From: DamascusCitizens
Sent: Friday, November 15, 2013 9:54 PM
To: ord.docket@epa.gov; Hanlon, Edward
Subject: Attention Docket ID No. EPA-HQ-ORD-2010-0674

Comment from:
On behalf of Damascus Citizens for Sustainability and NYH2O we are submitting the following information for consideration by the EPA Science Advisory Board and the researchers conducting the nationwide study of high volume hydraulic fracturing impacts.
B. Arrindell
Director
Damascus Citizens for Sustainability

This comment contains an attachment and citations for two USGS papers: one on Radium in oil / gas produced waters and the other (Ryder) on oil and gas movement

"First, these plots suggest that in both TPSs oil and gas migrated vertically at least 1,000 ft through relatively impermeable shale and carbonate, probably facilitated by fractures and faults. ...and migrated either into adjoining anticlines or updip as far as 50 miles. Furthermore, oil and gas generated and expelled from the Utica Shale in Ohio and Pa. suggest the following migration patterns: 1) westward across-dip migration for 30 to 80 miles through about 1,000 ft of underlying Ordovician carbonate rocks before entrapment in Cambrian reservoirs, and 2) vertical migration through about 1,500 ft of overlying Ordovician shale followed by updip migration as far as 50 miles before entrapment"

Citations:

Ryder, R.T., Trippi, M.H., Swezey, C.S., Milici, R.C., Repetski, J.E., Ruppert, L.F., and Rowan, E.L. A Regional Perspective of the Devonian Shale and Ordovician Utica Shale Total Petroleum Systems of the Appalachian Basin. 2011. American Association of Petroleum Geologists' Eastern Section Meeting, Washington, DC, USA, 25-27 September 2011.
http://www.gswweb.org/aapg/abstracts/Ryder.pdf

Rowan, E.L., Engle, M.A., Kirby, C.S., and Kraemer, T.F. Radium Content of Oil- and Gas-Field Produced Waters in the Northern Appalachian Basin (USA): Summary and Discussion of Data. 2011. United States Geological Survey Scientific Investigations Report 2011–5135.
http://pubs.usgs.gov/sir/2011/5135/pdf/sir2011-5135.pdf

Radium Content of Oil- and Gas-Field Produced Waters in the Northern Appalachian Basin (USA): Summary and Discussion of Data

Scientific Investigations Report 2011–5135

U.S. Department of the Interior
U.S. Geological Survey

Radium Content of Oil- and Gas-Field Produced Waters in the Northern Appalachian Basin (USA): Summary and Discussion of Data

By E.L. Rowan, M.A. Engle, C.S. Kirby, and T.F. Kraemer

Scientific Investigations Report 2011–5135

U.S. Department of the Interior
U.S. Geological Survey

U.S. Department of the Interior
KEN SALAZAR, Secretary

U.S. Geological Survey
Marcia K. McNutt, Director

U.S. Geological Survey, Reston, Virginia: 2011

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Units and Conversions

pCi/L – picocuries per liter

dpm – disintegrations per minute

Bq – becquerels

1 pCi = 0.037 Bq; 1 Bq = 27.03 pCi

1 pCi = 2.22 dpm; 1 dpm = 0.4505 pCi

Radium Content of Oil- and Gas-Field Produced Waters in the Northern Appalachian Basin (USA): Summary and Discussion of Data

By E.L. Rowan,[1] M.A. Engle,[1] C.S. Kirby,[2] and T.F. Kraemer[1]

Abstract

Radium activity data for waters co-produced with oil and gas in New York and Pennsylvania have been compiled from publicly available sources and are presented together with new data for six wells, including one time series. When available, total dissolved solids (TDS), and gross alpha and gross beta particle activities also were compiled.

Data from the 1990s and earlier are from sandstone and limestone oil/gas reservoirs of Cambrian-Mississippian age; however, the recent data are almost exclusively from the Middle Devonian Marcellus Shale. The Marcellus Shale represents a vast resource of natural gas the size and significance of which have only recently been recognized. Exploitation of the Marcellus involves hydraulic fracturing of the shale to release tightly held gas. Analyses of the water produced with the gas commonly show elevated levels of salinity and radium.

Similarities and differences in radium data from reservoirs of different ages and lithologies are discussed. The range of radium activities for samples from the Marcellus Shale (less than detection to 18,000 picocuries per liter (pCi/L)) overlaps the range for non-Marcellus reservoirs (less than detection to 6,700 pCi/L), and the median values are 2,460 pCi/L and 734 pCi/L, respectively. A positive correlation between the logs of TDS and radium activity can be demonstrated for the entire dataset, and controlling for this TDS dependence, Marcellus shale produced water samples contain statistically more radium than non-Marcellus samples. The radium isotopic ratio, Ra-228/Ra-226, in samples from the Marcellus Shale is generally less than 0.3, distinctly lower than the median values from other reservoirs. This ratio may serve as an indicator of the provenance or reservoir source of radium in samples of uncertain origin.

Introduction

Radium forms naturally from the decay of uranium and thorium, elements that commonly occur in sandstones and shales in sedimentary environments. Radium has been documented in the formation waters in many sedimentary basins (for example, Fisher, 1998). In the northern Appalachian Basin, radium has been measured in the water co-produced with gas and oil (that is, produced water[3]) from reservoirs of Cambrian-Mississippian age. Radioactive isotopes are commonly quantified in terms of "activity concentration" or simply "activity," which in this context refers to a number of disintegrations per unit time. For consistency with the studies cited, activity units of picocuries per liter (pCi/L) are used here to define the activity of radium in produced water samples.

In surface and shallow subsurface environments, radium can be relatively soluble and, therefore, mobile in groundwater over a range of pH and Eh (redox) conditions (Langmuir and Riese, 1985; Sturchio and others, 2001). Radium also may be adsorbed onto clay particles or onto oxide grain coatings (Krishnaswami and others, 1982; Ames and others, 1983; Sturchio and others, 2001). As a radioactive element, radium may represent a potential health hazard if released into the environment. The half-lives of the two principal isotopes of radium, Ra-226 and Ra-228, are 1,600 and 5.75 years, respectively (Akovali, 1996; Artna-Cohen, 1997), and approximately 10 half-lives are required for a radioactive element to decay to negligible quantities. Chemically, radium behaves in a manner similar to calcium and is capable of bioaccumulation in plants and animals. There is a significant body of research aimed at quantification of radium uptake in crops and livestock that make up the human food chain (for example, Tracy and others; 1983; Bettencourt and others, 1988; Linsalata and others,

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[3]The term "produced water" in this report represents water produced from an oil or gas well at any point during its life cycle. The term, therefore, includes waters produced immediately after hydraulic fracturing, with compositions close to those of the injected fluid, as well as waters produced after months or years of production, whose compositions resemble formation water.

1989). Most of these studies were conducted in areas where uranium mining had previously taken place; however, it is not known whether similar investigations have been conducted in regions where oil- and gas-field produced waters are the source of radium. The purpose of this report is to compile and present data from multiple sources to facilitate ongoing research.

Activity data for radium-226 (Ra-226) and radium-228 (Ra-228) in oil- and gas-field produced waters from New York and Pennsylvania have been compiled from publicly available sources and combined with new data for six wells (tables 1 and 2, p. 19–31). Measurements of total dissolved solids (TDS) and of gross alpha and beta activities were also tabulated when available. Unstable (radioactive) isotopes decay by emitting alpha and beta particles; therefore, alpha and beta activities can serve as rough indicators of the presence of radioactive elements.

The publicly available radium data were obtained from the New York State Department of Environmental Conservation (NYSDEC), the Pennsylvania Department of Environmental Protection (PA DEP), and the Pennsylvania Geological Survey. Most of these data are available online, although the most recent Marcellus Shale produced water data were available only from the regional PA DEP offices. Three of the studies, Gilday and others (1999), Pennsylvania Department of Environmental Protection (1992), and Dresel and Rose (2010), provide data from wells producing from reservoirs of Cambrian-Devonian age. In contrast, the analyses reported by the New York State Department of Environmental Conservation (2009) and by the Pennsylvania Department of Environmental Protection (unpub. data, 2009–2010) are for produced waters predominantly from the Devonian Marcellus Shale.

Background

The Appalachian Basin comprises a vast accumulation of sedimentary rock west of the Appalachian Mountains, extending from Quebec and Ontario south through New York, Pennsylvania, Ohio, West Virginia, to Alabama. Hydrocarbons are produced throughout the basin from reservoirs of Cambrian-Pennsylvanian age (Legall and others, 1981; Milici and others, 2003). In recent years, however, the Middle Devonian Marcellus Shale has become the focus of gas exploration and production, particularly in Pennsylvania, New York, and West Virginia.

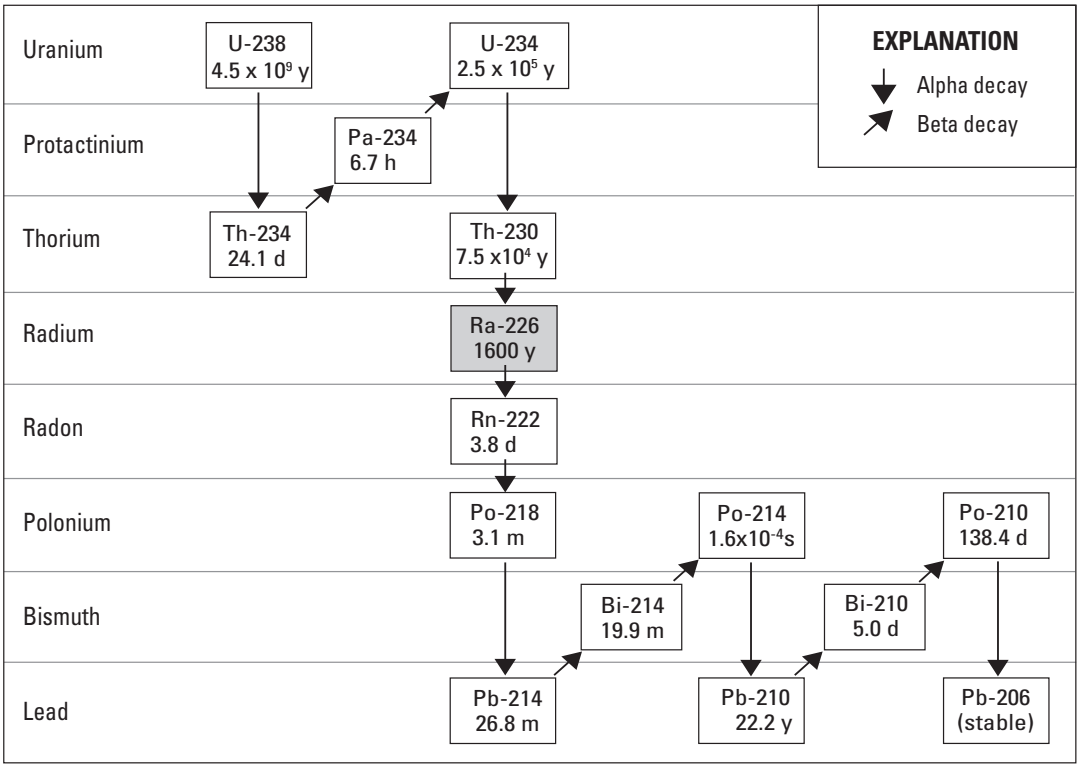
A regional comparison of produced water salinities indicates that Appalachian Basin salinities are high relative to other oil- and gas-producing basins in the United States (Breit, 2002). The compilation yielded a median TDS of about 250,000 milligrams per liter (mg/L) for the Appalachian Basin (USA), which was exceeded only by the median salinity for the Michigan Basin (about 300,000 mg/L). The data presented here indicate a wide salinity range for water produced from the Marcellus Shale, from less than 1,500 mg/L to greater than 300,000 mg/L. The lower salinities may be attributed in part to dilution with less saline fluid injected during hydraulic fracturing, but the upper end of the salinity range is comparable to the waters produced from the underlying Lower Devonian and older reservoirs as well as some of the overlying Devonian reservoirs (Rowan and others, 2010).

The Marcellus Shale is an organic-rich shale that is both the source rock and the reservoir for an extensive natural gas resource (Harper, 2008). Shale-gas accumulations, such as the Marcellus, are termed “unconventional” or “continuous” because the gas is dispersed within a stratigraphic interval rather than confined by a conventional structural or stratigraphic trap. The process of “hydraulic fracturing” commonly is used to access the gas in a continuous reservoir. In this process, water is pumped into a well at pressures high enough to fracture the rock, and the newly created fracture network allows gas that is tightly held in micropores or adsorbed onto clay particles to be released. The injected fluid may be freshwater or relatively dilute, or alternatively, it may have been recycled, that is, produced from one well and then used to hydraulically fracture a new well. The water flowing from hydraulically fractured wells initially reflects the composition of the injected fluid, but with time shifts toward salinities and inorganic chemical compositions similar to the fluids in adjacent formations (for example, Rowan and others, 2010). Hayes (2009), for example, examined the chemistry of produced water samples collected from 12 Marcellus Shale wells at 1-, 5-, 14-, and 90-day intervals following hydraulic fracturing. The water injected into these wells was essentially fresh, with a median TDS of less than 1,000 mg/L, but within 90 days, the salinities had increased to a median value exceeding 200,000 mg/L TDS.

Ra-226 and Ra-228 are the decay products of U-238 and Th-232, respectively (fig. 1; Ivanovich, 1992). Once formed, radium may remain within the original host mineral or other solid phase, or may be released into the adjacent pore water. Lithologies that contain substantial amounts of uranium and (or) thorium can, therefore, have measurable amounts of radium dissolved in their pore waters. The data compiled in this report span most of the oil- and gas-producing regions of the Appalachian Basin in Pennsylvania and New York (fig. 2), and show significant levels of radium in produced water samples from Cambrian-Mississippian reservoirs.

Dissolved radium occurs predominantly as the Ra^{+2} ion, but also forms complexes with chloride, sulfate, and carbonate ions (Rose and Korner, 1979; Kraemer and Reid, 1984; Langmuir and Riese, 1985; Sturchio and others, 2001). Aqueous radium may remain in solution, be adsorbed from pore water onto oxide grain coatings or clay particles by ion exchange, or may substitute for cations, such as Ba^{+2}, Ca^{+2}, and Sr^{+2}, during precipitation of mineral phases, such as barite, anhydrite, and calcite. Radium sulfate ($RaSO_4$) is much less soluble than barite, anhydrite, and other sulfate minerals, but rarely occurs as a separate mineral phase. When alkali earth sulfates precipitate, however, radium present in solution

***A.* Uranium-238**



***B.* Thorium-232**

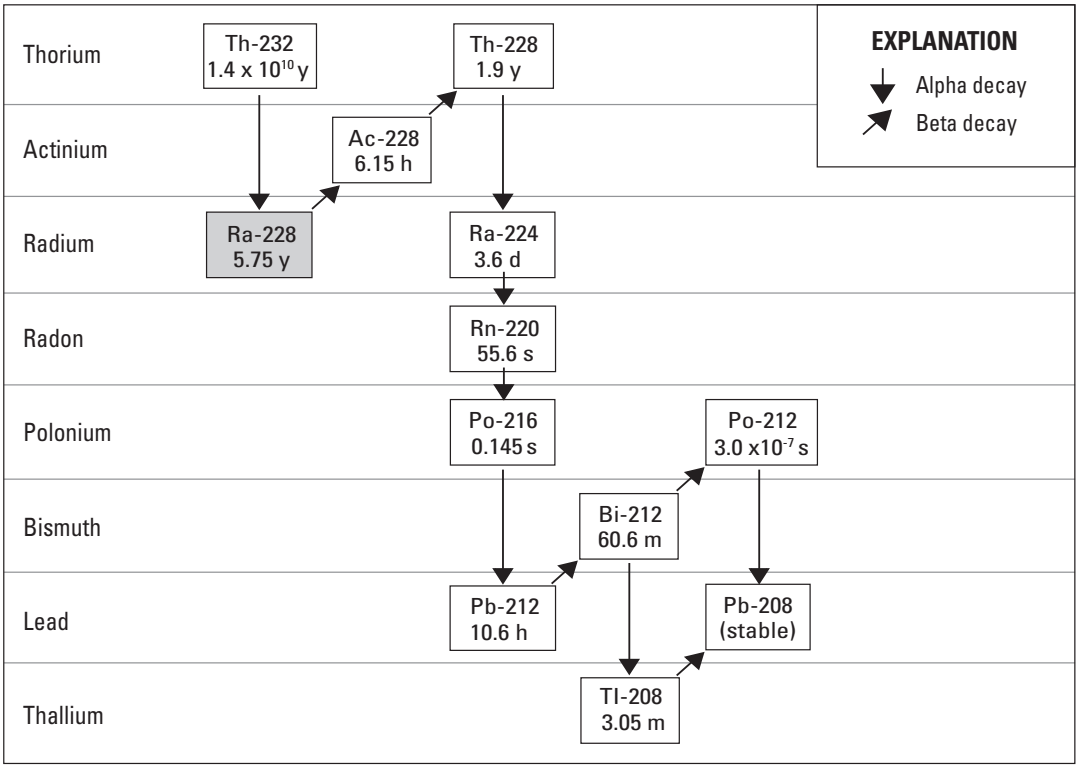


Figure 1. Radioactive decay chains for (*A*) U-238 and (*B*) Th-232. Times shown are half-lives: y, years; d, days; h, hours; m, minutes; s, seconds. Ra-226 and Ra-228 (shaded) are the primary isotopes of interest in this study. Half-lives were obtained from the National Nuclear Data Center (*http://www.nndc.bnl.gov/chart/*).

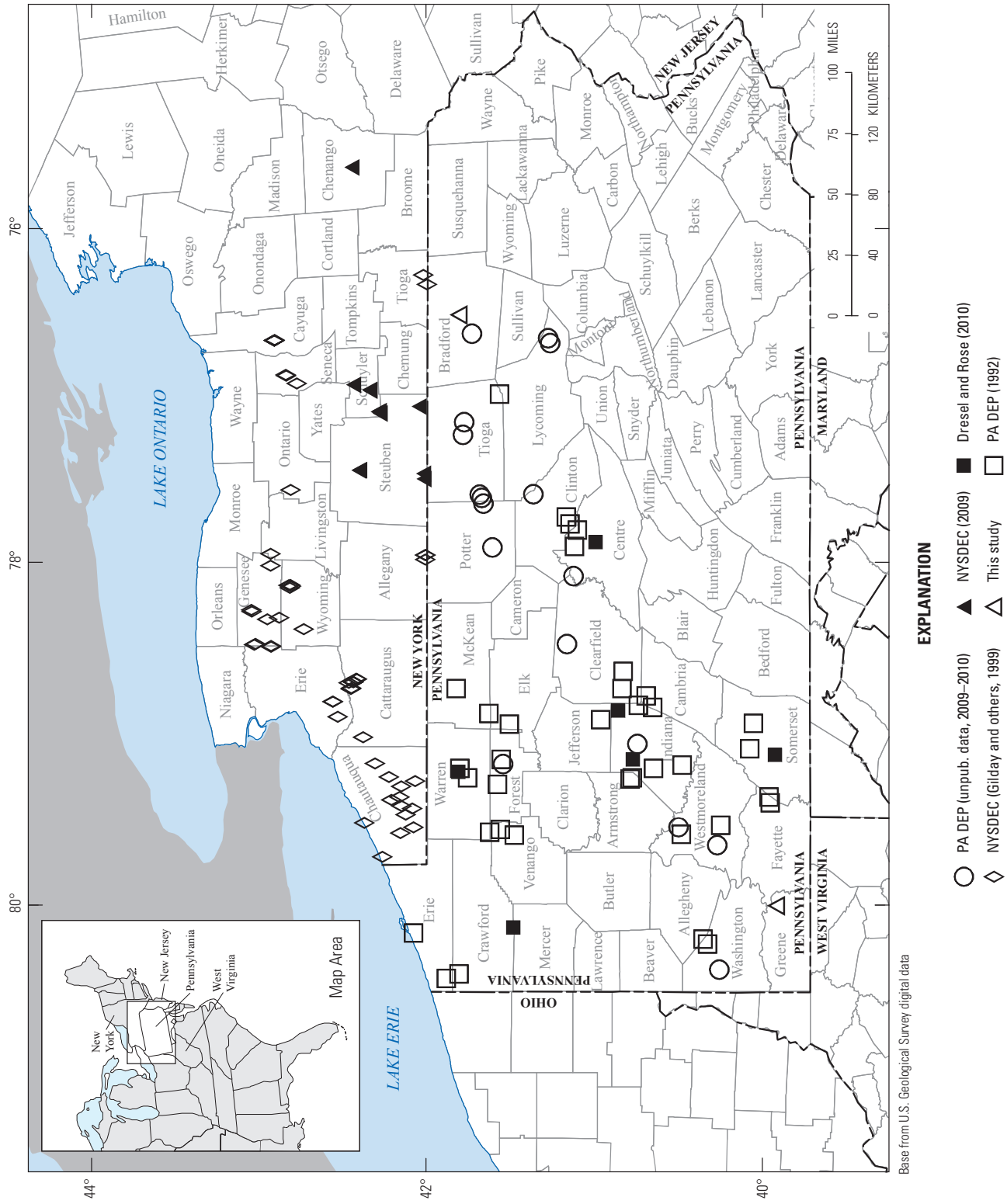


Figure 2. Locations of wells with data compiled for this study.

coprecipitates as a solid-solution, preferentially enriching the solid phase and depleting the solution of radium (Langmuir and Riese, 1985).

Data Sources and Analytical Methods

The sources of data in this report (tables 1 and 2) are discussed below together with the available information on quality assurance/quality control (QA/QC), analytical methods, and uncertainty. The U.S. Environmental Protection Agency (USEPA) method codes refer to standard analytical procedures defined by the USEPA (Krieger and Whittaker, 1980; Eaton and others, 2005).

New York State Department of Environmental Conservation Report (Gilday and others, 1999)

The New York State Department of Environmental Conservation (NYSDEC) conducted a study titled "An Investigation of Naturally Occurring Radioactive Materials (NORM) in Oil and Gas Wells in New York State," in which produced water, oil, sludge, and other waste materials were sampled from oil and gas wells in New York State (Gilday and others, 1999). Analyses were reported for a total of 57 brine samples collected from 48 oil or gas well sites, with 9 duplicate or replicate samples (table 1). The NYSDEC report indicates that the brines were sampled from storage tanks, but the length of time between production and sample collection is unknown. The wells in this study produced hydrocarbons and water from formations of Cambrian through Lower Devonian age, with one sample of possible mixed Lower Silurian and Upper Devonian reservoir origin (table 1). Several of the wells produced from the Lower Devonian Oriskany Sandstone and Helderberg Limestone. Silurian reservoirs provided samples from the Akron Sandstone, Bass Islands Dolomite, Medina Sandstone, and Rochester Shale. Ordovician reservoirs included sandstones within the Queenston Shale.

Analyses of radium activity in the NYSDEC report were determined using gamma-spectrometry as well as alpha-spectrometry in some cases. Gamma-spectrometry compares the gamma-ray wavelengths emitted by radioactive material with the emission spectra of known radioactive elements. In some instances, the signal emitted by a daughter product can be more accurately identified and quantified than that of its parent isotope. Laboratories may therefore elect to report a daughter product activity as representative of its radium parent's activity in an appropriately prepared sample. Gilday and others (1999) considered that the Ra-226 daughter products Pb-214 and Bi-214 were the most reliable indicators of Ra-226 activity, and they selected the larger of the Pb-214 and Bi-214 values to represent the Ra-226 activity. Gilday and others (1999) considered Ac-228 activity to be the most reliable indicator of Ra-228 activity, and Pb-212, which occurs lower on the decay chain (fig. 1), was seldom used.

The values listed in table 2 are consistent with the approach of Gilday and others (1999), but several instances differ from the values highlighted in their report as representative of a given sample. At one well (no. 76), the Pb-212 activity was anomalously high, 23,900 pCi/L, relative to a corresponding Ac-228 activity of 1,500 pCi/L. Gilday and others (1999) concluded that the Pb-212 value was erroneous, although this was the value they highlighted as representative of the sample. At a second well (no. 82), a Pb-212 activity of 7,650 pCi/L also appeared to be anomalously high relative to the Ac-228 activity of 1,110 pCi/L. In both instances, the Ac-228 rather than the Pb-212 activities are used to represent Ra-228 in table 2. Pb-212 activities were used in only five instances where Ac-228 was not reported. In wells where duplicate analyses were available, (nos. 38, 56, 79, and 80), the averages are given in table 2.

All of the samples collected by Gilday and others (1999) were analyzed by an outside contract laboratory, and a subset of nine samples was also analyzed by the NYSDEC Bureau of Pesticide and Radiation laboratory. Some interlaboratory comparison and QA/QC information was provided in that report and is discussed below. Ideally, metrics of both analytical accuracy (proximity of measured value to the "true" value) and precision (measurement reproducibility) are presented. Because no analyses of reference materials or other standards were reported, the analytical accuracy for the included data is unknown. Sample precision was examined by comparing data for analyses of duplicate[4] and replicate[5] samples (fig. 3). Despite the reported "internally consistent results" from each laboratory, the measurement uncertainty ranges did not overlap in five out of nine brine samples analyzed by both laboratories. A single outlier exhibited an exceptionally high difference of 143 percent between replicate analyses for Ra-226.

These findings indicate that sample precision is generally better (less than 20 percent discrepancy between duplicate or replicate samples) for samples that contained greater than 500 pCi/L, but poor agreement in interlaboratory comparisons indicates there may be bias between data sources. The magnitude of the biases, however, appears to be in the tens of percents while radium activities in brine samples range over more than four orders of magnitude. This comparison suggests that even the higher end of analytical imprecision observed in the data does not significantly affect the magnitude of radium activities reported.

[4]Duplicate refers to individual samples from a single source collected at the same place and time.

[5]Replicate refers to a repeat analysis made on the same sample or aliquots of the same sample.

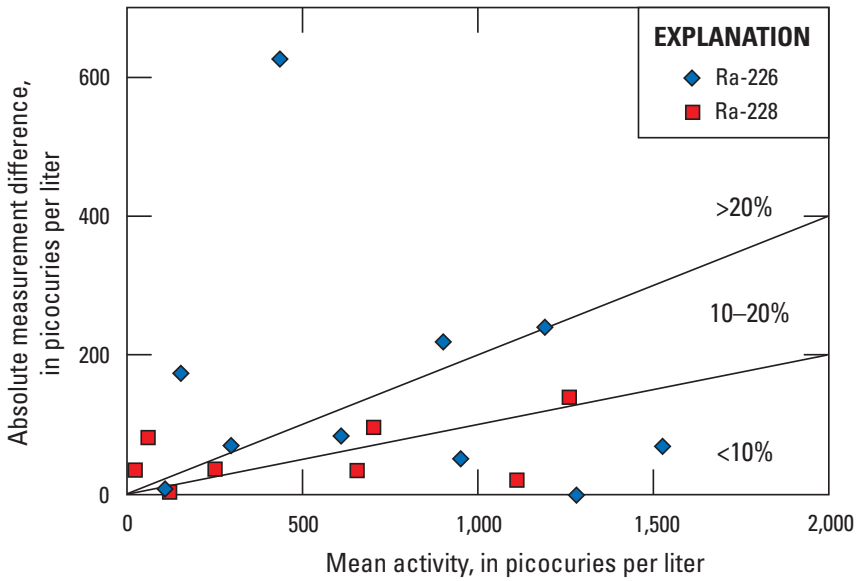


Figure 3. Differences between measurements of duplicate and replicate analyses of Ra-226 and Ra-228 in produced water samples in relation to the mean activity of the sample for data from Gilday and others (1999). The solid lines represent 10 percent and 20 percent relative difference between duplicates/replicates using the method of Thompson and Howarth (1978). Samples with higher radium activities generally have better measurement precision, that is, lower percentage differences.

New York State Department of Environmental Conservation, Draft Supplemental Generic Environmental Impact Statement (2009)

In 2009, the NYSDEC released a study titled "Draft Supplemental Generic Environmental Impact Statement related to Marcellus Shale Gas Development" (New York State Department of Environmental Conservation, 2009). Appendix 13 of the document, "NYS Marcellus Radiological Data from Production Brine," lists gross alpha, gross beta, and activities of Ra-226 and Ra-228 for water samples collected from 12 gas-producing Marcellus Shale wells in New York State. Appendix data were presented in table form without accompanying text, information relating to QA/QC, or analytical methods. However, well lease names and API numbers, towns, and counties were provided, allowing well locations and related information to be obtained from the State database (*http://www.dec.ny.gov/*; fig. 2; tables 1 and 2). Activities of uranium, thorium, and the anthropogenic isotopes, cesium-137, cobalt-60, ruthenium-106, and zirconium-95, were listed in the appendix, but are not compiled in this report.

Pennsylvania Department of Environmental Protection Report (1992)

In 1991, the PA DEP conducted field work for a study of salinity and radium activities in produced waters, sludge, and other related waste from oil and gas wells in Pennsylvania. The results were compiled in a report titled "NORM Survey Summary" and released the following year (Pennsylvania Department of Environmental Protection, 1992). The wells sampled for the study produced hydrocarbons and water from Lower Silurian–Upper Devonian Formations, with one sample thought to be from an Ordovician reservoir. Although the Marcellus Shale falls within this stratigraphic interval, the study long pre-dated the recent (2005–present) focus on the Marcellus Shale as an unconventional gas resource. Among the most commonly sampled reservoirs were sandstone in the Silurian Medina Group, the Lower Devonian Oriskany Sandstone, Huntersville Chert, and Onondaga Limestone, as well as Upper Devonian sandstones (table 1). About three-fourths of the samples were taken from storage tanks, or separator tanks, and the remaining samples were collected from surface pits or diked areas (table 1). The length of time between hydrocarbon production and sample collection is unknown, and therefore, Ra-228 activity may have been markedly reduced by natural decay. Brines that accumulated in open pits presumably would have been subject to evaporation and (or) dilution by rain.

In addition to brine samples, samples of sludge, drill cuttings, and pipe scale from brine treatment facilities, pipe yards, disposal wells, and other facilities were analyzed, but these results were not compiled in this report. No information on the laboratory, analytical methods, uncertainties, or QA/QC was included with the PA DEP (1992) report.

Pennsylvania Department of Environmental Protection Reports (Unpublished Data, 2009–2010)

A number of the annually filed "Form 26R" (Chemical Analysis of Residual Waste, Annual Report by Generator) waste reports related to shale gas production were obtained from the PA DEP. The forms and accompanying chemical analyses are filed annually with the PA DEP by generators of liquid or solid waste, including oil and gas well operators. The 26R forms can be viewed at the DEP regional offices by appointment, or photocopies can be requested from the DEP. The DEP offices in Williamsport and Pittsburgh were visited during the spring and summer of 2010, and the available 26R forms pertaining to liquid waste generated at gas well sites were electronically scanned. Additional data were obtained by correspondence with the Meadville, Pa., office. Radium activities from the 26R forms were included in this report only when the well name and related information could be obtained for a given sample. Information obtained from 26R forms filed with the PA DEP during 2009–2010 for a total of 23 wells was compiled and included in tables 1 and 2. In most instances, the TDS values of the samples were also available. Time series data were available for four wells (table 2). When duplicate analyses were provided, the average value is shown in table 2.

Laboratory notes accompanying 26R forms reported to the PA DEP varied substantially between individual wells, but all included the laboratory name and, in some cases, the analytical method and QA/QC information. Despite the numerous different reporting entities, the radiochemical data reported in the 26R forms were obtained from only four different laboratories, and all are accredited in accordance with the National Environmental Laboratory Accreditation Program (NELAP).

Gross alpha and beta emission measurements included in the PA DEP 26R forms were determined by methods that include standard and modified versions of EPA Method 900.0 (Gross Alpha and Gross Beta Radioactivity in Drinking Water) and Standard Method 7110C (Eaton and others, 2005). No duplicate samples, replicate analyses, or other QA/QC information were available for either the gross alpha or beta results.

When methods for radium analysis were reported, Ra-226 activity typically was measured using gamma-spectrometry, and in some cases by alpha-spectrometry, using standard USEPA methods: EPA Method 901.1 (Gamma Emitting Radionuclides in Drinking Water), EPA Method 903.0 (Alpha-Emitting Radium Isotopes in Drinking Water), and EPA Method 903.1 (Radium-226 in Drinking Water Radon Emanation Technique). Radium-228 was analyzed using similar methods: EPA Method 901.1 (Gamma Emitting Radionuclides in Drinking Water) and EPA Method 904.0 (Radium-228 in Drinking Water). For the four sets of duplicate Ra-226 and Ra-228 analyses, the discrepancies were less than 7 percent, with one exception: Ra-226 analyses in duplicate samples from well no. 1 differed by 72 percent.

Dresel and Rose (2010)

A recent publication by Dresel and Rose (2010) reports the produced water analyses originally conducted as part of a Master's thesis at Pennsylvania State University (Dresel, 1985). Of the 40 samples collected, Ra-226 analyses are reported for six wells producing hydrocarbons and water from Lower Silurian–Upper Devonian sandstone reservoirs; Ra-228 values are not reported. Most of the samples in this study were collected from the wellhead rather than secondary storage units (table 1). The Ra-226 activities reported were determined by measurement of radon-222 activity at secular equilibrium (Rose and Korner, 1979), using a method equivalent to USEPA Method 903.1 (Krieger and Whittaker, 1980). Detailed QA/QC information was not available.

This Study

Radium activities have been determined at the U.S. Geological Survey (USGS) for samples from six additional Marcellus Shale gas wells in Pennsylvania. Samples were collected from five of the wells (nos. 127–131, tables 1 and 2) as part of a study by Pritz (2010). The precise localities of these wells in Bradford County are confidential, and they are represented in figure 2 by a single point. Well no. 132 was sampled jointly by the USGS, the Department of Energy, and industry collaborators on successive dates, thus providing time series information. Analyses of the samples were conducted at the USGS radiochemistry laboratory in Reston, Virginia. Two to four duplicates of each sample from well no. 132 were prepared and analyzed, and the average values are reported in table 2.

In the samples from well no. 132, radium was chemically separated from the water by coprecipitating it with barium sulfate. The precipitate was then placed in the well of a high purity germanium detector, and quantitative analysis of the Ra-226 and Ra-228 content of the precipitate was performed by gamma-spectrometry using a technique adapted from Moore (1984). As discussed above for the New York State data of Gilday and others (1999), Ra-228 was quantified by measuring the intensity of gamma rays emitted by Ac-228, and Ra-226 was quantified by measuring the intensity of the gamma rays emitted by Pb-214 and Bi-214. As described in Kraemer (2005), the gamma-ray spectrometry systems were calibrated using standardized radium isotopic solutions.

Uncertainties for these analyses are listed in table 2 as +/– one standard deviation from the mean peak intensity and represent the "counting error" for a specific analysis. When duplicate samples were prepared, that is, reprecipitated, and analyzed, the range of the discrepancies matched closely with the range for the counting error: 0.2–8.5 percent. However, the discrepancies between analyses of duplicate samples were most commonly 2–4 percent higher than the counting error. In all cases, the maximum error did not exceed +/– 8.5 percent.

Results

Salinity and Radium

Salinities, reported as TDS, were available for approximately one-half of the produced water samples and ranged from 1,470 to 402,000 mg/L with a median of 157,000 mg/L TDS (table 2). The median total radium (defined here as Ra-226 + Ra-228) activity for the non-Marcellus Shale produced water samples is 1,011 pCi/L compared with 2,460 for Marcellus Shale produced water samples and 5,490 pCi/L for the Marcellus produced water data for New York State (fig. 4; New York State Department of Environmental Conservation, 2009; Pennsylvania Department of Environmental Protection, unpub. data, 2009–2010; this study). For comparison, the total radium limit for industrial effluent is 60 pCi/L, and the drinking water limit is 5 pCi/L (U.S. Environmental Protection Agency, 1976; Hess and others, 1985; U.S. Nuclear Regulatory Commission, 2011).

In the NYSDEC (2009) study, salinities were not reported; however, two wells, no. 28 and no. 33, were resampled and analyzed by Osborn and McIntosh (2010), yielding respective salinities of 206,446 and 205,102 mg/L TDS. Samples at two additional wells, no. 24 and no. 25, both from depths of approximately 2,600 feet (ft), exhibited very low total radium activities (less than 1 pCi/L), although the activities of the remaining sites exceeded 1,900 pCi/L (fig. 4; table 2). The reason for the low radium content of these samples is unknown, but they may have been composed largely of water injected for hydraulic fracturing, which often is of lower salinity and radium content than the formation water.

In Pennsylvania, the range of total radium activities for the Marcellus Shale samples (Pennsylvania Department

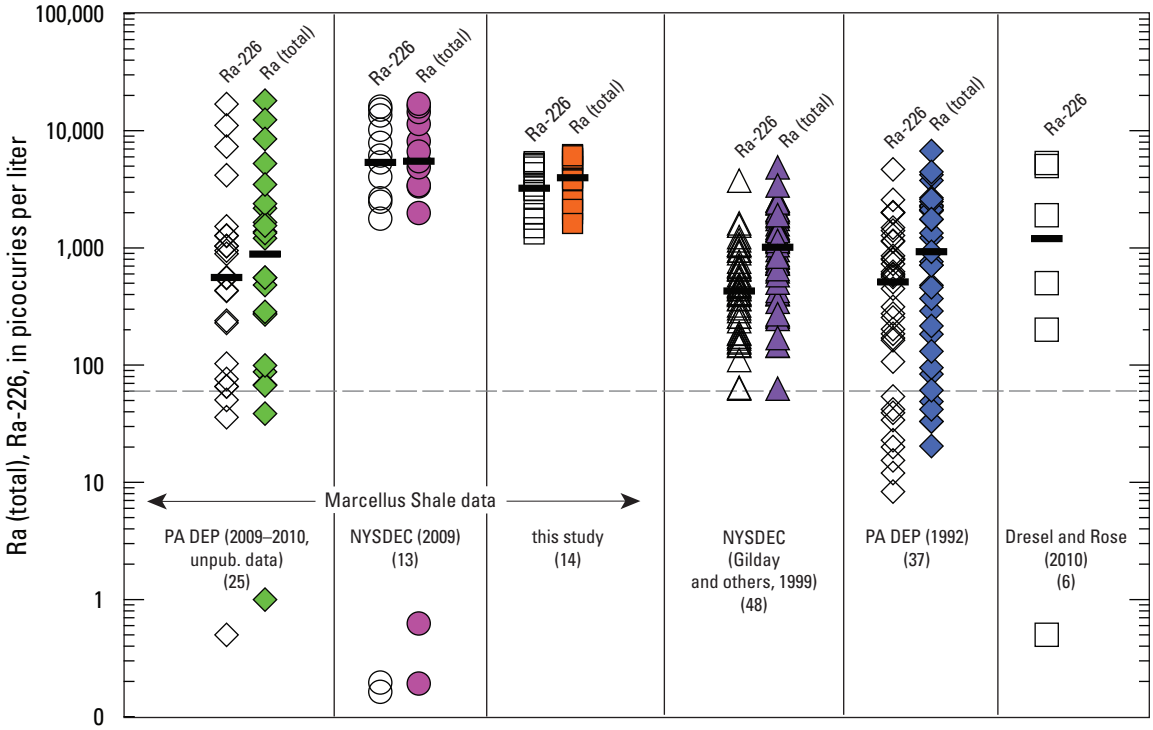


Figure 4. Measured activities for total radium (Ra-226 + Ra-228) and Ra-226 for each of the data sources used in the study. The three datasets for produced water from Marcellus Shale wells are shown on the left; the remaining three datasets are for non-Marcellus Shale wells. The number of points in each dataset is shown in parentheses, and the median values are plotted as heavy black lines. For reference, the dashed line shows the industrial effluent discharge limit (60 pCi/L) for Ra-226 (U.S. Nuclear Regulatory Commission, *http://www.nrc.gov/reading-rm/doc-collections/cfr/part020/appb/Radium-226.html*).

of Environmental Protection, unpub. data, 2009–2010) is similar to the Marcellus data from New York but is more evenly distributed (less clustered) over the range. Dilution of formation water with the relatively freshwater from the hydraulic fracturing process may have been an important factor influencing the distribution of both salinity and radium content. The time interval between hydraulic fracturing and sample collection is known in only a few cases.

Gross Alpha and Beta Particle Emissions

Emission of alpha and beta particles accompanies the decay of Ra-226 and Ra-228, respectively (fig. 1), and the USEPA has established the measurement of gross alpha and beta as a method of screening samples for the presence of radium (Hess and others, 1985; Buckwalter and Moore, 2007, p. 48). Gross alpha and beta data were available for two datasets (New York State Department of Environmental Conservation, 2009; Pennsylvania Department of Environmental Protection, unpub. data, 2009–2010) and are plotted with Ra-226 and Ra-228, respectively (figs. 5*A*–*B*). On log-log scales, gross alpha and gross beta activities are linearly correlated with Ra-226 and Ra-228, confirming their value as indicators of radium activity. Although these isotopes are unlikely to be the only sources of alpha and beta particles, the correlations shown in figures 5*A*–*B* suggest that they are likely to be the dominant sources for these samples.

Discussion

Salinity and Dilution

Several studies of Appalachian Basin formation water chemistry have shown general trends of increasing salinity with depth and age of the reservoir (for example, Stout and others, 1932; Poth, 1962; Breen and others, 1985); however, high salinities can occur even at relatively shallow depths. A salinity-depth curve for Mississippian-Devonian formation waters in eastern Ohio showed greater than 100,000 mg/L TDS at 1,000 ft (Stout and others, 1932, p. 18). Poth (1962, p. 37–38, table 6) noted that on the basis of a limited set of samples, an equilibrium salinity had apparently been reached in Middle Devonian and older reservoirs, and water produced from these units have a dissolved solids content of about 300,000 mg/L. In the dataset compiled here, produced water salinities from the Middle Devonian Marcellus Shale commonly range from 100,000 to 200,000 mg/L TDS (table 2).

Despite the presence of evaporites in the Silurian Salina Group in parts of New York and Pennsylvania, many authors consider that the salinity of the produced waters in much of the Appalachian Basin originated from the evaporative concentration of seawater (for example, Stout and others, 1932; Sanders, 1991; Dresel and Rose, 2010; Osborn and McIntosh, 2010). Brines derived from seawater evaporation are enriched in bromide and can be distinguished from brines formed by dissolution of evaporites on the basis of relations among Na, Cl, and Br (Walter and others, 1990). Brines produced with gas from Marcellus Shale wells after salinities have reached a plateau share similar major ion chemistries with formation waters from the overlying and underlying Devonian formations and show similar Na-Cl-Br relations (Osborn and McIntosh, 2010; Rowan and others, 2010). On the basis of these chemical similarities, a similar origin for the salinity of waters produced from the Marcellus Shale and from adjacent overlying and underlying formations can be hypothesized.

Blauch and others (2009), however, reported small lenses of halite and other salts in core from the Marcellus Shale and suggested that dissolution of these minerals contributed to the salinity of the produced waters. They also described minor volumes of salts, but noted that similar occurrences have not previously been reported in the literature on the Marcellus. Where present, salt lenses would contribute to total salinity, but it is difficult to assess their distribution or quantify their contribution to total fluid salinity. The elevated bromide concentrations and Na-Cl-Br relations suggest that the dominant source of salinity for Marcellus Shale waters, and for other formations in the stratigraphic section, originated as evaporatively concentrated seawater.

Dilution of formation water with relatively freshwater injected during the hydraulic fracturing may account for some of the lower salinity values. For example, in well no. 11 salinities were measured 14 and 90 days after hydraulic fracturing and showed an increase with time (fig. 6*A*; table 2). In well no. 5, successive salinity measurements made 17 days apart also showed increased salinity with time (table 2). In a more detailed study by Hayes (2009), repeated measurements of produced water salinity up to 90 days after hydraulic fracturing showed increases in salinity with time from less than 1,000 mg/L to greater than 100,000 mg/L TDS. The marked increase in salinity with time is interpreted to represent a decreasing proportion of the lower salinity injected fluid and an increasing proportion of the saline formation water returning to the surface. As mentioned previously, dissolution of mineral phases such as halite, if present, could also contribute salinity. For data compiled from the PA DEP 26R forms, when the sample collection date occurred less than 90 days from the initiation date of drilling, it seems plausible that salinities less than 100,000 ppm TDS may have been affected by dilution of the formation water with the water injected during hydraulic fracturing.

Like salinity, radium in the produced waters increases with time following hydraulic fracturing. A well producing gas from the Marcellus Shale in Greene County, Pa. (no. 132), was sampled daily for the first 5 days, then on days 7, 15, and 20 following hydraulic fracturing. The radium data for these samples are discussed below, but the salinity data are not yet available. The hydraulic-fracturing supply water was a mix of water recycled from similar gas wells nearby and more dilute surface water. Following hydraulic fracturing, the total radium activity in the produced water increased sharply

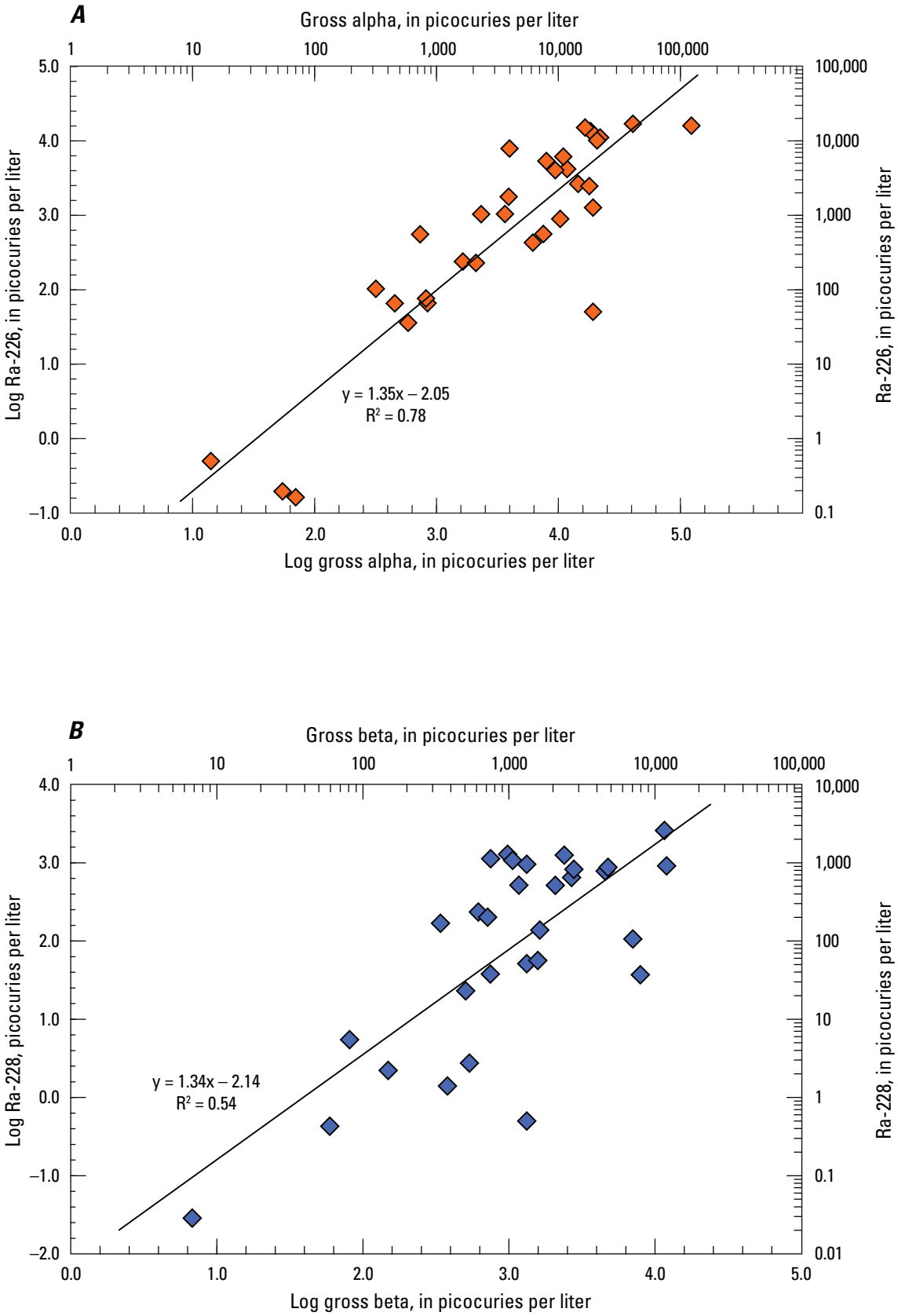


Figure 5. (*A*) Log of gross alpha particle activity in relation to the log of Ra-226 activity and (*B*) log of gross beta particle activity in relation to the log of Ra-228 activity. Gross beta activities below the reported detection limit in well nos. 17 and 33 are not plotted or used in the best fit line.

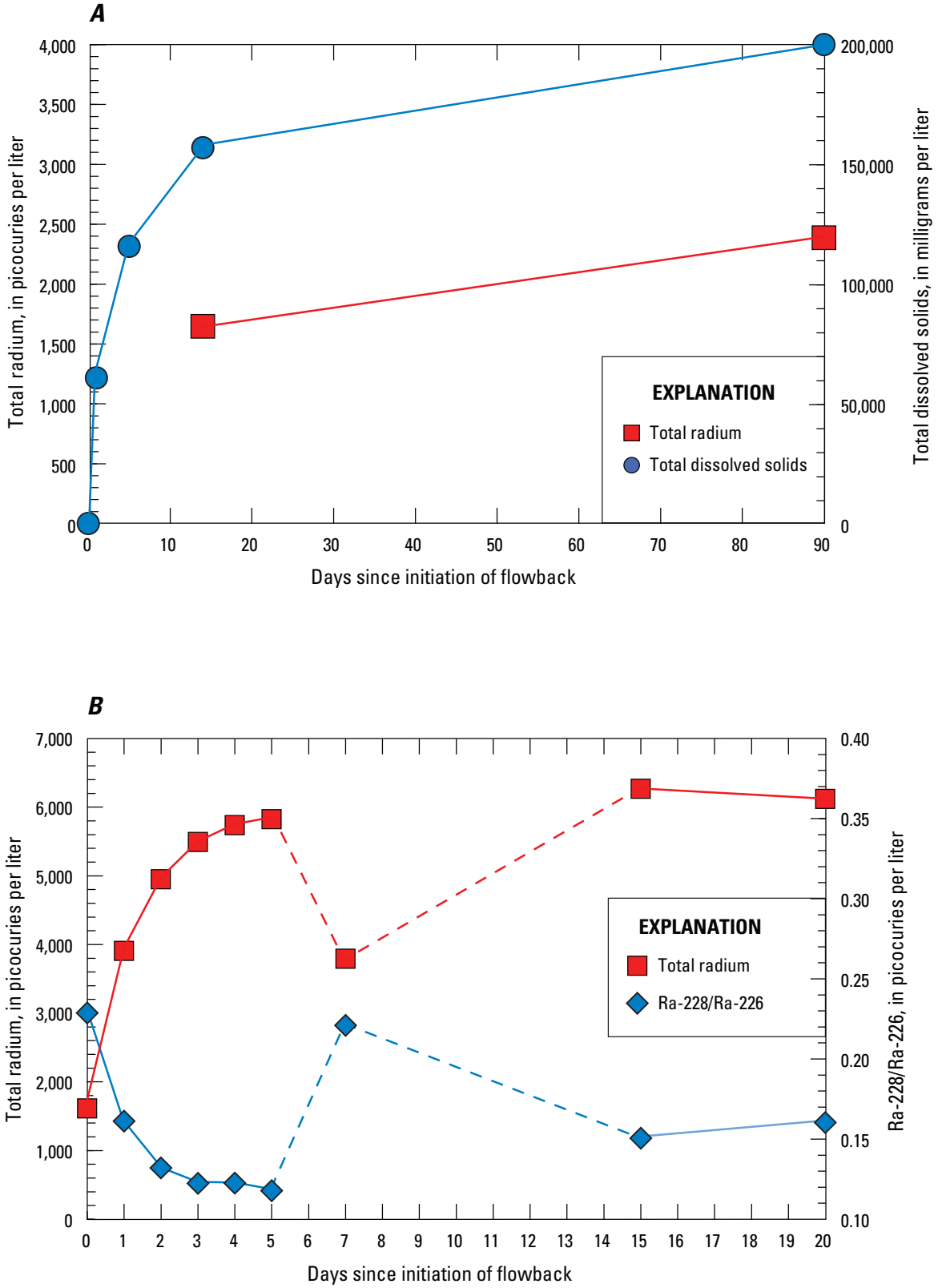


Figure 6. (*A*) Total radium activity and total dissolved solids related to time since initiation of flowback for well no. 11, Washington County, Pa. (*B*) Total radium activity (left axis, squares) and Ra-228/Ra-226 (right axis, diamonds) related to time since initiation of flowback for well no. 132, Greene County, Pa.

during the first week from the activity of the supply water (about 1,600 pCi/L) to a plateau at about 6,100 pCi/L (fig. 6*B*). The increase in radium activity is interpreted as the result of equilibration between the injected water, whose radium activity is relatively low, and the radium that is present in the reservoir, either adsorbed onto mineral surfaces or dissolved in pore water. An anomalously low value on day 7 remains as yet unexplained; close agreement between multiple repeat analyses of the original sample conducted on different dates rules out an instrumental or analytical error as an explanation.

The Ra-228/Ra-226 ratio for this fluid decreased from the initial value of 0.23 in the injected water to 0.12. Following an unexplained increase on day 7, the ratio returned to about 0.16. Low isotopic ratios reflect the low Th/U ratio that generally characterizes the Marcellus Shale. The evolution of total radium and Ra-228/Ra-226 with time displays a fairly consistent pattern, with the exception of day 7. The area being drained by the well on this day may have intersected a "pocket" of chemically distinct water, possibly a sandy horizon in the shale or a fracture intersecting a distinct facies.

Radium Activities in Context

In a study of NORM (naturally occurring radioactive material) in oil- and gas-producing regions, Fisher (1998) compiled radium activity data for nine sedimentary basins in the United States and Europe. In separate studies, Ra-226 activities were reported for formation water samples from clastic aquifers in the U.S. Gulf Coast (Kraemer and Reid, 1984) and from carbonate aquifers in the U.S. midcontinent (Sturchio and others, 2001). Radium activity ranges for these regions generally are comparable to those compiled here for the Appalachian Basin. The highest reported values found in literature are from the Donieper-Donets Basin, Ukraine (Gutsalo, 1964, cited in Kraemer and Reid, 1984) and from the Texas Panhandle (see references cited in Fisher, 1998); the high end of the Ra-226 activities ranges exceeded 4,500 pCi/L in the Ukraine and 5,000 pCi/L in the Texas Panhandle. In this report, several Ra-226 activities of approximately 4,000 pCi/L have been compiled for samples from non-Marcellus reservoirs, but the Marcellus Shale data range higher, with several activities exceeding 10,000 pCi/L (table 2; fig. 7).

Relationships between salinity and radium activity have been documented in a number of studies (for example, Kraemer and Reid, 1984; Fisher, 1998; Sturchio and others, 2001). Fisher (1998), however, pointed out that chloride or TDS concentrations "best predict radium activity in waters from reservoirs that are lithologically relatively homogeneous." Despite their origin in reservoirs of varying ages and lithologies, the non-Marcellus Shale data indicate a positive correlation between salinity and both total radium and Ra-226 activities. Salinity (TDS) is plotted with total radium and Ra-226 in figures 7*A*–*B*. Use of the longest-lived isotope, Ra-226, may reduce some of the scatter induced in the data by sample collection at varying, or unknown, time intervals since brine production at the wellhead. A year after production at the wellhead, for example, the activities of the shorter-lived Ra-228 isotope would be reduced by approximately 11 percent because of natural decay.

Analysis of covariance (ANCOVA[6]) was used to statistically examine the effect of Marcellus versus non-Marcellus sample origin on the linear relationship between TDS and radium activity. The resulting linear regression models yield nearly identical slopes in the trends of log total radium in relation to log TDS and log Ra-226 in relation to log TDS. However, the regression intercepts for the Marcellus Shale data are 0.4 and 0.55 log units (2.5 and 3.5 times, respectively) higher for total radium and Ra-226, respectively, than for the non-Marcellus samples. Produced water samples from the Marcellus Shale are, therefore, enriched in radium to a statistically significant degree ($p<0.05$) relative to samples from other formations in the basin. This relative enrichment is also illustrated in figure 8*A*.

An important mechanism by which salinity controls radium activity involves competition between Ra^{+2} and other multivalent ions for adsorption sites primarily on clay minerals (Kraemer and Reid, 1984). In low salinity fluids, radium tends to be adsorbed onto mineral surfaces and with increasing salinity radium is progressively desorbed and released into solution. Sturchio and others (2001) presented a salinity-dependent distribution coefficient (K) between dissolved radium and radium adsorbed onto clay particles and oxide grain coatings. The logs of K and TDS show a linear relationship with negative slope, indicating less adsorption of radium at higher salinities and, therefore, more radium in solution (Sturchio and others, 2001, fig. 7).

In a study of saline groundwater systems in the mid-continent with TDS concentrations reaching 250,000 mg/L, Sturchio and others (2001, fig. 5) used equilibrium speciation calculations to demonstrate that Ra^{+2} was the predominant dissolved radium species, independent of salinity. In their analysis, Ra^{+2} never accounted for less than 77 percent of the total dissolved radium over a range of fluid chemistries. The next most abundant species, $RaCl^{+}$, gained significance with increasing salinity and Cl^{-}/SO_4^{-2} ratios. Additionally, Sturchio and others (2001) reported that radium forms strong organic complexes at elevated salinities, which may be significant because limited, unpublished data for organic compounds in Appalachian Basin produced waters have shown significant concentrations of acetate and other anions of carboxylic acids.

Dissolved radium measured in produced water samples originates from the decay of the parent isotopes, U-238 and Th-232, in uranium- and thorium-bearing minerals or organic material contained in the host formation. The physical transfer of radium from the solid to the aqueous phase is discussed and illustrated in Fisher (1998, fig. 2) and Sturchio and others (2001). Uranium and thorium, unlike radium, are poorly soluble in the oxygen-poor, reducing conditions that

[6]See Crawley (2007), for example, for additional discussion of the ANCOVA and related statistical methods.

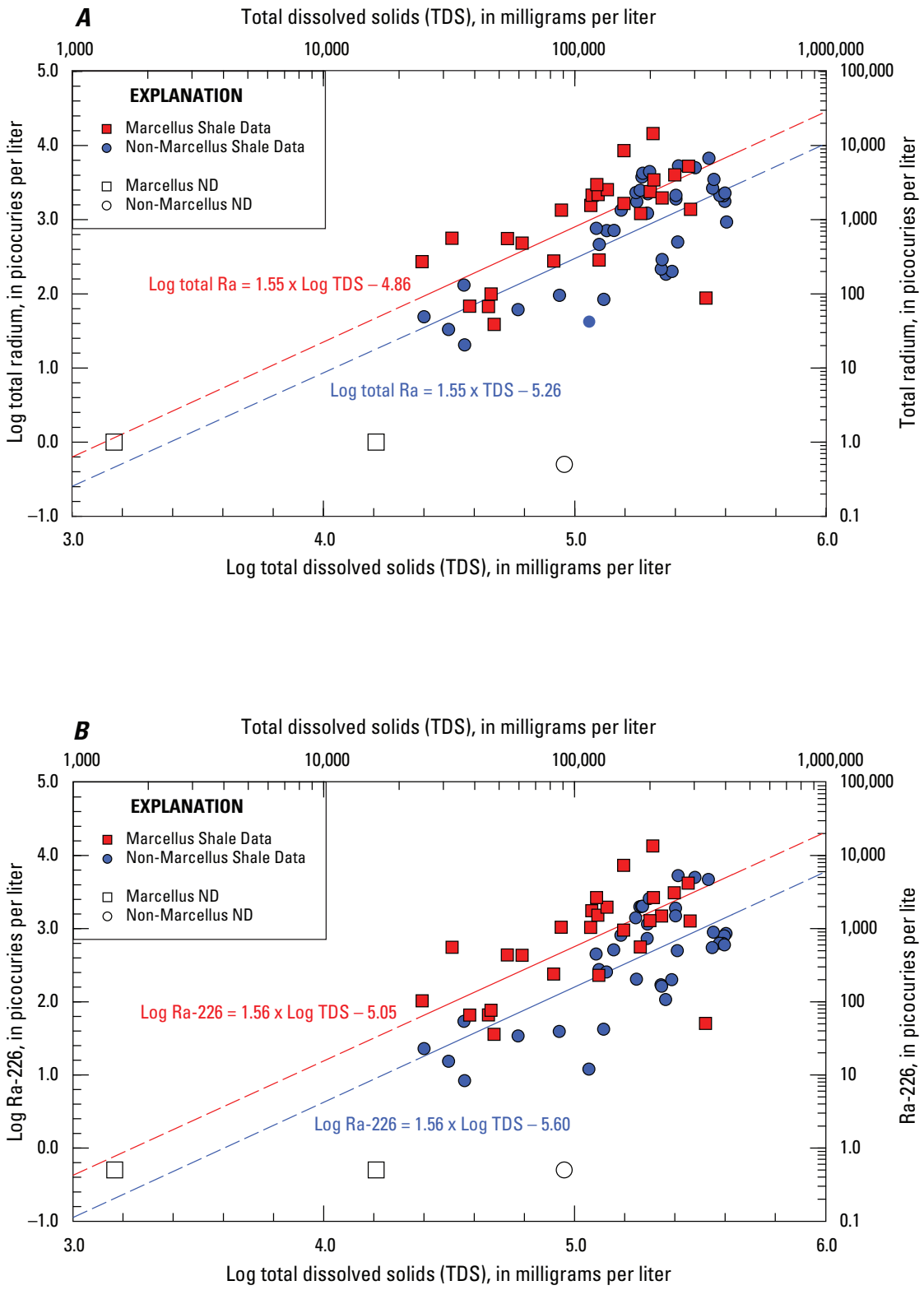


Figure 7. (*A*) Log activity of total radium (Ra-226 + Ra-228) in relation to log total dissolved solids and (*B*) log activity of Ra-226 in relation to log total dissolved solids. Linear regression lines are shown for data from the Marcellus Shale (red), and for non-Marcellus Shale (blue) data; the lines are solid over the range of the data and dashed where extrapolated. Radium analyses listed as "ND" or not detected (well nos. 2 and 14) indicate values below the reported level of quantification (LOQ), 1 pCi/L. These points were replaced by one-half of the LOQ, or 0.5 pCi/L, and are plotted as open squares, but are not included in the regression. A reported Ra-226 activity of 0 (well no. 123) was replaced by 0.5 pCi, and is plotted as an open circle, but was not included in the regression.

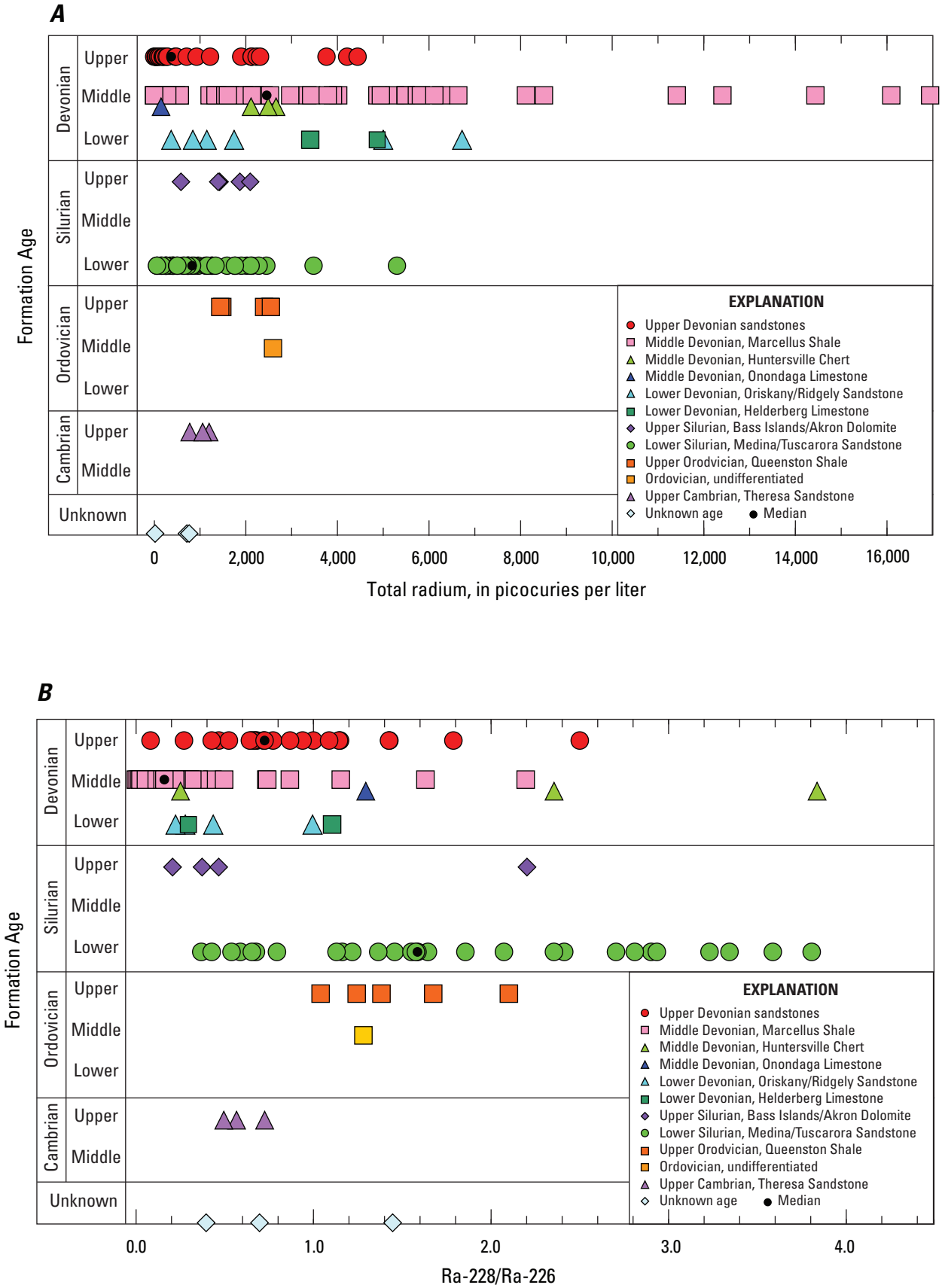


Figure 8. (*A*) Total radium and (*B*) Ra-228/Ra-226 plotted against the age of the producing formation. For formations with large numbers of analyses, the median is shown as a solid black dot.

are typical of oil- and gas-producing horizons and are likely to be more concentrated in mineral phases or organic matter than in solution (Langmuir and Herman, 1980; Kraemer and Reed, 1984; Fisher, 1998; Sturchio and others, 2001). Thus, dissolved radium that is in secular equilibrium with its parent isotopes of uranium and thorium at depth in a reservoir may be isolated from its parents when pumped to the surface.

In addition to displaying higher radium activities for a given salinity, produced water samples from the Marcellus Shale have distinctly lower Ra-228/Ra-226 ratios (median of 0.16) than those of non-Marcellus samples (median of 1.1; figs. 8*A*–*B*), reflecting the Th/U ratio of the reservoir lithologies. Organic carbon has long been known to play a role in concentrating uranium (Swanson, 1960, 1961; Szalay, 1964), and recent work by Bank and others (2010) has documented a close spatial association between the organic matter and uranium in the Marcellus Shale. As an organic-rich black shale, the Marcellus is readily identified on geophysical logs by its high gamma-ray signal (Schmoker, 1981; Harper, 2008).

In eastern Pennsylvania, numerous minor occurrences of uranium have been reported in upper Paleozoic sandstones surveyed at roadside outcrops (Klemic, 1962). If the uranium enrichment is assumed to extend into the subsurface, it provides a potential source for the radium reported in non-Marcellus Shale produced waters, and as discussed above, high formation water salinity can account for elevated radium activities in solution.

The Ra-228/Ra-226 ratios span a wide range among the non-Marcellus produced waters, and the highest values occur in samples from the Lower Silurian Medina Group/Tuscarora Sandstone. The median ratio for Medina/Tuscarora produced water samples is 1.61 (fig. 8*B*). Interestingly, this ratio is consistent with the data compiled by Vengosh and others (2009, fig. 4), which suggest an average isotopic ratio of approximately 1.6 for sandstones worldwide. The sandstones of the Upper Devonian Bradford Group have lower isotopic ratios (median, 0.78), possibly because of the interfingering of sandstone with siltstone and shale beds in this interval.

Summary

Produced water salinities from reservoirs in rocks of Cambrian-Devonion age in the Appalachian Basin commonly exceed 100,000 mg/L, and far exceed the salinities of many other oil- and gas-producing regions in the United States, including basins in California, the Great Plains, and Colorado Plateau. In many basins, radium activity is correlated with salinity, and particularly among samples from lithologically homogeneous reservoirs, salinity may be used as an indicator of radium activity. The data compiled for Pennsylvania indicate a relationship similar to that described in other basins; total radium and Ra-226 activities are linearly correlated with TDS. Salinity was not reported in the datasets for New York.

The radium activities in non-Marcellus produced waters in this report are broadly comparable to those reported in other studies of deep sedimentary basins with highly saline formation water. In the produced water dataset for the Marcellus Shale in New York, total radium activities have a distinctly higher median (5,490 pCi/L) than reported for other formations in the Appalachian Basin, and range to higher values than reported in other basins. Produced waters from the Marcellus in Pennsylvania have similar ranges to the New York data, but a lower median value (1,727 pCi/L), interpreted as being due, at least in part, to dilution of formation water by formation water injected for hydraulic fracturing.

In the data compiled here, Ra-228/Ra-226 ratios in produced water from the Marcellus Shale are most commonly less than 0.3, and samples from non-Marcellus reservoirs generally have Ra-228/Ra-226 ratios greater than 1. Elevated total radium activities combined with low Ra-228/Ra-226 ratios characterize produced waters from the Marcellus Shale, and these characteristics might be used to constrain the origin of samples of unknown provenance.

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Well / Sample ID	Sample collection date	State	County	Township	Longitude	Latitude	Description of sample site	Producing formation	Producing formation age	Well type
Source: PA DEP (2009–2010)										
1	11/18/2009	PA	Clinton	Chapman	–77.56	41.37	Storage tank	Marcellus Sh.	Devonian, M.	Gas
2	11/20/2009	PA	Clinton	Beech Creek	–77.68	41.20	Storage tank	Marcellus Sh.	Devonian, M.	Gas
3	6/1/2009	PA	Bradford	Burlington	–76.60	41.74		Marcellus Sh.	Devonian, M.	Gas
4	8/24/2009	PA	Lycoming	Penn	–76.63	41.28		Marcellus Sh.	Devonian, M.	Gas
5.1	3/18/2009	PA	Lycoming	Penn	–76.66	41.27		Marcellus Sh.	Devonian, M.	Gas
5.2	3/30/2009	PA	Lycoming	Penn	–76.66	41.27		Marcellus Sh.	Devonian, M.	Gas
6	12/21/2009	PA	Tioga	Charleston	–77.21	41.79		Marcellus Sh.	Devonian, M.	Gas
7	12/21/2009	PA	Tioga	Richmond	–77.13	41.78		Marcellus Sh.	Devonian, M.	Gas
8	9/8/2009	PA	Centre	Burnside	–78.05	41.13	Impoundment	Marcellus Sh.	Devonian, M.	Gas
9	1/8/2010	PA	Forest	Jenks	–79.16	41.55	Tank or lined pit	Marcellus Sh.	Devonian, M.	Oil
10	12/30/2009	PA	Potter	East Fork	–77.88	41.61		Marcellus Sh.	Devonian, M.	Gas
11.1	4/9/2009	PA	Washington	Cross Creek	–80.39	40.26		Marcellus Sh.	Devonian, M.	Gas
11.2	6/29/2009	PA	Washington	Cross Creek	–80.39	40.26		Marcellus Sh.	Devonian, M.	Gas
12	12/30/2009	PA	Tioga	Gainesville	–77.56	41.69		Marcellus Sh.	Devonian, M.	Gas
13	12/30/2009	PA	Tioga	Gainesville	–77.58	41.68		Tuscarora Fm.	Silurian, L.	Gas
14	1/7/2010	PA	Potter	West Branch	–77.62	41.67		Marcellus Sh.	Devonian, M.	Gas
15	12/16/2009	PA	Clearfield	Lawrence	–78.45	41.17		Marcellus Sh.	Devonian, M.	Gas
16	12/22/2009	PA	Westmoreland	Washington	–79.57	40.49		Marcellus Sh.	Devonian, M.	Gas
17	12/7/2009	PA	Westmoreland	Washington	–79.56	40.50		Marcellus Sh.	Devonian, M.	Gas
18	11/13/2009	PA	Westmoreland	Bell	–79.55	40.51		Marcellus Sh.	Devonian, M.	Gas
19	9/18/2009	PA	Westmoreland	Hempfield	–79.65	40.28		Marcellus Sh.	Devonian, M.	Gas
20	7/16/2009	PA	Westmoreland	Hempfield	–79.57	40.50		Marcellus Sh.	Devonian, M.	Gas
21	7/23/2009	PA	Indiana	Rayne	–79.04	40.75		Marcellus Sh.	Devonian, M.	Gas
22	7/31/2009	PA	Westmoreland	Washington	–79.58	40.50		Marcellus Sh.	Devonian, M.	Gas
23	8/13/2009	PA	Westmoreland	Bell	–79.54	40.50		Marcellus Sh.	Devonian, M.	Gas

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Well / Sample ID	Sample collection date	State	County	Township	Longitude	Latitude	Description of sample site	Producing formation	Producing formation age	Well type
Source: NYSDEC (2009)										
24	4/1/2009	NY	Steuben	Avoca	–77.41	42.40		Marcellus Sh.	Devonian, M.	Gas
25	4/1/2009	NY	Steuben	Avoca	–77.42	42.41		Marcellus Sh.	Devonian, M.	Gas
26	4/2/2009	NY	Chenango	Oxford	–75.61	42.45		Marcellus Sh.	Devonian, M.	Gas
27.1	10/7/2008	NY	Steuben	Caton	–77.04	42.05		Marcellus Sh.	Devonian, M.	Gas
27.2	4/1/2009	NY	Steuben	Caton	–77.04	42.05		Marcellus Sh.	Devonian, M.	Gas
28	10/8/2008	NY	Schuyler	Orange	–77.08	42.28		Marcellus Sh.	Devonian, M.	Gas
29	4/1/2009	NY	Steuben	Woodhull	–77.44	42.02		Marcellus Sh.	Devonian, M.	Gas
30	4/1/2009	NY	Steuben	Troupsburg	–77.47	42.02		Marcellus Sh.	Devonian, M.	Gas
31	4/6/2009	NY	Schuyler	Dix	–76.94	42.34		Marcellus Sh.	Devonian, M.	Gas
32	4/6/2009	NY	Schuyler	Dix	–76.94	42.34		Marcellus Sh.	Devonian, M.	Gas
33	3/26/2009	NY	Schuyler	Orange	–77.07	42.29		Marcellus Sh.	Devonian, M.	Gas
34	4/6/2009	NY	Schuyler	Reading	–76.91	42.44		Marcellus Sh.	Devonian, M.	Gas
35	10/8/2008	NY	Schuyler	Orange	–77.06	42.29		Marcellus Sh.	Devonian, M.	Gas
Source: NYSDEC (1999)										
36		NY	Cattaraugus		–78.66	42.42	Brine tank	Medina Gp.	Silurian, L.	Gas
37		NY	Cattaraugus		–78.66	42.42	Brine tank	Medina Gp.	Silurian, L.	Gas
38		NY	Cattaraugus		–78.68	42.44	Brine tank	Medina Gp.	Silurian, L.	Gas
39		NY	Cattaraugus		–78.68	42.47	Brine tank (rusted)	Medina Gp.	Silurian, L.	Gas
40		NY	Cattaraugus		–78.72	42.46	Brine tank	Medina Gp.	Silurian, L.	Gas
41		NY	Cattaraugus		–78.71	42.46	Brine tank	Medina Gp.	Silurian, L.	Gas
42		NY	Erie		–78.47	42.93	Bottom of brine tank	Medina Gp.	Silurian, L.	Gas
43		NY	Genesee		–78.46	42.93	Bottom of brine tank	Medina Gp.	Silurian, L.	Gas
44		NY	Genesee		–78.46	43.03	Brine tank, subsurface	Medina Gp.	Silurian, L.	Gas
45		NY	Genesee		–78.45	43.02	Brine tank	Medina Gp.	Silurian, L.	Gas
46		NY	Genesee		–78.25	43.05	Brine tank	Medina Gp.	Silurian, L.	Gas

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Well / Sample ID	Sample collection date	State	County	Township	Longitude	Latitude	Description of sample site	Producing formation	Producing formation age	Well type
Source: NYSDEC (1999)—Continued										
47		NY	Genesee		–78.26	43.05	Brine tank, subsurface	Medina Gp.	Silurian, L.	Gas
48		NY	Genesee		–78.30	42.95	Brine tank, subsurface	Medina Gp.	Silurian, L.	Gas
49		NY	Genessee		–78.30	42.88	Brine tank	Medina Gp.	Silurian, L.	Gas
50		NY	Wyoming		–78.10	42.82	Brine tank (rusted)	Theresa Ss.	Cambrian, U.	Gas
51		NY	Wyoming		–78.10	42.83	Brine tank	Theresa Ss.	Cambrian, U.	Gas
52		NY	Wyoming		–78.12	42.82	Brine tank	Medina Gp.	Silurian, L.	Gas
53		NY	Wyoming		–78.11	42.81	Brine tank	Theresa Ss.	Cambrian, U.	Gas
54		NY	Wyoming		–78.36	42.74	Brine tank	Medina Gp.	Silurian, L.	Gas
55		NY	Cayuga		–76.64	42.91	Spigot, base of brine tank	Queenston Sh.	Ordovician, U.	Gas
56		NY	Cayuga		–76.65	42.91	Spigot, base of brine tank	Queenston Sh.	Ordovician, U.	Gas
57		NY	Seneca		–76.86	42.84	Spigot, base of brine tank	Queenston Sh.	Ordovician, U.	Gas
58		NY	Seneca		–76.85	42.84	Spigot, base of brine tank	Queenston Sh.	Ordovician, U.	Gas
59		NY	Seneca		–76.90	42.78	Spigot, base of brine tank	Rochester Sh.	Silurian, L.	Gas
60		NY	Genesee		–77.98	42.93	Brine tank	Medina Gp.	Silurian, U.	Gas
61		NY	Livingston		–77.91	42.94	Brine tank	Medina Gp.	Silurian, L.	Gas
62		NY	Ontario		–77.54	42.81	Brine tank	Medina Gp.	Silurian, L.	Gas
63		NY	Chautauqua		–79.52	42.38	Bottom of brine tank	Medina Gp.	Silurian, L.	Gas
64		NY	Chautauqua		–79.72	42.27	Bottom of brine tank	Medina Gp.	Silurian, L.	Gas
65		NY	Chautauqua		–79.58	42.16	Brine tank	Medina Gp.	Silurian, L.	Gas

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Well / Sample ID	Sample collection date	State	County	Township	Longitude	Latitude	Description of sample site	Producing formation	Producing formation age	Well type
Source: NYSDEC (1999)—Continued										
66		NY	Chautauqua		–79.54	42.08	Brine tank	Bass Islands Dolo.	Silurian, U.	Oil
67		NY	Chautauqua		–79.43	42.09	Brine tank	Medina Gp.	Silurian, L.	Gas
68		NY	Chautauqua		–79.47	42.13	Brine tank	Bass Islands Dolo.	Silurian, U.	Oil
69		NY	Chautauqua		–79.42	42.16	Brine drain tank	Bass Islands Dolo.	Silurian, U.	Oil
70		NY	Erie		–78.89	42.54	Brine tank	Onondaga Ls.	Devonian, M.	Gas
71		NY	Chautauqua		–79.01	42.38	Brine tank	Medina Gp.	Silurian, L.	Gas
72		NY	Chautauqua		–79.16	42.31	Brine tank	Medina Gp.	Silurian, L.	Gas
73		NY	Chautauqua		–79.19	42.31	Bottom of stock tank	Devonian, U., undiv.	Devonian, U.	Gas–Oil
74		NY	Chautauqua		–79.24	42.23	Bottom of stock tank	Bass Islands Dolo.	Silurian, U.	Oil
75		NY	Chautauqua		–79.36	42.17	Brine tank	Bass Islands Dolo.	Silurian, U.	Gas–Oil
76		NY	Chautauqua		–79.38	42.22	Brine tank	Medina Gp.	Silurian, L.	Gas
77		NY	Chautauqua		–79.30	42.16	Brine tank	Medina Gp.	Silurian, L.	Gas
78		NY	Chautauqua		–79.27	42.07	Brine tank	Medina Gp.	Silurian, L.	Gas
79		NY	Erie		–78.79	42.56	Brine tank	Medina Gp.	Silurian, L.	Gas
80		NY	Allegany		–77.92	42.01	Brine tank	Oriskany Ss.	Devonian, L.	Gas
81		NY	Allegany		–77.95	42.01	Brine tank	Oriskany Ss.	Devonian, L.	Gas
82		NY	Tioga		–76.26	42.03	Brine tank	Helderberg Ls.	Devonian, L.	Gas
83		NY	Tioga		–76.31	42.00	Brine tank	Helderberg Ls.	Devonian, L.	Gas

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Well / Sample ID	Sample collection date	State	County	Township	Longitude	Latitude	Description of sample site	Producing formation	Producing formation age	Well type
Source: PA DEP (1992)										
84		PA	Allegheny	S. Fayette	–80.21	40.36	Diked area			Gas
85		PA	Armstrong	Cowanshannock	–79.26	40.79	Tank		Devonian, U.	Gas
86		PA	Armstrong	Cowanshannock	–79.25	40.78	Tank	Catskill/Lock Haven Fms.	Devonian, U.	Gas
87		PA	Cambria	Susquehanna	–78.76	40.70	Tank	Venango Gp.	Devonian, U.	Gas
88		PA	Cambria	Barr	–78.83	40.66	Tank	Lock Haven Fm.	Devonian, U.	Gas
89		PA	Centre	Curtin	–77.77	41.11	Tank	Catskill/Lock Haven Fms.	Devonian, U.	Gas
90		PA	Centre	Burnside	–77.87	41.12	Tank	Catskill/Lock Haven Fms.	Devonian, U.	Gas
91		PA	Clearfield	Jordan	–78.61	40.83	Tank	Catskill/Lock Haven Fms.	Devonian, U.	Gas
92		PA	Clearfield	Burnside	–78.72	40.84	Tank	Lock Haven Fm.	Devonian, U.	Gas
93		PA	Clinton	Beech Creek	–77.70	41.17	Tank	Lock Haven Fm.	Devonian, U.	Gas
94		PA	Clinton	Beech Creek	–77.74	41.15	Tank	Medina Gp.	Silurian, L.	Gas
95		PA	Crawford	Beaver	–80.41	41.81	Tank	Medina Gp.	Silurian, L.	Gas
96		PA	Elk	Highland	–78.83	41.54	Separator tank	Bradford Gp.	Devonian, U.	Oil
97		PA	Elk	Highland	–78.93	41.51	Tank		Devonian	Oil
98		PA	Erie	Millcreek	–80.17	42.08	Tank	Medina Gp.	Silurian, L.	Gas
99		PA	Erie	Conneaut	–80.44	41.89	Tank	Huntersville Chert	Devonian, M.	Gas
100		PA	Fayette	Springfield	–79.36	39.97	Tank	Oriskany Ss.	Devonian, L.	Gas
101		PA	Fayette	Springfield	–79.40	39.96	Tank	Huntersville Chert	Devonian, M.	Gas
102		PA	Forest	Howe	–79.14	41.56	Tank battery	Warren sand	Devonian, U.	Oil
103		PA	Forest	Kingsley	–79.29	41.58	Tank	Devonian, U., undiv.	Devonian, U.	Oil
104		PA	Indiana	Cherryhill	–78.82	40.74	Tank	Kane sand	Devonian, U.	Gas

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Well / Sample ID	Sample collection date	State	County	Township	Longitude	Latitude	Description of sample site	Producing formation	Producing formation age	Well type
Source: PA DEP (1992)—Continued										
105		PA	Indiana	Burrell	–79.17	40.48	Tank	Fifty Foot sand	Devonian, U.	Gas
106		PA	Indiana	White	–79.19	40.65	Tank			Gas
107		PA	Jefferson	Bell	–78.90	40.97	Tank	Devonian, U., undiv.	Devonian, U.	Gas
108		PA	McKean	Wetmore	–78.87	41.63	Separator pit		Unknown	Oil
109		PA	McKean	Lafayette	–78.72	41.83	Pit	Oriskany Ss.	Devonian, L.	Oil
110		PA	Somerset	Middlecreek	–78.92	40.06	Tank	Oriskany Ss.	Devonian, L.	Gas
111		PA	Somerset	Lincoln	–79.07	40.08	Tank	Huntersville Chert	Devonian, M.	Gas
112		PA	Tioga	Union	–76.96	41.57	Drill pit		Ordovician	Gas
113		PA	Venango	Cornplanter	–79.59	41.48	Separator	Red Valley sand	Devonian, U.	Oil
114		PA	Venango	Allegheny	–79.55	41.57	Pit	Venango Gp.	Devonian, U.	Oil
115		PA	Warren	Pleasant	–79.19	41.81	Pit	Medina Gp.	Silurian, L.	Oil
116		PA	Warren	Southwest	–79.57	41.63	Tank	Medina Gp.	Silurian, L.	Gas
117		PA	Warren	Watson	–79.25	41.76	Pit	Devonian, U., undiv.	Devonian, U.	Oil
118		PA	Washington	Cecil	–80.24	40.33	Separator		Devonian, U.	Oil
119		PA	Westmoreland	Washington	–79.58	40.49	Tank	Venango Gp.	Devonian, U.	Gas
120		PA	Westmoreland	Hempfield	–79.53	40.26	Tank	Venango Gp.	Devonian, U.	Gas
Source: Dresel and Rose (2010)										
121	1982	PA	Indiana	Banks	–78.85	40.87	Wellhead	Devonian, U., undiv.	Devonian, U.	Gas
122	1982	PA	Indiana	South Mahoning	–79.14	40.78	Wellhead	Devonian, U., undiv.	Devonian, U.	Gas
123	1982	PA	Warren	Pleasant	–79.21	41.82	Wellhead	Glade sand	Devonian, U.	Oil
124	1982	PA	Crawford	Fairfield	–80.14	41.49	Wellhead	Medina Gp.	Silurian, L.	Gas
125	1982	PA	Centre	Boggs	–77.84	41.00	Separator	Tuscarora Fm.	Silurian, L.	Gas
126	1982	PA	Somerset	Black	–79.11	39.93	Separator	Ridgeley Ss.	Devonian, L.	Gas

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Well / Sample ID	Sample collection date	State	County	Township	Longitude	Latitude	Description of sample site	Producing formation	Producing formation age	Well type
Source: Pritz (2010), this study										
127	04/09	PA	Bradford					Marcellus Sh.	Devonian, M.	Gas
128	04/09	PA	Bradford					Marcellus Sh.	Devonian, M.	Gas
129	04/09	PA	Bradford					Marcellus Sh.	Devonian, M.	Gas
130	04/09	PA	Bradford					Marcellus Sh.	Devonian, M.	Gas
131	04/09	PA	Bradford					Marcellus Sh.	Devonian, M.	Gas
Source: This study										
132.1	12/8/2010	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas
132.2	12/29/2010	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas
132.3	12/30/2010	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas
132.4	12/31/2010	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas
132.5	1/1/2011	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas
132.6	1/2/2011	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas
132.7	1/4/2011	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas
132.8	1/12/2011	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas
132.9	1/17/2011	PA	Greene		–80.05	39.88	Separator	Marcellus Sh.	Devonian, M.	Gas

Table 2. Ra-226, Ra-228, gross alpha, and gross beta activities measured in samples of produced water for wells listed in table 1. Analytical uncertainties are included when known.

[TDS, total dissolved solids; mg/L, milligram per liter; pCi/L, picocurie per liter; ND, not detected]

Well / Sample ID	TDS (mg/L)	Gross alpha (pCi/L)	+/–	Gross beta (pCi/L)	+/–	Ra-226 (pCi/L)	+/–	Ra-228 (pCi/L)	+/–	Total radium (pCi/L)	Ra-228/ Ra-226	Method, method codes
Source: PA DEP (2009–2010)												
1	54,000					436	32.2	121	8.2	556	0.28	SM2540C; EPA904.0, 903.0
2	16,200	14	2	1,322	86	ND	1.8	ND	0.3			SM2540C, 7110C; EPA 900.0, 903.0, 904.0
3	333,000	19,220	2,843	7,944	1,320	50	1.3	37	3.3	87	0.73	SM2540C; EPA 900.0 903.0, 904.0
4	61,800	6,159	743	1,325	190	430	11.0	51	8.9	482	0.12	SM2540C; EPA 900.0, 903.0, 904.0
5.1	38,200	454	126	149	78	66	4.0	2.2	0.9	68	0.03	SM2540C; EPA 900.0, 903.0, 904.0
5.2	82,600	1,644	371	745	242	239	9.7	38	6.3	277	0.16	SM2540C; EPA 900.0, 903.0, 904.0
6		40,880	7,512	750	732	16,920	3,283	1,125	227	18,045	0.07	EPA 903.1, 904.0
7		21,960	4,074	980	757	11,120	2,204	1,287	261	12,407	0.12	EPA 903.1, 904.0
8	124,000					1,525	110	657	76	2,182	0.43	SM18 2540C; EPA 901.1 Mod.
9	284,000	11,810	2,482	1,060	759	4,184	789	1,074	202	5,258	0.26	SM20 2540C; EPA 903.1, 904.0
10	157,000					7,330	460	1,180	180	8,510	0.16	SM18 2540C; EPA 901.1 Mod.
11.1	157,000					951	86	703	69	1,654	0.74	SM18 2540C; EPA 901.1 Mod.
11.2	200,000					1,280	130	1,110	120	2,390	0.87	SM18 2540C; EPA 901.1 Mod.
12	183,000	7,530	1,141	2,683	372	562	26	648	67	1,210	1.15	SM18 2540C; EPA 900.0, 903.0, 904.0
13	358,000	10,356	2,186	11,595	723	892	32	2,589	128	3,481	2.90	SM18 2540C; EPA 900.0, 903.0, 904.0
14	1,470	ND	3	78	4	ND	0.31	ND	0.39	1.00		SM2540C; EPA 900.0, 903.0, 904.0
15	288,900	19,240		7,049		1,268		106		1,374	0.08	SM2540C
16	24,700	318	453	340	590	103	24	168	32	271	1.63	SM2540C; EPA 900.0Mod., 903.1, 904.0
17	88,500	3,640	1,004	ND	631	1,042	197	298	59	1,340	0.29	SM2540C; EPA 900.0Mod., 903.1, 904.0
18	116,000	2,320	800	2,077	929	1,037	200	515	97	1,552	0.50	SM2540C; EPA 900.0Mod., 903.1, 904.0
19	32,500	733	175	81	61	554	104	5.5	1.9	559	0.01	SM2540C; EPA 900.0Mod., 903.1, 904.0
20	45,400	845	213	379	116	66	4.05	1.4	0.3	67	0.02	SM2540C; EPA 900.0, 903.0, 904.0
21	46,460	820	249	505	140	76	2.7	23	2.4	99	0.30	SM2540C; EPA 900.0, 903.0, 904.0
22	47,800	585	163	536	83	36	1.75	2.7	0.2	39	0.08	SM2540C; EPA 900.0, 903.0, 904.0
23	125,100	2,103	631	1,574	335	229	6.8	56	6.5	285	0.25	SM2540C; EPA 900.0, 903.0, 904.0

Table 2. Ra-226, Ra-228, gross alpha, and gross beta activities measured in samples of produced water for wells listed in table 1. Analytical uncertainties are included when known.—Continued

[TDS, total dissolved solids; mg/L, milligram per liter; pCi/L, picocurie per liter; ND, not detected]

Well / Sample ID	TDS (mg/L)	Gross alpha (pCi/L)	+/–	Gross beta (pCi/L)	+/–	Ra-226 (pCi/L)	+/–	Ra-228 (pCi/L)	+/–	Total radium (pCi/L)	Ra-228/ Ra-226	Method, method codes
Source: NYSDEC (2009)												
24		70	48	7	54	0.163	0.20	0.029	0.22	0.192	0.175	
25		54.6	37	59	58	0.195	0.16	0.428	0.34	0.623	2.195	
26		3,914	813	715	202	1,779	343	201	39	1,980	0.113	
27.1		17,940	8,634	4,765	3,829	2,472	484	874	174	3,346	0.354	
27.2		3,968	1,102	618	599	7,885	1,568	234	51	8,119	0.030	
28	206,446	14,530	3,792	4,561	1,634	2,647	494	782	157	3,429	0.295	
29		9,426	2,065	2,780	879	4,049	807	826	160	4,875	0.204	
30		7,974	1,800	1,627	736	5,352	1,051	138	37	5,490	0.026	
31		10,970	2,363	1,170	701	6,125	1,225	516	99	6,641	0.084	
32		20,750	4,117	2,389	861	10,160	2,026	1,252	237	11,412	0.123	
33	205,102	18,330	3,694	ND	654	13,510	2,655	929	179	14,439	0.069	
34		16,550	3,355	1,323	711	15,140	2,989	957	181	16,097	0.063	
35		123,000	23,480	12,000	2,903	16,030	2,995	912	177	16,942	0.057	
Source: NYSDEC (1999)												
36						669	88	1,100	250	1,769	1.644	γ-spectrometry
37						402	68			402		γ-spectrometry
38						1,164	93	429	27	1,593	0.369	γ-spectrometry
39						398	64	234	182	632	0.588	γ-spectrometry
40						259	47			259		γ-spectrometry
41						409	60			409		γ-spectrometry
42						413	61	856	222	1,269	2.073	γ-spectrometry
43						260	43	703	194	963	2.704	γ-spectrometry
44						63	71			63		γ-spectrometry
45						169	86	565	254	734	3.343	γ-spectrometry
46						306	126	568	248	874	1.856	γ-spectrometry
47						175	100	255	179	430	1.457	γ-spectrometry
48								347	55	347		γ-spectrometry
49						290	50	460	172	750	1.586	γ-spectrometry

Table 2. Ra-226, Ra-228, gross alpha, and gross beta activities measured in samples of produced water for wells listed in table 1. Analytical uncertainties are included when known.—Continued

[TDS, total dissolved solids; mg/L, milligram per liter; pCi/L, picocurie per liter; ND, not detected]

Well / Sample ID	TDS (mg/L)	Gross alpha (pCi/L)	+/–	Gross beta (pCi/L)	+/–	Ra-226 (pCi/L)	+/–	Ra-228 (pCi/L)	+/–	Total radium (pCi/L)	Ra-228/ Ra-226	Method, method codes
Source: NYSDEC (1999)—Continued												
50						764	81	433	242	1,197	0.567	γ-spectrometry
51						450	66	326	319	776	0.724	γ-spectrometry
52						477	65	651	306	1,128	1.365	γ-spectrometry
53						708	71	350	297	1,058	0.494	γ-spectrometry
54						238	117	269	52	507	1.130	γ-spectrometry
55						1,240	100	1,290	130	2,530	1.040	γ-spectrometry
56						823	69	1,333	450	2,156	1.620	γ-spectrometry
57						557	67	933	230	1,490	1.675	γ-spectrometry
58						465	65	977	230	1,442	2.101	γ-spectrometry
59						ND						γ-spectrometry
60						369	61	890	227	1,259	2.412	γ-spectrometry
61						538	72	625	207	1,163	1.162	γ-spectrometry
62						146	92			146		γ-spectrometry
63						187	20	80	29	267	0.427	γ-spectrometry
64						324	36	503	30	827	1.552	γ-spectrometry
65						444	47	1,690	55	2,134	3.806	γ-spectrometry
66						1,550	110	319	60	1,869	0.206	γ-spectrometry
67						366	45	1,660	47	2,026	4.536	γ-spectrometry
68						654	36	1,440	50	2,094	2.202	γ-spectrometry
69						1,040	40	387	32	1,427	0.372	γ-spectrometry
70						64.2	27.0	83	12	147	1.294	γ-spectrometry
71						148	17	100	23	248	0.676	γ-spectrometry
72						160	35	574	31	734	3.588	γ-spectrometry
73						185	20	86	25	271	0.467	γ-spectrometry
74						953	64	444	45	1,397	0.466	γ-spectrometry
75						585	67			585		γ-spectrometry
76						951	109	1,500	170	2,451	1.577	γ-spectrometry
77						484	32	1,420	50	1,904	2.934	γ-spectrometry
78						156	35	504	36	660	3.231	γ-spectrometry

Table 2. Ra-226, Ra-228, gross alpha, and gross beta activities measured in samples of produced water for wells listed in table 1. Analytical uncertainties are included when known.—Continued

[TDS, total dissolved solids; mg/L, milligram per liter; pCi/L, picocurie per liter; ND, not detected]

Well / Sample ID	TDS (mg/L)	Gross alpha (pCi/L)	+/–	Gross beta (pCi/L)	+/–	Ra-226 (pCi/L)	+/–	Ra-228 (pCi/L)	+/–	Total radium (pCi/L)	Ra-228/ Ra-226	Method, method codes
Source: NYSDEC (1999)—Continued												
79						111	35	60	23	170	0.538	γ-spectrometry
80						901	52	250	42	1,151	0.278	γ-spectrometry
81						691	26	154	30	845	0.223	γ-spectrometry
82						3,760	100	1,110	60	4,870	0.295	γ-spectrometry
83						1,620	110	1,790	60	3,410	1.105	γ-spectrometry
Source: PA DEP (1992)												
84	143,432					512		202		714	0.395	
85						315		165		480	0.524	
86						20		13		33	0.650	
87	175,296					1,408		904		2,312	0.642	
88	195,404					1,154		1,083		2,237	0.938	
89	222,672					163		126		289	0.773	
90	252,980					1,489		636		2,125	0.427	
91	185,146					2,015		1,749		3,764	0.868	
92	230,924					107		77		184	0.720	
93	194,902					731		491		1,222	0.672	
94	153,096					811		530		1,342	0.653	
95	396,012					599		1,683		2,282	2.810	
96	31,502					15		18		33	1.150	
97	25,159					23		26		49	1.143	
98	390,928					628		1,478		2,106	2.355	
99	378,148					588		1,483		2,072	2.521	
100	341,918					4,685		2,038		6,723	0.435	
101	354,034					566		2,110		2,676	3.728	
102	130,588					42		42		84	1.000	
103	86,988					39		56		95	1.427	
104	186,736					2,019		2,196		4,215	1.088	
105	198,668					2,575		1,866		4,441	0.725	
106	121,928					450		313		763	0.696	

Table 2. Ra-226, Ra-228, gross alpha, and gross beta activities measured in samples of produced water for wells listed in table 1. Analytical uncertainties are included when known.—Continued

[TDS, total dissolved solids; mg/L, milligram per liter; pCi/L, picocurie per liter; ND, not detected]

Well / Sample ID	TDS (mg/L)	Gross alpha (pCi/L)	+/–	Gross beta (pCi/L)	+/–	Ra-226 (pCi/L)	+/–	Ra-228 (pCi/L)	+/–	Total radium (pCi/L)	Ra-228/ Ra-226	Method, method codes
Source: PA DEP (1992)—Continued												
107						1,280		848		2,128	0.663	
108	36,470					8		12		20	1.446	
109						185		184		370	0.995	
110	176,676					203		1,543		1,746	7.601	
111	182,274					1,988		499		2,487	0.251	
112						1,137		1,457		2,594	1.281	
113	114,208					12		30		42	2.500	
114	36,282					54		77		131	1.426	
115	59,554					34		27		61	0.794	
116	395,440					795		968		1,763	1.219	
117	125,264					275		187		462	0.680	
118	134,164					255		456		711	1.788	
119	221,134					170		46		216	0.271	
120	402,148					857		71		928	0.083	
Source: Dresel and Rose (2010)												
121	253,000					1,900				1,900		
122	244,000					200				200		
123	91,000					0				0.0		
124	257,000					500				500		
125	259,000					5,300				5,300		
126	302,000					5,000				5,000		
Source: Pritz (2010), this study												
127	122,527					2,653	11	318	22	2,971	0.120	γ-spectrometry
128	250,112					3,082	21	935	27	4,018	0.303	γ-spectrometry
129	134,880					1,958	26	572	37	2,530	0.292	γ-spectrometry
130	222,681					1,486	13	472	23	1,957	0.317	γ-spectrometry
131	117,259					1,756	6	377	10	2,133	0.215	γ-spectrometry

Table 2. Ra-226, Ra-228, gross alpha, and gross beta activities measured in samples of produced water for wells listed in table 1. Analytical uncertainties are included when known.—Continued

[TDS, total dissolved solids; mg/L, milligram per liter; pCi/L, picocurie per liter; ND, not detected]

Well / Sample ID	TDS (mg/L)	Gross alpha (pCi/L)	+/–	Gross beta (pCi/L)	+/–	Ra-226 (pCi/L)	+/–	Ra-228 (pCi/L)	+/–	Total radium (pCi/L)	Ra-228/ Ra-226	Method, method codes
Source: This study												
132.1						1,312	19	300	23	1,612	0.229	γ-spectrometry
132.2						3,363	21	542	19	3,905	0.161	γ-spectrometry
132.3						4,372	39	577	19	4,949	0.132	γ-spectrometry
132.4						4,892	38	599	29	5,491	0.122	γ-spectrometry
132.5						5,110	18	626	20	5,736	0.123	γ-spectrometry
132.6						5,210	31	614	32	5,824	0.118	γ-spectrometry
132.7						3,105	26	686	30	3,791	0.221	γ-spectrometry
132.8						5,446	28	820	17	6,266	0.151	γ-spectrometry
132.9						5,272	88	846	33	6,118	0.160	γ-spectrometry

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