# RADIOACTIVITY IN MARCELLUS SHALE
# CHALLENGE FOR REGULATORS AND WATER TREATMENT PLANTS

**Marvin Resnikoff**[a]

[a]Radioactive Waste Management Associates, Bellows Falls, VT 05101, USA

## ABSTRACT

Studies by the U.S. Geological Survey and gamma logs of drillers show radium concentrations up to 32 times background concentrations in the Marcellus shale formation. Brought to the surface in rock cuttings, drilling fluids, flowback water and brine, radium can enter the environment in several forms. A fraction will be reinserted into deep disposal wells, or go to water treatment plants. Because of its high salinity, a portion will be spread on highways in the winter. The rock cuttings will go to solid waste landfills. Over the production cycle, a portion of Marcellus radium plates out on pipes. And an inert radioactive gas, radon, will enter homes when natural gas is used for heating and cooking.

This paper examines the fate and transport of radionuclides brought to the surface, and the environmental impact in the environment. Regulators have the task of developing regulations that protect the health and safety of the population. The water treatment industry must develop processes for separating radium from large volumes of waste waters so that surface streams meet regulatory limits. This paper explores these different waste streams and potential resolutions of these differing problems.

## INTRODUCTION

Geologists consider the Marcellus shale formation to be relatively highly radioactive and regionally extensive. Radioactivity in the Marcellus shale results from the high content of naturally occurring radioactive uranium and thorium, including their decay products, and potassium elements in the rock. In New York State the formation ranges from 25 to over 100 feet in thickness and the depth of the base varies from an outcrop to 1000-foot depth by Syracuse to 4000-foot depth by the border with Pennsylvania (Hill, 2994).

The areal extent of Marcellus shale is shown in Figure 1. As seen, the formation underlies the States of New York, Pennsylvania, eastern Ohio, West Virginia, Virginia, all the way into Alabama.

