fracturing fluids can contain chemical additives (acids, bactericides, breakers, corrosion inhibitors, cross-linkers, emulsifiers, flocculants, foaming agents, proppants, scale inhibitors, surfactants) and cuttings (rock, soil and metal shavings excavated by the drill bit) which can contain technologically enhanced naturally occurring radioactive material (TENORM) [1, 2]. Previous research has identified ^{226}radium (^{226}Ra), ^{228}radium (^{228}Ra), and radon gas (^{222}Rn) as the predominant radionuclides in natural gas wastes from oil and gas drilling. The focus of existing regulation guidelines has been related to ^{226}Ra and ^{228}Ra, which have the potential to release radon gas into the atmosphere when these radioactive nuclides are brought to the surface through the oil and gas extraction processes [3]. The long half-lives of these two radium isotopes (^{226}Ra, 1,600 years; ^{228}Ra, 5.8 years) are particularly concerning given that they have been identified as abundant in saline and chloride-rich produced waters [4]. To date, few other radionuclides have been identified as associated with natural gas extraction, and fewer still have had regulatory guidelines developed for occupational or public health exposures.

Naturally occurring radioactive material (NORM) is terrestrial radiation distributed by nature throughout natural geologic formations. It is undisturbed radioactive material that exists in nature as background material, or at its in-situ location, whether at the earth's surface or subsurface. TENORMs are when naturally occurring radionuclides are transported by anthropogenic activity to where humans are present, thereby increasing exposure potential, which may result in concentrations enhanced above natural background levels [5]. As such, NORM transported or concentrated during exploration and mining of oil and gas is thereby reclassified, according to regulatory definition, as TENORM.

Both NORM and TENORM are clearly defined and distinct from radionuclides that are produced through nuclear reactions, nuclear explosions or nuclear accelerators (commonly referred to as "man-made, artificial, or anthropogenic"). The term NORM is often misused when applied to radioactive material introduced into the human environment by oil and gas exploration and mining processes.

Estimates of water needed for unconventional natural gas extraction are reported to range from one to five million gallons per well for initial well completion [6]. The use of up to 12 million gallons per well completion (one million gallons per stage) has been documented for the 12-stage open-hole completion systems [7]. Disposal of large quantities of chemical- and radionuclide-laden materials in wastewater is a known problem [8]. Reserve pits are commonly

found in agricultural areas where the potential for crop and animal contamination is high. Animals drinking pit water, dust particles blowing onto soil and crops, and berms breaching (thus contaminating adjacent croplands) are all potential exposure pathways. If reserve pits are built with an aerator, aerosolized radioactive material can be further dispersed onto soil, crops, livestock, and humans. Deposition of reserve pit contents in county landfills and municipal water treatment facilities has elicited a public outcry of concern for environmental contamination and potential human exposure to harmful radioisotopes often present in the drilling mud and cuttings, since these facilities do not have the capability to test for or remove radioactive material from the waste stream [9-11]. Incorporation of reserve pit material into the earth's surface either by draining and leveling the reserve pit where it exists, and/or land farming the material into the ground in place or at other locations, may increase the potential for surface and drinking water contamination from percolation or migration of radionuclides into water bodies. A better understanding is needed to assess the potential effects that radionuclides may have on the health of cattle, on cattle productivity, and on agricultural products. The potential exposure to humans is from reserve pit contents via wind, and by consumption of crops and animal products that have taken up radioactivity, has not been established [12-17].

The purpose of this article is to present laboratory analysis of water and soil (sludge) analyzed for the presence of TENORM, obtained from two unrelated reserve pits located on agricultural land in the Barnett Shale (located in Texas) and used as holding ponds for unconventional natural gas mining and extraction processes. This study originated as part of a field study conducted as a preliminary exploratory investigation (Phase II) during a property transaction to ascertain if, in fact, any regulatory impact existed (such as the presence of radioactive materials in the reserve pits). Comparison of study findings to state and federal guidelines for TENORM material identifies the complexity in regulatory reporting and guidelines, and current voids in regulatory oversight.

EXPERIMENTAL METHODS

Field Sampling

Soil and water matrices from reserve pits in the core area of the Barnett Shale East Newark Field were obtained and analyzed for the presence of radionuclides (TENORM). Soil and water was collected from two separate site locations: 1) farmland that was once a reserve pit, which had been drained and leveled to the surrounding elevation; and 2) a reserve pit that, at the time of sampling, held drilling mud, water for hydraulic fracturing, processed water and/or cuttings. For the purpose of this report the drained reserve pit has been identified as Reserve Pit #1 (RP1) and the pit with fluid has been identified as Reserve Pit #2 (RP2). In total, four separate samples of water and soil were obtained, two from

each sampling location, and identified by the laboratory as sludge due to high water content. Water was collected in clear plastic 500-ml containers with no preservative. Two sample points were selected for each pit based on each pit's use and the most likely impact resulting from surrounding exploration and extraction activities.

