Review of Pennsylvania Department of Environmental Protection Technologically Enhanced Naturally Occurring Radioactivity Materials (TENORM) Study Report

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By Marvin Resnikoff, Ph.D.

Radioactive Waste Management Associates Box 105 Bellows Falls, VT 05101

> For Delaware Riverkeeper Network Bristol, Pennsylvania

Introduction

It has been known since the 1960's that the Marcellus shale formation is radioactive¹. Drilling logs by gas companies² and reports by USGS³ show that underground/subsurface radium concentrations in the Marcellus shale are up to 32 times surface background concentrations. More recent measurements by New York State DEC show radium in rock cuttings over 200 times background concentrations⁴. Drilling and natural gas production brings this radioactivity to the surface in the form of solids (rock cuttings), liquids (drilling fluids, flowback water and brine), and gas (radon).

This is not alchemy, where lead is magically turned into gold, or in the case of Marcellus shale, where radioactivity below ground, magically disappears when brought to the surface. Contaminated liquids, gases and solids will enter the accessible environment and be taken in by the public, increasing the likelihood of cancers. Radium-226 has a half-life of 1,600 years, so it will be present in the environment for thousands of years. It is also water soluble, meaning it easily travels with water via streams and rivers. One of its decay products, radon, is an inert gas, allowing it to travel with natural gas and enter homes through kitchen stoves,⁵ and from fugitive gas emissions throughout the natural gas distribution network. As we discuss later, recent studies show radon concentrations in homes in Pennsylvania are on the rise.

The Pennsylvania Department of Environmental Protection (DEP) undertook a study (the "DEP Study") to assess the environmental and public health impact of technologically enhanced naturally occurring radioactive material (TENORM) related to oil and natural gas production in Pennsylvania. DEP appointed Perma-Fix Environmental Services, Inc. (PESI) to undertake the study on its behalf. The report was issued in January 2015.⁶ PESI operates five nuclear waste treatment facilities throughout the country. In my professional opinion, an independent consultant without investments in an industrial activity that is regulated by DEP would have been a more appropriate choice without the risk of bias or conflict of interest.

This report critiques DEP and PESI's study of TENORM related to oil and gas development, by examining its methodology and the available data. It is Delaware Riverkeeper Network's (DRN) position that DEP has not released all of the data gathered for this study, so our review is limited to the data released to DRN through the Right to Know Law and information published on DEP's website.^{7 8}

¹ Swanson, 1960.

² Resnikoff, 2010.

³ Ibid.

⁴ Allied, 2012.

⁵ Resnikoff, 2012.

⁶ <u>http://bit.ly/1Yeb6BW</u>

⁷ Department of Environmental Protection v. Delaware Riverkeeper Network, 1373 C.D. 2014 (Pa. Cmwlth. 2015).

⁸ The opinions, including the conclusions, in this report, stated to a reasonable degree of scientific certainty, are those of the author. As more information becomes available, I reserve the right to supplement this report.

Hydraulic fracturing or "fracking" in Pennsylvania started in earnest in 2008, however DEP did not begin its study of the radioactive waste produced there until 2013. The first Marcellus shale wells numbered about 375 wells from 2004-2007, but drilling began in earnest in 2008. ⁹ Since gas operators began drilling, over nine thousand unconventional Marcellus Shale wells have been drilled¹⁰; and the DEP has issued over 16,000 drilling permits, and along with those, over 5,000 on-site drilling violations have been issued.¹¹

What is hydraulic fracturing of Marcellus Shale?

The Marcellus shale formation lies at depths of 4,000 to 8,500 feet¹³ below the Earth's surface and ranges from West Virginia through eastern Ohio, across Pennsylvania, and into southern New York. The new technology has allowed this major expansion of gas wells and involves directional drilling, specifically horizontal drilling into shale formations, with high pressure, and has opened shale formations and allowed the release of natural gas. New York State presently has a moratorium on high volume hydraulic fracturing in tight shale formations, including Marcellus Shale, but drilling is extensive in Ohio, West Virginia and primarily Pennsylvania. Differing from older gas and oil wells, wells in the Marcellus shale formation are drilled a mile or more vertically to reach the shale formation and then drilled a mile or more horizontally through the shale formation. After casing the well bore, holes are punched in the horizontal tubing with explosives, and fluid is pumped under high pressure, fracturing the tight shale geologic formation to release the gas trapped in the rock.

The use of fracking has increased dramatically in the U.S. In the years 2011 to 2014, roughly 25,000 to 30,000 new oil and gas wells were hydraulically fractured each year. For Pennsylvania, specifically, the number of hydraulically fractured gas wells that were producing waste fluid was estimated as 1,232 in 2010, but increased to a total of 5,015 in 2013,¹⁴ and 6,987 by March 2014.¹⁵

Fracking requires a large quantity of water to complete the drilling process - on average 5 million gallons of water per well¹⁶. In terms of millions of gallons of wastewater produced in Pennsylvania: in 2010, 180 million gallons, in 2011, 740 million gallons, and in 2013, 1300 million gallons were produced. Drilling fluid is used to remove the rock cuttings from horizontal wells in the Marcellus shale formations and to transport the drill cuttings to the well surface.

⁹ <u>http://geology.com/articles/marcellus-shale.shtml.</u>

¹⁰ DEP counts wells according to conventional and unconventional. These definitions are in the state regulations and are based on depth of formation and drilled horizontally within the formation—Marcellus wells are unconventional wells. The definition of unconventional well is found at 25 Pa. Code, Ch. 78.1.

¹¹ Total Unconventional Wells as of Mar. 27, 2015", 04.24.2015, provided by The FracTracker Alliance on FracTracker.org.

¹²<u>http://www.depreportingservices.state.pa.us/ReportServer/Pages/ReportViewer.aspx?/Oil_Gas/Wells_Drilled_By_County</u>.

¹³ EPA, 2015, p. 2-5.

¹⁴ EPA, 2015, p. 8-8.

¹⁵ Total Unconventional Wells as of Mar. 27, 2015", 04.24.2015, provided by The FracTracker Alliance on www.FracTracker.org.

¹⁶ Iowa, 2014.

Chemicals and proppants are added to the fluids to stimulate and facilitate gas extraction; while many of the formulas are proprietary, it is well known that the mixtures contain hazardous and toxic substances.¹⁷

There are two types of liquid waste water generated in the process: flowback water and produced water or brine. Ten to forty percent¹⁸, or an average of 25%¹⁹ of the drilling fluid plus interstitial Marcellus liquid is returned within a few days; this is called flowback water and is typically released to lined holding ponds near the drill site. Over time, particles settle out in the pond and form a sludge at the lined pit bottom. Some operator use tanks to hold the flowback water and particles settle to form a sludge at the bottom of the tank. When the gas well is in production, additional water is brought up with natural gas and is separated at the well site and moved into condensate tanks, or is trucked to central waste treatment (CWT) or deep wells for disposal. This produced water has high concentrations of radium-226 and its decay products, up to 18,000 pCi/L or more radium-226 and 2,500 pCi/L radium-228²⁰, a decay product of thorium-232. While no one is drinking this salty solution, as a yardstick, drinking water standards are 5 pCi/L combined Ra-226 and Ra-228.

How are these drilling wastes disposed? Solids are separated from liquids at the drill site. The contaminated solids are shipped to municipal landfills, occasionally tripping portal monitors because the radioactivity is above permissible levels. Liquid from the settling ponds has been generally transferred to local publicly owned wastewater treatment plants known as POTWs or to CWT facilities, and then released to surface waterways.²¹

Flowback and produced water are injected into deep wells for long term storage or transported to centralized water treatment facilities or CWTs that discharge to surface water. Transportation to deep wells or centralized treatment facilities is generally by truck. Flowback water is also recycled in hydrofracking operations at well sites and may be transferred from one well site to another by pipeline. Some solids such as rock cuttings and residual waste such as materials remaining in a pit after wastewater is removed, if they meet certain standards and permitting requirements, can be buried on the well site or applied to the land. Some cuttings or other solids are trucked to landfills or, if containing regulated materials, to treatment facilities.

¹⁷ EPA recently issued a list of 692 chemicals, many are regulated toxics. NYSDEC RDSGEIS also lists chemicals, many are toxic and have adverse health effects. EPA citation: <u>http://www2.epa.gov/hfstudy/appendix-chemicals-identified-hydraulic-fracturing-fluids-and-wastewater-excel-file</u> or, as they show it: US EPA. 2012. Study of the Potential Impacts of Hydraulic Fracturing on Drinking Water Resources: Progress Report. EPA 601/R-12/011. Available at http://www.epa.gov/hfstudy.

¹⁸ Urbina, 2011a.

¹⁹ Haluszczak, 2012.

²⁰ USGS, 2011.

²¹ US EPA has proposed effluent regulations for oil and gas extraction that prohibits the discharge of unconventional oil and gas wastewater to POTWs; the rule is expected to be adopted as final in 2016. Available at http://www2.epa.gov/eg/unconventional-extraction-oil-and-gas-industry.

What did PESI seek to measure?

As we discuss in detail below, under the DEP plan, the following waste streams were to be sampled for radioactivity: holding ponds, streams, flowback water, brine, influent and effluent at water treatment plants, rock cuttings and radon gas at the well head. We sought to investigate these waste streams and compare what PESI measured with what has been previously measured elsewhere and compare the results to safe levels.

In April 2013, PESI prepared field sampling and quality assurance plans for the Pennsylvania DEP. These plans outlined a "comprehensive study investigating the NORM and TENORM related to the oil and gas exploration activities including conventional and unconventional drilling through geological formation(s) and associated waste water operations throughout the Commonwealth of Pennsylvania (PA)." This ambitious study plan presented a wide array of components associated with drilling activities and included an evaluation of TENORM in ambient air, drilling cuttings, natural gas, natural gas processing pipes and equipment, waste water generated on drilling sites, sludge resulting from the processing of waste water from the well pad development process and landfill leachate.

PESI intended to take samples related to the operations, equipment, and features pertaining to "the drilling and production of natural gas from these geologic units and also in the transfer of water to POTWs and CWTs for processing. Landfill leachate would also be sampled to study whether radium had migrated from POTW and CWT sludge to the landfill leachate." PESI took samples related to:

- Vertical and horizontal drill cuttings
- Onsite pits containing cuttings
- Production and Flowback water
- Filter socks and filter presses
- Compressed gas lines
- Off gassing
- Well pads
- Centralized impoundments
- Waste water facility sludge
- Waste water facility influent and effluent water
- Fresh proppant sands
- Drilling muds
- Piping and casing scale

Sampling locations are shown below in Fig. 1. Below the figure is a 2013 preliminary summary of the type of sampling data collected; additional data were collected in 2014. It is unclear whether DEP has reported all the data collected, or just a selected sample. We have identified gaps in DEP's data numbering system and sampling locations were not provided for all sites. Delaware Riverkeeper Network attempted to obtain all records through a Right to Know Request.



Figure 1. Locations of Sampling Sites. The red dots indicate well sites where data were collected.

The number of samples and types are listed below, as reported to the Citizens Advisory Council, February 2014:

The field work portion of the Study commenced on April 15, 2013. As of December 31, 2013, 184 field visits had taken place at 114 individual locations across the state. These efforts resulted in the collection of 911 samples that have been processed by the DEP Laboratory. The field work was conducted at a variety of facility types including the following:

- > 20 well pads were visited through the various phases of development on 41 occasions.
- > 3 rounds of waste water treatment plant sampling at 25 facilities were completed on December 16, 2013.
- > 13 sites were sampled in the northern part of the state where brine from conventional gas wells was applied for dust suppression.
- > A facility that stores well casings from decommissioned conventional wells was visited for the collection of radiation surveys of that material.
- ➢ 7 facilities that compress, store, and utilize natural gas for electricity generation were visited for data collection.
- > 48 landfills across the state were visited for sampling.
- A shipment of waste water treatment plant sludge was surveyed at its originating location, followed to a landfill, and surveyed again to assess the effect of transport on the radiological characteristics of that material.

Significant field work remains to be completed in 2014 along with final report preparation. DEP estimates 51 data collection visits to 49 sites remain to be conducted to complete the field work portion of the Study. The majority of these visits are scheduled to take place at well pads to collect samples during the production phase of operations. The balance will include visits to well pad wastewater impoundments, beneficial use sites, gas operations facilities, landfills, and sludge shipment sampling opportunities. The majority of the field work is planned for completion by the end of February. March through August will be primarily dedicated to data analysis, report preparation, peer reviews, and edits. The final report is planned for release in 2014.

Concerns with the PESI Study

For the different waste streams, we discuss the data PESI gathered for each waste stream according to the TENORM Study Scope of Work²² and compare the PESI results with measurements obtained elsewhere and regulatory safe levels. We compare the data reported by DEP with data previously reported by respected authorities such as the USGS, and we identify any weaknesses of the study. We also examine whether the data are sufficient to quantify releases to the environment. We reserve the right to amend this report if DEP makes more data available.

PESI's sampling plan discussed the sampling of many different sites and equipment, most of which were on private property. The plan was not forthcoming regarding how and why PESI chose their sample sites. It is unclear whether PESI had the freedom to choose any site in the field, or whether DEP sampled specific sites based on industry's approval. PESI's study outline also failed to discuss why some drill sites with high radiation readings were not included in the study. For example, many spills at gas wells have been reported in Pennsylvania, and radiation alarms have been set off at Pennsylvania landfills, but it is unclear whether these sites were sampled.

²² PESI Scope of Work.

Another concern is that many measurements taken by unbiased agencies/entities lie outside the data range PESI has measured. For example, the New York State DEC^{23} has measured rock cuttings from Susquehanna County, Pennsylvania that far exceed the range of values PESI has measured. And Duke University²⁴ measured sediments downstream from a Pennsylvania Publicly Owned Treatment Works that far exceeded the range of values found by PESI. Interestingly, EPA gathered pre-fracking measurements of radon concentrations at wellheads throughout the nation ²⁵ and found about the same concentration as those found by PESI. This is interesting as Marcellus shale is much more radioactive and therefore radon levels would be expected to be much higher.

