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Naturally Occurring Radioactive Materials and Chemical Analysis

of

Produced Water Samples

submitted by

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Abstract

Naturally occurring radioactive materials (NORM) are everywhere and we are constantly exposed to it. Natural radiation has been around since the beginning of the Earth and is found in our bodies, our food, and in the products we use. This study investigates the presence and activity of naturally occurring radioactive materials contained in produced water which is associated with oil and gas exploration. Specifically the principal radionuclide used to measure NORM was radium. We also measured , where possible, other parameters which may have had an effect on the NORM activity.

Produced water samples from two different oil field sites, Fausse Pointe and North Broussard, have been used as test sites for this water analysis study. The overall purpose was to determine the specific, selected parameters associated with the produced waters, which are defined as the waters which are brought to the surface along with oil and natural gas. This study is also a precursor in an attempt to predict the activity of naturally occurring radioactive materials and the other factors which may affect the activity and/or occurrence of NORM in produced water samples. A control site used for comparison, was Lake Kernan on the Southern University Campus.

The average reading for the first site, Fausse Point, was pH 6.39, with the total dissolved solids, and conductivity readings off scale for our system. The radium analysis of the Fausse Point site (avg value of 0.5291 pCi/mL) indicates a relatively Ahot@ NORM area of activity. This activity indicates a potential, future NORM regulatory problem for this site.

The average reading for the second site, North Broussard, was pH 7.39, with the total

dissolved solids, and conductivity readings again being very high. The radium analysis of the North Broussard site (avg. of 0.4697 pCi/mL) also indicates a relatively Ahot@ NORM area of activity.

The average readings for our control site, Lake Kernan, were pH 7.29, total dissolved solids 62.5 milligrams per liter and conductivity, 141.7 microsiemens, respectively. The radium activity of the Lake Kernan site was, as expected, negligible. Metals analysis for each of the sites was also performed.

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INTRODUCTION

This research explores the occurrence of NORM and other parameters associated with produced water. The purpose of this study is to ultimately lead to methods for predicting NORM activity which, in time, will ultimately allow the oil and gas industry to consider disposal options and costs prior to exploration. NORM is becoming an ever increasing concern to the oil and gas industry as the State of Louisiana, and other States, move towards regulating the disposal of NORM. As a result, this and other studies of this type, are needed to minimize, and in some cases eliminate future problems.

The North Broussard and Fausse Point sites, have been used as the sampling sites for this produced water study. Water samples are subject to have a number of different substances dissolved within them. These dissolved substances can affect the properties of water and also the properties of other substances within water specimens. In addition, the quality of water samples are also subject to environmental factors, such as the geology of the immediate area.

Produced waters (water associated with oil and gas exploration) is the major focus of this study. In oil and gas wells, there are three major components present: natural gas which is the top layer, oil, which is the middle layer, and finally water which is the bottom layer. This layer of water is called formation water and is subjected to the environment surrounding it. As gas and oil are removed from wells, formation water begins to rise in wells. This formation water generally contains numerous dissolved substances, some of which are metals and some which may be radioactive.

Radioactive components in formation waters arise primarily from the presence of

Uranium-238 and Thorium-232, and these elements decay further to other radioactive species. Radioactive decay products of primary concern in this study are Radium-226 which is derived from the decay of Uranium-226, and Radium-228, which is a decay product of Thorium-232.

Ultimately, this study is to determine specific parameters of produced waters. These produced waters may or may not contain various levels of naturally occurring radioactive materials (NORM). In an attempt to apply developed water analysis procedures and techniques, the two sites were selected for testing due to their close proximity and small size. A control site, Lake Kernan, was also used as a natural water system.

Another concern of NORM is that it is deposited in scales and sludges throughout oil and gas production and processing equipment. The deposition of NORM is usually associated with processed water because the radium is dissolved in the water that comes out of the ground. NORM levels in produced waters may be affected by a number of water parameters. These parameters include total dissolved solids, dissolved oxygen, salinity and heavy metal concentration. NORM does not include source, by-product, or special nuclear material (terms defined by law and referring primarily to uranium, thorium and nuclear fuel cycle product); or commercial products containing small quantities of natural radioactive materials (i.e. phosphate fertilizer, potassium chloride for road de-icing) or natural radon in buildings.

REVIEW OF LITERATURE

Naturally Occurring Radioactive Materials (NORM) are of increasing public health and worker safety concern for the oil and gas industry. NORM is deposited in scales and sludges

throughout oil and gas production and processing equipment. The deposition of NORM is usually associated with processed water because the radium is dissolved in the water that comes out of the ground. NORM levels in produced waters may be affected by a number of water parameters. These parameters include total dissolved solids, dissolved oxygen, salinity and heavy metal concentration. The enhancement has occurred when a naturally occurring radioactive material has its composition, concentration, availability or proximity to people altered by human activity. The term is usually applied when the naturally occurring radionuclide is present in sufficient quantities or concentrations to require control for purposes of radiological protection of the public or the environment. NORM does not include source, by-product, or special nuclear material (terms defined by law and referring primarily to uranium, thorium and nuclear fuel cycle product); or commercial products containing small quantities of natural radioactive materials (i.e. phosphate fertilizer, potassium chloride for road de-icing) or natural radon in buildings.

Naturally occurring radioactive materials are everywhere. Radioactive isotopes of uranium, thorium, carbon, potassium, polonium, lead, radon, and more than a dozen other elements are present in rocks, soils, building materials, consumer products (e.g. law fertilizers), food, and even in our own bodies.

One particular place NORM is present in, is in the oil and gas industry, and it is a major concern for these industries in that radionuclides are known to be associated with organic materials in nature. Therefore, oil, gas, and oil field brines frequently contain radioactive materials. These materials accumulate in piping used to remove and process petroleum and natural gas. The Environmental Protection Agency estimates that about eight million metric ton

of NORM waste will be produced by the gas and oil industry over the next twenty years.

NORM in oil and gas fields results from accumulation of Radium (Ra) isotopes, which are decay products of parent uranium and thorium. Radium is a radioactive metallic element, discovered in pitchblende in 1898 by Pierre and Marie Curie. It is a rare, lustrous, white alkaline earth metal that resembles barium in its chemical properties. Radium compounds are found in uranium ores. In its radioactive decay, radium emits alpha, beta, and gamma rays and produces heat. Ra isotopes in reservoir fluids are not supported by uranium (U) and thorium (Th) parent isotopes in the fluids. Ra in solution is not produced by U and Th in solution. To predict areas likely to have high potential for NORM, therefore, it is necessary to understand the geologic and lithologic distribution of the parent isotopes and the mechanisms by which daughter products escape to the fluid phase. Two hypotheses regarding NORM in produced water and oil- producing and gas- processing equipment scale can be evaluated. The first hypothesis is that NORM is produced by locally high U and Th concentrations in the reservoir rocks. If this is generally true, NORM scale will be largely controlled by geologic formation and lithology. The second hypothesis is that NORM is released from ordinary geologic media during normal geologic processes. If this is generally true, the potential for NORM scale precipitation can be predicted largely from basin setting and history. Determining which of these hypothesis prevails will enable us to improve our ability to predict where NORM scale is most likely to accumulate in amounts that produce a health hazard.

NORM shows up with natural gas in co-produced waters. Since the Gulf Coast reservoirs of natural gas are entirely water-saturated, co-production of water bearing NORM is a serious and

regulated issue for all producers in the area. With Natural Gas supplies balancing

Table I. Principal Natural Radionuclide Decay Series

Nuclide	Half-Life	Major Radiations
Uranium-238	4.47 billion years	alpha, x-rays
Thorium-234	24.1 days	beta, gamma, x-rays
Protactinium-234	m1.17 minutes	beta, gamma
Uranium-234	245,000 years	alpha, x-rays
Thorium-230	77,000 years	alpha, x-rays
Radium-226	1600 years	alpha, gamma
Radon-222	3.83 days	alpha
Polonium-218	3.05 minutes	alpha
Lead-214	26.8 minutes	beta, gamma, x-rays
Bismuth-214	19.7 minutes	beta, gamma
Polonium-214	164 microseconds	alpha
Lead-210	22.3 years	beta, gamma, x-rays
Bismuth-210	5.01 days	beta
Polonium-210	138 days	alpha
Lead-206	stable	
Thorium-232	14.1 billion years	alpha, x-rays
Radium-228	5.75 years	beta
Actinium-228	6.13 hours	beta, gamma, x-rays
Thorium-228	1.91 years	alpha, gamma, x-rays
Radium-224	3.66 days	alpha, gamma
Radon-220	55.6 seconds	alpha
Polonium-216	0.15 seconds	alpha
Lead-212	10.64 hours	beta, gamma, x-rays
Bismuth-212	60.6 minutes	alpha, beta, gamma, x-rays
Polonium-212	0.305 microseconds	alpha
Thallium-208	3.07 minutes	beta, gamma
Lead-208	stable	
Potassium-40	1.28 billion years	beta, gamma
Argon-40	stable	
Calcium-40	stable	
Rubidium-87	4.7 billion years	beta
Strontium-87	stable	

Source: NRC 1994

demand, a reduction in risk related to production over-costs is of vital importance. Risks may be associated with unexpected disposal costs when high NORM containing waters are produced or when other chemical composition declare the waste hazardous. Of particular interest in this study, are NORM levels associated with produced waters. NORM is rapidly becoming the "hot" waste of concern [U.S. Water News, 1991]. Studies by Hanan [Hanan, 1981] and [Reid, 1984], indicate produced waters, worldwide, generally contain high Radium 226 levels.

Radium 226 is the NORM material of major interest because it is an intermediate member of the Uranium- 238 series, is water soluble, and therefore is classified as a water borne pollutant. Millions of gallons of produced water containing NORM contamination are generated every year in Louisiana with their fate mainly unknown [Bohlinger, 1990]. In 1990, the Louisiana Department of Environmental Quality, LADEQ, requested NORM data from produced water dischargers in the State of Louisiana. Subsequent analysis of the 403 sites responding [Schlenker and St. Pe', 1990] indicated a mean Radium 226 activity of 175 pCi/L. These levels are compared to the baseline activities of less than 1 pCi/L for surface waters [Louisiana Department of Environmental Quality, 1989], and 0.8 to 2.8 pCi/g for saline, brackish and fresh marshes reported by DeLaune [DeLaune, et.al., 1986]. Both the Nuclear Regulatory Commission and the State of Louisiana have set regulatory limits [Wagner, 1990], for Radium at 60 pCi/L, above which the waste is considered hazardous and requires expensive disposal procedures.

One innovative means of reducing these disposal costs is by prediction of NORM levels prior to drilling. The study by Schlenker and St. Pe' indicates that Radium 226 activities in produced water effluents vary from site to site. A study by Kramer and Reid [Kraemer and Reid 1984],. attributes the variation in site effluent Radium activities to differences in the mineral

composition of geologic formations. This study also suggested that Radium 226 was leached from the surrounding subsurface formations. Accordingly data relating NORM levels with geologic information and ultimately prediction of NORM activities based on geologic formations would be very valuable.

NORM in oil and gas field operations occurs as produced water; sludges, sands, minerals and scale precipitated in down hole or aboveground piping, valves, or gauges; or as thin platings on the inner surfaces of gas processing and transporting equipment; or as a gas. (Smith, 1987; Baird and others, 1990; White, 1992). NORM levels in produced water range from essentially background values to activities as high as several thousand picocuries per liter (pCi/L) (White, 1992).

Ra has an ionic charge (+ 2) and radius similar to that of other Group 2A elements (1.48 angstroms in 8-fold coordination, versus 1.12, 1.26, 1.42 angstroms for calcium (Ca),strontium (Sr), and barium (Ba), respectively (Shannon, 1976). Of the Group 2A elements Ca is the most abundant in the tissues of living organism (biomass), and therefore the effects of Ra substitution for Ca are of the greatest concern. Ingested Ra has been associated with bone cancer, bone sarcoma, and head carcinoma, the last of which is presumably caused by production of Rn gas that accumulates in head cavities (Mays and others, 1985). Rn, a decay product of Ra, exists as a gas and is associated with occurrences of lung cancer (Wanty and Schoen, 1993).

The first hypothesis is that NORM is produced by locally high U and Th concentrations in the reservoir rocks. If this is generally true, NORM scale will be largely controlled by geologic formation and lithology. The second hypothesis is that NORM is released from ordinary geologic

media during normal geologic processes. Many workers have noted a correlation between Ra concentrations and total salinity in ground water and formation water (for example, Dickson, 1990). High salinity appears to be a requisite for high Ra concentrations (Dickson, 1990), although not all saline waters support high Ra activities.

RESULTS AND DISCUSSION

In addition to the geological weathering of rock and soil [20], NORM concentrations vary because of physical and chemical processes, both natural and anthropogenic. If mobilized, the NORM radioisotopes are available for transport. When radionuclides are dissolved in groundwater, the isotopes tend to travel with the water until redeposition takes place. The average nuclide activity found in the North Broussard and Fausse Point sites are listed in Table II and III below. For this study the control group used was Lake Kernan on the Southern University Campus. The Lake Kernan average for pH, total dissolved solids (TDS) and conductivity readings were 7.29, 62.5 milligrams per liter, and 141.7 microseimens respectively. The reason for using the Lake Kernan site was to compare produced water readings to the readings obtained from a natural water system. The values obtained for Lake Kernan all fall within the ranges expected for natural water systems.

The average reading for the Fausse Point site was pH 6.39, with the TDS, and conductivity readings off scale for our system. The radium analysis of the Fausse Point site (avg value of 0.5291 pCi/mL) indicates a relatively hot NORM area of activity. This activity indicates a potential, future NORM regulatory problem for this site. The average reading for the North Broussard site

was pH 7.39, with the TDS, and conductivity readings again being off scale for our system. The radium analysis of the North Broussard site (avg. of 0.4697 pCi/mL) indicates a relatively a hot NORM area of activity.

TABLE II: Average Nuclide Activity (North Broussard)

NUCLIDE	AVG. ACTIVITY (pCi/mL)
Cr-51	7.306E-02
Co-60	6.697E-03
Sr-85	2.177E-03
Ra-226	**4.697E-01**
Sn-113	1.835E-02

Table III: Average Nuclide Activity (Fausse Point Site)

NUCLIDE	AVG. ACTIVITY (pCi/mL)
Cr-51	5.753E-02
Co-60	6.409E-03
Sr-85	4.020E-03
Ra-226	**5.291E-01**
Sn-113	9.056E-02

The major radioactive isotope of interest, Radium-226, was chosen due to its mobility as a result of being water soluble and also because of its significant half life of 1,526 years. This long half life poses a potential threat in that NORM levels in produced water range from essentially background values to activities as high as several thousand picocuries per liter (pCi/L). This is a result of the fact that, redeposition, involves the same factors as does mobility. Changes in any of the parameters of a stream of material may result in reduced mobility and subsequent redeposition. These processes may also take place preferentially; and ultimately increased concentrations of specific minerals may occur. Examples include: chlorination of metallic ores as one step in metal production, mobilizes radium, which accompanies uranium in the ore. The high solubility of RaCl2 relative to other species leads to extraction of radium wherever the parent mineral is exposed to chloride ions. Production of brine or brine-contaminated oil includes dissolved radium as well, since the brine contains chloride ions.

Groundwater chemistry may change as the water reaches the surface or as it passes through different strata and the dissolved minerals form at the surface. Changes in pH, oxidation state, or chemical equilibrium may result in precipitation of dissolved minerals. This mechanism accounts for the existence of many ore bodies. Addition of alum and softening chemicals in drinking water treatment plants similarly precipitates radium with the other minerals. Uncontrolled discharges from tailings piles may contain extracted radionuclides (as well as other heavy metals).

Oxidation-reduction potential can affect solubility. Variation in oxidation state affects solubility since complex formation depends strongly on oxidation state. For example, water exposed to sulfur-bearing minerals generally exhibits reducing potential, which may alter the oxidation state of other minerals in contact with the water [21].

Low solubility of alkaline earth SO_4^{2-} (sulfate) species, is also a factor in redeposition of NORM. Movement of sulfuric acid solutions through piping in mineral extraction processes is known to cause precipitation of scale containing high concentrations of radium. Production of water containing sulfate-bearing solutions can also cause precipitation of pipe scale containing elevated concentrations of uranium.

Adsorption depends on the substrate and the specific species in question. Clays are known to adsorb some chemical species preferentially over others; passage of groundwater through a layer of clay may strip out NORM species that would otherwise travel with the water. Adsorption of radon on activated charcoal is an equilibrium process. Desorption can occur if the ambient radon concentration drops; saturation can prevent further radon removal.

Ion exchange is used to control water chemistry, typically to remove contaminants, soften potable water, or remove radium. Ion exchange does not cause mobilization of radionuclides, but once mobilized in water, any subsequent treatment by ion exchange has clear potential for reconcentration.

Temperature-dependent variations in solubility may also result in increased concentrations of radionuclides, together with other elements in geothermal waters. Thermal processes can mobilize radionuclides. Combustion of coal or lignite volatilizes some isotopes (thorium, uranium, radium, and bismith). Subsequent redeposition may occur in process equipment, in pollution control equipment, or in the environment. Minerals dissolved in naturally occurring geothermal waters typically plate out as the temperature drops. Deposits of scale containing substantial concentrations of radionuclides may result.

As the water comes to the surface and the partial vapor pressure drops, radon dissolved in water partitions into the air. In open air, dilution, convection, and diffusion minimize increases in concentration, but in caves or buildings, higher concentrations of radon and its progeny may result. Brines exhibit similar behavior. Oil mixed with brine brings radium with it; as the chemical and physical conditions change in the pipe string and at the well head, the radium precipitates with other minerals and forms scale inside the piping.

Another consideration when dealing with NORM is the term TENORM, which stands for technically enhanced naturally occurring radioactive materials. TENORM is found in many waste streams; for example, scrap metal, sludges, slags, fluids, and is being discovered in industries traditionally not thought of as affected by radionuclide contamination.

The majority of radionuclides in TENORM are found in the uranium and thorium decay chains. The decay products of Radon are the largest source of natural radioactivity we are exposed to. We will briefly review other radionuclides occurring in nature (carbon, potassium and rubidium) that contribute primarily to background doses. It again should be noted that radium and radon are the principal radionuclides used to measure NORM as well as TENORM in the environment. Proposed regulations are moving from concentration-based standards to dose-based standards. The Health Physics Society and the Council of Radiation Control Program Directors are working on standards for States to adopt.

Radioactivity in oil and gas production and processing equipment is of natural origin and is now known to be widespread, occurring throughout the world. Estimates suggest that up to 30 % of domestic oil and gas wells may produce some elevated TENORM contamination. The geographic

areas with the highest recorded measurements were northern Texas and the gulf coast crescent from southern Louisiana and Mississippi to the Florida panhandle. Very low levels of TENORM radioactivity were noted in California, Utah, Wyoming, Colorado, and northern Kansas fields. Uranium and thorium compounds are mostly insoluble and as oil and gas are brought to the surface, remain in the underground reservoir. As the natural pressure within the bearing formation falls, formation water present in the reservoir will also be extracted with the oil and gas. Some radium and radium daughter compounds are slightly soluble in water and may become mobilized when this production water is brought to the surface. The precipitate consists principally of barium sulfate ($BaSO_4$), calcium sulfate ($CaSO_4$), and calcium carbonate ($CaCO_3$). Because the chemistry of radium is similar to that of barium and calcium (all are Group IIA elements), radium may also precipitate to form complex sulfates and carbonates.

As a result it was decided to determine the metal concentrations of the produced water samples. The selected metals analysis are indicated in Tables V and VI and were chosen due to their being a Group II metal or having a common $+2$ ion.

Table IV: Average Metal Concentrations (Fausse Pointe)

METAL	Fausse Pointe	Lake Kernan
	Conc. (ppm)	Conc. (ppm)
Copper	0.221	0.00
Iron	12.33	1.506
Lead	2.858	0.258
Barium	106.4	0.000
Strontium	68.94	0.224

Table V: Average Metal Concentrations (North Broussard)

METAL	North Broussard	Lake Kernan
	Conc. (ppm)	Conc. (ppm)
Copper	0.000	0.000
Iron	0.782	1.506
Lead	1.221	0.258
Barium	19.54	0.000
Strontium	7.152	0.224

As the produced water is subjected to changes in temperature and pressure, dissolved solids may precipitate out of solution and deposit sludge within the oil production system. These deposits are generally in the form of oily, loose material. Sludge often contains silica compounds, but may also contain significant amounts of barium. Some of the solids in the original product stream are removed in the separator and accumulate there as sludge. As the stream is further treated using heater/treaters to separate oil from water, sludge is also separated and allowed to accumulate. The largest volumes of sludge settle out of the production stream and remain in the oil stock and water storage tanks. Radionuclide concentrations in sludge vary from background levels to several hundred pCi/g, with the highest concentrations in the separator and collection areas near the separator (drains, etc.) (Table VIII). The levels deposited in heater/treaters and in sludge holding tanks are about a factor of 10 less than those found in the separator. TENORM concentrations in sludge deposits in heater/treaters and tanks are generally around 2.78 Bq/g (75 pCi/g).

For comparison purposes, radioactivity of some natural and other materials are as follows: 1 adult human 7000 Bq, 1 kg of coffee 1000 Bq,1 kg superphosphate fertiliser 5000 Bq, 1 household smoke detector 30,000 Bq, Radioisotope for medical diagnosis 70 million.

NORM Geology

Originally this study was to correlate NORM activity with the specific geological formations of the wells in which the produced water samples were obtained. However required

budget revisions, well sales, and the inability to obtain the required information from well owners, did not allow for this to occur.

CONCLUSION

The results of this study indicates that there may be a potentially serious problem with the NORM associated with oil and gas exploration. As more and more States move towards regulating the disposal of NORM contaminated wastes, both liquid and equipment, the oil and gas industry has to address the disposal problems associated with exploration.

Based on the results of this project, the two sites selected, North Broussard and Fausse Point, are sites which have NORM levels which will be above most, if not all, current and/or future regulatory limits. More work must be done to establish the effects of other parameters, such as the metal content of produced waters, before more definitive conclusions can be reached.

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