

# Radioactivity and Unconventional Gas

## Author Biographical Sketch

My name is Brian Gaulke. I have a Bachelors of Science degree in Physics from the University of Washington and did graduate work in plasma physics at Princeton University. I recently retired from a 30 year career as a health physicist in the U.S. and Canada. I spent over 10 years as a Health Physicist at Puget Sound Naval Shipyard in Bremerton, Washington, primarily involved with reactor defueling and refueling activities. After moving to Canada in 1992 I worked as a Health Physicist in the Radiation Protection Branch at the Chalk River Laboratories of AECL for 4 years with responsibilities including waste management areas, training, program documentation development, decommissioning, MAPLE-X reactor commissioning team, and Recycle Fuels Fabrication Laboratory rehabilitation and commissioning. Following that I was the Dosimetry Section Head with Health Canada's National Dosimetry Services for 11 years. Lastly, I worked as Health Canada's Atlantic Region Radiation Specialist for over 6 years. During this time I was responsible for implementation of Health Canada's radon program throughout the Atlantic Region, had responsibilities for nuclear emergency preparation and response, and worked with other Specialists whenever issues arose involving radiation and radioactivity. I have been a Certified Health Physicist (certified by the American Board of Health Physics in the comprehensive practice of Health Physics) since 1994.

## Introduction

Unconventional gas is present in layers of shale which are typically deep underground, or in coal seams. These deposits tend to have elevated levels of naturally occurring radioactive material, which inevitably results in radioactivity reaching the surface in a variety of ways and forms. The source, nature and impacts of this radioactivity are discussed in this submission.

## Naturally Occurring Radioactive Material (NORM)

Naturally occurring radioactive material consists essentially of three components: uranium and its decay chains, thorium and its decay chain, and  $^{40}\text{K}$ . The  $^{238}\text{U}$  decay chain consists of 19 radioactive species (although 4 are present at very low concentrations relative to all the others), the  $^{235}\text{U}$  chain of 14 radioactive species (although 3 are present at very low concentrations relative to all the others), and the  $^{232}\text{Th}$  decay chain consists of 11 radioactive species. Altogether this involves 44 isotopes of 12 different elements.<sup>[1]</sup> Radium is a notable member of all three of these decay chains, being present as  $^{226}\text{Ra}$ ,  $^{223}\text{Ra}$ , and both  $^{228}\text{Ra}$  and  $^{224}\text{Ra}$  respectively.  $^{40}\text{K}$  decays directly to stable  $^{40}\text{Ca}$  or  $^{40}\text{Ar}$  and hence there is only one radioactive species. There are many other naturally occurring radioactive species such as tritium ( $^3\text{H}$ ) and  $^7\text{Be}$ , but they are present at much less significant levels than these three components.

While NORM consists of this large number of radionuclides, those of concern mainly include isotopes of uranium, thorium, radium, radon, lead and polonium. Of these, radium is of particular importance owing to the presence of radium isotopes in all three natural decay series, the relatively long half-lives and short lived progeny of two of the isotopes ( $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ), the high mobility of radium in the environment under a number of common environmental conditions and the tendency of radium to accumulate in bone following uptake into the body.<sup>[2][3]</sup>

$^{40}\text{K}$  is the largest source of natural radioactivity in the human body, that is, it is responsible for the largest number of radioactive decays. However, due to the fact that its content in the human body is under strict homeostatic control<sup>[4]</sup>, it will not be considered further.

NORM is found everywhere in both bedrock and surficial materials, but the concentrations vary from place to place. In sedimentary rocks, uranium tends to be concentrated in shale or clay rich layers. Sediments rich in bituminous material (coal or peat) are often favourable locations for uranium to be present at elevated concentrations. It should be noted that sedimentary rocks are formed at or near the earth's surface where uranium is more chemically mobile than is thorium. This greater mobility (of uranium) is exhibited by a low thorium/uranium ratio. This low ratio occurs as a result of enrichment in uranium rather than a depletion of thorium.<sup>[5]</sup>

## **Health Effects of NORM Radionuclides**

Contaminants reach human receptors through environmental pathways, namely air, soil, water, food and (in the case of radioactive contaminants) external exposure. Each contaminant or stressor has specific sources, transport, exposure mechanisms, and biochemistry; and each can impact health both directly and indirectly. NORM is often associated with unconventional oil and gas activities, equipment, and products because NORM exists naturally in below-ground hydrocarbon pools (see below). NORM emits ionizing beta ( $\beta$ ), or alpha ( $\alpha$ ) particles, and gamma ( $\gamma$ ) rays. Studies in northeastern BC have shown Rn and gamma rays associated with, and emitted from, scale and impurities in equipment, gas streams, and pipelines, sometimes at levels that exceed certain “acceptable risk” measures. Radon (an  $\alpha$  particle emitting gas) is considered to be a primary risk factor for lung cancers.<sup>[47]</sup> Depending on the density and proliferation of well pads, pipelines and other facilities, radiation exposure may be a threat to the public as well as workers. Radiation exposure is more likely when outside of populated areas (i.e., in the ‘bush’ at remote storage and processing operations); however, activities including hunting, fishing, snowmobiling, etc. are common in Nova Scotia’s back country where inadvertent exposure may occur. While the skin is able to shield some radiation (in particular  $\alpha$  and low energy  $\beta$  radiation), direct inhalation and ingestion and skin absorption expose internal organs to radiation. Exposure to NORM, particularly gamma rays, radium and radon, is known to cause various cancers and genetic mutations.<sup>[49]</sup>

The primary target for uranium is believed to be the kidneys. Kidney damage has been seen in humans and animals after inhaling or ingesting uranium compounds. Ingesting water-soluble uranium compounds will result in kidney effects at lower doses than following exposure to insoluble uranium compounds. Inhaled insoluble uranium compounds can also damage the respiratory tract. No health effects, other than kidney damage, have been consistently found in humans after inhaling or ingesting uranium compounds or in soldiers with uranium metal fragments in their bodies. Rats ingesting uranium over a long time had neurobehavioral changes and changes in the levels of certain chemicals in the brain. Uranium has been shown to decrease fertility in some studies of rats and mice; other studies have not found this effect. Very soluble uranium compounds on the skin caused skin irritation and mild skin damage in animals. Neither the National Toxicology Program (NTP), International Agency for Research on Cancer (IARC), nor the EPA have classified natural uranium or depleted uranium with respect to carcinogenicity.<sup>[8]</sup>

Studies on thorium workers have shown that breathing thorium dust may cause an increased chance of developing lung disease and cancer of the lung or pancreas many years after being exposed. Changes in the genetic material of body cells have also been shown to occur in workers who breathed thorium

dust. Liver diseases and effects on the blood have been found in people injected with Thorotrast. Many types of cancer have also been shown to occur in these people many years after thorium was injected into their bodies. Since thorium is radioactive and may be stored in bone for a long time, bone cancer is also a potential concern for people exposed to thorium. Animal studies have shown that breathing in thorium may result in lung damage. Other studies in animals suggest drinking massive amounts of thorium can cause death from metal poisoning. The presence of large amounts of thorium in your environment could result in exposure to more hazardous radioactive decay products of thorium, such as radium and thoron, which is an isotope of radon. Thorium is not known to cause birth defects or to affect the ability to have children.<sup>[7]</sup>

Radium occurs widely in water and occasionally in air as a dust or aerosol. As with all radionuclides, it can be inhaled (via pathways such as shower mist), ingested, or absorbed through intact or damaged skin. It can stay in bones for many years and can deposit there at a higher rate during periods of rapid growth, potentially affecting children more than adults. In addition to being a carcinogen and mutagen, it has also been linked with adverse reproductive, developmental, liver, kidney, and eye effects. Phosphate mining and fertilizer production can increase concentrations and exposures to radium and its daughter compound radon.<sup>[50]</sup>

Radon is a widespread, naturally occurring gas that is soluble in water, and is released into the air through showers and running faucets. In most parts of the United States radon-in-water is considered a minor contributor to indoor radon-in-air concentrations, compared with other sources such as soils and rocks beneath a building, but it may be a concern in some instances. Inhaled radon is a leading cause of lung cancer, a risk exacerbated for those who smoke. Health Canada has determined that about 16% of lung cancer in Canada can be attributed to radon exposure.<sup>[47]</sup> In addition, ingested radon may cause stomach cancer.<sup>[54]</sup>

<sup>210</sup>Pb is a naturally occurring radionuclide that occurs in air, soils, rocks, and water. Its chemical toxicity is the same as that of stable lead, which is widely acknowledged as causing a range of harmful effects even at very low concentrations. The prevalence of <sup>210</sup>Pb relative to stable isotopes of lead is quite low, and the risk posed as a carcinogen therefore is the main item of concern for this radionuclide. In addition to its carcinogenicity, <sup>210</sup>Pb has been found to be strongly elevated in the brains of Alzheimers and Parkinsons disease patients, especially in smokers.<sup>[9]</sup>

Polonium is a highly radioactive carcinogen, but hasn't received widespread attention as a drinking water contaminant. However, <sup>210</sup>Po has been found in drinking water in at least five U.S. states (Florida, Louisiana, Maryland, Nevada, and Virginia), sometimes at elevated concentrations (in the context of the limited data available). <sup>210</sup>Po can deposit in the liver, kidney, bone marrow, spleen, gastrointestinal tract, and gonads, and has been linked with adverse reproductive effects. Chronic intake can lead to increased absorption through the digestive tract, possibly due to intestinal wall damage caused by alpha particle emissions.<sup>[52]</sup>

Maximum allowable concentrations for naturally occurring radionuclides are found in [57] with technical guidance provided in [56].

## **Why is NORM Present at Elevated Levels in Oil and Gas Reservoirs?**

For decades the oil and gas industry has been using natural radioactivity to locate potential sites for drilling. In 1981 operators in the North Sea first began to identify scale deposits as being radioactive.<sup>[64]</sup> Scale is the accumulation of mineral deposits, such as calcium carbonate and barite, that precipitate

out of water and onto the inside of pipes and other equipment. Measurements of scale from the North Sea contained  $^{226}\text{Ra}$  at high concentrations.  $^{226}\text{Ra}$  was also present in sludges (muds resulting from the mixture of brine and fracturing fluids with rock dust and chips).<sup>[55][61]</sup>

Black shale, such as the Horton Bluff shale play in Nova Scotia, often contains trace levels of  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{40}\text{K}$ , and  $^{232}\text{Th}$  in higher concentrations than found in less organic-rich grey shales, sandstone, or limestone. This is because: 1)  $^{238}\text{U}$  and  $^{235}\text{U}$  preferentially bond to organic matter, such as algae that die and settle to the bottom of the ocean; and 2)  $^{40}\text{K}$  and  $^{232}\text{Th}$  preferentially bond to clays, which compose much of the sediment at the ocean floor.<sup>[62]</sup> Ultimately, because “black shales” contain more organic matter and clays, they are generally more radioactive than other shales or sedimentary rocks.<sup>[63]</sup> This radioactivity can be measured with sensitive equipment at black shale outcrops or equipment can be lowered into an uncased well, with radioactivity readings used to identify, map, and determine the thickness of the black shale facies.<sup>[55][61]</sup>

The end result is: radioactivity = organic richness = gas. The oil and gas industry uses a number of geophysical logging tools to characterize the subsurface rocks. The most commonly run logging tool, the gamma-ray log, is a very sensitive radiation detector (typically a scintillation detector) that measures the natural low-level radioactivity inherent in almost all sedimentary rocks. Most of the radiation emitted by these rocks is due to  $^{40}\text{K}$  which is found in feldspars, micas, clay minerals, and other common and abundant silicate minerals. On gamma-ray logs, shales can be differentiated from other rocks such as clean sandstones and limestones because shales have higher concentrations of  $^{40}\text{K}$ -bearing minerals. Organic-rich shales have higher radioactivity responses than typical shales because the organic matter tends to concentrate uranium ions that otherwise would be scattered throughout the sediment.<sup>[70][71]</sup> As a result, many organic-rich shales have uranium and thorium contents that can range from 10 to 100 ppm or higher (compared with a crustal average of 2-3 ppm), which will show up on a gamma-ray log as higher-than-normal gamma-ray responses. Comparisons of gamma-ray logs with drill cuttings show a fairly strong correlation between higher-than-normal radioactivity and black color in shales, derived from the organic content. To put it simply, black coloration generally correlates with organic richness, which correlates with high gamma-ray response. Finally, a number of studies have indicated an empirical relationship between high gamma-ray response and both gas production and total gas content in organic-rich shales. In other words, higher than normal gamma-ray response also equates to gas-production potential. The correlation is not 100 percent, but it is very high.<sup>[48]</sup>

Thus, in attempting to understand the oil yield and uranium content of black shales, the following points should be considered: 1. The organic matter in black shales accounts for all the oil yield and for most of the uranium content. In some shales, a clear-cut positive relation can be shown between oil yield and uranium content, but in others such a relation is lacking or is even inverse. 2. Oil is derived directly from organic matter, whereas most of the uranium is not in the original organic matter but is later attached to or precipitated in the presence of organic matter. 3. Two main types of organic matter in black shales, the sapropelic and humic, should be distinguished. The sapropelic type is derived from algae, spores, pollen, resins, cuticles, and analogous plant and animal remains. The humic type is derived from cellulose, lignin, and analogous woody parts of plants. Both types are present in varying proportions in most black shales. 4. The sapropelic type of organic matter yields 4 to 5 times more oil than does the humic type, whereas the humic type contains far more uranium than does the sapropelic type. 7. Other factors being constant, only where the proportion of sapropelic to humic type of organic matter in a shale remains constant will the oil yield and the uranium content have a high positive correlation. Because the humic type of organic matter is largely derived from land plants, this proportion would logically change with paleogeographic position, and the ratio of sapropelic to humic matter would increase with increasing distance from the shore.<sup>[6]</sup>

## **NORM and Shale Gas Exploration and Production**

In shale gas development, NORM can be found in drill cuttings, flowback and produced waters and natural gas. NORM is more noticeable in areas such as equipment, pipes and storage tanks where sediments or precipitates tend to accumulate, and as a result, exposure may occur when repair work is performed. Public concerns about water quality from horizontal gas well development include: aquifer and drinking water well contamination; waste storage pit leakage; spills of hydraulic fracturing fluids; handling of flowback and produced water streams; water use and supply; drilling waste disposal; stormwater runoff; and blowouts. These concerns stem from two related activities: 1) well development and completion, and 2) management of water and waste streams (handling, storage and disposal). Casing defects or damage, and cement failure to properly bond the well annulus, can result in upward migration of gas and fluids into shallow drinking water aquifers.<sup>[50]</sup>

A review of the literature on trace radioactivity in natural gas and natural gas products and the consequent radioactivity concentrations showed that dose rates due to natural radioactive elements in the natural gas produced from shale wells appear to fall within the range of those found in conventional gas wells. Also fracturing techniques do not appear to raise the relative concentration of radon in natural gas relative to conventional wells.<sup>[60]</sup> Consequently, the concentrations of radioisotopes, and specifically radon, are determined by the concentrations in the shale.

Some of the radioactivity is dissolved in water found in the gas deposits, and this 'formation water' becomes part of the 'flowback water' which comes out of the well during the fracking process and is the primary constituent of the produced water which comes up later in the life of the well. The flowback and produced water can then contaminate aquifers through multiple routes.

First, loss of zonal isolation in a well can result in direct contamination of aquifers. This can occur in a variety of ways. Gas flow that occurs immediately after cementing or before the cement is set is typically referred to as annular gas flow, or annular gas migration. This flow is generally massive and can be inter-zonal, charging lower-pressure formations, or can reach the surface and require well-control procedures. Flow to the surface occurring later in the life of a well is known as sustained casing pressure. This later flow can also be from gas-bearing formations to formations of lower pressure, generally at shallower depths. Causes of annular flow or sustained casing pressure include tubing and casing leaks, poor mud displacement during cementing, improper cement-slurry design, or cement damage after setting.<sup>[68]</sup>

Located between the casing and the well bore, a cement sheath is expected to provide zonal isolation throughout the life of a well. But its ability to do this depends on the proper placement of the cement, the mechanical behaviour of the cement and the stress conditions in the well bore. Even if the slurry was properly placed, changes in downhole conditions can induce sufficient stresses to destroy the integrity of the cement sheath. Over time, stresses are imposed on the cement by pressure integrity tests, increased mud weight, casing perforation, stimulation, gas production or a large increase in well bore temperature (which can occur as a result of production). Any of these events can damage the sheath. Often, damage to the cement sheath resulting from these forces shows up as microannuli. Even the smallest microannulus can be large enough to provide a pathway for fluid migration. In addition, drilling a well disturbs the stresses in the formation. Drillers must compensate for this disturbance, to the degree possible, by using drilling fluids (muds) to exert hydrostatic pressure on the formation. However, this pressure may be insufficient to maintain equilibrium with the far-field stresses, and the formation in which the well bore is located will deform. Draining fluids from a formation during production may also change formation pore pressure and related stresses. Within the rock, the resulting

increased loading can lead to varying degrees of deformation or failure that can cause cement to fracture or lose its bond at the interface with the formation rock. Production-induced stresses can also result in reservoir compaction, which may lead to shearing of a tubular and even buckling of a complete component.<sup>[69]</sup>

An assessment was performed of all the reliable data sets (25) on well barrier and integrity failure available in the published literature and online. These data sets include production, injection, idle and abandoned wells, both onshore and offshore, exploiting both conventional and unconventional reservoirs. The data sets have a wide range of numbers of wells examined, as well as ages of wells and study designs. For these reasons the percentage of wells that have had some kind of well barrier or integrity failure varies a lot between the studies (1.9%-75%). For example, of the 8030 wells in the Marcellus shale play inspected in Pennsylvania between 2005 and 2013, 6.3% have been reported to the authorities for infringements related to well barrier or integrity failure. In a separate study of 3533 Pennsylvanian wells monitored between 2008 and 2011, there were 85 examples of cement or casing failures, 4 blowouts and 2 examples of gas venting. In the UK, 2152 hydrocarbon wells were drilled onshore between 1902 and 2013 mainly targeting conventional reservoirs. UK regulations, like those of some other jurisdictions, include reclamation of the well site after well abandonment. Thus, there is no visible evidence of 65% of the well sites on the surface and monitoring is not carried out. The ownership of up to 53% of wells in the UK is unclear; this review estimates that between 50 and 100 are orphaned. Of 143 active UK wells that were producing at the end of 2000, one has evidence of a well integrity failure.<sup>[67]</sup>

In a study carried out in the Basses-Terres du Saint-Laurent, 130 residential wells, observation or municipal, were sampled in an area covering 14,000 km<sup>2</sup> between Montreal and Leclercville and the foothills of the Appalachians and Trois Rivières. Of these 130 wells, 18 had concentrations of methane exceeding 7 mg/l, which is the groundwater alert threshold proposed in the draft regulation on water extraction and protection, published May 29, 2013 in the official Gazette of Quebec. In about 95% of the wells containing high concentrations, methane is of biogenic origin, i.e., it is produced by bacteria in surface sediments. However, six wells have a mixture of biogenic and thermogenic (i.e., produced by cracking of hydrocarbons at depth) gas, and one well near Plessisville shows a signature which is clearly thermogenic. The helium, dissolved methane and to a lesser extent the radon show a certain relationship with distance from major faults in the region. The origin of this relationship between faults and the presence of natural gas in the groundwater of the region needs to be further explored in future work.<sup>[30]</sup>

Communication with older wells, whose well casing seals are more likely to have failed can also occur. This occurs deep underground, below the level of typical aquifers, but flow up the older well can, due to any of the issues discussed above, result in indirect contamination of aquifers.

In one study, geochemical evidence was collected in northeastern Pennsylvania showing that pathways, unrelated to recent drilling activities, exist in some locations between deep underlying formations and shallow drinking water aquifers. Integration of chemical data (Br, Cl, Na, Ba, Sr, and Li) and isotopic ratios (<sup>87</sup>Sr/<sup>86</sup>Sr, <sup>2</sup>H/H, <sup>18</sup>O/<sup>16</sup>O, and <sup>228</sup>Ra/<sup>226</sup>Ra) from this and previous studies in 426 shallow groundwater samples and 83 northern Appalachian brine samples suggest that mixing relationships between shallow ground water and a deep formation brine causes groundwater salinization in some locations. The strong geochemical fingerprint in the salinized (Cl > 20 mg/l) groundwater sampled from the Alluvium, Catskill, and Lock Haven aquifers suggests possible migration of Marcellus brine through naturally occurring pathways. The occurrences of saline water do not correlate with the location of shale-gas wells and are consistent with reported data before rapid shale-gas development in the region; however, the presence of these fluids suggests conductive pathways and specific

geostructural and/or hydrodynamic regimes in northeastern Pennsylvania that are at increased risk for contamination of shallow drinking water resources, particularly by fugitive gases, because of natural hydraulic connections to deeper formations.<sup>[22]</sup>

In 130 years of exploration, there have been 129 wells drilled in Nova Scotia, of which only 37 went more than 1000 m. 27 of these wells were drilled in the past 13 years, at locations in five different counties (Antigonish, Colchester, Cumberland, Hants, Pictou), although 17 of them were in Hants County. All wells drilled prior to 1995 are listed as plugged and abandoned. Of the newer wells, 14 are listed as suspended, and one as “location.”<sup>[66]</sup> The plugging and abandoning of the older wells occurred at a time when the regulatory process wasn't yet mature. Thus the integrity of the plugs in these wells should be considered suspect.

When flowback water comes to the surface it is stored in ponds, or sometimes tanks, which can leak. Ponds have also been known to overflow due to rainfall.

Most impoundments are lined with plastic sheeting. Pennsylvania requires that pit liners for temporary impoundments and disposal have a minimum thickness of 30 mils and that seams be sealed to prevent leakage.<sup>[40]</sup> Ohio's only requirement is that pits must be “liquid tight.”<sup>[40]</sup> However, improper liners can tear,<sup>[36]</sup> and there have been reports of pit liners tearing and pits overflowing in Pennsylvania and elsewhere.<sup>[41]</sup>

Soil and water (sludge) obtained from reserve pits used in unconventional natural gas well drilling and stimulation were analyzed for the presence of NORM. Samples were analyzed for total alpha, beta and gamma radiation, and specific radionuclides including  $^{210}\text{Pb}$  and  $^{214}\text{Pb}$ ,  $^{212}\text{Bi}$  and  $^{214}\text{Bi}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , thorium and uranium. Laboratory analysis confirmed elevated beta readings. Specific radionuclides present in an active reserve pit and the soil of a leveled, vacated reserve pit included  $^{232}\text{Th}$  decay series ( $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{208}\text{Tl}$ ), and  $^{226}\text{Ra}$  (from  $^{238}\text{U}$ ) decay series ( $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ,  $^{210}\text{Pb}$ ) radionuclides.<sup>[33]</sup>

In addition, there have been documented cases where companies have deliberately dumped waste water on fields or in waterways.

Oil-field equipment can contain radioactive scale and scale-bearing sludge, both of which form as coatings or sediments. The scale precipitates from flowback and produced water in response to changes in temperature, pressure, and salinity as the water is brought to the surface and is processed to separate coexisting natural gas. The scale is typically a mixture of carbonate and sulfate minerals. One of these sulfate minerals is barite (barium sulfate), which is known to readily incorporate radium. Many studies of radioactive scale from oil-field equipment have documented that barite is the primary host of oil-field NORM and that the radioactivity is from isotopes of radium and their decay products. The two radium isotopes present in produced water and barite scale are  $^{226}\text{Ra}$  (half-life = 1,600 years) and  $^{228}\text{Ra}$  (half-life = 5.8 years). The shorter lived radium isotopes normally decay before the scale is measured or handled. The concentration of dissolved radium is influenced by the abundance of uranium and thorium in reservoir rock and by the accessibility of water to the sites containing uranium and thorium. When radium is brought to the surface in produced water, the concentration of radium that is incorporated in barite scale is largely a function of (1) the concentration of dissolved radium and (2) the amount of produced water that moves past the site of barite precipitation. Radium tends to be more abundant in the more saline and chloride-rich varieties of these produced waters.<sup>[59]</sup> In this context, it should be recalled that the fracking waste water from the wells in Kennetcook, Nova Scotia were notable for their high salinity.

After hydraulic fracturing in the Marcellus and some other gas producing shales, the concentration of

dissolved salts in flowback and production waters increases dramatically with time. This has been thought to be due to dissolution of constituents from the shale by the water injected during hydraulic fracturing. However, an alternative origin of the high salinity is release of in situ brines (formation water) similar to those that are found to be produced from most oil and natural gas wells. Trends and relationships in brine composition indicate that (1) increased salt concentration in flowback is not mainly caused by dissolution of salt or other minerals in rock units, (2) the flowback waters represent a mixture of injection waters with highly concentrated in situ brines, and (3) these waters contain concentrations of radium and barium that are commonly hundreds of times drinking water standards.<sup>[24]</sup> As injection waters are depleted and diluted in the formation waters, the salinity increases. There is every reason to believe that this will be true of gas producing shales in Nova Scotia, which are of similar origin.

Drill cuttings, if improperly managed, also pose a risk to water quality. Drill cuttings brought to the surface during oil and gas development may contain NORM according to an industry report presented at the Society of Petroleum Engineers Annual Technical Conference and Exhibition.<sup>[42]</sup> According to the report, drill cuttings are stored and transported through steel pipes and tanks—which shield the bulk of the radiation. However, improper transport and handling of drill cuttings could result in water contamination or other exposure scenarios. For example, NORM concentrations can build up in pipes and tanks, if not properly managed, and the general public or water could come into contact with them, according to an EPA fact sheet.<sup>[43][36]</sup>

Even in the best case, when none of the above problems occurs, the water still has to be treated. Radioactivity cannot be neutralized or made non-radioactive. It can only be removed from the bulk of the material by methods such as filters or reverse osmosis, resulting in highly concentrated radioactive waste, which must be disposed of, typically at special licensed facilities, which don't exist in Nova Scotia.

Research published in December 2013 suggests one potential new treatment for radioactivity in fracking waste.<sup>[46]</sup> Vengosh and colleagues combined various proportions of flowback water with acid mine drainage (AMD) to test the possibility of using the latter as an alternative source of water for fracking. AMD—acidic leachate from mining sites and other disturbed areas—is an important water pollutant in some regions. Laboratory experiments showed that mixing flowback water with AMD caused much of the NORM in the flowback to precipitate, leaving water with radium levels close to EPA drinking water standards. The authors suggest the radioactive precipitate could be diluted with nonradioactive waste to levels appropriate for disposal in municipal landfills. If it can be brought to industrial scale, Vengosh says, this method could provide a beneficial use for AMD while reducing the need for freshwater in fracking operations and managing the inevitable radioactive waste.

Blending of acid mine drainage (AMD) with hydraulic fracturing flowback fluid results in precipitation of secondary minerals (e.g., celestine, barite and iron-bearing minerals) and sequestration of toxic levels of radium, barium, and strontium. The mixing experiments revealed the optimal conditions in which the NORMs and toxic metals can be removed, controlled by both the pH and sulfate concentration of the solution. The optimization of the NORM sequestration resulted in low NORM in solution but with high NORM levels in residual solids that need to be adequately managed and disposed of. While the laboratory tests have shown that it is technically possible to generate water effluents suitable for hydraulic fracturing, field-scale tests are necessary to confirm this feasibility under operational conditions.<sup>[55]</sup>

A preliminary radiological dose assessment of equipment decontamination, subsurface disposal, landspreading, equipment smelting, and equipment burial was conducted to address concerns regarding

the presence of naturally occurring radioactive materials (NORM) in production waste streams. The most significant conclusion of this report was: Assuming conservative residential land-use (i.e., individuals living and growing their own food on-site), landspreading with dilution presents the highest potential dose to the general public, on the order of 30 mSv/yr in the worst-case scenario of all of the methods assessed in this study. External irradiation, which contributes 80% of the total dose, is the exposure pathway of greatest concern. Arguments that the radium is virtually immobile in natural soils cannot be used to justify landspreading as a safe disposal method for NORM-contaminated wastes. While the relative immobility of radium in soils may be protective of groundwater, the other exposure pathways result in a total dose that is unacceptable.<sup>[58]</sup> So even assuming no mobilization of the NORM, land-spreading should not be performed.

However, the conventional wisdom about radium's stability in landfills rests on an assumption regarding its interaction with barite (barium sulfate), a common constituent in drilling waste. However, Charles Swann of the Mississippi Mineral Resources Institute and colleagues found evidence that radium in waste spread on fields may behave differently in soil than expected. When they mixed scale comprising radium and barite with typical Mississippi soil samples in the laboratory, radium was gradually solubilized from the barite, probably as a result of soil microbial activity. "This result," the authors wrote, "suggests that the landspreading means of scale disposal should be reviewed."<sup>[44]</sup> Radium is known to bioaccumulate in invertebrates, mollusks, and freshwater fish,<sup>[45]</sup> where it can substitute for calcium.

Concern has been raised in the scientific literature about the environmental implications of extracting natural gas from deep shale formations, and published studies suggest that shale gas development may affect local groundwater quality. The potential for surface water quality degradation has been discussed in prior work, although no empirical analysis of this issue has been published. A large-scale examination of the extent to which shale gas development activities affect surface water quality has been conducted. Focusing on the Marcellus Shale in Pennsylvania, it estimated the effect of shale gas wells and the release of treated shale gas waste by permitted treatment facilities on downstream concentrations of chloride (Cl<sup>-</sup>) and total suspended solids (TSS), controlling for other factors. Results suggest that (i) the treatment of shale gas waste by treatment plants in a watershed raises downstream Cl<sup>-</sup> but not TSS, and (ii) the presence of shale gas wells in a watershed raises downstream TSS but not Cl<sup>-</sup>.<sup>[26]</sup>

The discharge of effluent from a treatment facility has been found to increase downstream concentrations of bromide above background levels (in addition to Cl<sup>-</sup>). Barium and radium were substantially (>90%) reduced in the treated effluents compared to concentrations in the produced waters. Nonetheless, <sup>226</sup>Ra levels in stream sediments at the point of discharge were ~200 times greater than upstream and background sediments and were above radioactive waste disposal threshold regulations, posing potential environmental risks of radium bioaccumulation in localized areas of shale gas wastewater disposal.<sup>[45]</sup>

Natural gas contains varying amounts of <sup>222</sup>Rn which becomes dispersed in homes when natural gas is used in unvented appliances. Although there are few empirical data available, the natural gas industry has not been concerned about radon reaching its consumers in significant amounts, in part because of radon's short half-life and because much of it is released to the atmosphere at the wellhead.<sup>[43]</sup>

Radon decays to alpha-emitting daughter products which can contribute to lung cancer when inhaled and deposited in the respiratory system. For the average use of unvented kitchen ranges and space heaters, the tracheobronchial dose equivalent to individuals was estimated as 15 and 54 mrem/yr respectively, or 2.73 million person-rem/yr to the US population. A review of exposure conditions,

lung model parameters, dose conversion factors, and health effect factors indicated this population dose equivalent could potentially lead to 15 deaths a year from lung cancer in the US.<sup>[21]</sup> While these numbers are not up to date, they could indicate the order of magnitude of this exposure effect.

Risk Sciences International, of Ottawa, Ontario, performed an analysis of lung cancer risk due to radon in natural gas used for cooking for residents of New York State. They found an excess lifetime lung cancer risk of about 2 per 100,000.<sup>[27]</sup> This estimate was based on radon levels measured by Bowser Morner (<http://www.bowser-morner.com/radonlabfaq.html>). It is not indicated in the report how many samples were measured, where the samples were taken, or what the source of the measured natural gas was. However, the report's methodology is unaffected by this. It is only the magnitude of the lung cancer risk which would be in question. The radon level used in deriving the estimate was at the low end of the range of measurement values produced by Bowser Morner, and is probably too low for gas sourced within NY state.

In addition to the radon carried in gas to homes and other buildings, radon escapes with gas from many points in the distribution system from the well head out. Actual <sup>222</sup>Rn measurements made about 200 feet (60 m) from one well pad (with five well heads) prior to, during, and after fracking showed levels around 3 pCi/l, or 111 Bq/m<sup>3</sup>. A second test series at several three-phase separation/storage facilities and a recently completed well pad, also in northeast Colorado, indicated outdoor radon levels around 4.5 pCi/l, or 166.5 Bq/m<sup>3</sup>. Additional monitoring needs to be done to better characterize the radon levels released during the fracking process and at the many separation/storage facilities in the rather large geographical area.<sup>[28]</sup>

If one assumes that there is no wind during these measurements, and that radon transport from the well heads and other gas facilities is therefore radially uniform, the radon concentration near the well heads in the first measurement would be on the order of 10-100 kBq/m<sup>3</sup>. This is within the range of soil gas radon concentrations commonly encountered<sup>[29]</sup>, but is significantly higher than the 17 pCi/l (630 Bq/m<sup>3</sup>) assumed in the RSI analysis. The lifetime excess lung cancer risk is linear with respect to the radon concentration. This together with these higher radon concentrations suggest the possibility that the actual lifetime excess lung cancer risk for Nova Scotians cooking with natural gas sourced within the province may be 10 to 100 times higher than the 2 per 100,000 calculated by RSI. As a Canadian reference, the Health Canada guideline for radon-in-air in homes and workplaces is 200 Bq/m<sup>3</sup>. In addition, the average residential radon level in Canada is about 78 Bq/m<sup>3</sup> and Health Canada has calculated that this level causes about 16% of all lung cancer in Canada, i.e., over 4000 lung cancer cases per year.<sup>[47]</sup>

In an Australian study conducted in a coal seam gas field, an ~3 fold increase in maximum <sup>222</sup>Rn concentration was observed inside the gas field compared to outside of it. There was a significant relationship between maximum and average <sup>222</sup>Rn concentrations and the number of gas wells within a 3 km radius of the sampling sites. The hypothesis was that the radon relationship was a response to enhanced emissions within the gas field related to both point (well heads, pipelines, etc.) and diffuse soil sources.<sup>[25]</sup> This could also be expected to be true for shale gas fields.

Radon soil gas data are severely lacking for Nova Scotia. Limited, but very focused radon data, collected by mineral exploration companies engaged in the search for uranium deposits during the 1970s and early 1980s, are tabulated in [05]. The association of radon and uranium is well understood. Naturally occurring concentrations of uranium have been detected in all types of geochemical sample media (soil, till, lake bottom sediment, stream sediment, stream water, humus, vegetation and rock) throughout Nova Scotia and analyzed by the Nova Scotia Department of Natural Resources and, previously, the Nova Scotia Department of Mines and Energy. Uranium concentrations range from

parts per billion (ppb) up to percentages in mineralized environments, depending on the sample medium analyzed. A compilation of airborne radiometric surveys covering Nova Scotia has been completed by the Geological Survey of Canada and demonstrates that uranium exists throughout the province. A total of 72 sites were sampled for radon soil gas concentrations across Nova Scotia during the 2007 and 2008 field seasons resulting in an overall average sampling density of approximately 1 sample per 800 km<sup>2</sup>. Radon in soil gas was detected at all sample sites (except one near Meaghers Grant, discussed below) tested during the 2007 and 2008 sampling program, regardless of the soil type and conditions, or the underlying bedrock geology. At the Meaghers Grant site, extraction of soil gas from each of the five probes was extremely difficult. With the exception of the Meaghers Grant sample site, radon soil gas concentrations (calculated as the mean of the concentrations from the five probes) ranged from a low of 0.1 kBq/m<sup>3</sup> to a high of 207.0 kBq/m<sup>3</sup> with a mean of 25.3 kBq/m<sup>3</sup> (median of 20.8 kBq/m<sup>3</sup>). It is important to stress that the radon soil gas concentrations represent natural background conditions and are not related to what would be termed uranium occurrences. It is also important to stress that the raw field data were used to calculate the mean radon level for each site.<sup>[29]</sup>

## **Regulatory Environment**

Nova Scotia doesn't have any regulations dealing with radioactivity and the environment or the public. What regulations exist deal only workers and workplaces. Even those regulations only involve an implicit reference to guidelines developed by the Federal-Provincial-Territorial Radiation Protection Committee, which do not carry the force of law. This means that right now, if a company were to leave a pond full of contaminated water behind at a well site, the province has to make up rules on the fly, which inevitably leads to problems. An example of this is the water currently being stored in Kennetcook and Debert. To fix this situation the province will need to adopt appropriate regulations and hire trained staff who, in the event of large scale fracking operations, would have to be dedicated to this specific regulatory task.

## **Conclusion**

There are known risks from radioactivity associated with the fracking process. What is more troubling however, is what we don't know, which far exceeds what we do know. For example, we don't know what radioactivity levels exist in any shale or coal gas deposits in Nova Scotia. The only information we have on this comes from the waste waters stored in Debert and Kennetcook, and that information is both incomplete and probably not representative of actual conditions downhole. The tested samples were only taken long after the flowback was collected and in the interim, much of the radioactivity would have decayed and/or settled into untested sludge. Radon testing was never performed in the vicinity of the well heads, or by sampling any gas produced.

As a health physicist, my work has been heavily influenced by the precautionary principle, which denotes a duty to prevent harm, when it is within our power to do so, even when all the evidence is not in. To prevent harm, the first question one should ask is, is this practice necessary, or is there an available alternative which doesn't involve the same risks? Clearly there are many risks associated with fracking, and radioactivity and radiation are among them. Equally clearly, there are available alternatives which don't involve the same risks. I urge the Commission to find that Nova Scotia should implement a ban on fracking.

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*This review evaluates risks to public health from chemical and nonchemical stressors associated with UNG, describes likely exposure pathways and potential health effects, and identifies major uncertainties to address with future research. Overall, the current literature suggests that research needs to address these uncertainties before we can reasonably quantify the likelihood of occurrence or magnitude of adverse health effects associated with UNG production in workers and communities.*

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*The purpose of this document is to provide guidance and highlight industry recommended practices for well construction and integrity for those wells that will be hydraulically fractured. The guidance provided here will help to ensure that shallow groundwater aquifers and the environment will be protected, while also enabling economically viable development of oil and natural gas resources.*

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*I was hired to assess the hydraulic fracturing process and inspect the integrity of the Drake Drilling Corporation's machines, storage tanks, drills and created wells. My findings would then lead me to either pass the company's inspection or require additional improvements to continue production. The continuation of hydraulic fracturing by the Drake Drilling Corporation would mean an eventual increase in health related problems in surrounding areas to the fracking site, and overall complete contamination of the surrounding environment. The choice that I was asked to make by the CEO of the DDC would mean the violation of many ethical codes of not only engineers but me as a person in society. The consequences of continued production without the problems being fixed would be so great, that even I as a person could not live with it. For that reason, and many others, I have decided to deny the offer of the CEO, and fail the inspection of The Drake Drilling Corporation. My decision will most likely provide difficult times for the DDC, but more notably, ensure a safer environment for the public. Engineers around the world are required to abide not only by codes of ethics set by society, but also their own code of ethics. Toxic gases, radioactive material, hazardous byproduct, and environmental destruction are some of the many consequences of hydraulic fracturing which are no longer major problems due to my decision.*

Goodwin, T.A., et al, *Radon Soil Gas in Nova Scotia*, Mineral Resources Branch, Report of Activities, 2008, pp. 25-34, ed. MacDonald, D.R., <http://www.envnetwork.smu.ca/documents/RadonSoilGasinNovaScotia.pdf>

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*The potential for groundwater contamination is higher when producing natural gas and oil from unconventional sources, including shale and tight sandstones. These reservoirs require horizontally-completed wells and massive hydraulic fracturing that injects large volumes (up to 50,000 m<sup>3</sup>/well) of high-pressured water with added proppant, and toxic organic and inorganic chemicals. Recent results show that flow back and produced waters from Haynesville (Texas) and Marcellus (Pennsylvania) Shale have high salinities ( $\geq 200,000$  mg/L TDS) and high NORM (up to 10,000 picocuries/L) concentrations. A major research effort is needed worldwide to minimize all potential environmental impacts, especially groundwater contamination and induced seismicity.*

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