Measurement of Radon Gas at the Wellhead

Radon gas is a natural by-product of radium-226 and thus is present in the Marcellus Shale formation. It is a chemically inert, but radioactive gas. Radon-222 has a short half-life, 3.8 days. Radon gas, which is the 2nd leading cause of lung cancer worldwide, occurs with natural gas. When natural gas is used in home furnaces or stoves, it is released and creates an increase in radon in the home, exposing citizens and creating an increased risk of lung cancers²⁶. PESI did not conduct any sampling of radon in homes, yet reached the conclusion that radon exposure in homes is not an issue.

Radon was not an important issue when natural gas was transported via pipelines to the Northeast from the Gulf Coast because of the time required to transport the gas from hundreds of miles away. At a speed of ten to eleven mph^{27} , much of the Gulf Coast radon had time to decay during the long transport period. But the Marcellus shale formation is close to Northeast metropolitan areas such as Philadelphia and New York City. Thus, the expected radon concentrations will be higher. For this reason, it would be important to measure radon gas at the well head²⁸; if the concentrations were high, as we expect, then a delay time should be built into delivery to gas customers in the Northeast. The PESI Study measured radon at the well head.

Seventeen radon samples were collected in eight counties from Marcellus shale formations. These samples were correctly taken between the well head and the separator units²⁹. The results ranged between 3.0 and 147.5 pCi/L, with a median Rn concentration of 40.8 pCi/L.

PESI's results are concerning for two reasons; the results are inconsistent with prior studies and our own calculations. PESI's results are highly suspicious when compared to EPA's measurements prior to the use of hydraulic fracturing in deep formations, as explained further below. The average radon in natural gas, based on a survey conducted 42 years ago, in 1973, by

²³ Allied, 2012.

²⁴ Duke, 2013.

²⁵ Johnson, 1973.

²⁶ Resnikoff, 2012.

²⁷ Johnson, 1973.

Natural gas delivered to customers is mixed from Pennsylvania and Gulf Coast sources. To understand the radon contribution from Marcellus shale sources, we need to measure natural gas at the wellhead. ²⁹ DEP, p. 3-8.

the EPA³⁰ was 37 pCi/L, with a range of 1 to 79 pCi/L. This survey predates hydraulic fracturing and gas extraction from the Marcellus shale formation.

We have carried out theoretical calculations that take into account a range of radium concentrations in the Marcellus formation to calculate radon at the well head. We factored in the distance between the well head and metropolitan areas, the mixing of radon with air in apartments and the likelihood of developing lung cancer by inhaling radon gas. Our peer-reviewed article³¹ shows that up to 30,000 additional lung cancers could be caused by radon from the Marcellus formation, assuming the radium-226 concentrations are as high as 30 pCi/g. Since the article was published, NYSDEC found radium-226 concentrations up to 204 pCi/g from rock cuttings in Pennsylvania³², demonstrating that radon concentrations could be higher than our original calculations, as we discuss shortly.

A recent study by researchers at John Hopkins University's Bloomberg School of Public Health confirms the increased impact of fracking on radon in homes in Pennsylvania³³. The Bloomberg study reported that 42 percent of the readings were higher than what is considered safe by federal standards. The study was based on a review of DEP's database of radon concentrations in 860,000 buildings between the years 1989 to 2013. Radon levels are often assessed when property is being bought or sold and these records are kept by DEP.

Radon concentrations began an upward trend between 2004 and 2012 as more unconventional wells were drilled in Pennsylvania, "with higher levels in counties with greater than 100 drilled wells versus counties with none."³⁴

Because this is such a critical issue, all wells, including those with high radium-226 concentrations, should have been sampled for radon gas.

Under section 2g of the Scope of Work, PESI coordinated with DEP's Radon Division and well operators to perform radon sampling of gas as appropriate. The fact that the average radon concentrations were about the same as reported by the EPA³⁵ in 1973, in pre hydraulic fracturing days indicates to us that wells were not chosen at random, but specific wells were chosen by DEP. The EPA's 1973 data of radon concentrations were obtained from conventional wells, in geologic formations primarily from the Gulf Coast and Oklahoma. It is highly unlikely the more radioactive Marcellus shale formation would have radon concentrations equal to 1973 concentrations unless the wells were conventional. This highlights the fact that the method for choosing the wells and all of the raw data gathered therefrom, such as the radium-226 concentrations, are crucial to understanding the radon results. Additionally, because radon in homes is such a critical issue, wells with high radium-226 concentrations should have been sampled for radon gas.

- ³² Allied, 2012.
- ³³ Schwartz, 2015.

³⁰ Johnson, 1973.

³¹ Resnikoff, 2012.

³⁴ Casey, 2015.

PESI also only sampled radon at two natural gas plants, and those samples were further limited because of "wind and rain".

The methodology employed by PESI³⁶, is standard. It is not clear why 10% of the samples taken by PESI were chosen for further analysis, what were the criteria for selection, what were the radium concentrations in each well, and the locations of all of the wells chosen for original sampling is also unknown.

Measurement of Scale in Production Pipes

During production of natural gas, radium, which is dissolved in brine in Marcellus shale, deposits or "plates out" inside the production pipes and inside above-ground feeder lines, and separator and condensate tanks. The chemical form of this scale is a complex of primarily barium sulfate and also barium carbonate.

Pennsylvania state regulations require that the direct gamma emanating from production pipes be less than 50 μ R/hr. This direct gamma radiation limit will not be protective of the public, the environment, or workers because in order to yield a direct gamma level of 50 µR/hr outside the pipe, the radium-226 and radium-228 concentrations within the pipe, must be on the order of 1500 pCi/g and 500 pCi/g, respectively.

We have observed radium-226 and radium-228 concentrations of 6,000 pCi/g and 2,000 pCi/g respectively, from pipes withdrawn after 15 years in production³⁷. Production pipes are generally removed after they become clogged and restrict oil or gas flow. If a pipe is cut open for removal, the radium may be released from its confines within the pipe, exposing both the workers involved in cutting up the pipes and the environment to high concentrations of radium. EPA radium concentration limits in soil³⁸ are 5 pCi/g for the top 15 cm of soil and 15 pCi/g below the top 15 cm of soil. That is, radium released from pipes that met external gamma limits would exceed EPA cleanup standards and would not be safe. The regulatory requirements are shown in the box below.

³⁶ Using a Lucas cell, which essentially consists of placing a radon sample in a ZnS-lined bag and counting the light pulses with a photomultiplier tube.

³⁷ See James McAllen's successful suit against Forest Oil Corp. in 2008 for damages from contamination and personal injury from post-production pipe radiation. ³⁸ 40 CFR §192.32(b)(2).

Pertinent Radiation Regulations

Maximum yearly radiation to the public

Whole body dose (total effective dose equivalent) to individual members of the public from a facility cannot exceed 100 millirem in a year. (10 CFR 20.1301(a)(1) The whole body dose is the sum of direct gamma radiation and the dose from inhaled and ingested radioactive materials. A rem is a unit of tissue dosage, in terms of energy per mass. A whole body X-ray is 4 to 10 millirem.

Drinking water standards

The maximum contaminant limit for drinking water in a public water system is 5 picoCi/L for combined Ra-226 + Ra-228. (40 CFR 141.66(b)).

Maximum ground contamination

The maximum ground contamination limit for combined Ra-226 + Ra-228 is 5 pCi/g in the top 15 cm of soil and 15 pCi/g below 15 cm (40 CFR 192.32(b)(2)).

DEP's Study again falls short as it does not consider the eventual cutting and possible reuse of production and other pipes. In Radioactive Waste Management Associates' experience with natural gas fields in Texas, over half of the pipes that were used in production were removed after fifteen years and had high direct gamma levels which exceeded 50 μ R/hr. These contaminated pipes were subsequently cut up and re-used. After being cut up for use as fence posts and gates, the radium was released from the pipes and spread on the ground, contaminating a rancher's property. The use of an acetylene torch to cut the pipes vaporized the radium, presenting an inhalation hazard. Similar situations occurred in Martha, Kentucky, where oil field pipes were used to build a corral, and in Allegany County, New York, where radioactive pipes were used to construct playground equipment.

Natural gas production pipes have not been in use in Pennsylvania for enough time for scale to build up to the point where pipes must be removed from operation. DEP failed to account for this serious issue in its study; eventually production pipes will become clogged with scale and must be removed from use and either cleaned or disposed of.

Under Section 2g, PESI sought to, "if possible, collect and screen samples of solids," from production equipment. It is unlikely this will be feasible for this study and should be reserved for a future study.

Radium in Rock Cuttings

Under Section 2a of the Scope of Work, PESI intended to sample rock cuttings, and conduct laboratory analysis for eighteen samples from ten well sites. The exact locations and methods of selection have not been released by DEP.

Studies by the USGS present a range of radium-226 concentrations³⁹. Prior to the PESI sampling program, Dr. David Allard, Director of the DEP's Bureau of Radiation Protection, stated that radium-226 in rock cuttings ranged from 3.4 to 34 pCi/g⁴⁰ based on USGS and drill logs. But NYSDEC's measurements of rock cuttings from a Cabot Oil and Gas Co. Marcellus shale well in Susquehanna County, PA⁴¹ reported considerably higher concentrations of radium-226 - 204 pCi/g. It is not possible to ascertain using the Fig. 1 map whether these two well sites were sampled or avoided, but, inexplicably, DEP found horizontal solid drill cuttings for the State as a whole to have a range⁴² from 0.092 pCi/g to 13.0 pCi/g Ra-226, with an average 5.22 pCi/g Ra-226.⁴³

We find these results completely out of line with measurements by the USGS and measurements by NYSDEC in 2012 of rock cuttings from Susquehanna County, Pennsylvania of Cabot Oil and Gas Co. from a Marcellus shale well.⁴⁴ High radium-226 concentrations of rock cuttings transported to a Niagara County landfill, up to 204 pCi/g Ra-226 in two train carloads of rock cuttings were returned to Cabot Oil and Gas Co.

Pennsylvania has 49 municipal waste landfills, 23 of which receive TENORM waste, as seen in Appendix A. Some Pennsylvania landfills have reported trucks carrying rock cuttings exceeding the alarm settings of portal monitors at waste dump sites. According to DEP, in 2008, TENORM triggered 423 alarms; by 2011, this number had risen to 798 alerts. In one such instance, the MAX Environmental Technologies landfill in South Huntingdon, PA reported direct gamma readings of 96 μ R/hr; almost 10 times background levels and 10 times the acceptable level at that particular landfill. An average reading of 5 pCi/g would not trigger radiation alarms, so again, it appears as though cuttings from lower-emitting, outlier wells were selected for sampling, or that the average was brought down by non-Marcellus cuttings.

In contrast, we compare the PESI results to those obtained by the USGS. The USGS analyzed seventeen cores from wells in Pennsylvania, New York, Ohio, West Virginia, Kentucky, Tennessee, and Illinois⁴⁵. Although the cores varied in thickness and in depth, geologists identified the Marcellus stratum in several cores using data on the organic, sulfur, and uranium content of the samples. Table 1 below summarizes the results from four cores that tapped into

⁴⁴ Allied, 2012.

³⁹ Leventhal, 1981.

⁴⁰ DEP, 2013a.

⁴¹ NYSDEC, 2012.

⁴² DEP, Table 3-7.

⁴³ DEP, p. 3-5.

⁴⁵ Leventhal, 1981.

the radioactive Marcellus formation. The depths at which the layer was found as well as the uranium measurements are presented. Radium-226 concentrations up to 30 pCi/g were found⁴⁶. As mentioned above, Ra-226 in one sample from Susquehanna County, PA, was measured by NYSDEC at 204 pCi/g. The measurements from NYSDEC and USGS⁴⁷ (Table 1 below) lead us to question how the PESI rock cuttings samples were chosen and whether they are representative.

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

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

Radium-226 in Brine

After wells are drilled and hydraulically fractured, the water returned within approximately the first two weeks is called flowback water.⁴⁸ Following this initial period, when gas wells are put into production, lesser amounts of water are separated from gas and placed into condensate tanks or trucks. This produced water or brine contains high concentrations of total dissolved solids (TDS), as seen in Table 2. Over the initial fourteen day period before the well goes into production, the TDS goes up dramatically, as seen in Table 2.

As additional wells have been drilled into Marcellus shale, TDS concentrations have risen in the Monongahela River, the correlation clearly showing a trend that indicates that gas companies may have been dumping wastewater into surface waters.⁴⁹

Measurements by Duke University scientists⁵⁰ that showed elevated levels of chloride and bromide, combined with the strontium, radium, oxygen, and hydrogen isotopic compositions reflect the effluents of Marcellus Shale produced waters. According to the study, the discharge of the effluent from the treatment facility increased downstream concentrations of chloride and bromide above background levels. In particular, Ra-226 concentrations in stream sediments at the point of discharge were 200 times greater than upstream and background sediments and above radioactive waste disposal threshold regulations.

⁴⁹ Urbina, 2011.

⁴⁶ Without going into the physics, Radium-226 in units pCi/g is approximately 1/3 total U content in units ppm, or up to 30 pCi/g.

⁴⁷ Leventhal, 1981.

⁴⁸ Veil, 2012.

⁵⁰ Warner, 2013.

Location	Day 0*	Day 1	Day 5	Day 14	Day 90
A	990	15,400	54,800	105,000	216,000
В	27,800	22,400	87,800	112,000	194,000
С	719	24,700	61,900	110,000	267,000
D	1,410	9,020	40,700		155,000
E	5,910	28,900	55,100	124,000	
F	462	61,200	116,000	157,000	
G	1,920	74,600	125,000	169,000	
н	7,080	19,200	150,000	206,000	345,000
1	265	122,000	238,000	261,000	
J	4,840	5,090	48,700	19,100	
к	804	18,600	39,400	3,010	
L	221	20,400	72,700	109,000	
M	371			228,000	
N	735	31,800	116,000		
0	2,670	17,400	125,000	186,000	
Р	401	11,600	78,600	63,900	0
Q	311	16,600	38,500	120,000	
R	481	15,100	46,900	20,900	
S	280	680	58,300	124,000	
			1		

Table 2. TDS (mg/L) as a Function of Time After Well Hydraulic Fractured⁵¹

* Day 0 sample was taken of the influent water plus additives without sand.

