Radioactivity and Uranium Content of Some Cretaceous Shales, Central Great Plains

By Harry A. Tourtelot

Trace Elements Investigations Report 298

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RADIOACTIVITY AND URANIUM CONTENT OF SOME CRETACEOUS SHALES, CENTRAL GREAT PLAINS*

By

Harry A. Tourtelot

January 1955

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*This report concerns work done on behalf of the Division of Raw Materials of the U. S. Atomic Energy Commission.

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TEPCO, RPS, Washington (Including master)	3
	53

CONTENTS

ILLUSTRATIONS

ILLUSTRATIONS

TABLES

RADIOACTIVITY AND URANIUM CONTENT OF SOME CRETACEOUS SHALES, CENTRAL GREAT PLAINS

By Harry A. Tourtelot

ABSTRACT

The Sharon Springs member of the Pierre shale of Cretaceous age, a hard black organic-rich shale similar to the Chattanooga shale, is radioactive throughout central and western South Dakota, most of Nebraska, northern Kansas, and northeastern Colorado. In the Missouri River valley, thin beds of the shale contain as much as 0.01 percent uranium. Beds as much as 20 feet thick or more have a radioactivity of about 0.01 percent equivalent uranium in southwestern Nebraska according to interpretation of gamma-ray well logs. The radioactivity and uranium content is highest in the Missouri River valley in South Dakota and in southwestern Nebraska where the shale rests disconformably on the underlying Niobrara formation of Cretaceous age. Near the Black Hills, and in the area to the north, the shale of the Sharon Springs member rests on a wedge of the Gammon ferruginous member of the Pierre, which is represented by a disconformity to the east and south, and the radioactivity of the shale is low although greater than that of overlying strata. The shale also contains a suite of trace elements in which arsenic, boron, chromium, copper, molybdenum, nickel, selenium, and vanadium are conspicuous. Molybdenum and tin are less abundant in the Sharon Springs than in similar shales of Paleozoic age and silver and selenium are more abundant.

In the Great Plains region, the upper 30-50 feet of Cretaceous shales overlain unconformably by the White River group of Oligocene age has been altered to bright-colored material. This altered zone is chiefly the result

of pre-Oligocene weathering although post-Oligocene ground water conditions also have affected the zone. The greatest radioactivity occurs in masses of unaltered shale measuring about 1 x 4 feet in cross section included in the lower part of the altered zone. Where the zone is developed on shale and marl of the Niobrara formation, parts of the included unaltered shale contains as much as 0.1 percent equivalent uranium and 0.03 percent uranium. The disequilibrium between equivalent uranium (radioactivity) and the uranium content of the shales is believed to be a surface feature caused by relatively recent leaching of uranium from the present outcrops. The co-extensive distribution of the altered zone of Cretaceous shales and strata of the overlying White River group suggest that most of the uranium in the small masses of unaltered marl in the altered zone has been derived from the White River group.

INTRODUCTION

A reconnaissance search for uranium in shale was made in August 1953, in western Nebraska and southern South Dakota as part of the Geological Survey's search for uranium in carbonaceous materials. The reconnaissance was made on behalf of the Division of Raw Materials of the U. S. Atomic Energy Commission. The objectives of the reconnaissance were to examine the Sharon Springs member of the Pierre shale of Cretaceous age, which published accounts indicated was similar to the Chattanooga and other uranium-bearing shales, and to examine Cretaceous shales where they were overlain by the White River group.

Radioactivity in the Sharon Springs member was thought likely to be supported by uranium that was incorporated with the shale when it was deposited. Radioactivity in the shales immediately beneath the White River group was thought likely to be related to geologic processes acting on the shales prior to the deposition of the White River group or during and afterwards.

SHARON SPRINGS MEMBER OF THE PIERRE SHALE

The Sharon Springs member is the basal member of the Pierre shale in Kansas and was named by Elias (1931) from exposures in Logan and Wallace Counties. At the type locality, the member is about 155 feet thick and consists chiefly of black organic-rich shale in which scales and bones of fish are abundant. In 1937, Dane, Pierce, and Reeside applied the name Sharon Springs member to about 500 feet of similar rocks in east central Colorado, and Griffitts (1949) extended the name to a similar zone of hard flaky shale in northeastern Colorado along the Front Range. Searight (1938) used the name Sharon Springs for the basal member of the Pierre in

central South Dakota and along the eastern margin of the Black Hills. Crandell (1950) reviewed the subsequent modifications of usage of the name in the Missouri River valley. The Sharon Springs member is correlated with the Mitten black shale member of the Pierre shale in the western and northern parts of the Black Hills (Cobban, personal communication), and in this region the Mitten lies about 800 feet above the base of the Pierre shale (Cobban, 1952). Strata assigned to the Sharon Springs are 7 to 35 feet thick in central South Dakota, but at least 65 feet of bituminous shale of the Sharon Springs member is present along the southeast flank of the Black Hills. The Mitten shale is about 150 feet thick on the north flank of the Black Hills (Cobban, 1952).

The distribution of the Sharon Springs member in parts of South Dakota, Wyoming, Nebraska, Colorado, and Kansas, as reported in the literature, is shown in figure 1. Stratigraphic sections of the shale in South Dakota are shown in figure 2.

The Sharon Springs member of the Pierre shale consists predominantly of hard laminated black shale that weathers to fissile silvery gray flakes. Because it is harder than overlying shales in the Pierre, the member forms a prominent scarp around the Chadron anticline and near the Black Hills. When burned, small flakes of the shale give off an oily odor. Fish scales and bone fragments are abundant throughout the unit. The shale contains unusual amounts of selenium (Moxon, Olson, and Searight, 1939) and arsenic (Moxon, and others, 1944).

Bentonite is conspicuous in thick beds near the Chadron anticline and the Black Hills. A 4-foot bed of bentonite near the base of the member in the Chadron anticline section (loc. 4, figs. 1 and 2) is correlated with a 3-foot bed near the base of the member in the Buffalo Gap section (loc. 5, figs. 1 and 2). Other bentonite beds as much as 1.5 feet thick are common

FIGURE 1 MAP SHOWING OUTCROP OF SHARON SPRINGS MEMBER OF PIERRE SHALE, AND LOCATION OF WELLS

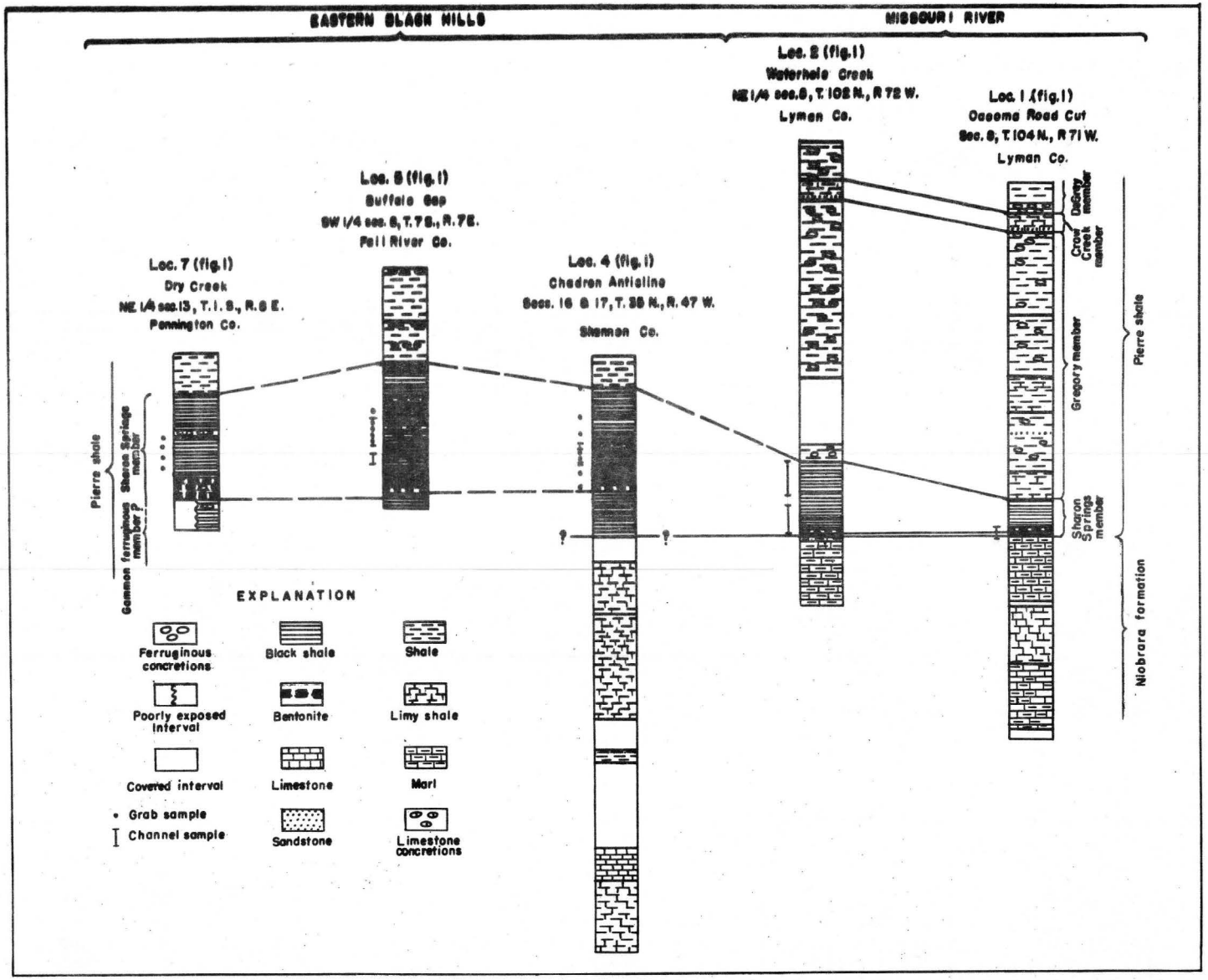

FIGURE 2– SECTIONS OF SHARON SPRINGS MEMBER OF PIERRE SHALE AND ASSOCIATED ROCKS IN SOUTH DAKOTA

in the lower parts of these sections and give the outcrops a banded appearance. At the Dry Creek section (loc. 7, figs. 1 and 2), a 20-foot unit at the base of the Sharon Springs consists of bentonite with only a few thin interbeds of shale. In the Missouri River valley (locs. 1 and 2, figs. 1 and 2), a bed of bentonite 0.3 foot thick lies 1.5 - 2.0 feet above the base of the Sharon Springs and is the thickest bed of bentonite observed in that area. Beds of bentonite a fraction of an inch thick occur in the upper part of the Sharon Springs and also in the upper few feet of the underlying Niobrara formation.

Pyrite is finely disseminated in the shale and weathers to form limonite-stained patches. Efflorescent iron sulfate minerals coat the lower 2 feet of the Sharon Springs member in the Missouri River valley where pyrite is particularly abundant. Some of the pyrite cubes are as much as a millimeter across. Crystals of selenite are abundant on most outcrops.

Limestone concretions as large as 3 feet in diameter and 1 foot thick are abundant at several horizons in the Buffalo Gap section but only a few limestone concretions occur in the upper part of the shale in the Dry Creek section. Some of the limestone concretions are veined with coarsely crystalline calcite. Ferruginous concretions as much as 4 feet in diameter and 2 feet thick are scattered in the lower part of the shale in the Chadron anticline section. The core of the ferruginous concretions appears to be siderite. Similar concretions are abundant in the soft shale units above the Sharon Springs member.

In the Missouri River valley sections, the Sharon Springs member rests directly on marl of the Niobrara formation; the basal contact is sharp and slightly irregular. In the vicinity of the Chadron anticline, along the Nebraska-South Dakota boundary, a few feet of soft shale separates the Sharon Springs from underlying limy shale that seems clearly to be part of

the Niobrara formation. This shale interval thickens to the northwest, near the Black Hills, where the Sharon Springs appears to lie 100 feet or more above the Niobrara formation. According to Cobban (personal communication), this soft shale between the Sharon Springs and the Niobrara may represent the Gammon ferruginous member of the Pierre shale. The Gammon is about 800 feet thick at the north end of the Black Hills and is overlain by the Mitten black shale member of the Pierre with which the Sharon Springs member is correlated (Cobban, 1952).

The upper boundary of the Sharon Springs is placed at a thin marl bed in the Oacoma road cut section (loc. 1, figs. 1 and 2), which is a fresh exposure. This marl was not recognized at the Waterhole Creek section (loc. 2, figs. 1 and 2) about 18 miles to the south, where the exposures are weathered. The upper boundary of the Sharon Springs is not sharply defined near the Chadron anticline and the Black Hills. The hard fissile-weathering shale grades upwards into soft shale that weathers to gumbo. At a few places, thin beds of hard black shale are interbedded with the soft shale 10 to 20 feet above the main mass of the hard shale.

Except for fish scales and bones, the Sharon Springs member is not generally fossiliferous. Scattered well-preserved bones of marine reptiles were found in the exposures at Buffalo Gap and the Chadron anticline. Elias (1931) also noted the abundance of reptilian remains in the Sharon Springs of Kansas. Molds of Inoceramus and other mollusks were found in limestone concretions at Buffalo Gap.

Radioactivity and uranium content

Shale in the Sharon Springs member of the Pierre shale contains as much as 0.01 percent uranium in the Missouri River valley (fig. 3). The shale is markedly radioactive in the vicinity of the Chadron anticline

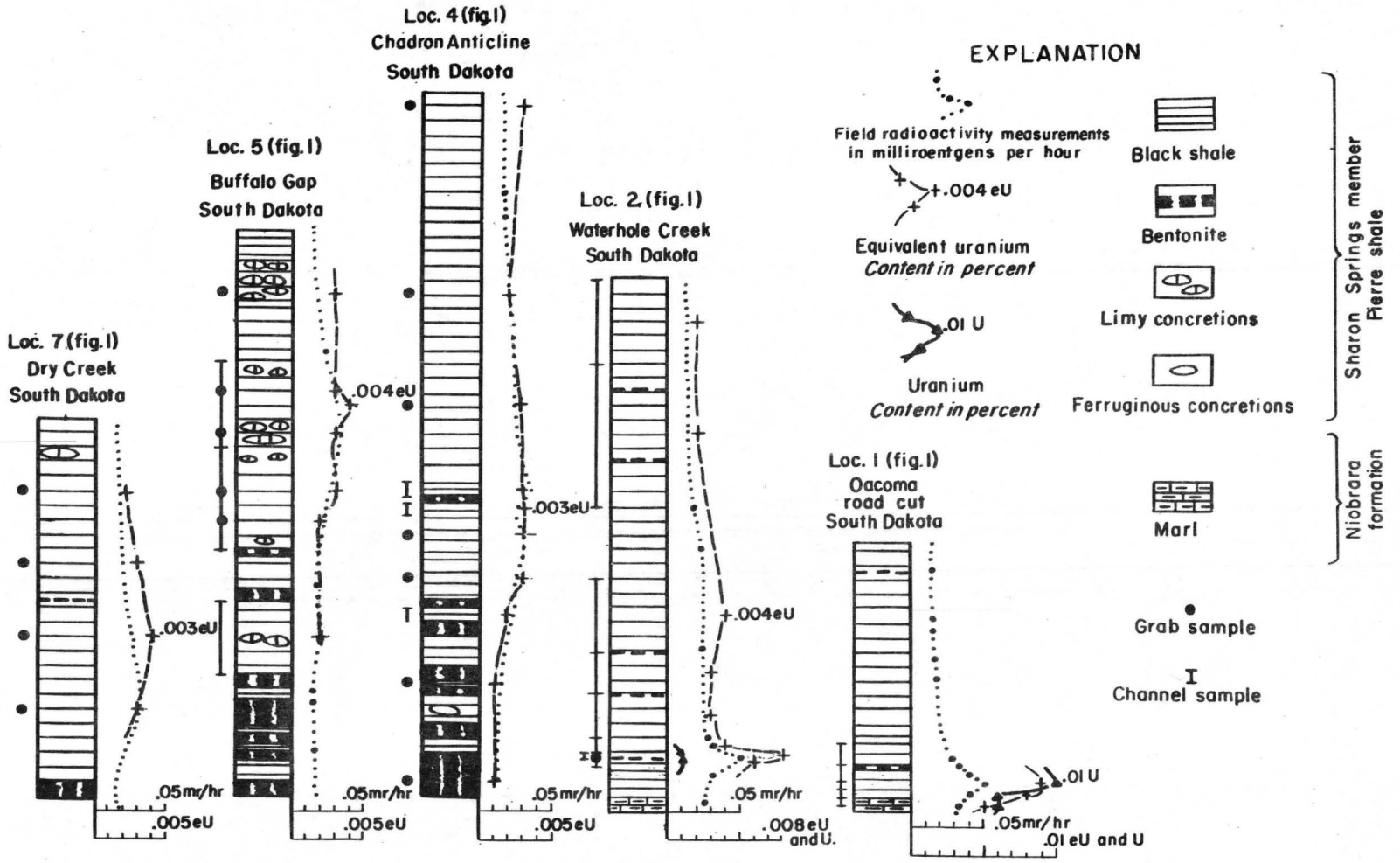

FIGURE 3 — RADIOACTIVITY AND URANIUM CONTENT OF SHARON SPRINGS MEMBER OF PIERRE SHALE SOUTH DAKOTA

and near the Black Hills, compared to the overlying and underlying strata, but the equivalent uranium1/ content there is not more than about 0.004

1/ Equivalent uranium is an expression of the radioactivity of a substance in terms of the amount of uranium in equilibrium with its daughter products that would produce the measured radioactivity.

percent in the sections examined. Gamma-ray logs of wells drilled for oil and gas in western Nebraska and northeastern Colorado show that the Sharon Springs also is radioactive in the subsurface. An equivalent uranium content of about 0.01 percent is indicated by the gamma-ray logs for the shale at some places. The soft shale above the Sharon Springs and the Niobrara formation below the Sharon Springs are estimated to contain less than 0.001 percent equivalent uranium at most places on the basis of scintillation detector readings in the field. However, the upper foot of the Niobrara at the Oacoma road cut section (loc. 1, figs. 1 and 3) contains 0.006 percent uranium.

Surface outcrops

The distribution of uranium and radioactivity in the measured sections is shown in figure 3. Scintillation detector readings were made at points along the sections, including sample points, and the readings plotted as curves. The sections also were scanned with the scintillation detector between the points where readings were recorded. Although the curves are not directly comparable with the continuously recorded curves of gamma-ray logs, the curves show the distribution of radioactivity in a similar way. Curves also were drawn by plotting points representing the equivalent uranium and uranium contents of analyzed samples. Chemical analyses for uranium were made only of samples that yielded an equivalent uranium determination of 0.005 percent or more.

The distribution of uranium in the Missouri River valley sections is different from that near the Black Hills and the Chadron anticline. In the Missouri River valley sections, about 1.5 feet of shale, including about 0.3 feet of bentonite, contains as much as .008 percent equivalent uranium. The 1 to 2 feet of shale between this zone of highest radioactivity and the underlying Niobrara formation contains about 0.006 percent uranium. Above the zone of highest radioactivity, the radioactivity decreases abruptly and scintillation detector readings indicate that the upper part of the Sharon Springs member contains not much more than 0.001 percent equivalent uranium. In the exposures near the Chadron anticline and the Black Hills, no sharp concentrations of radioactivity were detected. The curves show only a very broad peak from 8 to 15 feet above the prominent bentonite beds in the lower part of the shale. The maximum radioactivity of these sections is about 0.004 percent equivalent uranium.

In the Oacoma road cut section (loc. 1, figs. 1 and 3), which is a recent excavation for a highway, the radioactivity expressed as equivalent uranium content and the uranium content are about the same, 0.009 percent and 0.01 percent, respectively. At the Waterhole Creek section (loc. 2, figs. 1 and 3), which is a weathered stream bank exposure, the radioactivity is about 0.008 percent equivalent uranium, but the uranium content is only 0.001 percent. This disequilibrium between the equivalent uranium and uranium contents appears to be the result of leaching of uranium from the shale. Phair and Levine (1953) have shown that UO_3 is preferentially leached from pitchblende by sulfuric acid solutions with a residual concentration of radium salts in proportion to the uranium leached away. Although the form in which uranium occurs in organic shales is unknown, uranium is now being leached from shale outcrops by acid water derived from the weathering of pyrite, and residual concentration of radium salts seems to have

taken place. Water seeping through the shale at the Buffalo Gap section (loc. 2, figs. 1 and 2) has a pH of 3.0 as determined in the field with pH-indicator paper and contains 260 parts per billion uranium. The agreement in magnitude of the equivalent uranium determinations of the shale at the Oacoma road cut with that at the Waterhole Creek section suggests that the equivalent uranium content of weathered shale of the Sharon Springs member may be representative of the uranium content of some of the unweathered shale.

Subsurface data

Many wells drilled for oil and gas in western Nebraska and northeastern Colorado pass through the Sharon Springs member; radioactivity logs of some of these holes are made by oil industry service companies to provide information from which the position in the hole of stratigraphic units and their lithology can be interpreted. The commercial radioactivity log consists of two curves, one that records the gamma radiation emanating from the rocks penetrated in the hole, and another that records the neutrons emitted by the rocks after receiving strong gamma radiation from a source that is introduced down the hole. Only the gamma-ray curve that records the natural radiation of the strata is considered here.

Correlation of the length of deflections on gamma-ray logs with the equivalent uranium content of the strata has not been of much concern in the use of such logs to the oil and gas industry. The fact that organic shale, however, is in general the most radioactive of sedimentary rocks (Russell, 1945) is widely used in interpreting gamma-ray logs (Mercier, 1950). Gamma-ray logs are recorded at various sensitivity scales to permit good differentiation of changes in radioactivity without producing deflections within the width of the recording paper that are too large or too small.

Gott and Hill (1953) studied the relation between equivalent uranium content of the Weber formation from the Rangely oil field in northwestern Colorado and the deflections on gamma-ray logs produced by the Weber. They report that 1 inch of deflection on a 10-inch sensitivity scale is equal to approximately 0.0007 percent equivalent uranium. The application of this calibration factor to the Sharon Springs member is uncertain, and no analyses have yet been obtained of cores of the Sharon Springs from wells for which gamma-ray logs are available. However, use of the calibration factor of Gott and Hill indicates radioactivity in the Sharon Springs that is comparable to the equivalent uranium content of outcrop samples of the shale. The gamma-ray logs in figures 5, 6, and 7 have been converted to a 10-inch sensitivity scale from the several scales on which they originally were recorded. The magnitudes of deflection of the curves are thus directly comparable.

Figure 4 shows the radioactivity of the Sharon Springs member compared to the radioactivity of the underlying Cretaceous formations, of the overlying part of the Pierre shale, and of Tertiary rocks. The Sharon Springs is the most radioactive unit penetrated in the well and is half again as radioactive as thin units in the Dakota sandstone and in the upper part of the Pierre shale.

Gamma-ray logs for three wells drilled in north central Nebraska are shown in figure 5. The Sharon Springs member is most radioactive in the Shell Oil Co. Wildy No. 1 well (loc. 23, figs. 1 and 5). The log indicates that about 12 feet of shale has an equivalent uranium content of about 0.008 percent. In the R. C. Gangwer and Co. Roseberry No. 1 well (loc. 22, figs. 1 and 5), two zones of shale, one about 15 feet thick and the other about 10 feet thick, have an indicated equivalent uranium content of about 0.007 percent. The top of the Niobrara formation is placed by oil industry

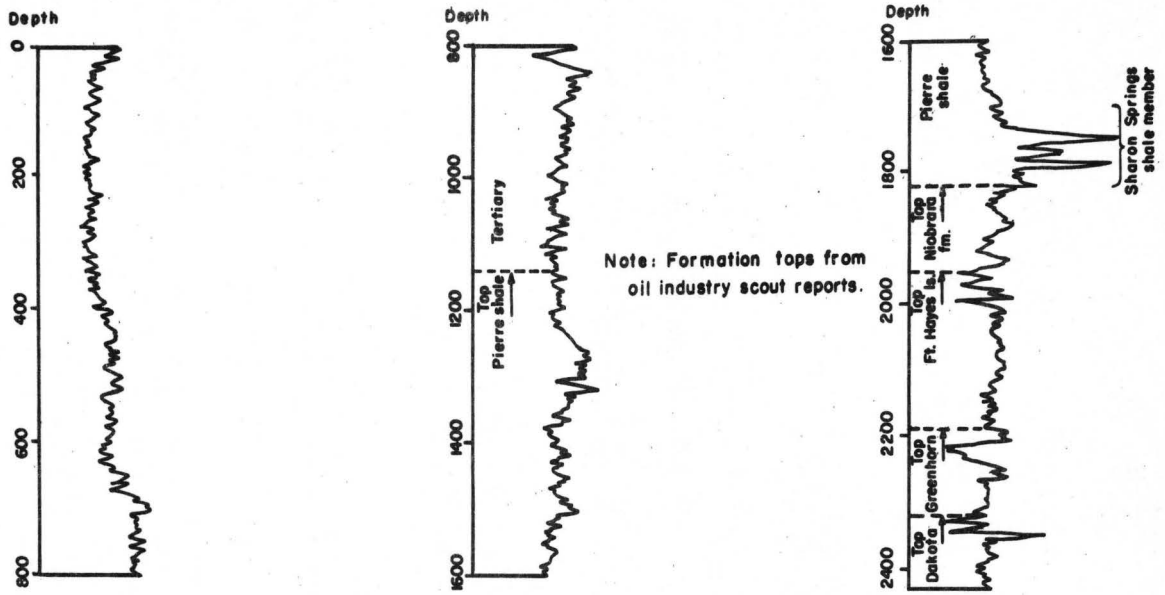

FIGURE 4 - GAMMA-RAY LOG OF R.C. GANGWER AND CO. ROSEBERRY NO. 1 (Loc. 22, fig. 1) CHERRY COUNTY, NEBRASKA SHOWING RADIOACTIVITY OF SHARON SPRINGS MEMBER OF PIERRE SHALE AND ASSOCIATED ROCKS

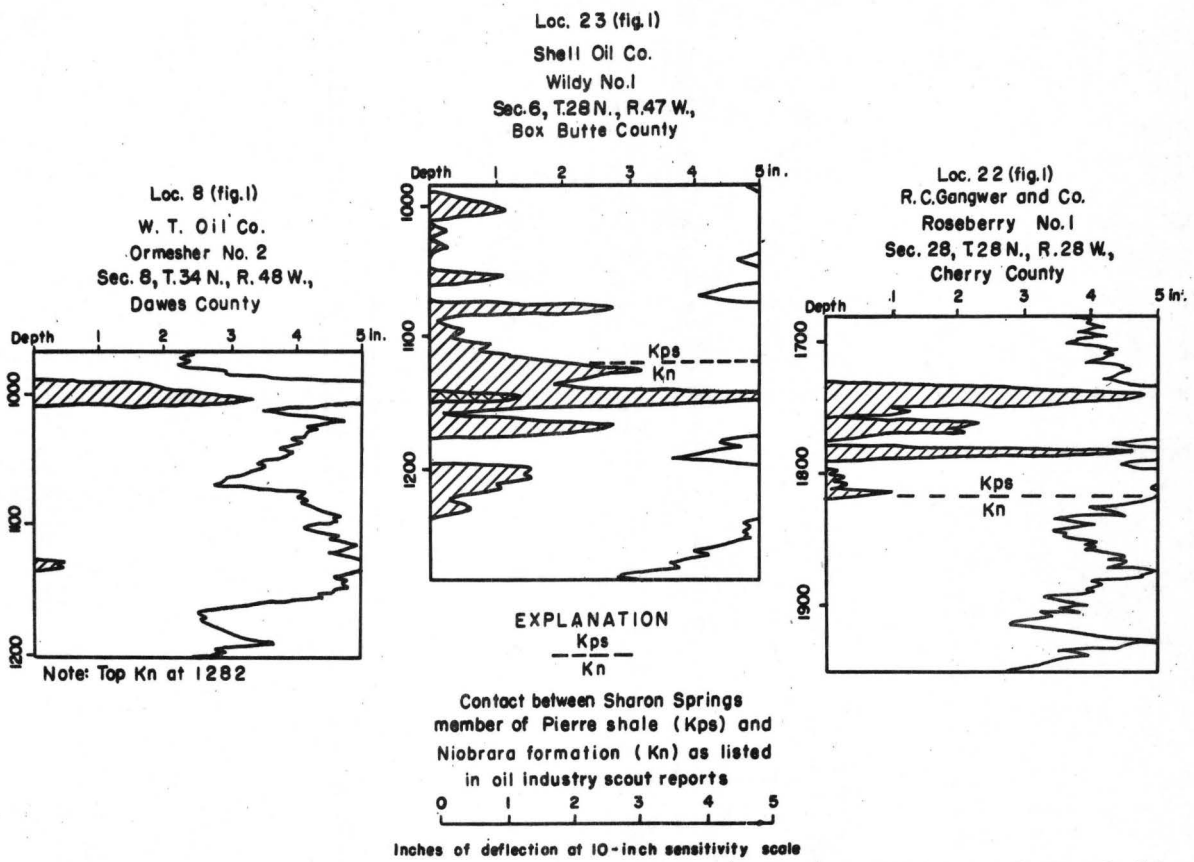

FIGURE 5 - GAMMA-RAY LOGS OF SHARON SPRINGS MEMBER OF PIERRE SHALE IN HOLES DRILLED FOR OIL AND GAS IN NORTH CENTRAL NEBRASKA.

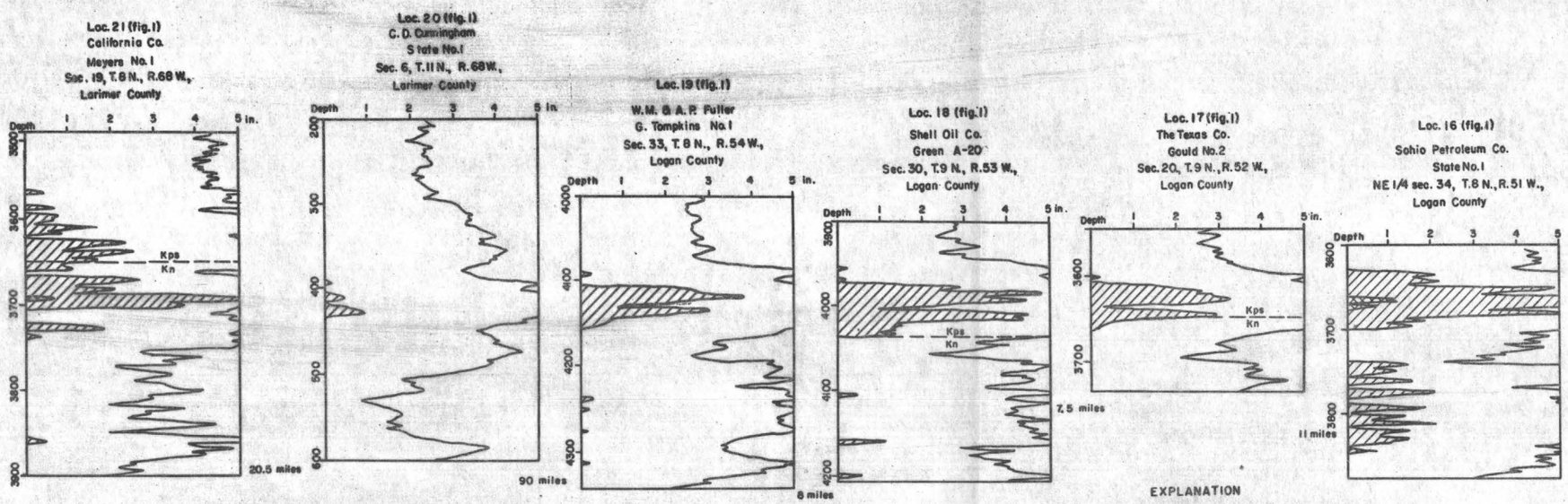

FIGURE 6—GAMMA-RAY LOGS OF SHARON SPRINGS MEMBER OF PIERRE SHALE IN HOLES DRILLED FOR OIL AND GAS IN NORTHEASTERN COLORADO

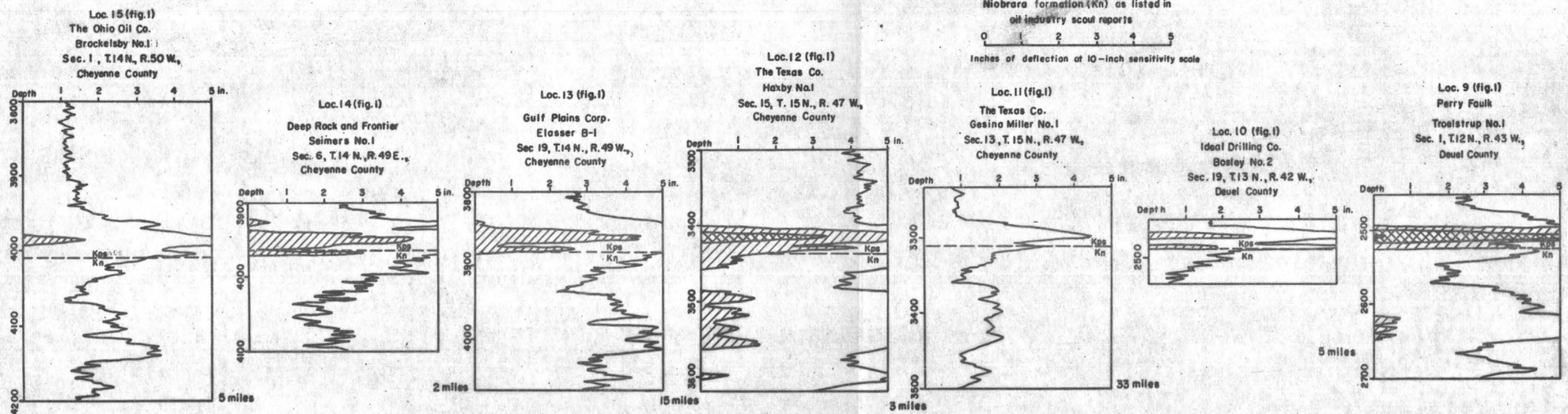

FIGURE 7—GAMMA-RAY LOGS OF SHARON SPRINGS MEMBER OF PIERRE SHALE IN HOLES DRILLED FOR OIL AND GAS IN SOUTHWESTERN NEBRASKA

scout reports at different positions in relation to the main mass of radioactive shale. Inasmuch as only the upper foot or so of unaltered shale or marl of the Niobrara formation has been found to be radioactive on the outcrop, it seems likely that the contact between the Niobrara and the Sharon Springs member has been placed about 50 feet too high in the Shell Oil Co. Wildy No. 1 well.

The Sharon Springs member in the W. T. Oil Co. Ormesher No. 2 well (loc. 8, figs. 1 and 5) is much less radioactive than in the two wells to the south and east. About 20 feet of the shale has an indicated equivalent uranium content of about 0.005 percent. The Ormesher No. 2 well is only 6 miles southwest of the Chadron anticline section (loc. 4, figs. 1 and 3) where about 13 feet of the shale has an average equivalent uranium content of about 0.003 percent. This difference in radioactivity may be real, or it may fall within the margin of error that is probable in the application of a calibration factor based on a lithology other than that typical of the Sharon Springs. The difference in radioactivity between the shale in Ormesher No. 2 well and the wells to the south and east is thought to be significant, however, because the radioactivity of the Sharon Springs increases away from the Black Hills and the apparent distribution of radioactivity in the subsurface is consistent with this relationship.

Figure 6 shows gamma-ray logs of wells drilled in northeastern Colorado. The logs of the four easternmost wells, all in Logan County, have deflections that indicate an equivalent uranium content of about 0.006 to 0.008 percent. The contact between the Sharon Springs member and the underlying Niobrara formation as listed in scout reports seem to be placed about 15 feet higher in the Texas Co. Gould No. 2 well than in the Shell Oil Co. Green A-20 well about $7\frac{1}{2}$ miles to the west. The position of the contact in the Green A-20 well appears to agree with radioactivity data

obtained from outcrops. The contact in the California Co. Meyers No. 1 well (loc. 21, figs. 1 and 6) is not consistent with the position of the contact in other wells and may have been placed as much as 80 feet too high, judging solely from the gamma-ray log.

A regional distribution of radioactivity in the Sharon Springs member is suggested by the data from wells in northeastern Colorado. There, the highest radioactivity is found in the eastern part of the area, and the lowest radioactivity is in the farthest northwest well, C. D. Cunningham State No. 1 (loc. 20, figs. 1 and 6), near the Wyoming-Colorado boundary. The Sharon Springs member has not been recognized in southeastern Wyoming and, presumably, the shale loses its characteristic lithology, of which the organic content is perhaps the most prominent feature. Typical Sharon Springs has been recognized to the south along the Front Range (Griffitts, 1949) and the radioactivity in the Seimers No. 1 well (loc. 21, fig. 1) is comparable to the radioactivity of the shale to the east. Although factors other than organic content could control the amount of uranium in shale, the decrease in radioactivity of the Sharon Springs from Colorado toward Wyoming seems consistent with the recognized association between uranium and the organic content of shales.

Figure 7 shows gamma-ray logs of oil wells in southwestern Nebraska. There, the equivalent uranium content of the shale indicated by the logs ranges from about 0.003 percent in the Texas Co. Gesina Miller No. 1 (loc. 11, figs. 1 and 7) to about 0.01 percent in the Perry Faulk Troelstrup No. 1 (loc. 9, figs. 1 and 7). The thickest, most radioactive unit indicated in the present study of the Sharon Springs member is in the Troelstrup No. 1 well where a thickness of about 30 feet has an indicated equivalent uranium content of about 0.01 percent.

The variation in radioactivity of the shale in wells shown on figure 7 is larger than in the wells shown on figure 6 and does not seem to conform to a geologic pattern. The variation may be real, or it may be the result of differences in logging techniques that are not detectable from the data given on the log. As pointed out by Gott and Hill (1953) such factors as the thickness and radioactivity of the bed, the fluid content of the well, differences in individual detecting instruments, and the rate of movement of the ionization chamber in the well all affect the magnitude of deflection of gamma-ray curves. It is not possible to analyze most of these factors in considering the apparently anomalous low radioactivity of the Sharon Springs in the Gesina Miller No. 1 well.

The position of the contact between the Sharon Springs member and the underlying Niobrara formation as listed in scout reports for wells in southwestern Nebraska is reasonably consistent with respect to features of the gamma-ray log.

Outcrop data and gamma-ray logs indicate that the Sharon Springs member, which is at the base of the Pierre shale throughout most of South Dakota, Nebraska, and Colorado, is conspicuously more radioactive than underlying or overlying Cretaceous rocks. The Sharon Springs thus provides a convenient marker on gamma-ray logs. Study of the Sharon Springs and the regional extent of its radioactivity map help greatly in relating the subdivisions of the Great Plains section of the Pierre shale to the equivalent rocks in the Williston Basin and areas to the west; to the Steele shale of eastern Wyoming, and to the Pierre shale in northeastern New Mexico. Gamma-ray logs suggest that units of the shale as much as 20 feet thick contain an average of about 0.006 percent equivalent uranium over wide areas. The shale appears to be most radioactive in its eastern and southern extent in the areas examined so far.

Content of other elements

The Sharon Springs member of the Pierre shale has a higher average arsenic content than any other Cretaceous sequence investigated by Moxon and others (1944), and its selenium content is notably and consistently high compared to other Cretaceous sequences (Moxon, Olson, and Searight, 1939). The occurrence of selenium in the shale first suggested that the shale might also contain uranium. Trelease and Beath (1949) summarized information on the selenium content of the Phosphoria formation of Permian age and the Morrison formation of Jurassic age in both of which uranium also is found (McKelvey and Nelson, 1950; Cannon, 1954). The uranium and selenium may not have the same kind of origin in the Morrison formation as they do in the Phosphoria. The Niobrara formation, beneath the Sharon Springs, also contains unusually large amounts of selenium and arsenic in South Dakota.

Moxon, Olson, and Searight (1939) report that samples of the Sharon Springs member contain 0.0003 to 0.0024 percent selenium_/. The highest

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_/ Results of analyses for selenium and arsenic were expressed in parts per million by Moxon, Olson, and Searight, 1939, and Moxon and others, 1944. One part per million equals 0.0001 percent.

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average selenium content, 0.0024 percent for a thickness of 20 feet, was found at the base of the Sharon Springs in exposures about halfway between the Oacoma road cut and Waterhole Creek exposures, (locs. 1 and 2, fig. 1). The highest average arsenic content of the Sharon Springs, about 0.0035 percent, was found at the same place at the same horizon (Moxon and others, 1944). The highest uranium content of the Sharon Springs also is in the lower part of the unit in this area. Near the Black Hills, the shale at Buffalo Gap (loc. 5, fig. 1) contains 0.0004 to 0.0013 percent selenium and 0.00047 to 0.0017 percent arsenic.

Samples collected in the present study for radioactivity measurements and uranium analysis were not analyzed for arsenic and selenium. The data cited from South Dakota workers indicate, however, that the Sharon Springs has its maximum known contents of arsenic and selenium in the area of the Missouri River valley, where the uranium content also is highest.

Samples of the Sharon Springs member of the Pierre shale and of the Niobrara formation were analyzed spectrographically and the results are shown in table 1, along with similar analyses of uranium-bearing black shales of Paleozoic age. Arsenic and selenium, as well as many other elements, are not shown in the table because they are present in the shale in amounts too small to be detected by the spectrographic method or are absent. The minimum concentrations of the elements that can be detected are shown in table 2. Table 3 was compiled from table 1 and shows the range in composition for each element in the shales of Cretaceous age (circles) and of Paleozoic age (dots). Each circle or dot represents an analysis and the range in composition of the shales is evident. The crosses in some of the element columns represent the mean values for that element in pelitic rocks as determined by Shaw (1954).

Semiquantitative spectrographic analysis gives bracketed values only. About 25 percent of the individual determinations may be actually one bracket higher or one bracket lower than the bracket reported for the determination (Claude L. Waring, personal communication). The analyses thus cannot be treated statistically, but the presentation of the analyses in a distribution diagram such as table 3 permits a visual interpretation of the concentrations of the elements in the shales.

(Text is continued on page 29.)

TABLE 1. TRACE ELEMENTS COMPOSITION OF BLACK SHALES OF CRETACEOUS AND PALEOZOIC AGES.[1/]

Sample	Be	B	Na	Mg	Al	Si	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Sr	Y	Zr	Mo	Ag	Sn	Ba	La	Yb	Pb	U[3/]
Cretaceous shales																														
95839	1	6	8	8	11	11	9	10	3	7	6	5	4	9	3	5	7	5	4	5	4	4	4	3	-[2/]	7	-	2	5	5 (0.03)
95842	1	7	7	7	10	11	9	8	-	8	6	5	4	9	3	4	4	-	3	5	-	4	5	1	3	7	-	-	3	2 (.001-)
95854	1	5	8	8	10	11	7	8	2	6	5	4	5	9	3	5	5	-	3	4	3	3	3	1	-	6	-	1	3	3 (.003)
95856	1	5	7	7	9	10	7	10	2	6	5	4	6	9	3	5	4	5	3	5	4	3	3	1	-	6	-	2	3	4 (.006)
95857	1	5	8	8	10	11	8	8	3	7	5	4	5	9	3	5	4	4	4	5	3	4	4	1	-	7	-	1	3	3 (.003)
95860	1	5	7	7	10	11	7	7	2	7	5	4	4	9	2	5	4	-	3	4	3	3	4	1	-	7	-	1	3	3 (.004)
Paleozoic shales																														
116642	1	5	7	8	11	11	9	8	3	8	6	5	5	10	3	5	6	-	3	5	4	5	5	-	3	7	5	2	3	3 (.004)
116639	2	6	7	9	11	11	8	9	3	8	7	6	5	10	5	7	7	-	3	5	4	4	7	1	4	7	5	2	5	5 (.01)
7516	1	5	8	8	11	11	9	8	3	7	5	4	4	10	4	5	5	-	3	5	4	3	6	-	3	6	-	2	5	4 (.008)
112836	1	6	8	8	11	11	9	8	3	7	5	5	5	10	5	6	6	5	4	5	4	4	6	-	3	6	-	2	3	4 (.008)
112842A	1	5	8	8	10	11	9	8	3	7	5	5	5	10	4	5	5	5	3	5	4	4	6	-	3	6	-	2	3	4 (.008)
112812	1	5	8	8	10	11	9	8	3	7	5	5	5	10	5	6	5	4	4	4	4	4	6	-	3	6	3	2	4	4 (.008)
114274	1	6	8	8	11	11	9	8	3	7	4	5	5	10	4	5	5	-	4	4	4	4	6	-	3	6	-	2	4	4 (.008)
114280	1	6	8	9	11	11	9	8	3	8	6	5	5	10	4	6	5	5	4	5	4	4	6	1	3	6	-	2	5	3 (.003)

Parts per million	Code
More than 100,000	11
50,000 - 100,000	10
10,000 - 50,000	9
5,000 - 10,000	8
1,000 - 5,000	7
500 - 1,000	6
100 - 500	5
50 - 100	4
10 - 50	3
5 - 10	2
1 - 5	1

1/ Semiquantitative spectrographic analyses by Mona Frank in the Washington Trace Elements Laboratory of the Geological Survey. See Table 2 for minimum detectable concentrations of the elements.

2/ - indicates element looked for but not found. Other elements listed in Table 2 were also looked for but not found in concentrations equal to or larger than the concentrations listed in the table.

3/ Uranium content determined chemically by H. Bivens and J. Siverly for Cretaceous shales and by Joan Smith and Audrey Smith for Paleozoic shales. Figure in parentheses is uranium content in percent. Cretaceous shale samples containing 0.005 percent equivalent uranium or less were not analysed chemically and equivalent uranium values are substituted. Equivalent uranium values are underlined and were determined by S. Furman.

See following pages for description and location of samples.

Table 1. Trace elements composition of black shales of Cretaceous and Paleozoic ages.--Continued

	Description and location of samples
95839	Grab sample of highly radioactive unaltered shale within altered zone in Niobrara formation (Cretaceous), SE¼ sec. 35, T. 36 N., R. 46 W., Shannon County, S. Dak. (loc. 3, fig. 1).
95842	Grab sample of bentonite near base of Sharon Springs member of Pierre shale (Cretaceous), Waterhole Creek section, NE¼ sec. 8, T. 102 N., R. 72 W., Lyman County, S. Dak. (loc. 2, fig. 1).
95854	Channel sample of 0.7 ft. shale and 0.3 ft. bentonite near base of Sharon Springs member, Oacoma Road Cut section, sec. 8, T. 104 N., R. 71 E., Lyman County, S. Dak. (loc. 1, fig. 1).
95856	Grab sample of 0.8 ft. marl at top of Niobrara formation, Oacoma Road Cut section, sec. 8, T. 104 N., R. 71 E., Lyman County, S. Dak. (loc. 1, fig. 1).
95857	Channel sample of 7 ft. of shale in lower part of Sharon Springs member, Buffalo Gap section, SW¼ sec. 8, T. 7 S., R. 7 E., Fall River County, S. Dak. (loc. 5, fig. 1).
95860	Channel sample of 6 ft. of shale in Sharon Springs member directly overlying sample D-95857 (loc. 5, fig. 1).
116642	Grab sample of basal 0.3 ft. of Big Stone Gap shale (Mississippian) of Swartz at northwest edge of town of Big Gap, Wise County, Va. Sample collected by W. Hass.
116639	Grab sample of shale of Devonian age, 4.9 miles south-southeast of Marble Falls, Doublehorn Creek area, Burnet County, Texas. Sample collected by P. E. Cloud, Jr., U. S. Geol. Survey, and Virgil E. Barnes, Texas Bureau Economic Geol.
7516	Channel sample of upper 6.25 ft. Chattanooga shale (Devonian), locality LC-201 (adit), 6 miles east of Smithville, DeKalb County, Tenn. This and following samples collected by T. Kehn.
112836	Core sample of upper 3.23 ft. Chattanooga shale, drill hole YB-36, 8 miles southwest of Smithville, DeKalb County, Tenn.
112842A	Core sample of upper 4.03 ft. Chattanooga shale, drill hole YB-28, 6 miles south of Smithville, DeKalb County, Tenn.
112812	Core sample of upper 5.71 ft. Chattanooga shale, drill hole YB-26, 3 miles southwest of Smithville, DeKalb County, Tenn.

Table 1. Trace elements composition of black shales of Cretaceous and Paleozoic ages.--Continued

Description and location of samples.--Continued

114274 Core sample of upper 6.64 ft. Chattanooga shale, drill hole YB-44, 2 miles west of Manchester, Coffee County, Tenn.

114280 Core sample of upper 4.0 ft. Chattanooga shale, drill hole WR-49, 3 miles southeast of Pikeville, Bledsoe County, Tenn.

Table 2.--Minimum concentrations of the elements detectable by the semi-quantitative spectrographic method, in parts per million. Revised January 13, 1954.

Element	Minimum Concentration	Element	Minimum Concentration	Element	Minimum Concentration
Ag	0.1	Hf	300	Re	400
Al	1	Hg	800	Rh	40
As	100	Ho	10	Ru	80
Au	10	In	4	Sb	100
B	50	Ir	300	Sc	10
Ba	10	K	3,000	Si	50
Be	0.5	La	30	Sm	80
Bi	50	Li	400	Sn	40
Ca	100	Lu	50	Sr	10
Cd	50	Mg	0.3	Ta	1,000
Ce	300	Mo	5	Tb	100
Co	80	Mn	7	Te	800
Cr	6	Nb	10	Th	800
Cs	8,000	Na	100	Ti	5
Cu	0.5	Nd	60	Tl	400
Dy	60	Ni	50	Tm	10
Eu	30	Os	1,000	U	800
Er	30	P	700	V	10
F	800	Pb	10	W	700
Fe	8	Pd	30	Y	30
Ga	40	Pr	100	Yb	3
Gd	60	Pt	30	Zn	80
Ge	10	Rb	70,000	Zr	8

TABLE 3. DISTRIBUTION OF ELEMENTS IN SHALES OF CRETACEOUS AND PALEOZOIC AGES

(Compiled from table 1)

Parts per million	Be	B	Na	Mg	Al	Si	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Sr	Y	Zr	Mo	Ag	Sn	Ba	La	Yb	Pb	U
More than 100,000					⁘ ∘	⁘ ∘																								
50,000 - 100,000					⁘ ∘	∘		∘						⁘		•														
10,000 - 50,000				⁘	∘		⁘ ∘	·						∘ ∘																
5,000 - 10,000			⁘ ∘	⁘ ∘			· ∘	⁘ ∘		⁘ ∘																				
1,000 - 5,000		∘	⁘ ∘	∘			∘	∘		⁘ ∘	·					·	· ∘						·			⁘ ∘				
500 - 1,000		⁘ ∘								∘	⁘ ∘	·	∘			⁘	⁘						⁘			⁘ ∘				
100 - 500		⁘ ∘									⁘+∘	⁘+∘	⁘ ∘		⁘	⁘ ∘	⁘ ∘	⁘ ∘		⁘ ∘		·	· ∘				⁘	⁘ ∘	· ∘
50 - 100											·	· ∘	· ∘		⁘	+∘	∘ ∘	· ∘	⁘ ∘	:+∘	⁘ ∘	⁘ ∘	∘ ∘		·				⁘	⁘ ∘
10 - 50									⁘+∘						⁘+∘		+		⁘+∘		∘ ∘	⁘+∘	∘	∘	⁘ ∘		·		⁘+∘	⁘ ∘
5 - 10	·								∘ ∘						∘						∘ ∘							⁘ ∘		∘
1 - 5	⁘ ∘																							⁘ ∘				∘ ∘		

EXPLANATION: · Analysis of shale of Paleozoic age. ∘ Analysis of shale of Cretaceous age. + Mean content of pelitic rocks (Shaw, 1954)

The trace elements composition of the shales of Cretaceous and Paleozoic ages is quite similar. Significant differences appear to exist only for molybdenum, silver, and tin. Molybdenum and tin appear to be less abundant in shales of Cretaceous age and silver is more abundant in shales of Cretaceous age than in shales of Paleozoic age. Lanthanum appears to be absent in the Cretaceous shales and only sporadically present in the Paleozoic shales. Chromium, cobalt, and copper may be less abundant in shales of Cretaceous age than in shales of Paleozoic age but this is not positively indicated by the data. The uranium content of the shales has a range comparable to that of most of the metallic elements and a somewhat more even distribution within its range. The uranium values are not as well clustered as vanadium, for instance.

Selenium and arsenic are not detectable in low concentrations by the spectrographic method and chemical analyses for arsenic in Paleozoic shales have not been made for the present study. Mr. L. C. Conant reports (personal communication) that five samples of Chattanooga shale contained less than 0.001 percent selenium. The Sharon Springs shale member contains more selenium and arsenic than other Cretaceous shales (Moxon, Olson, and Searight, 1944, and Moxon and others, 1939). The Sharon Springs contains more selenium than one sampled section of the Chattanooga shale and may contain more arsenic as well.

The shales of Cretaceous and Paleozoic ages, taken as a group, do not differ significantly in their content of scandium, vanadium, chromium, cobalt, gallium, strontium, zirconium and lead from the averages reported by Shaw (1954) for 67 samples of a black shale formation of Devonian age and its metamorphic equivalents. The shales of Cretaceous and Paleozoic age appear to contain significantly more copper than Shaw's average and they may contain more nickel. (See table 3.) The shales of Paleozoic age

may contain more cobalt, strontium, and zirconium than Shaw's average and the shales of Cretaceous age may contain less chromium than Shaw's average. The shale investigated by Shaw is not known to be uranium-bearing. It is suggested that the uranium-bearing shales may differ from average shales chiefly in their content of uranium, organic material, and possibly copper, nickel, selenium, and arsenic.

Origin of uranium in shale

In their analysis of the characteristics of marine uranium-bearing rocks, McKelvey and Nelson (1950) pointed out that the uranium-bearing black shales generally are black or dark, rich in organic matter and sulfides, low in carbonate, and relatively thin. The Sharon Springs thus resembles other known uranium-bearing black shales. The fact that uranium is concentrated only in shales having a high organic content suggests that the uranium possibly is chemically combined with organic compounds. Assuming this to be so, Swanson (1953) considered some of the factors operating at the time of deposition that would affect the degree of concentration of uranium in a black shale. These factors include length of time that a depositional surface was exposed on the sea bottom and the proportion of organic material present on the surface. Sufficient data are not yet available on the Sharon Springs member to do more than mention some aspects of the known distribution of uranium in the shale and their possible implications as to the origin of the uranium.

The highest uranium content of the shale, and probably the highest selenium and arsenic contents also, is in the Missouri River valley area. At the north end of the Black Hills the Sharon Springs member and its correlative, the Mitten black shale member of the Pierre shale, are separated from the Niobrara formation by the 800-foot thick Gammon ferruginous

member. The shale of the Gammon member disappears to the south and east and the Sharon Springs rests directly on the Niobrara formation. The Gammon member of the Pierre either was never deposited in the Missouri River valley or else was deposited there and eroded away prior to the deposition of the Sharon Springs. In either situation, the Sharon Springs was deposited on the surface of a disconformity in a region east and south of the Black Hills. Abnormal amounts of elements could have been concentrated by weathering on this surface prior to the deposition of the Sharon Springs. The elements then would be made soluble under the acidic and reducing conditions in which the pyrite-rich and organic-rich Sharon Springs was deposited and made available for absorption by the organic material or combination with it. This could explain the generally lower uranium content of the shale where it conformably overlies the Gammon member.

The Sharon Springs member contains an aggregate of about 20 feet of bentonite in exposures near the Black Hills but less than a foot of bentonite in the Missouri River valley exposures. Volcanic ashes are considered to be sources of small amounts of many different elements. The greater metal contents of the Sharon Springs are found, however, where the bentonites are least prominent. The volcanic ash deposited near the Black Hills probably would have released much of its metal content, including uranium, in the early stages of its alteration to bentonite and thus could have increased the metal content of the waters of the Sharon Springs sea. The greatest concentration of metals then would have taken place where the deposition of the Sharon Springs was slowest, assuming good circulation within the Sharon Springs sea. The Sharon Springs is 2 to 10 times as thick near the Black Hills as it is in the Missouri River valley. If these beds were deposited in about the same time interval, then the thinner sections of shale could be expected to contain the most uranium. That this simple

relationship is only partly the explanation is indicated by the fact that the highest known uranium content in surface outcrops of the Sharon Springs member in the Missouri River valley is in the basal few feet of the shale and is not distributed uniformly through the shale section. The overlying shale at most places is not greatly different in its content of uranium or other metals from the thick shale sections near the Black Hills.

Data from the gamma-ray logs indicate considerable differences in radioactivity for the shale from place to place. Although the distribution of radioactivity appears to make an areal pattern, the most that can be inferred from the logs now is that the most highly radioactive units are very thin compared to adjacent beds of lower radioactivity.

ALTERED SHALE

Throughout large areas of the Great Plains in Nebraska and South Dakota, the predominantly shaly marine strata of late Cretaceous age are overlain by tuffaceous sandstone and claystone of the Chadron formation at the base of the White River group of Oligocene age. A conspicuous feature of the unconformable contact between these two sequences is a brightly colored zone of altered Cretaceous shales beneath the Chadron.

The zone is as much as 50 feet thick and is present everywhere except where channels were cut into it prior to the deposition of the Chadron formation. The altered shale ranges in color from nearly white through shades of yellow to orange, brown, and purple. Red streaks are common and oxides of iron coat joints and bedding planes. Selenite is abundant on the outcrops. The intensity of the alteration decreases downwards from the base of the overlying Chadron formation and the altered shale grades into unaltered shale along the base of the zone. Alteration extends deeper along joint planes at many places. The base of the zone is irregular.

From place to place tongues of unaltered shale extend upwards into the altered shale at small angles to the general base of the zone. