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Marcellus Shale

THE SCIENCE BENEATH THE SURFACE

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Understanding Naturally Occurring Radioactive Material in the Marcellus Shale

A discussion of the naturally occurring radioactive material resulting from natural gas drilling in the Marcellus Shale.

DID YOU KNOW?

- The yearly radiation dose for a person who smokes 1.5 packs of cigarettes a day is equivalent to about 320 chest x-rays.
- In Brazil, monazite sand deposits found along certain beaches have radiation levels up to 5 mrad/hr, which is almost 400 times the normal background radiation in the US!
- The cosmic radiation dose rate doubles with each 2 km increase in altitude. So, the radiation dose a person receives while flying is about 1 millirem per 1000 miles flown.

Introduction

The Marcellus shale is a gas-bearing shale formation that ranges geographically from Ohio and West Virginia into Pennsylvania and southern New York. It was deposited around 390 million years ago in the shallow sea that once covered the region. Shale is composed of tiny mud particles and organic matter, which, because of the properties of radioactive elements in sea water, often contains concentrations of naturally occurring radioactive material (NORM). There has been concern that drilling in the Marcellus shale will bring NORM to the surface in a concentrated form, which could pose a radiation hazard. Some of the details surrounding this issue are addressed in this pamphlet.

What is NORM?

Naturally Occurring Radioactive Materials (NORM) are radioactive substances that exist in all natural media: soils, rocks, water, and even air.

NORM is often present in high concentrations in gas-bearing shale, and may be brought to the surface via drill cuttings and other waste from an oil or gas well. NORM can be concentrated by human actions (i.e., drilling and processing ores). This concentrated, technologically (human) enhanced naturally occurring radioactive material is called TENORM. This pamphlet will explain both NORM and TENORM, but will reference both as NORM.

What is Radioactivity?

Radioactivity is the spontaneous emission of energy (radiation)-and sometimes sub-atomic particles-by unstable atoms. Radiation becomes a concern when these particles and energy come in contact with molecules inside the human body, because this interaction can result in cell damage or death.

Radioactivity can be found everywhere in the universe - even you. The degree of radioactivity, however, varies greatly among different materials. Food, for example, is radioactive, but usually at extremely low levels that are not detectable unless measured with very sensitive laboratory instruments. In fact, 0.01% of all potassium, a mineral vital to all living things, is radioactive.

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Table 1. Natural Radioactivity in Food

Food	^{40}K pCi/g	^{226}Ra pCi/g
Banana	3.52	0.001
Brazil Nuts	5.60	1-7
Carrot	3.40	0.0006-0.002
White Potatoes	3.40	0.001-0.0025
Beer	0.390	--
Red Meat	3.00	0.0005
Lima Bean, Raw	4.64	0.002-0.005
Drinking Water	--	0-0.00017

Table 1. A summary of naturally occurring radioactivity in food.[1] ^{40}K stands for the radioactive isotope No. 40 of potassium. ^{226}Ra stands for the radioactive isotope No. 226 of radium. The table is measured in pCi/g (picocuries per gram), which is a conventional unit for measuring the amount of radioactive material contained in a gram of a substance.

In a given year, we are exposed to an estimated 620 millirems of radiation. This is considered "background" radiation.

A rem is a measurement of radiation dose; a millirem (mrem) is 1/1000 of a rem. Almost half of background radiation comes from radon gas, which is a product of uranium-238 (via radium-226, explained in more detail in the next sections), and is present throughout the Earth. An individual's radiation exposure varies from place to place and from activity to activity. Medical x-rays and airplane flights can dramatically increase yearly exposure; one CAT scan can give you several rem (and therefore several thousand mrem) of exposure. To calculate an estimate of your own yearly dose, visit EPA's website at: http://www.epa.gov/radiation/understand/calculate.html.

Federal and State radiation regulations allow up to 100 mrem/year radiation dose to members of the general public from the licensed use of radiation-producing equipment or radioactive materials. This level is adopted as a conservative limit, and is many times below levels demonstrated to cause measurable effects in a population.

What are the Different Types of Radioactivity?

The three most common types of radiation are alpha particles, beta particles, and gamma rays.

Alpha particle emission: Alpha particles (α) are highly charged, have a large mass compared to other radioactive particles, and consist of two protons and two neutrons. They are emitted from the nucleus of an unstable atom. The size and charge of alpha particles limits their penetration to very short distances. Most alpha particles are stopped by a few centimeters of air, a sheet of paper, or the outer (dead) layer of skin on our bodies. Though their size and energy make them potentially one of the more dangerous radioactive particles, they are easily blocked and usually not of much concern unless the material that emits them is ingested or inhaled.

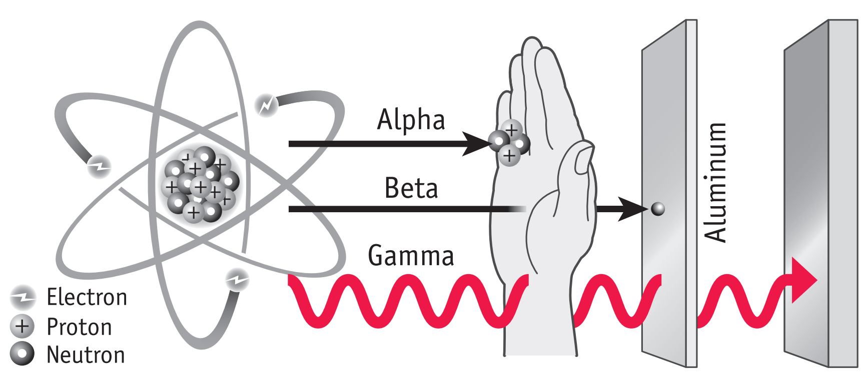
Beta particle emission: Beta particles (β) are the equivalent of energetic electrons and have significantly less mass and half the electrical charge of alpha particles. Because they are not as highly charged, beta particles do not have as many interactions with other particles. Therefore, they travel further before giving up all their energy and coming to rest. Beta particles have a limited penetrating ability. Their typical range in air is up to about 10 feet. In human tissue, beta particles can travel only a centimeter or so at most, so they are primarily of concern to eyes and skin as well as being an internal hazard if the material that emits them is ingested.

Gamma ray emissions: Gamma (γ), or x ray, radiation is made of photons, which travel at the speed of light and have no electrical charge or mass. As a result, they are less likely to interact with anything, giving them very high penetrating power and allowing them to travel great distances through different materials. Due to its high penetrating power, gamma radiation can result in radiation exposure to the whole body rather than a small area of tissue near the source.

Common Naturally Occurring Radioactive Elements

Some elements have isotopes, which occurs when a chemical element has the same number of protons (atomic number) as another, but a different number of neutrons, and thus a different mass. Radioisotopes are isotopes with an unbalanced number of protons and neutrons. This results in

Figure 1. The basic components of an atom, the building block of all matter, are pictured to the left. Radioactive particles emitted from an atom have different degrees of penetration, with alpha having the least and gamma the most, as shown to the right.



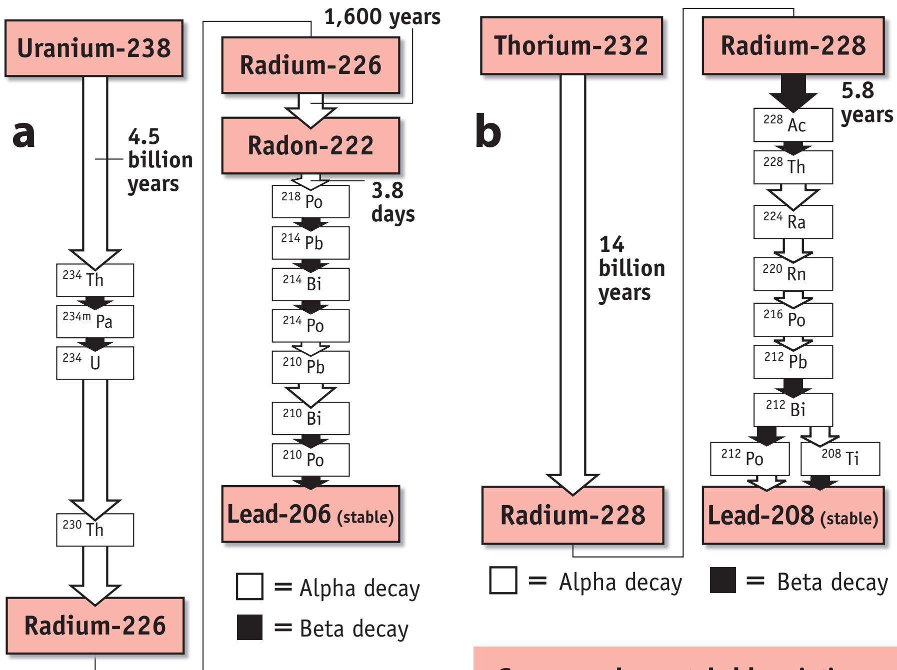


Figure 2. Decay sequence leading to the main elements associated with radioactivity in the Marcellus shale by means of a) ^{238}U and b) ^{232}Th decay.

Common elemental abbreviations

U = uranium

Th = thorium

K = potassium

Ra = radium

an unstable nucleus which undergoes radioactive decay until it is stable. For example, uranium-238 (^{238}U) and ^{235}U have similar chemical behavior, but have different atomic masses, and thus different physical and radioactive properties.

Primordial radioisotopes, such as ^{238}U, and thorium-232 (^{232}Th), are those that originated from the fusion of multiple elemental nuclei during one or more supernovae (star collapse) over 6 billion years ago. These isotopes have not all decayed because their *half-lives* (the amount of time it takes for half of a quantity of a given radioactive element to decay) are so long that there are still many atoms left to decay.

Secondary radioisotopes (also known as daughter isotopes), like radium-226 (^{226}Ra), are the decay products of some primordial radioisotopes. ^{238}U and ^{232}Th decay through a series of radioactive elements, including ^{226}Ra and ^{228}Ra respectively, while ^{40}K, another primordial radionuclide, decays directly to stable (non-radioactive) argon-40 (^{40}Ar) or calcium-40 (^{40}Ca).

Why does the Marcellus Shale have NORM?

Black shale, such as the Marcellus, often contains trace levels of ^{238}U, ^{235}U, ^{40}K, and ^{232}Th in higher concentrations than found in less organic-rich grey shales, sandstone, or limestone. This is because: 1) ^{238}U and ^{235}U preferentially bond to organic matter, like algae that die and settle to the bottom of the ocean; and 2) ^{40}K and ^{232}Th preferentially bond to clays, which compose much of the sediment at the ocean floor.[2] Ultimately, because "black shales" contain more organic matter and clays, they are generally more radioactive than other shales or sedimentary rocks.[3] This radioactivity can be measured with sensitive equipment at Marcellus outcrops or equipment can be lowered into an uncased well, with radioactivity readings used to identify, map, and determine the thickness of black shales below the surface.

Is NORM Unique to Marcellus Shale Drilling?

For decades the oil and gas industry has been using natural radioactivity to locate potential sites for drilling. In 1981, in the North Sea, operators first began to identify scale deposits as being radioactive.[4] Scale is the accumulation of mineral deposits, such as calcium carbonate, that precipitate out of water and onto the inside of pipes and other equipment. Measurements of scale from the North Sea contained ^{226}Ra at high concentrations between 2,700 and 27,000 pCi/g, as well as containing ^{226}Ra in sludges (muds resulting from the mixture of brine and fracturing fluids with the rock) from 130 to 1,300 pCi/g.[5]

Results from a 1999 investigation of NORM in oil and gas wells of NYS showed maximum ^{226}Ra concentrations of 11 pCi/g in scale (scale on the outside of a pipe) and about 7 pCi/g in sludge.[6]

The radioactivity in observed New York State samples for scale and sludge are about 100 to 1000 times less than the North Sea oil field samples.

The Barnett Shale, which underlies northern Texas, is the first location in which natural gas was extracted from a tight shale through horizontal drilling and hydrofracturing. Like the Marcellus, the Barnett is a deeply buried shale with natural gas trapped in unconnected pore spaces within the rock.[7] However, Barnett shale wells have been drilled longer, and more data is available on NORM concentrations. For the Barnett, ~97% of all 5900 NORM readings collected were below 0.05 mR/hr, overlapping measured Marcellus radioactivity values, with the remaining

Table 2. NYS NORM Data:

Native Rock NORM values	^{226}Ra	0.2-2.4 pCi/g
	^{228}Ra	0.2-0.8 pCi/g
	^{40}K	3.3-28 pCi/g
Native Soil NORM values	^{226}Ra	0.8-1.1 pCi/g
	^{228}Ra	1.3-1.6 pCi/g
	^{40}K	16-26 pCi/g

Table 2. NORM values found throughout New York State in native rocks and soils.[9]

3% ranging from 0.052 to 1.1 mR/hr.[8] Values of 0.1 mR/hr or greater have not been measured in the Marcellus, as seen in Figure 4, but were recorded in the Barnett.

Common units for measuring radioactivity include: pCi/g or pCi/L (picocuries per gram or liter), which are conventional units measuring the concentration of radioactive material in a substance, and R/hr or mR/hr (roentgens or milliroentgens per hour), units for measuring exposure, which are defined only for effect of gamma rays on air. Data has been collected in both Curies and Roentgens as well as other units, and unfortunately these units do not relate to each other well enough to allow for direct conversion.

Nearly all rocks and soil contain some levels of uranium & thorium, their daughter products, and ^{40}K. The concentrations of these isotopes vary based on the type of rock. In order to understand how elevated NORM is in the Marcellus shale, average New York State rock NORM values are needed for comparison. These measurements are reported in picocuries per gram, a conventional unit for measuring the amount of radioactive material in a gram of a substance. The radioactivity of New York State rocks and soils are shown in Table 2 and Figure 3. These numbers are based on a DEC sample set of 7 rocks from 4 different geologic units, and 4 soil samples.

How Radioactive is the Marcellus Shale?

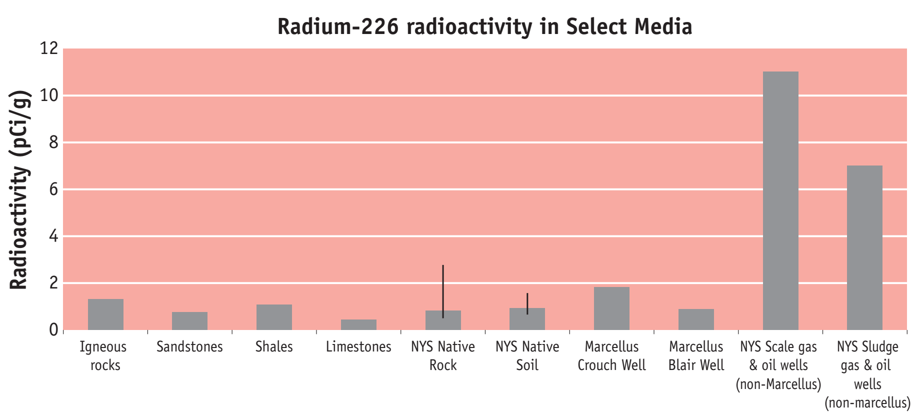
Primary radioactive materials found in the Marcellus shale include ^{40}K as well as ^{238}U and ^{232}Th and their decay products. These primary radioactive minerals have half-lives of a billion plus years and consequently are not very radioactive. Also, uranium and thorium are generally insoluble, more likely to adhere to rock and soil than to be carried along in fluids, making concentration of the elements unlikely.[10] The decay products of ^{40}K; ^{40}Ar and ^{40}Ca, are not a radioactive hazard either, as they are stable isotopes. ^{238}U, however, decays to more soluble ^{226}Ra, and ^{232}Th decays to more soluble ^{228}Ra, both radioactive products.

^{226}Ra has a half-life of 1,600 years, compared to 5.75 years for ^{228}Ra, and according to the DEC is the primary radioisotope of concern in shale-gas drilling.[11]

Because of their relative solubility over long time periods, ^{226}Ra and ^{228}Ra have a greater chance of being concentrated as they preferentially migrate into solutions like brine (salty, mineral-rich water trapped within the rock during its deposition), which can mix in with hydraulic fracturing fluids. A portion of this resulting mix is pumped back out of the well and is generally referred to as flowback water. There is not enough time to allow the direct migration of radium from the Marcellus shale to hydraulic fracturing fluids. Thus, radium is present in flowback water only if hydraulic fracturing fluids have mixed with brine already present in the formation. Concentrations of NORM in flowback water are often initially relatively low because proportionally more hydraulic fracturing fluid is being pumped out. However, NORM levels have the potential to climb over time as the fluid being extracted begins to contain proportionally greater amounts of brine.

Radioactivity measurements for the Marcellus shale are limited due to the relatively small number of wells that have been drilled in New York State and the small number of samples that have been analyzed and reported in all states with underlying Marcellus shale. In order to supplement this data, NORM measurements from other NYS wells will be presented, in addition to data

Figure 3. Average ^{226}Ra measurements from various samples including igneous rocks, sandstones, shales, limestones.[12]*, background samples of New York State rocks and soils, New York State gas well scale (mineral deposits) and sludge (muds)*[13]*, and two Marcellus shale wells.*[14] *Marcellus-specific data was very limited, due to the few wells drilled into the unit in New York. For comparison, North Sea radioactivity in oil and gas wells found to be a concern in the 1980s ranges from 2,700-27,000 pCi/g for scale and 130-1300 pCi/g for sludge.*[15]



from gas-shale in other localities.

Exposure to NORM

We know that NORM exists in the Marcellus Shale, but does that pose a hazard to workers and the general public? Using data from Marcellus shale cores, outcrops, and well sites, an estimate of the risk of radiation exposure can be calculated. For this data, the units used (mR/hr) only apply to gamma radiation. Since gamma radiation has the greatest penetration potential, this data can provide a benchmark estimate for continuous exposure to NORM in the Marcellus shale. In order to translate this data to dose equivalent rate (mrem), for which safety standards exist, we use a rough 1:1 conversion of milliroentgens to millirems. Again, this conversion is not exact, as rem factors in the biological effect of exposure while roentgen is a measure of energy.

Bringing Potentially Concentrated Marcellus NORM to the Surface

Natural gas drilling can bring NORM to the surface through cuttings, flowback fluid and produced brine, and scale accumulation in pipes and tanks.

1. Cuttings are chips of rock cut by the drill bit and brought to the surface by the drilling fluid. DEC analysis of samples of Marcellus shale cuttings suggest that their NORM levels are similar to those in other black shales, and are not likely to pose a problem. Additional samples analyzed by CoPhysics Inc of drill cuttings collected on site at various Pennsylvania Marcellus Shale wells and of those that have been transported to New York State landfills suggest that NORM in cuttings will not pose a public health threat. The rock cuttings are the same material as outcrops of the Marcellus shale.[17,18]

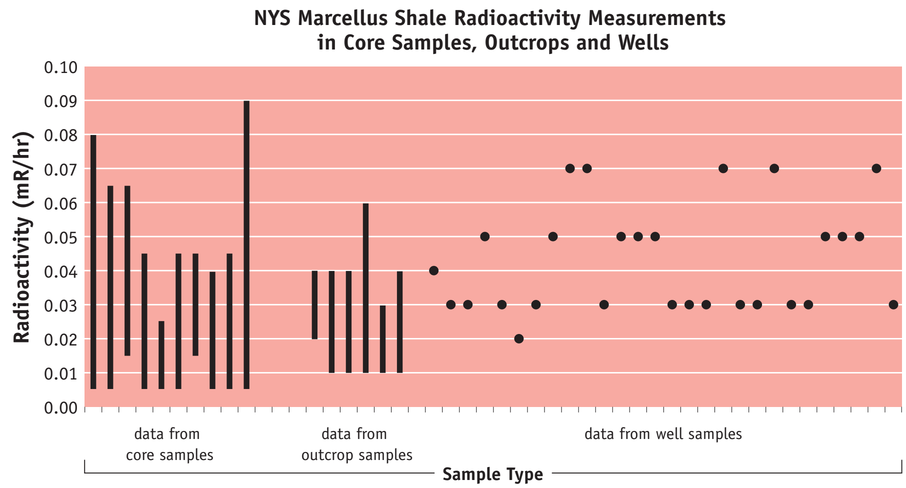


Figure 4. Specific measurements of radioactivity within the Marcellus Shale reported in mR/hr. The maximum recorded reading is 0.09 mR/hr ≈ 0.09 mrem/hr, and will be referenced as mrem/hr for subsequent exposure safety calculations. The government regulated additional continuous exposure rate for the general public is 0.011 mR/hr (100 mrem/year), while for occupational workers the limit is 2.5 mR/hr (5,000 mrem/year).[16]

2. Flowback fluid is a mix of hydraulic fracturing fluid and formation water that is pumped back out of the well during well development, after the hydraulic fracturing process is complete. Water is already present in the Marcellus formation from when the sediment was originally laid down by a shallow sea. Dissolved metals and the salt present in this water make it brine water. After a well is developed, the volume of fluid removed decreases and is made up of less hydraulic fracturing fluid and more formation water, which contains a higher proportion of NORM. Levels of NORM in flowback water are elevated above those from average rock background levels.

^{226}Ra and ^{228}Ra were measured in flowback water from13 different wells within the Marcellus shale. ^{226}Ra ranged from 0.163 to 16,030 pCi/L, while ^{228}Ra ranged from 0.0286 to 957 pCi/L.[19] Because the proportion of brine to hydraulic fracturing fluid cannot be determined without well data, the range of these values cannot be used to calculate average NORM values for the Marcellus.

The 1999 Investigation of NORM in oil and gas wells of NYS shows a broad range in radioactivity among 43 brine samples. Unlike the limited data reported from the Marcellus in NY, all of these samples are considered to be of produced water composed primarily of brine. ^{226}Ra varies from not detectible to 3,800 pCi/L and radium-228 from not detectible to 24,000 pCi/L (Figure 5). Only 2 of the 43 samples measured were over 10,000 pCi/L, so the 24,000 pCi/L value is anomalous. Note that the data from the 1999 Investigation of NORM does not include samples from the Marcellus, but from other comparable shales and sandstones in NYS. The little Marcellus-specific data that exists shows a range of ^{226}Ra and ^{228}Ra from 0 to 16,030 and 0 to 1,252 pCi/L, respectively[20] ^{226}Ra is elevated above readings from general NYS wells, and ^{228}Ra is similar. The Marcellus has elevated NORM levels because it is generally more organic-rich than other shales and radioactive elements bond

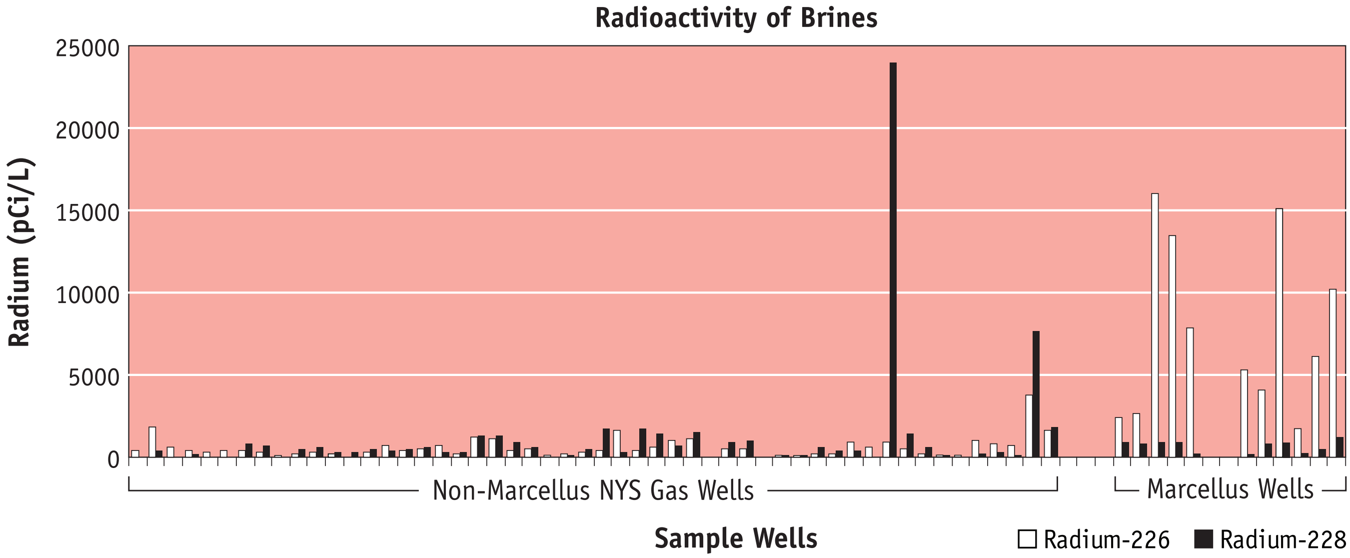


Figure 5. Radium isotope ranges in brines from Marcellus shale wells[22] and other gas wells found in NYS.[23] Marcellus shale wells show elevated ^{226}Ra values when compared to other NYS wells.

to organic matter.

3. Scale is a mineral deposit (for example, calcium carbonate) that precipitates out of water and adheres to the inside of pipes, heaters, and other equipment.

There is no available data on NORM in scale from the Marcellus Shale. However, measurements of 10 gas wells from other rock formations across New York show maximum ^{226}Ra concentrations of 11 pCi/g and ^{228}Ra of 3.8 pCi/g with average concentrations of 1.46 and 0.64 pCi/g.[21] These are low concentrations that are not hazardous. However, due to the elevated NORM levels found in some brines, it is possible that scale from Marcellus wells could show more elevated NORM levels. Scientific reviewers of this pamphlet expect this to be the only scenario that could possibly produce enough radiation to have a measurable exposure rate. Most states already involved in similar drilling processes (and NYS would likely follow suite) require periodic radiation monitoring of the pipes. If a certain level is reached a radioactive materials license is required and with that comes regulation for potential worker exposures, public exposure, and waste disposal.

What are the Health Risks Associated with Disposal of NORM-Concentrated Brine?

In the 1999 Investigation of NORM, the DEC Bureau of Pesticides & Radiation used the Residual Radioactive Material Guideline computer model (RESRAD) to construct a realistic NORM brine "beneficial use" disposal scenario involving road applications of brine to estimate the resulting radioactive dose to a member of the public. RESRAD was developed for the U.S. Department of Energy to calculate site-specific residual radioactive material guidelines, radiation doses, and the lifetime cancer risk to repetitively exposed on-site residents. The brine road application scenario was chosen as this is the most common method of brine disposal by New York State. In addition to being used on paved and unpaved roads for ice and snow control, brines are also occasionally sprayed in the summer on dirt roads to control dust.

The RESRAD parameters included a standard application rate of 1/3 gallon per square yard and two applications per each significant snow fall event. With an average of 20 snow fall events per year for 20 years, the resulting application would be 13.3 gallons per square yard per year or roughly 60 liters per square meter.

This scenario assumed no losses of radium due to runoff, which is unlikely but provides us with a worst case estimate, and a direct pedestrian exposure of two hours per day, 300 days per year. Assuming both an inhalation and direct ground hazard, the dose rates were calculated for the highest brine results measured in the study: 950 pCi/L ^{226}Ra and 24,000 pCi/L ^{228}Ra and 3,800 pCi/L ^{226}Ra and 7,700 pCi/L ^{228}Ra. The resulting dose equivalent after 20 years of application under this scenario was estimated at 2.9 mrem/yr and 1.7 mrem/yr respectively. This is the radiation equivalent of one dental x-ray or sleeping next to someone for 8 hours.

Using these "worst-case" assumptions, RESRAD showed that the dose rate upon the 20th year of exposure from all radium isotopes and their decay products would still be well below the DEC cleanup guideline of 10mrem/yr. Based on the very conservative nature of the model the DEC maintains that there is no public health or environmental threat.[27]

Coming into Contact with Radioactivity from the Marcellus Shale

When thinking about the hazard associated with NORM in the Marcellus shale it is important to distinguish between the radioactivity of the shale itself and the radioactive dose that a person in contact with the shale could receive. The following calculations provide some perspective on the risk from ongoing close exposure to the shale.

For the general public, the exposure limit for "licensed activities" set by the EPA is 100 mrem/yr of whole body exposure.[24] The maximum recorded reading from Marcellus shale to date is equivalent to 0.09 mrem/hr (see figure 4); this value is taken from direct measurements of the rock. Taken at face value, it may at first appear that this shale is well above the federal limit: 0.09 mrem x 24 hr/ day x 365.25 days/yr yields an annual value of nearly 800 mrem/yr (for comparison, average background radiation is about 620 mrem/yr).[25] It is important to note, however, that such a calculation is not technically correct, as it assumes full body contact with the shale over this entire interval. There is no feasible scenario in which either a member of the general public or a worker would receive either full body or year-long contact with the shale, much less both. The impact of radioactivity declines to almost zero within several inches (alpha particle) to feet (beta particles) of the shale. Gamma rays, which have a greater penetration, follow the inverse square law, where radiation at a given distance from a source varies inversely with the square of its distance. In other words, doubling the distance from a radiation source will reduce the dose to $1/4^{th}$ of its original value, and tripling the distance will reduce the dose to $1/9^{th}$ of its original value. Thus, those in the general public with the most contact with the Marcellus shale–those with homes on the Marcellus shale, or with shale outcrops on their property–are not likely to receive more than a few percent of the exposure limit from the shale itself.

Workers at drill sites may be more likely to be in regular contact with Marcellus shale, and the current regulatory occupational exposure limit in the United States for occupations working around radiation, as set by the Occupational Safety and Health Administration (OSHA), is 5,000 mrem per year (50 times the level for the general public).[26] Assuming constant exposure on the worksite for 2000 working hours per year (40 hrs/day at 5 days a week for 50 weeks), the exposure limit is about 2.5 mrem/hr. This is much higher than the Marcellus shale maximum of 0.09 mrem/hr.

In sum, radiation levels in the Marcellus shale itself are sufficiently low that they are not expected to affect the public or drill site workers. The primary radiation exposure concern from the drilling process is concentrating mechanisms, such as the build-up of scale-precipitation of salts from the brine within drilling equipment and the fate of concentrated NORM in drilling wastewater. For more on NORM and wastewater disposal, see Marcellus Issue Water: Out of the Wells.

As for the Marcellus formation, there have been a few cases of brine spreading from conventional Marcellus wells. However, this is relatively rare due to the large amount of dissolved solids, such as salts and minerals, in the brine make it generally unusable for road spreading. Since New York State does not currently have proper facilities to dispose of waste with even a minimal radioactive signature, it is likely that this waste will be shipped out of state. It is possible that brine from unconventional drilling in the Marcellus could still be disposed of by road spreading, but that is pending final revisions to New York's Generic Environmental Impact Statement (GEIS), and will be decided on a case by case basis by the DEC if analysis shows it would not pose a public health threat.

Summary

We encounter NORM on a daily basis, and the amount of NORM acceptable for an individual varies. The different types of radiation include alpha, beta, and gamma radiation, with gamma being of the greatest concern to the public from NORM. Black, organic-rich shales, like the Marcellus, can be more radioactive then other rock types because radioactive elements bond to clays and organic material within the rock. The radioactive decay products of uranium and thorium (radium-226 and radium-228) are of greatest concern in Marcellus Shale gas-drilling because of their solubility and potential for concentration in brines and scale. The effect on any individual person depends upon the mechanisms that concentrate NORM, as well as the type and period of exposure. As each person is different, the effect of radiation on them varies. It is not possible to determine what amount of radiation is "safe" so current regulations are designed to be conservative.

With limited data available on

NORM in the Marcellus, it is hard to completely understand the potential hazard; however, available data seems to show levels similar to that of the Barnett shale, and lower than values recorded in North Sea drilling in the 1980s. Marcellus Shale NORM values are elevated with respect to other New York State shales and sandstones. For non-Marcellus brine, computer models predict that even if NORM concentrated brines were disposed of on pedestrian-traveled roads, the radiation dose to the public would not be elevated above background levels. The potential for Marcellus brine disposal via road spreading from unconventional drilling is still pending governmental review and, if permitted by the DEC, will be on a case by case basis.

Overall, it is unlikely that the general public will ever come into contact with NORM in any significant concentrations.

Some workers at drilling sites may come into contact with NORM concentrated material (scale and production water). In such cases, measurements of the radioactive materials are monitored according to regulatory standards.

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Partnering Organizations include

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