The DEP Study sampled brine for radium content. In his public presentations, Dr. Allard previously stated that radium concentrations in brine could be as high as 11,000 pCi/L⁵², also NYSDEC has reported radium-226 concentrations over 15,000 pCi/L. PESI found radium-226 concentrations from 40.5 to 26,600 pCi/L⁵³ in unfiltered samples, and almost the same in filtered samples; 87.0 to 24,100 pCi/L. Clearly filtering was not removing the radium, which was in solution, not in particle form. Conventional water treatment plants which remove solids are not able to effectively remove radium from wastewater unless radium is converted to a solid. The difficulty of removing radium as a liquid is a major problem for the gas industry.

Brine used for Dust Suppression and De-icing

As reported to the Citizen Advisory Council in February, 2014, PESI sought to study thirteen sites where brine has been used for dust suppression. The brine was sourced from "conventional" gas wells; currently, only non-shale brine can be used for dust suppression and de-icing. In all, thirty-two O&G brine-treated roads were surveyed in the southwest, northwest, and north-central regions of the State, and eighteen reference background roads were surveyed. PESI tested for direct gamma radiation and the surveys included gross gamma radiation scans performed using 2-inch x 2-inch NaI detectors and a Ludlum Model 2221 scaler/ratemeter instrument. The surveys showed about half the roads were slightly above background.

⁵¹ Veil, 2012.

⁵² Allard, 2013.

⁵³ DEP, E-22.

As a practice, using brine fluid as a de-icer is unwise for health and safety reasons; it is a means of providing the **maximum** radiation dose to the population. The number of radiation pathways to humans is increased. This is because radium-226 decays to gamma emitters, such as bismuth-214, so spread brine is a gamma emitter. In addition, during dry periods, radium-226 in brine can be resuspended by car movement and be inhaled by the public. It can also be spread off roads onto nearby residential and agricultural lands. PESI did not analyze inhalation and incidental ingestion of radium; PESI also did not consider ingestion of food/plants grown near to treated roadways, in contaminated soil.

Wastewater at Water Treatment Facilities

While DEP has asked drillers to voluntarily cease sending wastewater directly from ponds or tanks at well sites to POTWs, some POTWs have continued the practice under a Consent Decree. This should cease when the proposed EPA effluent regulations are implemented, if it hasn't already.⁵⁴ It is not clear how pending federal regulations will affect effluent that has been pre-treated at CWTs. POTWs can accept wastewater from CWTs and this is current practice. In any case, neither POTWs nor currently operating CWTs are effectively removing radium in solution. PESI sampled a small fraction of influent and effluent wastewater at POTWs and CWTs; for example, only six POTWs were sampled. The locations of the POTWs and CWTs and the origin well sites are not stated.

As shown in Tables 4-8 through 4-15 of DEP's report, the average radium-226 concentration found in effluent is 129 pCi/L and the maximum is 363 pCi/L. The average radium-228 concentration found in effluent is 9.6 pCi/L and the maximum is 35 pCi/L. These concentrations are far above drinking water standards for POTWs, which is 5 pCi/L combined Ra-226 and 228. Also troubling is the fact that brine and produced water, according to PESI data, have Ra-226 concentrations up to 26,000 pCi/L. The low Ra-226 concentrations to POTWs are due to DEP's recommendation in 2011 that well sites transport brine to CWT's and to deep wells for disposal⁵⁵, but high radium concentrations can still be received by CWTs.

The PESI Study also sampled influent and effluent at CWTs. The effluent from CWTs may end up in one of three places: as influent to POTWs, released directly into the environment, or to outof-state injection wells. According to the Duke study⁵⁶, in 2011, about twenty percent of drilling fluids, eight percent of hydraulic fracturing flowback fluid, and fourteen percent of produced water (i.e., brine) from unconventional Marcellus Shale wells were treated at centralized waste treatment facilities and then discharged to local streams. The PESI Study showed the Ra-226

⁵⁴ It should be noted that EPA reports in its proposed hydraulic fracturing wastewater effluent rulemaking that EPA found no instances where onshore unconventional gas wells were still sending wastewater to POTWs. US EPA has proposed effluent regulations for oil and gas extraction that prohibits the discharge of unconventional oil and gas wastewater to POTWs; the rule is not yet in force and is expected to be adopted as final in 2016. Available at http://www2.epa.gov/eg/unconventional-extraction-oil-and-gas-industry.

⁵⁵ EPA, 2015, p. 8-17.

⁵⁶ Duke, 2011.

average and maximum effluents from CWTs as 1,840 pCi/L and 15,500 pCi/L, respectively. The PESI Study did not detail the ultimate disposition of this material.

PESI stated that twenty-two of the highest volume Marcellus shale waste water treatment facilities would be included in this study and that each would be sampled as often as three times to establish a trend. The sampling plan was more ambitious than the actual work performed. For example, of the ten POTWs sampled, only six were influenced by Marcellus shale input, i.e., by having received wastewater from the gas industry and only five were sampled in all three rounds. Three survey rounds were conducted at nine of the ten CWTs. Radiological surveys were also conducted at all nine zero liquid discharge (ZLD) plants.

As discussed above, it is clear from the data that the POTWs and the CWTs do not remove radionuclides in solution, such as radium, leaving open the question, where does this radium go? The data shows that treatment plants' effluent contains radium-226, which will be released into the environment. The radium-226 concentration found by PESI is high, as high as 26,600 pCi/L⁵⁷ and far above safe drinking water levels, which are 5 pCi/L for combined Ra-226 and Ra-228.

Ground Contamination and Worker Exposure

Open land areas around POTWs have become contaminated with radioactive materials, exposing workers. The highest average gamma radiation exposure rate found by PESI was 36.3 μ R/hr and the maximum gamma radiation exposure rate measured was 257 μ R/hr⁵⁸. For a 2,000 hr/yr. work period, the potential dose to a worker who remained at this location would be 514 mrem/yr., considerably above the allowable dose of 100 mrem/yr.⁵⁹. This is the dose due to gamma radiation alone; additional radiation pathways include incidental ingestion which can occur when a worker wipes contaminated hands across his or her mouth, or if he or she eats with contaminated hands. Alpha and beta emitting radiation can also be inhaled when contamination is resuspended. These pathways add to the direct gamma radiation dose.

In evaluating health impacts, DEP's Study considers only the immediate time period. Since radium-226 has a half-life of 1,600 years, it is vital to consider more than the immediate time period. Institutional controls may not be assumed to be in place for more than 100 years⁶⁰ after which landfills may be abandoned and monitoring ceased. For public safety, it should be assumed that residents live on the property full-time, build homes with basements and grow gardens. Farmers should be assumed to continue or resume agricultural practices on these lands. This kind of exposure was not studied or considered by DEP and PESI, thus calling into question the credibility of DEP's evaluation of health impacts and of long term pollution and degradation of the environment, e.g. legacy pollution.

⁵⁷ DEP, ES-22.

⁵⁸ DEP, p. 4-2.

⁵⁹ See Regulatory Box on p. 24.

⁶⁰ 10 CFR §61.59.

Landfills

According to the sampling plan, landfill leachate was sampled at each of the 51 active landfills and analyzed for gross alpha/beta and Ra-226/Ra-228 by gamma spectroscopy. The sampling was broken into two groups, 42 samples were taken at landfills receiving less O&G solid waste, and more extensive sampling was conducted at nine landfills taking more O&G waste. The terms "less" and "more" were not defined. We assume all these landfills tested by PESI were located in Pennsylvania, and not in New York State, Ohio and West Virginia, where rock cuttings have also been shipped. However, DEP did not provide the locations of the landfills tested. Radium was detected in all leachate samples.⁶¹ Sample results from the 42 landfills not selected for extensive sampling showed Ra-226 results that ranged from 54.0 to 416 pCi/L, with an average Ra-226 112 pCi/L. Radium-226 results from the nine selected landfills ranged from 85 pCi/L to 378 pCi/L with an average 106 pCi/L.

As a yardstick, these concentrations exceed drinking water regulations for POTW effluent into a public water supply⁶². One must also keep in mind that the above concentrations are a snapshot in time. When management at these landfills no longer exists and maintenance is not maintained, these concentrations may increase. For time periods greater than 100 years, management controls may no longer exist.

It is unclear whether effluent at all nine landfills was filtered, but for those that did filter effluent, radium was detected in all of the filter cake samples. Radium-226 results ranged from 8.73 to 53.0 pCi/g, with an average of 24.3 pCi/g. At three landfills that discharged effluent water directly to the environment, a sediment-impacted soil sample was collected at each of the three effluent outfalls. Radium-226 results ranged from 2.82 to 4.46 pCi/g with an average of 3.57 pCi/g. These results for downstream sediments are much lower than those found by Duke, which were 14.7 pCi/g to 237 pCi/g. Landfills that do not discharge effluent directly to the environment often direct the effluent to a POTW. The radium concentration at a POTW would be diluted with other influent.

Brine Trucks and Driver Radiation Exposures

PESI estimated the gamma radiation exposure for drivers transporting wastewater from well sites to wastewater treatment plants. PESI assumed the driver worked for ten weeks per year (rather than fifty weeks per year)⁶³ and the truck carried 3,800 gallons of wastewater with a radium-226 and progeny concentration of 18,400 pCi/L. We reviewed the Microshield calculations and except for the number of hours worked, they appear correct. However, PESI and DEP fail to analyze the safety implications of transporting this total inventory of brine in one shipment.

Brine is transported by truck from well sites either to CWTs or deep wells. These trucks must satisfy Federal DOT rules. Transported material that exceeds a total activity, in terms of total Curies of radioactivity, is classed as a hazardous, class 7, radioactive material by DOT. The

⁶¹ DEP, p. 5-1.

⁶² 40 CFR §141.66.

⁶³ DEP, p. 4-12.

specific limit for Radium-226 is 2.7×10^{-7} Curies^[47]. Below this total activity, the material is not classed as radioactive by DOT; above this amount, specific Federal regulations apply regarding design, packaging and labeling of transportation vehicles. For placarding, the NRC has even stricter limits 1×10^{-7} Curies (10CFR20, App. C). The above shipments contain 2.6 E-4 Ci Ra-226 and are therefore 1000 times above DOT limits. PESI does not discuss this aspect of transportation. The brine trucks we have seen on Pennsylvania highways have not satisfied federal DOT design, packaging and labeling requirements for class 7 radioactive materials. **This is a major oversight by Pennsylvania DOT and should be immediately corrected.**

Production Site Survey and Sampling

PESI intended to sample closed/reclaimed cuttings pits⁶⁴. PESI was to sample pits using portable survey meters, which is an effective method only if contamination is on the surface. If contamination is covered by more than one foot of soil, radium and its decay products will not be detected, as gamma radiation from TENORM is reduced by 98% by one foot of soil cover. In order to detect subsurface contamination, push probes and laboratory analysis of samples is necessary; PESI did not utilize this method of sampling. This is particularly concerning because buried TENORM can be exhumed by landowners when constructing homes or planting gardens; all buried TENORM must be found.

PESI intended to sample and analyze fresh proppant sands, drill muds, flowback and produced water on sites in accordance with the sampling plan⁶⁵. While the intent of the gas industry is to recycle seventy to eighty percent of drilling fluids, our concern is with the twenty to thirty percent that have become too concentrated in certain properties (such as salts) so they cannot be re-used. It is not clear whether these fluids that cannot be recycled and which contain buildups of toxic and radioactive materials have been sampled by PESI. Similarly, the sludges and residues from open pits, settling ponds and holding tanks at well sites, wastewater treatment plants or pre-treatment basins or vessels may also have not been sampled.

PESI also sampled temporary water storage vessels and recycle systems⁶⁶. PESI conducted gamma surveys and took wipe samples for removable alpha and beta radiation. The gamma surveys were sufficient to determine whether a gamma dose exceeded regulatory limits of 100 mrem/yr. for a member of the public. However, the gamma surveys, as outlined in the Statement of Work, were not sufficiently location-specific to determine whether particular locations were unusually radioactive, such as the gas/water separators, condensate tank bottoms or feeder lines. This would have yielded information on whether radium scale or sludge had built up in these components. Sludge would have built up in the condensate tank bottoms; scale would build up in all three components over time.

PESI intended to visit twenty well sites in various stages of development, out of over 5000 well sites in production in the State. These visits were for the purpose of taking radon samples, or

⁶⁴ PESI Scope of Work Section 2b.

⁶⁵ *Ibid* at Section 2c.

⁶⁶ *Ibid* at Section 2d.

gamma surveys. Many well sites in the State have had spills, and it is not clear whether any such wells were sampled for the study.

Sludge Sampling

Though PESI stated that sludge would be sampled, we cannot identify whether sludge was sampled. PESI sampled filter cake, sediment-impacted soil and high gamma rates on the exterior of condensate tanks, which may be due to sludge, but we cannot identify sludge itself being sampled. The maximum gamma radiation exposure rate measured at the POTWs was 257 μ R/hr on contact with the outside of a wastewater tank; this may be due to sludge settled at the bottom of the tank.

The sampling results of solids collected on filters or filter cakes are presented in Table 4-27. All the CWT filter cake samples contain elevated Ra-226. The maximum results were 305 pCi/g for Ra-226 and 177 pCi/g for Ra-228. The PESI Study should have detailed the fate and transport of the filter cake samples from the CWTs, but does not.

Three survey rounds were conducted at nine of the ten CWTs. Sediment-impacted soil was collected at the accessible effluent discharge points at the CWTs. Radium above typical soil background levels to a maximum of 508 pCi/g of total Ra was identified. This can be compared to EPA clean-up criteria of 5 pCi/g total Ra for surface soils and 15 pCi/g below 15 cm depth. The highest average gamma radiation exposure rate was 43.1 μ R/hr, and the maximum gamma radiation exposure rate measured was 445 μ R/hr. These are not safe levels. For example, if a permanent resident lived in an area with radium-226 soil concentration 508 pCi/g, and cultivated a garden, the annual radiation whole body dose would be almost 5000 mrem/y, due almost entirely to direct gamma and ingestion of contaminated food.⁶⁷

Conclusion

It is well known and has been demonstrated by several studies by reputable scientific bodies that the Marcellus shale formation, from which natural gas is drawn, is highly radioactive. This radioactivity does not disappear during drilling and when natural gas is brought to the surface. Our conclusion from our review of DEP's Study is that it failed to fully evaluate the full worker and public radiation exposure, TENORM disposal, and environmental impacts. Our review identified major gaps in data collection.

Radon: Radon is a radioactive chemically inert gas and a decay product of radium. It is also the second leading cause of lung cancer worldwide. As natural gas is released from the Marcellus shale formation, radon is released with it. The median concentration of radon found by DEP is highly suspect as it is almost identical to those levels found by the EPA in 1973, well before horizontal high volume hydraulic fracturing was used to extract natural gas. Studies show that

⁶⁷ We assumed a one foot contamination layer with no clean soil shielding, and employed the software RESRAD 7.0. The lung dose due to radon would be very high.

radon concentrations in Pennsylvania homes have been increasing in areas where well activity is the greatest. The findings in the DEP report cannot be accepted as an accurate assessment of radon from Marcellus shale.

Pipe Scale: During production of natural gas, radium, which is dissolved in brine in Marcellus shale, is deposited or "plates out" inside the production pipes and also inside of above-ground feeder lines, separator and condensate tanks. Eventually, pipes become clogged and must be removed. DEP's Study does not consider the fate of and resulting exposure to harmful levels of radiation from removal of these pipes. Since no pipes have been removed yet, none have been tested. DEP should plan a follow up study to sample this future exposure pathway. Until this follow up sampling and study is done, it cannot be assumed that scale will not present a significant pathway for exposure of workers and the public to radiation from Marcellus shale.

Rock Cuttings: Prior to the PESI sampling, Dr. David Allard, Director of the Bureau of Radiation Protection, stated⁶⁸ that radium-226 ranged from 3.4 to 34 pCi/g. This was our understanding as well, based on a previous USGS study and drill logs. But, surprisingly, horizontal solid drill cuttings in the DEP Study ranged from 0.092 pCi/g to 13.0 pCi/g Ra-226, with an average of 5.22 pCi/g Ra-226. Rock cutting measurements by New York State from the same formation were found to range up to 204 pCi/g Ra-226. The surprisingly low range found by DEP demonstrates either an anomaly or selective sampling. Either way, the findings cannot be accepted as a valid representation of the radioactive properties of Marcellus shale rock cuttings.

Wastewater: Wastewater, in the form of brine and flowback water, is highly radioactive, with concentrations that range up to 26,600 pCi/L⁶⁹ Ra-226. Workers at treatment plants have the potential to be exposed to high levels of radiation if they work with or around the filter cakes. Radium is in solution at CWTs and is either released directly to the environment and potentially taken in by surface water intakes that supply drinking water, or sent to POTWs as influent. But the average influent to POTWs is 129 pCi/L, i.e., nowhere near 26,600 pCi/L. The concentration 129 pCi/L is not a plausible measurement. These data must be re-examined based on the fact that currently employed wastewater treatment systems do not effectively remove radioactivity, so it is reasonable to examine the potential that radioactive contaminants are entering the environment and can be contaminating drinking water sources.

Landfills: Radium was detected in all landfill leachate samples gathered for DEP's study. Radium-226 concentrations ranged from 67.0 to 378 pCi/L for effluent samples. This can be compared to drinking water standards of 5 pCi/L for combined Radium-226 and 228. Workers at landfills have the potential to be exposed to high levels of radiation if they work with or around the filter cakes processing landfill leachate. Since leachate treatment does not remove Radium-226 and effluent discharges into waterways, downstream water intakes could be impacted. Radium-226 may not be sampled for or removed by water treatment systems, so downstream water systems could unknowingly pass these contaminants through to water users. Additionally,

⁶⁸ Allard, 2014.

⁶⁹ DEP, ES-22.

since radium-226 has a half-life of 1,600 years, this leaching will continue long past management oversight of these landfills. The issue of landfill leachate carrying Radium-226 into the environment and drinking water must be fully analyzed through a comprehensive examination of all point discharges of treated leachate from landfills that accept gas well drill cuttings.

Brine Transport: The study discusses the safety to truck drivers of transporting brine, but does not discuss the hazard to the general public of potential accidents and the need for proper packaging, placarding and insurance. DEP also falsely and inexplicably assumes that workers transporting brine work only ten weeks a year. The Commonwealth should refer for immediate action to Pennsylvania Department of Transportation the important issue of required placarding and the enforcement of transport regulations which apparently are not being enforced regarding Marcellus shale wastewater.

Production Site Survey and Sampling: PESI's method of sampling abandoned cuttings pits is ineffective because gamma surveys cannot detect contamination from pits covered by one foot or more of clean soil. Push probes and laboratory analysis are required to detect such buried contamination. Landowners may unearth this radioactive contamination, resulting in exposure, when constructing homes and buildings, or even planting a garden, or farmers may disturb and distribute the contamination through agricultural practices, exposing farm workers, the public, livestock and food supplies to potential contamination. DEP must further test well site pits and production areas through push probes and laboratory analysis to gather the data needed to draw a valid conclusion regarding the radioactivity of buried solids at Marcellus shale gas well sites.

Non-recycled drilling fluids and sludges: The industry's goal is to recycle seventy to eighty percent of drilling fluids, however it is not clear that the fluids that cannot be recycled, that have buildups of toxic and radioactive materials, have been sampled by PESI. Similarly, the sludges and residues from settling ponds and holding tanks at wastewater treatment plants or pre-treatment basins or vessels may also have not been sampled. We have not been able to identify from the DEP Study where this has been done. These sludges must be sampled to provide the data necessary for valid conclusions regarding residues from recycled drilling fluids.

Finally, stream water quality, sediments and instream habitats could be degraded by cumulative buildup of radioactivity in waterways. DEP's Study failed to analyze streams, sediments, and failed to sample fish flesh or mussels, for example. These should be analyzed to provide the evidence from which to draw valid conclusions regarding the environmental impacts of radioactive contaminants produced by Marcellus shale extraction.

References

(Allard, 2013) Allard, DJ, "Marcellus Shale and Tenorm," ISCORS Meeting, May 1, 2013.

(Allard, 2014) Allard, DJ, "TENORM Experiences in Pennsylvania," presentation before Mid-Atlantic States Rad Control DVSRS Meeting, March 25, 2014.

(Allied, 2012) NYSDEC, Division of Environmental Remediation, August 2012, re. Allied Landfill, Niagara County.