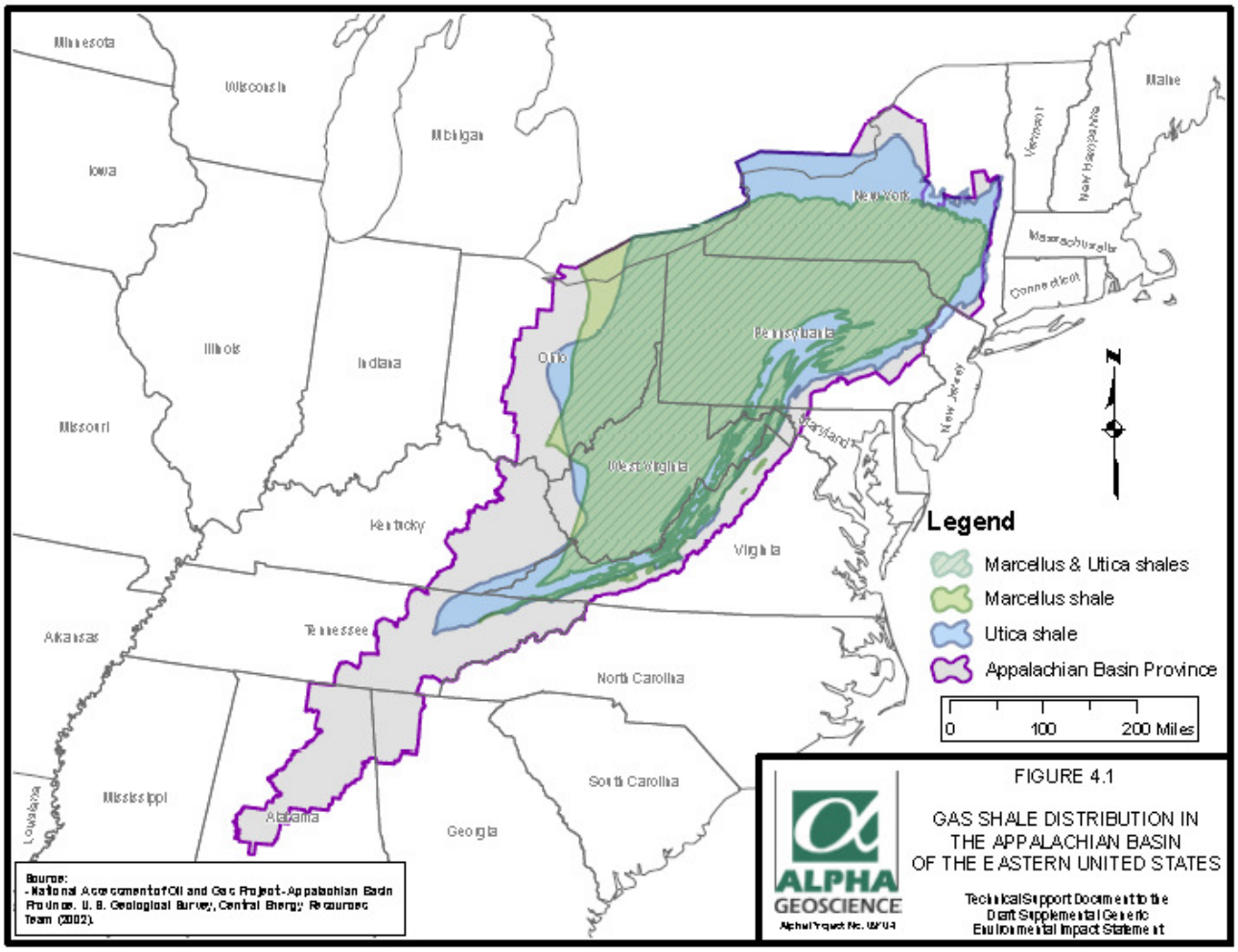


**Figure 1. Areal Extent of Marcellus Shale** (NYSERDA, 2009)

The depth to the top of the Marcellus formation in New York State is shown in Figure 2. As seen, the formation is deeper as one goes south.

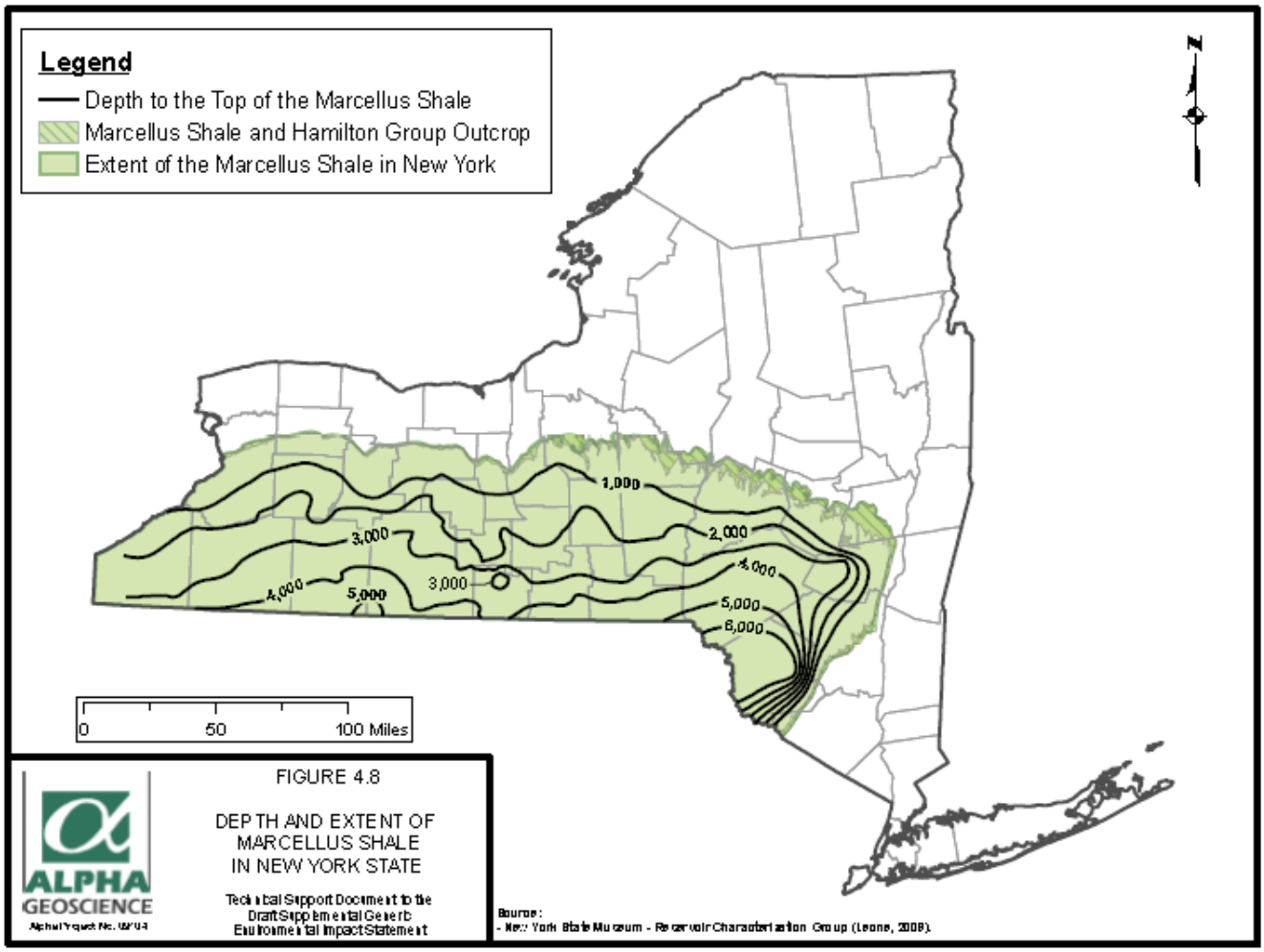


**Figure 2. Depth of Marcellus Shale in New York State** (NYSERDA, 2009)

In addition to the depth of the Marcellus formation, the thickness varies as well. In New York State the Marcellus shale thickness increases from less than 25 feet in the western section of the State, Chautauqua County, to 300 feet in the southeastern section of the State, as seen in Figure 3. The dots represent exploratory gas wells.

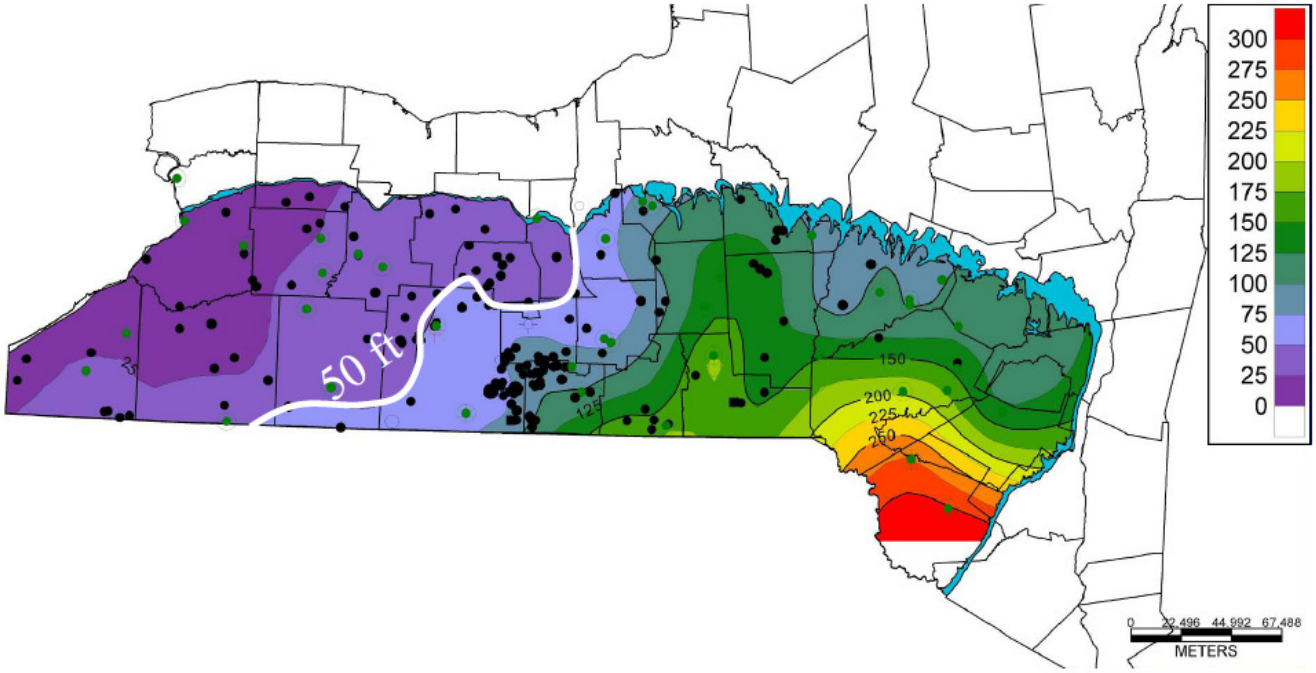


**Figure 3. Marcellus shale thickness in New York** (NYSERDA, 2009)

## RADIOACTIVITY IN MARCELLUS SHALE

Most disinterested scientists agree that the Marcellus shale contains uranium-238, thorium-232 and their decay products at concentrations significantly above background. The issue is: how much radioactivity above background? In general, the radioactivity throughout the vertical depth of rock cuttings appears to be equal to or less than 10 picocuries per gram, including K-40. However, at certain depths in each well, the activity is significantly higher. The gas industry knows it has reached the Marcellus shale horizon when cores show high gamma ray and high total organic carbon content (TOC). Consider the log from Beaver Meadows (Figure 4). This well is located near Norwich in Chenango County in upstate New York.

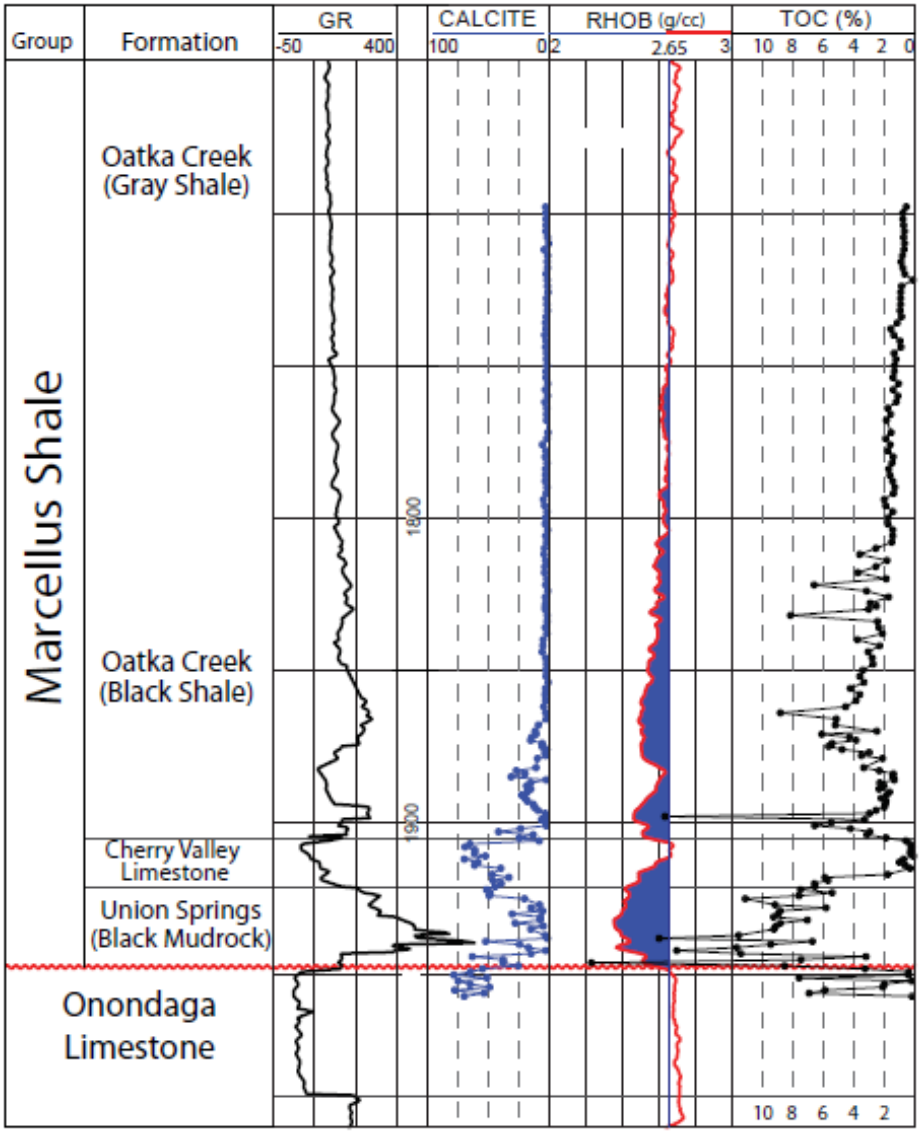


**Figure 4. Beaver Meadows Core** (AAPG, 2010)

As one goes from the shale layer (Union Springs) to the limestone layer (Onondaga Limestone), one sees a major drop in gamma ray and total organic content. Without going into a discussion of GAPI units, 16.5 GAPI represents 1 pCi/g radium, and 400 GAPI is 24.2 pCi/g radium. As you can see, the GAPI is much greater than 400 units, and this also corresponds to very high TOC. These radium concentrations are far higher than background radium concentrations in New York State (Myrik, 1981), which are 0.85 pCi/g. Other gamma ray logs wells down to the Marcellus shale horizon in New York State such as the Shiavone 2, WGI11 and Bergstrasser wells show similar results.

The logs mentioned above correspond to a geochemical study of trace elements and uranium in the Devonian shale of the Appalachian Basin (Leventhal, 1981) carried out in 1981 by the United States Geological Survey (USGS). The Devonian layer refers to sediment formed

350 million years ago from mud in shallow seas. Since the layers do not form in a line parallel to the ground surface, the depth at which Marcellus is found can vary from surface outcroppings to as deep as 7,000 feet or more below the ground surface along the Pennsylvania border in the Delaware River valley (NYDEC, 2011), and as deep as 9000 feet in Pennsylvania [2].

The USGS study analyzed seventeen cores from wells in Pennsylvania, New York, Ohio, West Virginia, Kentucky, Tennessee, and Illinois. The researchers collected a variety of geochemical data to be used for resource assessment and identification of possible environmental problems. Rather than direct gamma spectroscopy, uranium was measured in each core with a more appropriate and precise method, delayed-neutron analysis. I use data from the USGS since it is a reputable and objective government agency.

Although the cores varied in thickness and in depth, geologists identified the Marcellus Shale stratum in several cores using data on the organic matter (carbon), sulfur, and uranium content of the samples. Table 1 below summarizes the results from four cores that tapped into the radioactive Marcellus formation. The depths at which the layer was found as well as the uranium measurements are presented.

| Location of the Core | Depth of Sample (feet) | Uranium Content (ppm) |
|---|---|---|
| Allegheny Cty, PA | 7342 – 7465 | 8.9 – 67.7 |
| Tomkins Cty, NY | 1380 – 1420 | 25 – 53 |
| Livingston Cty, NY | 543 – 576 | 16.6 – 83.7 |
| Knox Cty, OH | 1027 – 1127 | 32.5 – 41.1 |

**Table 1. Uranium Content and Depth of Marcellus Shale in Four Cores**

The four cores were taken from different geographical locations, but the characteristics of the identified Marcellus shale layer, specifically the high uranium and carbon content, are consistent.

To compare the uranium content in parts per million (weight) to radioactive concentration in picocuries per gram, we use the correspondence (HPS, 2007):

$$2.97 \text{ ppm} = 1 \text{ pCi/g U-238}$$

Using this relationship, the U-238 ranges up to 28 pCi/g, or 32 times background for radium-226, assuming U-238 and Ra-226 are in secular equilibrium, as it is in the Marcellus Shale formation. That is, the USGS measurements and the GAPI logs are consistent. This is our starting point for the concentrations of Ra-226 in the natural Marcellus Shale formation. The radium itself is found in the pore water of the Marcellus Shale formation, since radium, under the temperature, pressure and chemical conditions, is preferentially dissolved in the pore water.

## FATE AND TRANSPORT OF RADIOACTIVE MATERIALS

If higher levels of radioactivity are present in the Marcellus shale formation, as shown by gamma logs, USGS measurements, and statements by DEC, where will this radioactivity appear in the aboveground environment and how will DEC regulate this radioactive waste?

To discuss where uranium, radium and other radionclides enter the environment, we consider two components of natural gas development: well development and production.

**Well Development**

We are concerned about the horizontal, not the vertical, component of natural gas wells. As seen in Figure 5, fracking consists of pumping a fluid, proppants and chemicals under high pressure to

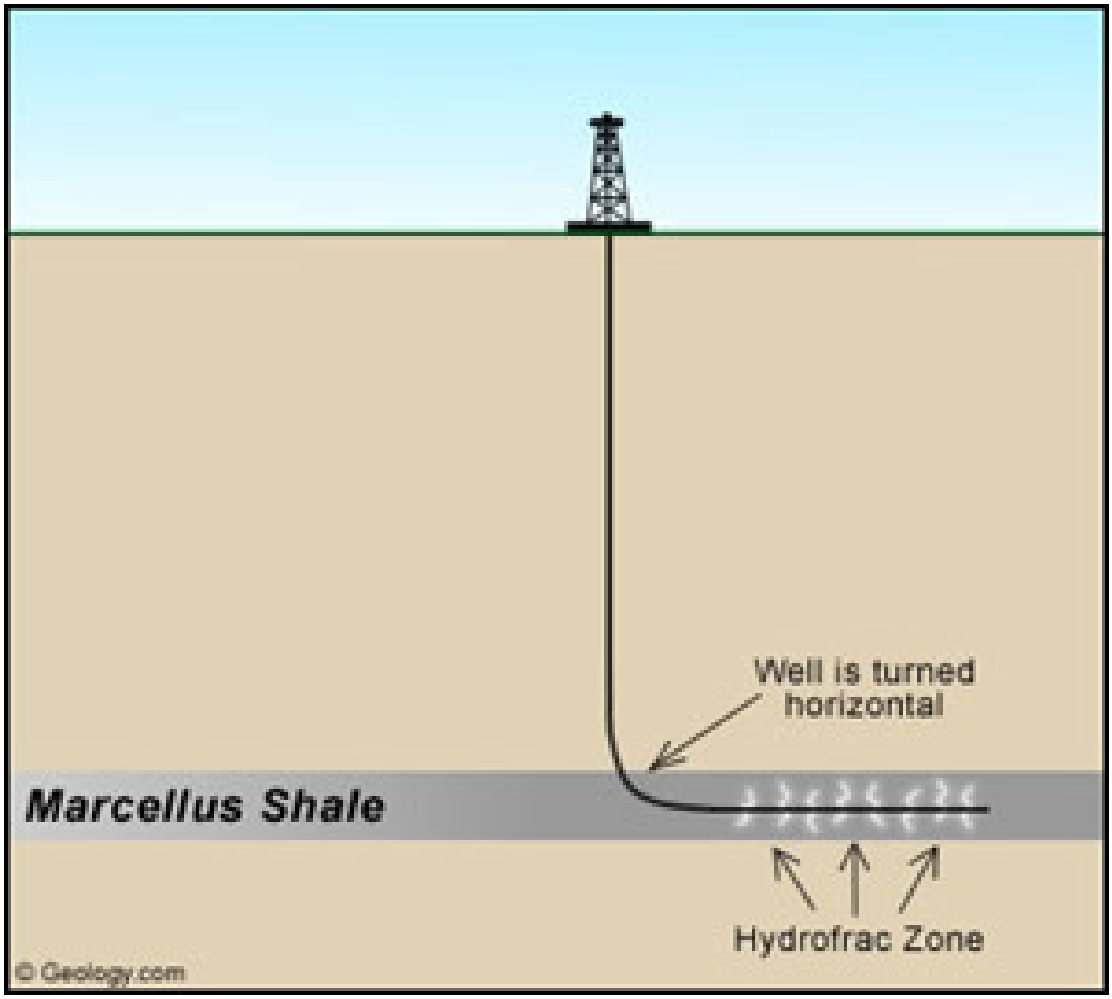


**Figure 5. Fracking**

create fractures along the horizontal region. I'm not going to discuss other chemicals that may contaminate properties, such as arsenic, mercury and hydrocarbons. The fracture zone may extend up to 200 foot radius from the horizontal bore. The bore itself may extend horizontally up to a mile. One 4,000-foot lateral wellbore undergoing hydraulic fracturing may require between 2.4 million and 7.8 million gallons of water. (NYDEC, 2009). During well

development, rock cuttings are produced. These are separated from the drilling fluid by shakers, shown in Figure 6.

**Figure 6. Rock cutting shakers**

**Rock Cuttings**

The drilling fluid itself may be partially recycled and the rock cuttings have been going to solid waste landfills. Radium is located in the water and not in the shale rock itself. But solid waste that goes to a solid waste landfill can be, according to NYS regulations, up to 20% water. An industry lab measured rock cuttings with a gamma detector; the rock cuttings were shown to be slightly radioactive. On the other hand, NYDEC, in their 2009 Draft Supplemental GEIS, reported radioactivity for two wells in Lebanon and Bath, NY, total radioactivity concentrations of 25.4 +/- 4.6 and 29.2 +/- 4.3 pCi/g, respectively, which is consistent with what we expect from the gamma logs and USGS findings. The EPA limits for radium in surface soil (EPA, 2011) are 5 pCi/g in the top 15 cm, and 15 pCi/g below 15 cm. These rock cuttings, above EPA limits, should be disposed of in a licensed facility for handling radioactive materials, such as Energy Solutions in Clive, Utah. DEC and the Ohio EPA believe they have no jurisdiction over NORM, but if drilling fluid is recycled, the NORM is technologically enhanced. I'm not a lawyer, but it appears to me that these agencies do have jurisdiction over TENORM, if recycled drilling fluids are intermixed with rock cuttings.

**Flowback Water**

After a well is drilled, part of the hydraulic fracturing fluid flows back to the wellhead. This flowback water constitutes about 10 – 40% of the original pumped volume. (NYTimes, 2011) Since Marcellus shale is of marine origin, it contains high levels of salt and NORM which is dissolved in the fluid. (Cornell, 2010) DEC found 13 samples of flowback water from vertical Marcellus shale wells in Schuyler, Chemung and Chenango counties to contain radioactive concentrations as high as 267 times the limit for discharge into the environment and thousands of times the limit for drinking water. (Davies, 2009) Flowback water from horizontal drilling will be much more radioactive. If flowback water is fed to a water treatment plant, the radium needs to be removed, as we'll discuss shortly.

**Well Production**

Once a natural gas well is producing, salt water or brine is brought up with the natural gas, and is separated out at the water gas separator. DEC has sampled brine for radium-226 and radium-228. The results are shown in Table 2 below.

| Well | Town | County | Ra-226 | Ra-228 |
|---|---|---|---|---|
| Maxwell 1C | Caton | Steuben | 7885 | 234 |
| Frost 2 | Orange | Schuyler | 2647 | 782 |
| Carpenter 1 | Troupsburg | Steuben | 5352 | 138 |
| Webster T1 | Orange | Schuyler | 16030 | 912 |
| Calabro T1 | Orange | Schuyler | 13510 | 929 |
| Schiavone 2 | Reading | Schuyler | 15140 | 957 |
| WGI 11 | Dix | Schuyler | 10160 | 1252 |

**Table 2. Radioactivity in Brine Water (pCi/L)**

The radium concentrations in brine are very high. While no one is drinking brine, just to compare these concentrations with drinking water standards, 5 pCi/L combined radium-226 and 228. That is, the radium concentrations in brine range up to 3000 times safe drinking water limits. Under the proposed DEC regulations (NYDEC, 2010), a drill applicant must have a plan to deal with flowback water and brine, and that is where water treatment specialists enter the picture. A practice, which has since been suspended, is to spread brine, as a de-icing solution, on highways in New York State.

Just to get a handle on the magnitude of the problem. If hydraulic fracturing were approved, DEC estimates 1600 drilling applications per year; each horizontal well will produce between 2.4

million and 7.8 million gallons of flowback water. So we estimate 3.8 to 12.5 billion gallons of contaminated water per year that has to be treated. This is a small volume compared to the water usage in New York State, but this type of waste poses additional problems.

In Pennsylvania, much of the 1.3 billion gallons of radioactively-contaminated water produced between 2007 and 2010 was sent to sewage treatment plants. Most sewage plants in the State are not required to monitor for radioactivity, so there are little data. According to sewage plant operators interviewed by the Times (NYTimes, 2011), these plants are not equipped to remove radioactive material.

As a potential method of treating flowback water and brine, one can investigate the method of treating uranium mine waste water, which has high concentrations of radium. Here radium is precipitated with barium chloride to produce barium sulfate $Ba(Ra)SO_4$. Radium is then a particulate that can be filtered. Water treatment plants may then meet SPDES permit limits, and the filters will have to be sent to a licensed landfill. Other methods include cation exchange softening, lime softening, and sorption onto $MnO_2$. A physical process being proposed by Atela, Inc and Casella Waste Systems is distillation, low pressure evaporation. The sludge would then have to be disposed of in a licensed landfill. Volatiles from this process would have to be captured. It is not clear whether municipal water treatment plants can handle the magnitude of the radioactive waste problem posed by flowback water and brine, so that will be a challenge to water treatment professionals, such as you.

**Radium scale buildup in gas equipment.**

During production, radium dissolved in water, is brought to the surface. Scale, radium sulfate, plates out on production pipe surfaces. Scale also appears in water/gas separators, feeder lines and condensate tanks. As DEC states, a high concentration of scale will result in an elevated radiation exposure level at the pipe exterior surface. (NYDEC, 2011) The NYS Department of Health (DOH) proposes a radioactive materials license when exposure levels exceed 50 microR/hr (µR/h). (NYDEC, 2012b) Our calculations show that the radium concentrations for an external dose rate of 50 µR/h are far higher than the EPA limit of 5 pCi/g. We have found that exposed workers have an increased risk of developing cancer. Workers at, and residents near, pipeyards that clean pipe scale will have an additional risk from inhaling radioactive dust. Based on our experience, the proposed DOH regulations are too lax, as we discuss below.

For DEC and DOH to grasp the magnitude of the problem, we provide one example. At one natural gas well in Texas, 388 pipe joints were pulled after 5 years service. Exposure levels exceeded 50 µR/h in 55% of the 30 foot joints (max, 150 µR/h) 38% were < 50 µR/h and 7% were free of NORM. Similarly, hundreds of pipes at each gas well in New York and Pennsylvania will be contaminated with radium scale. If thousands of gas wells are drilled in New York State, how will DEC and DOH have the resources to regulate the industry and track these contaminated pipes? In our experience, oil and gas producing pipes with high external exposure levels have been "donated" to city governments for playgrounds in Texas, or to farmers for use in animal corrals in Texas and Kentucky. They have been cut up with oxyacetylene torches and welded to fit their use. In the process, children and farmers have been directly

exposed to gamma, and inhaled radium. Workers at pipeyards that cleaned pipes have inhaled radium-contaminated dust and have developed cancer.

We find that direct gamma exposure levels of 50 μR/h are much too high. In order to determine the concentrations of Ra-226 and Ra-228 that correspond to a dose rate of 50 μR/hr, we employed the program MicroShield (Grove, 2008), a program used to estimate dose rates due to a specific external radiation source.

As inputs to MicroShield, we assumed an outer pipe diameter of 4 inches (10.16 cm), a scale thickness of 0.2 cm, and a pipe wall thickness of 0.91 cm, as suggested by the US EPA EPA, 1993b). We could also have used another standard pipe diameter, 2 7/8 inch. We assumed that each contaminated pipe is 30 feet long, and that radiation measurements had been taken at the center of the pipe, on contact with the outer pipe wall. From MicroShield, for a pipe with external gamma of 50 μR/h, we obtain a Ra-226 concentration in scale of 1,313.5 pCi/g, and a Ra-228 concentration in scale of 437.8 pCi/g that correspond to a dose rate of 50 μR/h. We assumed a 3 to 1 ratio of Ra-226 to Ra-228.

As DEC and DOH are well aware, the EPA cleanup standard for total radium on soil is 5 pCi/g for the first 15 cm depth and 15 pCi/g 15 cm or more below the surface. Pipes, with external radiation 50 μR/h greatly exceed this standard. If these pipes, with NORM <50 μR/h are released for general use, they will be cut up and welded and the scale will be accessible. Ground contamination could easily exceed 5 pCi/g or 15 pCi/g. Though the 50 μR/h limit has been instituted in several States, in my opinion the DOH-suggested dose rate of 50 μR/h is much too high.

While DEC and DOH intend to regulate joints with external radiation > 50 μR/h, in practical terms, it is not clear what the State agencies envisage will become of these radioactive pipes (and separators, feed lines and condensate tanks). Once the oil and gas industry begins production in New York State, this will be a major unresolved problem down the road.

**Radon in Natural Gas**

Yet another significant public health hazard associated with drilling for natural gas in the Marcellus Shale formation that should be seriously investigated by the New York State Department of Environmental Conservation (DEC) and other State agencies is the radon hazard. This hazard has the potential for large numbers of lung cancer among natural gas customers. This issue, which has been ignored in the DEC's Draft Supplemental Environmental Impact Statement (NYDEC, 2011), must be addressed in a revised Impact Statement and before DEC issues any hydraulic fracturing drilling permits in the Marcellus shale.

Unlike present sources for natural gas, located in Texas and Louisiana, the Marcellus Shale is considerably closer to New York consumers. In addition, the radioactive levels at the wellheads in New York are higher than the national average for natural gas wells throughout the United States.

In a paper we recently submitted to a journal (RWMA, 2012), RWMA calculated the wellhead concentrations of radon in natural gas from Marcellus Shale, the time to transit to consumers, particularly New York City residents, and the potential health effects of releasing radon, especially in the smaller living quarters found in urban areas.

It is well known that radon (radon-222) is present in natural gas (ATSDR, 1999) (NRC, 1988) and has been known since the early 1900's (Gesell, 1975). Published reports by R. H. Johnson of the U.S. Environmental Protection Agency (Johnson, 1973) and C. V. Gogolak of the U.S. Department of Energy (Gogolak, 1980) address this issue in depth, as we discuss below. Radon is present in natural gas from Marcellus Shale at much higher concentrations than natural gas from wells in Louisiana and Texas.

Since radon is a decay product of radium-226, to calculate radon levels it is necessary to know the concentrations of radium-226. Based on a USGS study (Leventhal, 1981) and gamma ray logs (also known as GAPI logs) that we have examined, the radium concentrations in the Marcellus Shale is 8 to 32 times background. This compares to an average radium-226 in surface soil in New York State of 0.81 picoCuries per gram (pCi/g).

Using this range of radium concentrations and a simple Fortran program that simulates the production of radon in the well bore, and transit to the wellhead, we calculate a range of radon concentrations at the wellhead between 36.9 picoCuries per liter (pCi/L) to 2576 pCi/L. The maximum calculated concentration in Marcellus shale, 2576 pCi/L, is higher than the maximum concentrations found in the Panhandle region of Texas, 1450 pCi/L. (Johnson, 1973) (Gogolak, 1980).

These wellhead concentrations in Marcellus shale are up to 70 times the average in natural gas wells throughout the U.S. The average was calculated by R. H. Johnson of the U.S. Environmental Protection Agency in 1973 (pre-fracking) to be 37 pCi/L (Johnson, 1973) to a maximum of 1450 pCi/L.

In addition, the distance to New York State apartments and homes from the Marcellus formation is 400 miles and sometimes less. This contrasts with the distance from the Gulf Coast and other formations, 1800 miles. At 10 mph movement in the pipeline, natural gas containing the radioactive gas, radon, which has a half-life of 3.8 days, will have three times the radon concentrations than natural gas originating at the Gulf Coast, everything else being equal, which it is not.

Being an inert gas, radon will not be destroyed when natural gas is burned in a kitchen stove.

We have examined published dilution factors and factored in numbers for average urban apartments where the dilution factor and the number of air exchanges per hour are less than those of non-urban dwellings. This analysis implies that the radon concentrations in New York City and urban apartments will be greater than the national average.

We assume 11.9 million residents in New York State are affected. This figure is calculated in the following manner: Based on U.S. Department of Energy figures our calculations assume 4.4

million gas stoves in New York State (EIA, 2009). This figure is multiplied by 2.69 persons per household to determine the number of residents affected: this number equals 11.9 million.

We calculate the number of excess lung cancer deaths for New York State. Our results: the potential number of fatal lung cancer deaths due to radon in natural gas from the Marcellus shale range from 1,182 to 30,448.

This is an additional number of lung cancer deaths due to radon from Marcellus Shale over deaths from natural radon already impacting New York State homes and their residents. The Draft Supplemental Environmental Impact Statement produced by the New York State Department of Environmental Conservation (NYDEC, 2011) needs to be revised to take into account this public health and environmental hazard. In the entire 1400 page statement there is only one sentence containing the word "radon" and no consideration of this significant public health hazard.

Further, NYDEC needs to independently calculate and measure radon at the wellhead from the Marcellus Shale formation in presently operating wells before issuing horizontal hydraulic drilling permits in New York State. The presence of processing plants and storage must also be taken into account, and can serve to reduce the radon concentrations in natural gas delivered to consumers. The present rdsGEIS by DEC discusses none of these issues and should be rewritten.

**CONCLUSIONS**

Studies by the U.S. Geological Survey and gamma logs of drillers show radium concentrations up to 32 times background concentrations in the Marcellus shale formation. Brought to the surface in rock cuttings, drilling fluids, flowback water and brine, radium can enter the environment in several forms. During well development, rock cuttings and flowback water containing radium will be bought to the surface. Flowback water from horizontal hydraulic fracturing in the Marcellus formation has high concentrations of radium that must be disposed in a deep well or properly separated at a water treatment facility; the radioactive filters or sludges must go to a licensed landfill. During well production, radioactively-contaminated brine, up to 3000 times safe drinking water limits, must also be treated or disposed in a deep well. Radium-contaminated scale will form on the down well pipes as well as feeder lines and condensate tanks and these must be safely decontaminated; direct gamma from these pipes will increase the radiation dose to workers and the general public. Workers at pipeyards have developed cancer from direct gamma radiation and inhaling radioactive dust. The scale must be sent to a licensed disposal facility. Radioactive radon gas in natural gas from the Marcellus shale formation, much closer to the end use market than natural gas from Texas and Louisiana, will enter homes through kitchen stoves. Unless this natural gas is treated or held up, increased radon concentrations in homes and urban apartments will cause an increase in lung cancer rates.

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