Samples in RP1 were obtained at a soil depth of 6 inches from the soil surface, since the RP1 pit had been drained and appeared to have the greatest potential to be relatively homogeneous from initial field investigation. This reserve pit was originally constructed with above-ground berms without any surface discharge outlet. Water could be pumped into the pit from an adjacent water well and could flow out of the pit only via its natural down-gradient seepage. Two samples were obtained along a line following the direction of the pit's down-gradient groundwater flow, which ultimately intersected with a flowing creek located near to and down-gradient from the pit.

RP2 is a typical triangular ranch pond with the triangle base side perpendicular to the downgradient flow line of the pond. A surface flow outlet is located at the center of the downgradient side. The samples were taken inside of the pond. Since cuttings and drilling mud settle to the bottom of ponds, efforts were made to obtain sludge/sediment samples from the pit bottom of RP2 along with water. Impact to or from the pit appeared to occur at either end of this down-gradient side (i.e., at the corners). Flow gradients dictated exploration and production impact would occur at the corners and then would flow from these corners down-gradient to the outfall. A sample was taken at one corner and a second sample was taken at the upstream pond side of the outfall. RP2 samples were collected from the pond's floor on the down-gradient side of the pit.

Initial observations indicated that impact from well mining extraction and injection materials appeared to be located on the upgradient side of each pond's downhill side. This observed material in the pit was considered likely to be from the geologic formations mined and materials injected. All samples were shipped to a certified radiological laboratory (American Radiation Services, Inc., Port Allen, LA) for analysis of radioactive isotopes by EPA method 901.1M (ARS-007/EPA901.1M). Radioisotope concentrations were reported in picocuries/gram (pCi/g). Reserve pit contents were analyzed for the radionuclides beryllium (^{7}Be), potassium (^{40}K), scandium (^{46}Sc), cobalt (^{60}Co), cesium (^{137}Cs), thallium (^{208}Tl), lead (^{210}Pb and ^{214}Pb), bismuth (^{212}Bi and ^{214}Bi), radium (^{226}Ra and ^{228}Ra), thorium (^{228}Th), uranium (^{235}U), strontium (^{89}Sr and ^{90}Sr), and total gamma, total alpha, and total beta radiation.

This study was designed to be an initial investigative field study performed for an industrial land transaction decision. Samples were not randomized, but selected to represent the most likely worst-case down-gradient impact point. Analysis of a control sample was not performed or authorized. Soil sample results were compared to findings of previous studies and to regulatory limits. However, inconsistencies in collection and analysis of specific radioisotopes in

previous studies made comparison difficult and it was not easy to ascertain in many cases whether the samples exceeded expected baseline concentrations.

Reserve Pit #1 (RP1)

The location identified as Reserve Pit #1 (RP1) had originally been part of a reserve pond, but at the time of sampling had been drained and leveled to the original ground surface grade. The original reserve pit was a manmade pond of approximately 2.9 acres, whose depth was increased with berms to a height of six to seven feet above ground level. Soil in the drained and leveled area sampled (RP1 location) appeared to have been undisturbed and the pond material allowed to drain and settle naturally, incorporating back into the existing soil rather than being removed and disposed of offsite. The RP1 sampling sites chosen were at one time the reserve pit bottom material. The remaining reserve pit was still present at the time of sampling and was still in use as a water reservoir for mining operations. Soil and water samples taken at this location were identified as RP1.1-West and RP1.2-East. The RP1.1-West sample was obtained approximately 15 feet from the edge of the existing pit berm, and the RP1.2-East sample was obtained approximately 75 feet from the edge of the existing pit berm. The purpose of obtaining soil from this location was to examine if any radioactivity in the soil existed after the reserve pit had been drained and the land left fallow. The adjacent land was used as agricultural land, which at the time of sampling was growing livestock feed. Field notes taken at RP1 locations identified the soil to be homogeneous black clay with very little organic matter and high water content, believed to be related to a precipitation event a few days prior to sampling. The U.S. Department of Agriculture Natural Resource Conservation Service defines black clay as having slow infiltration rates, high runoff potential when wet, and high shrink swell potential [18].

Reserve Pit #2 (RP2)

At the time of sampling, Reserve Pit #2 was being used as a water reservoir for natural gas extraction and mining operations and was believed to have been used to hold drilling mud, processed water, water for hydraulic fracturing operations, and drill cuttings. RP2 encompassed approximately 11.3 acres. This pit was originally a manmade pond at ground level. The water level was high due to recent precipitation events with an area overflowing the banks of the pit into a neighboring stream. The overflow area led to a creek and had been graded and cemented to provide a controlled exit for overflow water to minimize water breaching the pit berm at various locations. Two separate samples were obtained at RP2: one was obtained inside the pit along the east edge at the overflow location (identified as RP2.1-North), inside the pit along the northeast edge; the second sample was obtained on the south end of the pit closest to the well pad site inside the pit (identified as RP2.2-South). The samples taken in

the reserve pit consisted of both water, obtained from approximately 6 inches below the surface, and soil, obtained approximately 3 feet from the berm edge at the bottom of the pit.

The soil matrix at RP2 location was varied, with the presence of dark grey sticky clay soil, commonly referred to as black clay soils on the exterior of the pit and a light yellowish brown clay soil mixed with high very fine sand (<1 mm diameter) interior to the pit [20].

Field notes taken at the RP2 location identified a noticeable lack of any insects, fish, turtles, snakes or birds present in the or around the pit. The pit contained water grasses and reeds which are optimum breeding and cover areas for fish, snake and bird activity but no activity or signs of any feeding, nesting, or breeding activity were apparent.

RESULTS

Results of laboratory analysis of the four samples are presented in Table 1. The level of radioactivity is presented as pCi/g, and the minimum detection concentration (MDC) is the lowest concentration reliably detected by the laboratory equipment. The Analysis of Error is a numerical factor that represents error in the laboratory detection technique. This error factor is specific to each radionuclide and specific to each test. A zero is entered in the table if the radioactivity detected is below the MDC.

In general, specific radioisotopes detected included ^{40}K, elements of the ^{228}Th decay series (^{228}Th, ^{228}Ra, and ^{208}Tl), elements of the ^{226}Ra decay series (^{226}Ra, ^{214}Bi, ^{214}Pb, ^{210}Pb), and ^{90}Sr. With the exception of total alpha radiation for RP2-North, varying levels of total alpha, beta, and gamma radiation were detected in all samples. Interestingly, different portions of the same pit showed some differences in the radioactivity present.

It is important to note that not all radioisotopes present in sample RP1.1-West were also present in sample RP1.2-East, despite their close proximity and presumed homogeneous material. At the time of sampling, both locations had a high water content in the soil due to a recent precipitation event that may have been a contributing factor to variability in radioisotope concentrations. Sample RP1.2-East had a greater variety of isotopes recorded above laboratory minimum detection. Some of the isotopes present in this study are known to have very short half-lives (^{214}Bi, 20 minutes; ^{214}Pb, 27 minutes), and their presence is not easily captured. Their presence is likely to be due to the fact that they are part of a decay series and are continuously being generated. Other isotopes have longer half-lives and are more easily identified. In comparing results of the two RP1 locations, similar concentrations were noted for ^{40}K, ^{208}Ti, ^{214}Pb, ^{228}Ra, ^{228}Th. Notably, ^{210}Pb and ^{90}Sr were found in the RP1.1-West sample but not in the RP1.2-East sample, while ^{226}Ra was detected in the RP1.2-East sample but not the RP1.1-West sample. The gross gamma radiation (22.8 and 21.4 pCi/g),

Table 1. TENORMs Found in Reserve Pits

Isotope	RP1.1-west	RP1.2-east	RP2.1-north	RP2.2-soth
Beryllium (^{7}Be)	0 (0.45)	0 (0.48)	0 (0.45)	0 (0.53)
Potassium (^{40}K)	5.3 ± 1.3 (0.82)	5.5 ± 1.0 (0.41)	4.9 ± 1.1 (0.68)	3.6 ± 1.0 (0.67)
Scandium (^{46}Sc)	0 (0.076)	0 (0.078)	0 (0.064)	0 (0.58)
Cobalt (^{60}Co)	0 (0.090)	0 (0.064)	0 (0.10)	0 (0.69)
Cesium (^{137}Cs)	0 (0.086)	0 (0.062)	0 (0.72)	0 (0.62)
Thallium (^{208}Tl)	0.20 ± 0.07 (0.060)	0.27 ± 0.06 (0.041)	0.18 ± 0.06 (0.076)	0.19 ± 0.05 (0.04)
Lead (^{210}Pb)	1.7 ± 1.2 (1.4)	0 (0.94)	0 (1.1)	0.99 ± 0.65 (0.94)
Bismuth (^{212}Bi)	0 (0.56)	0 (0.46)	0 (0.56)	0 (0.54)
Bismuth (^{214}Bi)	0.45 ± 0.15 (0.17)	0.35 ± 0.30 (0.15)	0.36 ± 0.12 (0.15)	0.25 ± 0.12 (0.18)
Lead (^{214}Pb)	0.68 ± 0.63 (0.14)	0.70 ± 0.15 (0.14)	0.44 ± 0.12 (0.15)	0.40 ± 0.11 (0.13)
Radium (^{226}Ra)	0 (1.3)	2.4 ± 1.0 (1.2)	0 (1.5)	0 (1.1)
Radium (^{228}Ra)	0.66 ± 0.21 (0.26)	0.71 ± 0.13 (0.19)	0.51 ± 0.15 (0.25)	0 (0.24)
Thorium (^{228}Th)	0.72 ± 0.11 (0.087)	0.67 ± 0.11 (0.093)	0.64 ± 0.13 (0.12)	0.36 ± 0.10 (0.10)
Uranium (^{89}Sr)	0 (0.34)	0 (0.27)	0 (0.42)	0 (0.32)
Strontium (^{89}Sr)	0 (0.24)	0 (0.24)	0 (0.36)	0 (0.26)
Strontium (^{90}Sr)	0.30 ± 0.17 (0.24)	0 (0.24)	0.59 ± 0.26 (0.36)	0.29 ± 0.18 (0.26)
Total gamma	22.8	21.4	10.8	8.22
Total alpha	10.8 ± 3.3 (2.6)	16.4 ± 4.6 (3.1)	0 (3.6)	9.1 ± 3.4 (3.9)
Total beta	9.1 ±2.5 (1.8)	5.7 ± 2.0 (2.3)	1329 ± 311 (5.0)	5.8 ± 1.8 (1.7)

[a]The level of radioactivity is given in pCi/g and is shown with the analysis error. The numbers in parentheses are the Minimum Detection Concentrations (MDCs). In cases where the radioactivity measured was less than the MDC, a value of 0 is entered.

gross alpha radiation (10.8 ± 3.3 and 16.4 ± 4.6), and gross beta radiation (9.1 ± 2.5 and 5.7 ± 2.0) were not significantly different in the two RP1 samples.

Similar results were seen in individual radioisotopes in the second reserve pit RP2.1-North and RP2.2-South samples. ^{228}Ra was detected in RP2.1-North but not RP2.2-South, whereas ^{210}Pb was observed in RP2.2-South but not RP2.1-North. Total gamma radiation was similar in the two samples, but gross alpha radiation was observed only in RP2.2-South.

The most unexpected result of this study was the difference identified in gross beta radiation within the same pond. Gross beta radiation in the RP2.1-North sample was considerably higher than in the South sample (1329 ± 310 vs. 5.8 ± 1.8 pCi/g). The highest beta radiation levels were recorded near the spillway in pond RP2. Radionuclides are unstable isotopes of elements that undergo radioactive decay continually. Accumulation of sediment near the spillway may have accounted for the variability in beta radiation levels. Despite the close proximity of the soil samples within the pond, it is difficult to determine if the variability in concentrations reflects initial concentration in the soil, amount of material deposited in the pond, or lack of uniformity of soil chemistry. The fact that such variability can exist provides a complexity to single sample testing and may indicate that numerous samples within a single reserve pond are needed for accurate identification and quantification of TENORM, and proper representation of potential exposure to radioactive material.

DISCUSSION

Routine field study analysis of reserve pit contents from unconventional natural gas mining confirmed the presence of alpha, beta, and gamma radiation in the soil and water in reserve pits located on agricultural land. The specific gamma-emitting radionuclides identified included ^{40}K, ^{208}Tl, ^{210}Pb and ^{214}Pb, ^{214}Bi, ^{226}Ra and ^{228}Ra, ^{228}Th, and ^{90}Sr. Total beta radiation of 1329 pCi/g found in this study exceeded regulatory guideline values by more than 800 percent. Data from this limited field study showed elevated levels of alpha, beta, and gamma radiation to be present in reserve pit water/sludge material and also in the soil of a vacated reserve pit after draining and grading to original topographic levels. Based on the use of the pit, the presence of radioactive materials was not anticipated. Agricultural land adjacent to the drained reserve pit may have an increased potential for radioactive material taken up in livestock feed crops growing on the land due to wind transport, runoff, and migration of soil onto adjacent land. Deposition of radioactive material on land has been shown to have the potential to raise the radiation levels in soils above natural background levels increasing the potential for contamination of groundwater, soil, animals (domestic and migratory), and humans (through occupational and residential exposures). Historically, background levels of naturally occurring radiation prior to land use have not been measured, and little information on true

background radiation actually exists. Texas has a long history of oil and gas exploration, which has involved the practice of land farming and surface deposition of mining material. Further, for decades, unrefined oil has been deposited on roadways for dust control. Assessment of true background radiation levels may not be possible given this historical misuse of the land. Total radiation was found to be elevated above known background levels for radiation, but information is limited and exposure pathways poorly understood. Regulatory guidance documents currently do not address many of the radionuclides found in this study and provide few directives and little guidance in determining the potential synergistic or additive effects of exposure to several radionuclides simultaneously, or the potential for an increased incidence of disease in animals or humans due to simultaneous multiple exposures. Expansion of urban drilling and the practice of siting reserve pits within residential communities will increase the potential for radiation exposure to the general public. Health complaints related to low-level radiation sickness, common to occupational workers, may be overlooked by medical professionals who do not anticipate an industrial-type exposure to patients living within these communities. Stricter guidelines may be warranted in order to protect the general public from increased levels of radiation in soil, water, and air.

Radionuclide Decay

Radioactive decay releases three types of radiation: alpha (α), beta (β) and gamma (γ) emissions. All three types of radiation are known to present health hazards. The radionuclides in TENORM that present the most concern in the human environment due to potential health impacts are isotopes of radium, thorium, and uranium and their decay products. ^{238}U decays by alpha emission into ^{234}Th, and ^{234}Th decays by beta emission to protactinium and then ^{234}U. ^{226}Ra, ^{214} Bi, and ^{210}Pb are all daughter isotopes of ^{238}U. ^{234}U decays by alpha emission into ^{230}Th, which decays by alpha emission into ^{226}Ra, ultimately decaying by beta emission into products seen in this study: ^{214}Pb, ^{214}Bi, and ^{210}Pb.

Environmental and Health Impact of Exposure to TENORM

There are numerous potential pathways of exposure to radioactive material from wastes extracted by natural gas exploration and mining. This study attempts to investigate only one form of waste, reserve pit contents. However, there are several potential pathways of exposure from this one waste form alone. The potential exposures to humans directly, whether occupational or residential, include: ground-water contamination, soil contamination, windborne particulates and aerosolized material, and fugitive air emissions from industrial processes. Another secondary potential exposure pathway exists in the ingestion of agricultural products (vegetables, dairy, and meat products) that may

contain these radionuclides. This is an area that has received little attention or investigation.

The complexity in examining potential exposure is in quantifying how much radiation one has been exposed to, and the dose absorbed due to the exposure, and in accurately assessing the potential health impacts from multiple pathways. In order to properly assess exposure, exposures to all forms of radiation (alpha, beta, gamma) as well as to specific radioisotopes must be quantified and a thorough human health risk assessment performed. This is rarely done unless concentrations of a single radionuclide, for which regulatory guidelines have been established, greatly exceed those guideline levels; and for many radionuclides, no regulatory guideline levels have been established. Since many radionuclides have not been identified to be present in reserve pit wastes until recently, regulatory guidelines have not been established for non-occupational exposure limits.

The radionuclides discussed below were found in the samples taken in this study. Evaluating the potential health impacts of each radionuclide individually is important, in addition to evaluating the total decay (alpha, beta, and gamma) radiation, as the target organs and sites of damage can differ.

Health Effects of Potassium (^{40}K)

Potassium can be taken into the body through ingestion (food or water) or inhalation. ^{40}K is a naturally occurring radioisotope of potassium and widely distributed in nature (although normally at very low levels—0.015% in soil). It has a very long half-life of 1.3 billion years and decays primarily to ^{40}Ca by beta emission. External exposure to ^{40}K is generally to gamma radiation as ^{40}K decays to ^{40}Ar. Internal exposure to ^{40}K can pose a health hazard from ionizing beta and gamma emissions as it decays, with the potential to cause cell damage [19].

Health Effects of Radium (^{226}Ra, ^{228}Ra)

According to a U.S. Geological Survey (USGS) study (2009), little data exists on natural background concentrations of radium in the environment. Levels have been documented to increase as a result of human activity [20]. Radium levels in drinking water can become elevated in areas of mining. Exposure to radium may result in a variety of health effects such as tooth fractures, anemia, and cataracts. Chronic exposure to radium is known to increase the incidence of cancer in humans [21, 22]. Gamma radiation from radium is able to travel long distances through air before expending its energy, thus increasing exposure to the general population [23]. Radium is the radionuclide on which most of the drinking water and air regulations are set. It is the primary radionuclide identified in the past as a potential source of exposure to radon, a decay product of radium and a known lung carcinogen.

Health Effects of Strontium (^{90}Sr)

^{90}Strontium is a manmade isotope of strontium. ^{90}Sr is used as a subsurface radioactive tracer in mining processes and has a half-life of 29.1 years [24]. It is also present at low levels in surface soil due to fallout from previous atmospheric nuclear tests. It is hydrophilic, easily moving into and through the environment, adding to its ability to contaminate aquifers and drinking water sources [25]. It is known to be dangerous to the health of animals and humans. Exposure to ^{90}Sr can occur by inhalation of dust, eating food, or drinking water contaminated with the radionuclide. Grains, leafy vegetables, and dairy products can contain significantly high levels of ^{90}Sr [26]. The primary target organ for ^{90}Sr is bone. Strontium competes with calcium taken up in bone and can damage bone marrow, causing anemia. It can also cause cancer as a result of damage to cellular genetic material [27].

Health Effects of Thallium (^{208}Ti)

Thallium is absorbed by the human body through inhalation of dust particles and through ingestion of food and water. The nervous system is the primary target organ for thallium, which is known to cause trembling, nerve pains, paralysis, and behavioral impacts. Tiredness, depression, lack of appetite, and hair loss are all symptoms of chronic low-level Ti exposure. Thallium exposure to the fetus has been known to cause congenital disorders [28].

Health Effects of Thorium (^{228}Th)

Inhalation of thorium can adversely impact the respiratory system, causing damage that can eventually culminate as lung cancer. Exposure to thorium is known to cause pancreatic cancer, and thorium can be stored in bone, leading to bone cancer years after the initial exposure. People living in industrial areas near hazardous waste sites and near waste materials may be exposed to higher concentrations of thorium from wind-blown dust and consumption of food contaminated by the radionuclide [29].

Potential for Plant and Animal Exposure to TENORM

Contamination of soil and water from TENORM can expose workers and the general public to increased levels of radiation above normal background levels. Other important aspects of environmental contamination are through radiation taken up by the soil-plant system and exposure to animals through feedstock. Radionuclides in the soil can be directly intercepted by crops, which are then used as livestock feed, further increasing the potential for human exposure to increased levels of radiation through ingestion of milk and meat products.

In 2009, the U.S. Fish and Wildlife Service identified the importance of protecting migratory birds from exposure to reserve pit contents which can

contain diesel, glycols, and heavy metals, but failed to recognize the potential for bird populations to be exposed to radioactive material deposited in reserve pits [30]. Some states with oil and gas regulations recommend netting or screening of pits or open tanks to prevent contamination of birds and wildlife. For example, Texas Administrative Code, Title 16, Part 1, Chapter 3, Rule §3.22(b) Protection of Birds requires that an operator "screen, net, cover or otherwise render harmless to birds" specific tanks and pits with "frequent surface film or accumulation of oil," but does not address the potential exposure of birds or cattle to radioactive materials. Proper reserve pit management techniques include fencing cattle out of areas to prevent livestock from drinking reserve pit contents. Consumption of reserve pit fluids by livestock has been documented to cause poisoning, abortions, birth defects, weight loss, contaminated milk, and death [31, 32].

Proper public health protection may involve stringent quality controls upon agricultural and farm practices, to prevent exposure to reserve pit waste materials, and controls on harvest and food movement to prevent exposures to workers and the public. The presence of radioactive materials in agricultural soils and food products can create financial hardship and a significant psychological impact for communities whose economic base consists of agricultural and food products. Many of the radionuclides have long half-lives, which can result in contamination of the soil for decades. This ultimately could affect the marketability of both the land and any products produced from the land for decades.

Federal Regulatory Oversight

Neither the U.S. Environmental Protection Agency (EPA) nor the U.S. Nuclear Regulatory Commission (NRC) has established federal regulations that directly govern NORM waste from the oil and gas industry. In fact, wastes containing NORM are generally not regulated by federal agencies with one exception, transportation. NORM-containing wastes with a specific activity greater than 2,000 pCi/g (70 Bq/g) are subject to U.S. Department of Transportation (DOT) regulations governing transport of radioactive materials [33]. The Occupational Safety and Health Administration (OSHA) has promulgated rules specific to occupational exposure to ionizing radiation [34] , which may be applicable to petroleum industry NORM management activities.

By definition, oil and gas industry NORM that does not exceed 0.05 percent uranium or thorium by weight or any combination, is not subject to regulatory control under the Atomic Energy Act of 1954 due to the fact it is not a source material, special nuclear material, or by-product material [35].

The Low-Level Radioactive Waste Policy Act as amended in 1986 provides guidance to states on disposal of low-level radioactivity material, like the waste material generated from oil and gas activities, but does not include oil

and gas NORM waste. NORM wastes generated during the exploration, development, and production of crude oil, natural gas, and geothermal energy have been categorized by the EPA as "special wastes" and are currently exempt from federal hazardous waste regulations under Subtitle C of the Resource Conservation and Recovery Act (RCRA) by the Bevill Amendment and are not considered a listed or characteristic waste. The Superfund Amendments and Reauthorization Act listed none of the constituents of NORM as "extremely hazardous substances." The Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) lists radionuclides as hazardous substances because the CAA (Clean Air Act) lists them as hazardous air pollutants. Oil and gas waste streams that may contain NORM are exempt under RCRA and therefore considered not hazardous substances under CERCLA, although individual radioisotopes might be. Reportable Quantities (RQs) are one pound of radionuclides (cumulative), or concentrations expressed in curies for individual radionuclide, whichever is less (40 CFR 302.4).

In 1989 EPA issued a final regulation covering RQs for radionuclides. EPA used 1, 10, 100, 1000, and 5000 pounds as RQs for non-radionuclides and 0.001, 0.01, 0.1, 1, 10, 100, and 1,000 Curies (Ci) as RQs for radionuclides. Release values for approximately 760 radionuclides were calculated for each of four human health intake pathways. The lowest pathway release value for each radionuclide was selected and then rounded down to the nearest decade to set the RQ for each radionuclide. Radionuclides not having published intake limits were assigned an RQ of 1 Ci, based on the observation that 91 percent of the radionuclides being studied were below the 1 Ci level [36]. These RQ are not applicable to oil and gas exploration as a result of the RCRA Bevill Amendment and its relationship to CERCLA.

The EPA under the CAA developed National Emission Standards for Hazardous Air Pollutants (NESHAPs) specific to radionuclide emissions for several sources, but not for industrial activities that include NORM generated by the oil and gas industry.

The EPA under the provisions of the Safe Drinking Water Act (SDWA) regulates the following radionuclides in drinking water: (adjusted) gross alpha emitters, beta particle and photon (gamma) radioactivity, ^{226}Ra and ^{228}Ra (combined), and uranium. The EPA established drinking water standards for several types of radioactive contaminants: $^{226/228}$Ra (5 pCi/L); beta emitters (4 mrems); gross alpha standard (15 pCi/L); and uranium (30 μg/L).

State Regulatory Oversight

NORM is subject primarily to individual state radiation control measures and varies across the nation. "Section 651(e) of the Energy Policy Act of 2005 gives NRC jurisdiction over discrete sources of NORM by redefining the definition of source material" [37]. For example, the State of Texas has three

agencies are responsible for regulating different aspects of NORM. In Texas, NORM is regulated under the Texas Radiation Control Act (TRCA) as follows:

- The Texas Department of State Health Services (TDSHS), Radiation Control, has jurisdiction over the receipt, possession, use, treatment and storage of NORM (TDSHS NORM Licensing).
- "The Railroad Commission of Texas (RRC) has jurisdiction of handling and disposal of NORM wastes produced during the exploration and production of oil and gas (RRC rules for NORM)" [37], and disposal by the owner through on-site land farming and/or injection well. "The Texas Commission on Environmental Quality (TCEQ) has jurisdiction over the disposal of other NORM wastes" [37].

Under such a system, the Texas Administrative Code (TAC) defines exemptions for persons (parties/agencies) who receive, possess, use, process, transfer, transport, store, and commercially distribute NORM; that is, an exemption does not need to be licensed or is not regulated since NORMs are not hazardous waste streams. Often these exemptions are based on the NORM concentration of the waste stream being below a certain activity level (pCi/g) or radiation level (microRoentgens per hour μR/hr). Radium radionuclides are generally the measured standard for multiple radionuclide waste streams, while a higher exemption threshold is used for an individual radionuclide. This system requires the determination of nuclide concentration or emission only when a disposal permit is sought. Ponds used to store and receive waters from drilling, well rework, and hydraulic fracturing operations can be filled without determining radionuclide release or impact since they are not technically considered hazardous waste and no disposal permit is required.

The environmental management of lands contaminated with naturally occurring radioactive materials will require threshold guidance levels to be established to indicate when action is required. Successful management will need federal and state authority to enforce such threshold guidance levels. Unless regulatory loopholes are closed, testing, monitoring, and reporting of radionuclide release to the environment above existing background will continue, resulting in more human and environmental exposure. Guidelines for NORM/TENORM should correspond to levels of naturally occurring radionuclides in the environment at which it is practical to distinguish the radionuclides resulting from human activities from those in the undisturbed natural background. In 2008, the National Council on Radiation Protection and Measurements summarized the issue of radiation exposure and public health in the following statement: "There is a need to address public health concerns and to provide guidance on the cleanup and potential reuse of lands contaminated with NORM or technologically-enhanced NORM (TENORM). Although there are environmental cleanup standards in place for manmade radioactive contamination, there are no consistent federal or state regulatory controls or environmental

management policies for NORM or TENORM contamination resulting from industrial practices associated with processing natural metal and mineral resources" [35].

Recommendations

Historically, ^{226}Ra and ^{228}Ra have been tested for in water and guidance levels set with the intention of protecting people from exposure to radon gas. The findings of this study raise the question of whether radium, a single radionuclide, is the proper indicator for assessing radiation exposure levels to the general public, given the potential for the vast amount of radioactive waste, and number of radionuclides, produced from oil and natural gas exploration and mining that may be present in reserve pits. Current regulations require that ^{226}Ra and ^{228}Ra combined exposure levels not exceed 5 pCi/g, averaged over 100 m^2, identifying radon as the primary emission of concern [39]. The Texas RRC Commission can issue a permit for the burial of oil and gas NORM waste "if, prior to burial, the oil and gas NORM waste has been treated or processed so that the radioactivity concentration does not exceed 30 pCi/g ^{226}Ra and ^{228}Ra or 150 pCi/g of any other NORM nuclide" [40]. These limits were not established with the support of public health/medical professionals nor based on potential human health impacts of cumulative exposures to multiple radionuclides. The total beta radiation found in one sample (RP2.1-North) of this study of 1329 pCi/g exceeds regulatory guideline values by more than 800 percent. However, individual radionuclides did not exceed existing regulatory guidelines. Data from this limited field study showed that elevated levels of alpha, beta, and gamma radiation were present in reserve pit water/sludge material and also in the soil of a decommissioned reserve pit. Evaluating the single radionuclide radium as regulatory exposure guidelines indicate, rather than considering all radionuclides, may indeed underestimate the potential for radiation exposure to workers, the general public, and the environment.

Limitations to this study include the small sample size and limited analysis of reserve pit contents. The study does not make the assumption that all reserve pits contain radioactive materials. The study does not imply that all reserve pit contents are disposed of by land farming (either onsite or offsite) or postulate the extent to which contaminated material is incorporated back into the earth. Comparison of radionuclide levels found in this study to existing regulatory levels was difficult since regulatory guidelines have been established for only a few radionuclides. Furthermore, TENORM waste has been excluded from many regulatory guidelines and from regulatory oversight. Future studies are needed to evaluate what percentage of reserve pits are actually used for deposition of radioactive materials. Further studies are needed to understand how radioactive materials transfer to vegetation and animal products and the uptake mechanisms of those materials through the food chain. The long half-lives that

are intrinsic to many radionuclides are a major concern for future generations. Further research needs to be done to understand what exposure levels can be anticipated given the complex interactions within the physical and chemical components of soil and the lack of uniformity of soil chemistry.

As the United States goes forward with the expansion of drilling natural gas reservoirs (especially drilling in shale, which requires hydraulic fracturing with millions of gallons of water and producing nearly equal amounts of flowback), it is imperative that we obtain better knowledge of the quantity of radioactive material and the specific radioisotopes being brought to the earth's surface from these mining processes. Proper regulation of surface deposits and disposal of wastes can prevent elevation of natural levels of radiation and increased exposure of animals and humans to potentially harmful levels of radioactivity. It is essential that the public health community be consulted when establishing future regulatory guidelines. Materials classified as exempt under current regulations should be reviewed given the potential for adverse health effects from radiation exposure to the general public and with continued growth of urban drilling.

AUTHORS' BIOGRAPHIES

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