(Casey, 2015) Casey, JA, et al, "Predictors of Indoor Radon Concentrations in Pennsylvania, 1989 – 2013", Environmental Health Perspectives, vol 123, November 2015.

(DEP, 2013) Pennsylvania Department of Environmental Protection, "Technologically Enhanced Naturally Occurring Radioactive Materials (Tenorm) Study Report, Scope of Work," April 10, 2013.

(DEP, 2015) Pennsylvania Department of Environmental Protection, "Technologically Enhanced Naturally Occurring Radioactive Materials (Tenorm) Study Report," January 2015, prepared by Perma-Fix Environmental Services, Inc.

(DRN, 2015) Department of Environmental Protection v. Delaware Riverkeeper Network, 1373 C.D. 2014 (Pa. Cmwlth. 2015)

(Duke, 2013) Warner, RN, *et al*, "Impacts of Shale Gas Wastewater Disposal on Water Quality in Western Pennsylvania," Division of Earth and Ocean Sciences, Duke University, Environmental Science and Technology, October 2, 2013.

(Haluszczak, 2012) Haluszczak LO, Rose AW, Kump LR (2012) Geochemical evaluation of flowback brine from Marcellus gas wells in Pennsylvania, USA. Appl. Geochem, http://dx.doi.org/10.1016/j.apgeochem.2012.10.002

(Iowa, 2014) Schultz, MK and Nelson, AW, "Radioactivity and Unconventional Drilling," University of Iowa, for the U.S. Department of Energy, National Analytical Management Program (NAMP)

(Johnson, 1973) Johnson, RH, *et al*, "Assessment of Potential Radiological Health Effects from Radon in Natural Gas," Environmental Protection Agency, EPA-520/1-73-004, November 1973.

(King, 2008) King, HM, <u>"Marcellus Shale Gas: New Research Results Surprise Geologists!"</u>. *geology.com*. Retrieved 2008-05-03.

(Leventhal, 1981) Leventhal, J. S., J. G. Crock, and M. J. Malcolm. "Geochemistry of trace elements and uranium in Devonian shales of the Appalachian Basin." USGS Open File Report 81-778. (1981).

(PESI Scope of Work) TENORM Study Scope of Work. (2013) Available at files.dep.state.pa.us/.../TENORM-Study_SoW_04_03_2013_FINAL.pdf.

(Resnikoff, 2010) Resnikoff, M, Alexandrova, E, and Travers, J. "Radioactivity in Marcellus Shale." Report Prepared for Residents of for the Preservation of Lowman and Chemung (RFPLC). (2010)

(Resnikoff, 2012) Resnikoff, M, "Radon in Natural Gas from Marcellus Shale," Ethics in Biology, Engineering & Medicine - An International Journal, 2(4): 317–331 (2011).

(Schwartz, 2015) Schwartz, Brian S, et al, "Predictors of Indoor Radon Concentrations in Pennsylvania 1989-2013," Dept. Envtl. Health Sciences, Johns Hopkins U, April 9, 2015.

(Swanson, 1960) Swanson, VE, "Oil Yield and Uranium Content of Black Shales," USGS paper 356-A (1960)

(Urbina, 2011) Urbina, I, "Regulation Lax as Gas Wells' Tainted Water Hits Rivers", NY Times, February 27, 2011.

(US EPA, 2012). Study of the Potential Impacts of Hydraulic Fracturing on Drinking Water Resources: Progress Report. EPA 601/R-12/011. Available at <u>http://www.epa.gov/hfstudy</u>.

(USEPA, 2015, "Assessment of the Potential Impacts of Hydraulic Fracturing for Oil and Gas on Drinking Water Resources," EPA/600/R-15/047a, June 2015.

(USGS, 2011) Rowan, EL, *et al.*, "Radium Content of Oil- and Gas-Field Produced Waters in the Northern Appalachian Basin (USA), USGS Scientific Investigations Report 2011-5135" (2011)

(Veil, 2012) Veil, J, "Overview of Shale Gas Water Issues," WEFTEC 2012, New Orleans, LA, October 2012.

(Warner, 2013) Warner, NR, et al, "Impacts of Shale Gas Wastewater Disposal on Water Quality in Western Pennsylvania," Enviro Science and Technology, Oct 2, 2013, pp. 11849

Appendix A Municipal Landfills in Pennsylvania

DRAFT - Pennsylvania Landfill TENORM Acceptance

The following list represents the Pennsylvania Landfills that have accepted TENORM-containing waste between January 1, 2012 and February 28, 2014:

- 1. Southern Alleghenies Landfill
- 2. Arden Landfill
- 3. Tervita Sanitary Landfill
- 4. Valley Landfill
- 5. Lake View Landfill
- 6. McKean County Landfill
- 7. Seneca Landfill
- 8. Chestnut Valley Landfill
- 9. Evergreen Landfill
- 10. Northwest Sanitary Landfill
- 11. South Hills Landfill
- 12. Monroeville Landfill
- 13. Imperial Landfill
- 14. Kelly Run Sanitation Landfill
- 15. Alliance Sanitary Landfill
- 16. Greentree Landfill
- 17. Shade Landfill
- 18. Tullytown Landfill
- 19. Laurel Highland Landfill
- 20. Mostoller Landfill
- 21. GROWS North Landfill
- 22. White Pines Landfill
- 23. Phoenix Resources

2008, 200	9, 2010 and 2011
(Rad-226/Nat. F	adium 226) Yearly Hits

2008	2009	2010	2011
1 1 1 1 9 2 5 32 20 17 23 20 7 31 14 21 12 1 21 12 1 21 12 1 21 12 1 21 12 1 21 12 1 21 14 21 1 21 21 21 21 21 21 21 21	7 7 1 2 1 17 24 21 30 26 30 27 20 30 8 1 3 1	$\begin{array}{c} 2010\\ 2\\ 3\\ 9\\ 6\\ 2\\ 3\\ 9\\ 6\\ 27\\ 3\\ 3\\ 8\\ 4\\ 29\\ 00\\ 32\\ 27\\ 33\\ 0\\ 26\\ 24\\ 2\\ 2\\ 2\\ 1\\ 1\\ 1\\ 1\\ 1\\ 1\\ 1\\ 1\\ 3\\ 1\\ 23\\ 20\\ 23\\ 25\\ 9\\ 2\\ 1\\ 13\\ 2\\ 53\end{array}$	$\begin{array}{c} 2011\\ 19\\ 16\\ 32\\ 7\\ 7\\ 1\\ 2\\ 1\\ 9\\ 8\\ 36\\ 29\\ 27\\ 33\\ 5\\ 14\\ 3\\ 6\\ 11\\ 20\\ 7\\ 7\\ 1\\ 1\\ 3\\ 1\\ 1\\ 3\\ 1\\ 1\\ 3\\ 28\\ 24\\ 16\\ 2\\ 4\\ 38\\ 37\\ 37\\ 38\\ 37\\ 37\\ 38\\ 37\\ 37\\ 12\\ 1\\ 1\end{array}$
423	256	562	798

Total: