Naturally Occurring Radioactive Materials (NORM) in Arizona

by

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INTRODUCTION

Uranium-238 and other radioactive isotopes (radioisotopes) are present in small concentrations in all geologic materials. In a tiny fraction of these materials, such as uranium ore, radioisotope concentrations are greatly elevated. A much larger fraction of geologic materials contain slightly elevated concentrations of radioisotopes, but the radiation produced by these materials is far less than for uranium ore. The purpose of this brief report is to summarize the distribution, geologic association, and uranium concentration of naturally occurring radioactive materials (NORM) in Arizona, with emphasis on those materials that contain only slightly elevated concentrations of uranium.

The bedrock geology of Arizona is characterized by a great diversity of rock types that were produced by a complex geologic history over the past 1.8 billion years. The concentrations of all of the chemical elements are highly variable in these different rock types. Elements that are usually present in small concentrations, known as trace elements, include the radioactive elements uranium and thorium.

RADIOISOTOPES

Most of the radioactivity in Earth's crust is produced by uranium, thorium, their radioactive daughter products, and potassium. Potassium, a common and widespread element, is only very weakly radioactive and does not decay into radioactive daughter isotopes. Thorium is generally more abundant in crustal rocks than uranium, but its distribution and significance in Arizona are not as well understood as for uranium. Because of past exploration for uranium deposits, and its importance for atomic energy and weapons, many uranium deposits have been discovered and mined in Arizona, and many other sub-economic deposits are known. Radon gas, a uranium-decay product, seeps into homes and other buildings from underlying soil and rock. Inhalation of airborne radon-decay products causes most human radiation exposure from natural sources. Many studies have been done on radon and uranium as a result of this exposure. The following overview of NORM focuses exclusively on uranium because it is well studied and is the source of most radioactivity in typical NORM. This is not meant to imply, however, that thorium is not a significant source of natural radioactivity in Arizona.

Uranium-238, uranium-235, and thorium-232 have half lives measured in hundreds of millions to billions of years. When an atom of one of these isotopes decays it begins a relatively quick sequence of 10 to 14 decays before becoming a non-radioactive isotope of lead. These decay-series isotopes are the source of most of the radioactivity emanating from geologic materials. About 99.3% of naturally occurring uranium is the isotope uranium-238. When an atom of uranium-238 decays, it begins a series of 5 decays that, after a few hundred thousand years, reach the highly radioactive isotope radium-226, which has a half life of 1600 years. Radium behaves chemically like calcium and, if ingested, can be absorbed by bone where it is a hazardous internal source of radiation. It is commonly monitored in water supplies and is evaluated through measurement of alpha decay (radioactive decay by emission of alpha particles). Radium-226 decays into radon-222, a chemically inert but highly radioactive gas that can accumulate in homes and buildings. Most human radiation exposure is due to inhalation of the shortlived, airborne daughter isotopes produced by the decay of indoor radon-222 gas derived from NORM. Virtually all of this exposure is to the lung, and it is thought that thousands of lung cancer deaths occur each year in the United Stated as a result of this exposure.

BACKGROUND RADIOACTIVITY FROM NURE SURVEY

During the late 1970s, the U.S. Department of Energy conducted a National Uranium Resource Evaluation (NURE) that included an aerial radiation survey of most of the United States (Duval and Riggle, 1999). The survey airplanes carried gamma-ray spectrometers that measured the radioactivity of one of the radioactive daughter products (bismuth-214) of the isotope uranium-238. This survey provided a good indication of the regional distribution of uranium in the top 10 to 100 cm of the Earth's surface over the 96 percent of Arizona that was surveyed. Potassium and thorium were also surveyed. The NURE data were used to generate a digital map of uranium levels in the conterminous United States. The Arizona part of this map is shown in Figure 1. To produce this map, Arizona was divided into 2 x 2 km squares (4 square kilometers, or 1.5 square miles, for each square), each of which was assigned a numerical value for average uranium level based on the average bismuth-214 concentration measured in the airborne survey. The map data, along with similar data for thorium and potassium, are available at U.S. Geological Survey web site ftp://musette.cr.usgs.gov/pub/GEOPHYSICAL_DATA/cdrom_DDS-9/GRIDS/ARC_INFO/. (Also released on CD-ROM by Duval and Riggle, 1999).

Calculating uranium concentration based on bismuth-214 concentration is done with the assumption that uranium-238 decay series isotopes that are parent isotopes of bismuth 214 have not been removed from or added to surficial earth materials. For example, radon gas that escapes from the ground and is deposited elsewhere will reduce measured bismuth-214 levels. Calculated uranium concentration, based on bismuth-214 radioactivity, will be lower than actual levels in this situation. Because of uncertainty in the relationship between bismuth-214 and uranium-238, uranium concentrations calculated from bismuth-214 concentrations are referred to as "equivalent" uranium concentrations, or "eU". This calculation is usually done assuming "secular equilibrium", which means that it is assumed that decay series isotopes have not been lost from or added to the earth materials.

Map figure 1 shows that background radiation levels from uranium are highly variable. Analysis of the data set used for this map indicates that about 98.3 percent of Arizona has uranium concentrations below 4.5 ppm (Table 1). The survey did not detect small areas of elevated uranium concentration, however. For example, a limestone in southwestern Tucson that contains up to 40 ppm uranium over an area of about 0.12 km^2 (0.047 mi^2) did not produce an identifiable anomaly on the map. Failure to detect this area of elevated uranium concentrations probably occurred because the area is small and it fell between flight lines, or because it did not cover a large enough area to significantly influence the background radiation levels at the elevation of the survey airplane.

BACKGROUND RADIOACTIVITY FROM SURVEYS BY THE ARIZONA GEOLOGICAL SURVEY

During the 1980s it was recognized that radon gas is the major source of human radiation exposure. As a result, the U.S. Environmental Protection Agency (EPA) established a program to assist States in evaluating indoor-radon levels, reducing radon exposure, and determining the natural distribution of uranium in areas that are populated or that might be developed in the foreseeable future. The Arizona Geological Survey (AZGS) participated in the State Indoor Radon Grant (SIRG) program for 9 years. During this time, numerous surveys were done to determine uranium levels in geologic materials. The surveys were done using a portable gamma-ray spectrometer that, like the instruments used during the NURE aerial survey, measured the radioactivity of bismuth-214, a daughter isotope of uranium-238.

Statistical analysis of hundreds of uranium measurements indicated that average uranium levels for all types of rock and soil in Arizona, except rocks known to contain elevated uranium levels, was about

1.6 ppm uranium. The standard deviation of measurements was about 2 ppm uranium. Two standard deviations above the mean is about 5.6 ppm uranium. For the purpose of identifying areas where indoor radon is likely to be a problem in a significant percentage of homes, areas with greater than 6 ppm uranium were considered anomalous (Duncan and Spencer, 1993a).

NATURALLY OCCURRING RADIOACTIVE MATERIALS (NORM)

Areas where uranium levels are greater than approximately 4.5 ppm as determined by the NURE aerial surveys are considered here to be anomalous and to represent areas of *naturally occurring radioactive materials* (NORM). The NURE aerial survey identified no areas with uranium concentrations greater than 6 ppm, but found that about 1.7 percent of Arizona is underlain by materials with average uranium concentrations greater than 4.5 ppm (4988 km^2, or 1925 mi^2; Table 1). In contrast to the NURE results, Arizona Geological Survey SIRG program surveys and other land-based surveys found many areas with average uranium levels greater than 6 ppm (papers *in* Spencer, ed., 1993b). The discrepancy between the aerial and ground-based surveys is attributed to the fact that the airborne survey data represent average uranium levels over 4 km^2 areas, whereas most of the areas with elevated uranium levels surveyed in ground-based studies are much smaller. The NURE survey reveals, however, large areas of moderately elevated uranium that possibly contain smaller areas of higher concentrations of uranium.

In a preliminary analysis, the NURE NORM areas were plotted on a geologic map and divided into five types based on evaluation of their geologic associations (Table 2): (1) About a third of the NORM area, covering a total of about 1356 km^2 (523 mi^2), is underlain primarily by granite. (2) Another third is underlain primarily by volcanic rocks. (3) Eleven percent is underlain by the Triassic Chinle Formation, a sedimentary rock unit on the Colorado Plateau that contains numerous uranium deposits and historic uranium mines. (4) Eight percent is associated with mixed rock types where one dominant rock type was not identified. (5) The remaining 15 percent consists of small anomalies, generally less than 10 km^2, that were not associated with a specific geologic feature but that possibly could be with more analysis.

Uranium deposits on the Colorado Plateau have yielded almost all of the approximately 9000 tons of uranium that have been mined in Arizona (Table 3). Most of this production was from bedded deposits in the Chinle and Morrison Formations, and from the Orphan Mine, a breccia pipe deposit near the south rim of the Grand Canyon (Keith et al., 1983). The NURE aerial survey did not reveal the areas where two thousand tons of uranium were mined from the Morrison Formation in northeastern Arizona (Table 3). These areas undoubtedly would be considered NORM areas if they were surveyed in detail. Most likely they did not show up on the NURE aerial survey because the uranium associated with these deposits is not at the Earth's surface or is concentrated in small geographic areas. The Orphan Mine, which yielded two thousand tons of uranium, also does not appear on the NURE aerial survey, probably for the same reasons.

Of the five categories of NORM areas outlined above, only the areas of granitic rocks have been identified as a source of radon gas in homes, probably because significant numbers of homes have not been built on the other types of NORM. In addition, Cenozoic lakebed sediments are known to be a major source of elevated radon levels in homes. These lakebeds do not appear as significant on the NURE aerial survey map, but were surveyed extensively by the Arizona Geological Survey as part of its radon investigations because many homes are built on or near these materials.

The following are brief descriptions of some of the areas where granitic rocks and lakebed sediments include naturally occurring radioactive materials. The descriptions focus primarily on areas that have not been the target of uranium mining but contain enough uranium to represent NORM. Numerous other NORM areas of these types are present in Arizona but are not described here. The intention here is to briefly describe the character of these NORM areas where they have been studied (see papers *in* Spencer, 1993b, for outlines of many NORM areas). Uranium deposits on the Colorado Plateau are not reviewed because they are described by an extensive scientific literature (see review by Wenrich et al., 1989).

NORM areas underlain by volcanic rocks are not reviewed as very little is known about the low-level uranium anomalies in these rocks.

Granitic NORM

Dells granite. The Dells granite is exposed over about 10 km^2 (4 mi^2) northeast of Prescott in Yavapai County. Its average uranium content is ~9 ppm, but uranium concentrations are highly variable and range up to 40 ppm (Proctor et al., 1993). Mean thorium concentration is 27 ppm. Uranium and thorium levels are somewhat correlated. Silver et al. (1980) concluded that uranium and its decay products had remained in the specific minerals in which they were originally trapped when the granite crystallized 1.4 billion years ago. However, Proctor et al. (1993) concluded that (1) the large variability in uranium concentration in the Dells granite was the result of variable weathering and oxidation of uranium-bearing minerals, and that the distinctive and clearly visible iron-oxide banding in the granite was evidence of this oxidative weathering, (2) measurements of relatively high uranium concentrations reflect original uranium concentration before oxidation and leaching of uranium from the rock, and (3) approximately 6 thousand metric tons of uranium had been leached from the granite and had possibly been deposited in undiscovered uranium deposits in the subsurface. Sediments derived from the Dells granite and now located near but downslope from the granite do not contain elevated uranium levels (Duncan and Spencer, 1993d), which supports the interpretation of Proctor et al. (1993) that uranium has been leached from the granite by water and not merely displaced with disaggregated-granite sediment.

The Dells granite was identified as a possible source of radon gas in homes built on the granite. A survey revealed that roughly half the homes built on the granite had radon levels above 4 pCi/l (picocuries per liter; Kearfott, 1989), which is the guideline level above which the U.S. Environmental Protection Agency recommends remedial action to reduce radon levels. In contrast, similar testing statewide revealed that less than 6% of homes have levels above 4 pCi/l (Spencer, 1993a). One small home was built around a 73 m deep water well. The well casing extended upward through the floor of the home and exited through a wall. An opening between the floor and the casing allowed ready entry of air that had resided between the well casing and the granite beneath the house. A box was placed over the area where the casing entered the home. A charcoal-canister test of the air in the box revealed a radon level of 11,000 pCi/l (Kearfott, 1989), which was thought at the time to be the highest radon level ever measured in a home.

Lawler Peak granite. The Lawler Peak granite, exposed over about 28 km^2 (10 mi^2), is located in western Yavapai County about 70 km (43 mi.) west of Prescott (Anderson et al., 1955). The granite is next to the Bagdad copper mine, a large porphyry copper mine that is associated with a much younger, unrelated granite. Although average uranium levels in the Lawler Peak granite are about 16 ppm, several samples had significantly higher levels, including one at 551 ppm uranium (May et al., 1982). Silver et al. (1980) concluded that as much as 100,000 tons of uranium had been leached from the granite between about 75 and 230 million years ago, and that undiscovered uranium deposits produced by precipitation of this uranium could be buried in the area.

Lakebed NORM

Verde Valley. Verde Valley is a basin about 140 km (100 mi) north of Phoenix that was occupied by a lake several million years ago. Sediments of the Verde Formation that were deposited in this lake are widely exposed on the floor and low on the flanks of the valley. These sediments include limestone, silty limestone, calcareous siltstone and mudstone, and gypsum (Nations and Ranney, 1989; Wadell, 1972) that are exposed over an area of roughly 350 km^2 (135 mi^2). These sediments were divided into a mudstone unit and a limestone unit by Nations and Ranney (1989).

As with many lakebeds, the lake sediments in Verde Valley contain anomalously high uranium levels. A gamma-ray spectrometer survey revealed that the mudstone unit of the Verde Valley lake sediments contained an average of 11.5 ppm uranium, whereas the limestone unit contained an average of

4.6 ppm uranium (Duncan and Spencer, 1993b, f). Two samples of the mudstone contained over 40 ppm uranium. Three of 40 homes in Verde Valley that were tested for radon during a statewide survey contained >10 pCi/l radon, which is probably due to home placement on lakebed NORM (Duncan and Spencer, 1993e).

White Eagle Mine Formation. The White Eagle Mine Formation, which consists of calcareous lake sediments similar to the Verde Formation, is exposed in a 20-km-long belt near Cave Creek north of Phoenix. The total area of exposure is small, however, as the rocks are only discontinuously exposed (Duncan and Spencer, 1993c). Uranium concentrations averaged 8.4 ppm, with one measurement of 33 ppm. However, at the west end of the belt levels were much higher, with measured uranium levels of 247 ppm at one location and 261 ppm at another.

Southwest Tucson. Lakebed limestone is exposed over about 0.12 km^2 (0.047 $mi.^2$) near the intersection of Cardinal Avenue and Valencia Road in southwestern Tucson. The limestone contains the uranium mineral uranophane and had been prospected for uranium when the U.S. Atomic Energy Commission surveyed the area (Miller, 1955). The Arizona Atomic Energy Commission (AAEC, now the Arizona Radiation Regulatory Agency), after being notified that the Cardinal and Valencia area was being developed by a home builder, obtained permission to carry out a detailed radiation survey of the Chastain housing development property (Ochoa et al., 1976). The AAEC concluded that the "Chastain property yearly total [radiation] was calculated to be 0.22380 rem. AAEC permissible yearly total is 0.5 rem. No remedial action is recommended by the AAEC" (Ochoa et al., 1976; Spencer et al., 1993).

Recognition of the significance of radon in human radiation exposure led to another survey of the Cardinal and Valencia limestone deposit in the 1980s. It was found that background radiation levels associated with the limestone were up to 14 times background. A survey of indoor radon levels in the homes built on the limestone recorded radon levels up to 40 pCi/l. High radon levels were associated with areas where background radioactivity was high (Spencer et al., 1993).

Anderson Mine Formation. The Anderson Mine Formation is located in a remote area in west-central Arizona northwest of Wickenburg. It consists of limestone, siltstone, sandstone, and related fine grained and calcareous sediments, including coal seams and carbonaceous mudstone (Sherborne et al., 1979; Otton, 1981). These sediments extend southward in the subsurface beneath the margin of Date Creek Basin (Lease, 1981; Mueller and Halbach, 1983). Uranium concentrations are substantially elevated in carbonaceous mudstone and related calcareous and tuffaceous rocks, which are exposed over about 5 km^2 (Sherborne et al., 1979). Unoxidized sediments contain 250 to 750 ppm uranium, and oxidized uranium ore contains up to 25% uranium (Sherborne et al., 1979).

CONCLUSION

Geologic materials with greater than 4.5 ppm uranium, present at numerous locations in Arizona, are defined herein as naturally occurring radioactive materials (NORM). NORM could also include areas of elevated thorium concentration and, perhaps, other naturally occurring radioisotopes, but these were not considered in this report because there is less available information on other radioisotopes. The size, concentration, and geologic setting of NORM varies greatly in Arizona. An aerial survey conducted by the National Uranium Resource Assessment (NURE) program determined that about 1.7 percent of Arizona is underlain by materials with greater than about 4.5 ppm uranium.

Areas of granitic NORM and lakebed NORM are present at many widely spaced locations in Arizona. Some of these areas, which were studied as part of the EPA’s SIRG program, are associated with elevated radon levels in overlying homes (Spencer, 1993a). The granitic NORM were detected by the NURE survey, but the lakebed NORM were largely undetected by this survey. The fact that different survey methods identified different NORM types and areas probably indicates that our present knowledge of the significance and character of NORM in Arizona remains incomplete.

Aerial surveys such as the NURE surveys reveal variations in uranium concentration over length scales of kilometers. However, uranium concentrations in geologic materials can vary greatly over length scales that are microscopically small. Most surveys done by the Arizona Geological Survey under the

SIRG program used a portable detector that, with each measurement, determined average uranium levels over several square meters. Individual measurements were made at stations that were typically tens to hundreds of meters apart. In general, the SIRG surveys determined that uranium levels could vary greatly over distances of tens to hundreds of meters, but no effort was made in these surveys to specifically evaluate these variations. Furthermore, no effort was made to evaluate uranium-concentration variations over distances of meters. Evaluating anthropogenic enhancement or depletion of radioisotopes in geologic materials will require an understanding of both pre-existing concentrations and the significance of aerial variations in radioisotope concentration over intermediate length scales of meters to tens of meters. At present, the intermediate-scale heterogeneity of uranium in NORM is poorly understood and has not been specifically evaluated. Even less is known about intermediate-scale heterogeneity of other radioisotopes in NORM.

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Table 1: Survey of uranium content in Arizona from NURE airborne survey					
rank	U content	count	area (sq km)*	% area	% area excluding areas of no data**
0	no data	3038	12152	4.149	
1	0.0 - 0.5	18	72	0.025	0.026
2	0.5 - 1.0	585	2340	0.799	0.834
3	1.0 - 1.5	5285	21140	7.218	7.530
4	1.5 - 2.0	11615	46460	15.863	16.550
5	2.0 - 2.5	16016	64064	21.874	22.820
6	2.5 - 3.0	14161	56644	19.340	20.177
7	3.0 - 3.5	11344	45376	15.493	16.163
8	3.5 - 4.0	6363	25452	8.690	9.066
9	4.0 - 4.5	3549	14196	4.847	5.057
10	4.5 - 5.0	1195	4780	1.632	1.703
11	5.0 - 5.5	52	208	0.071	0.074
total		70183	292884		
*cell size = 4 km sq					
**area with data = 280732 sq km					
data from USGS GRID file, analysis by Ann Youberg, Sept. 2002					

Table 2: Preliminary geologic analysis of NORM areas*

NURE data summary	map squares	km^2	mi.^{2}	% of AZ	% of NORM
Arizona total area		295045	113860	100	
no NURE data	2613	16331	6302	5.54	
NORM areas	650	4063	1568	**1.38**	**100.0**
granite					
Hualapai Mountains	82	513	198	0.174	12.6
North Dome Rock Mountains	55	344	133	0.117	8.5
Poachie Mountains	43	269	104	0.091	6.6
North Mohave Mountains	18	113	43	0.038	2.8
Kitt Peak	8	50	19	0.017	1.2
Cochise Stronghold	7	44	17	0.015	1.1
Lawler Peak	4	25	10	0.008	0.6
total granite	217	1356	523	**0.460**	**33.4**
volcanic rocks					
Trigo Mountains	163	1019	393	0.345	25.1
Santa Rita Mountains	28	175	68	0.059	4.3
Sullivan Buttes	15	94	36	0.032	2.3
Galiuro Mountains	6	38	14	0.013	0.9
total volcanic rocks	212	1325	511	**0.449**	**32.6**
Chinle Formation					
Chinle Fm., Cameron area	49	306	118	0.104	7.5
Chinle Fm., Holbrook area	20	125	48	0.042	3.1
total Chinle	69	431	166	**0.146**	**10.6**
mixed rock types					
Atascosa Mountains	21	131	51	0.044	3.2
Patagonia Mountains	12	75	29	0.025	1.8
Quijotoa Mountains	11	69	27	0.023	1.7
Hopi Buttes / Bidahochi Fm	9	56	22	0.019	1.4
total mixed	53	331	128	**0.112**	**8.2**
total identified anomalous	551	3444	1329	1.167	84.8
unidentified anomalous	99	619	239	0.210	15.2

*Analysis of earlier reduction of NURE airborne data in which radiation measurements were averaged in 2.5 x 2.5 km squares and NORM was defined as >5.5 ppm eU. Revised data processing by USGS reduced NORM materials by about 1 ppm eU. Only this table uses old USGS data (the rest of this report uses new data reduction).

Table 3: Colorado Plateau Uranium Production*		
District (Chinle Fm.)	**Production (lbs)**	**Production (tons)**
Cameron	1216200	608.1
Cane Valley	5445500	2722.8
Chinle	0	0.0
Holbrook	420	0.2
Monument Valley	3225500	1612.8
Nakai Mesa	120500	60.3
Nazlini	0	0.0
Petrified Forest	6400	3.2
Rainbow	175	0.1
Stinking Springs	69	0.0
Vermillion Cliffs	6100	3.1
Warhoop	1516	0.8
Winslow	30	0.0
total	10022410	**5011.2**
District (Morrison Formation)	**Production (lbs)**	**Production (tons)**
Black Rock Point	89100	44.6
Cove Mesa	173300	86.7
Lukachukai	3483300	1741.7
N.E. Carrizon Mtns.	70400	35.2
Red Rock	9550	4.8
West Carrizo	2520	1.3
total	3828170	**1914.1**
District (breccia pipe)	**Production (lbs)**	**Production (tons)**
Bentley	0	0.0
Breccia Pipe	0	0.0
Chapel	9	0.0
Copper Mtn.	0	0.0
Francis	0	0.0
Grandview	0	0.0
Hacks Canyon	4800	2.4
Havasu Canyon	0	0.0
Orphan	4360000	2180.0
Pigeon Pipe	0	0.0
Prospect Canyon	42	0.0
Valle	0	0.0
total	4364851	**2182.4**
District (misc)	**Production (lbs)**	**Production (tons)**
Hopi Buttes	576	0.3
Black Mountain	57600	28.8
total	58176	**29.1**
District (Transition Zone)	**Production (lbs)**	**Production (tons)**
Salt River	390	0.2
Sierra Ancha	106300	53.2
total	106690	**53.3**
Grand Total	18380297	9190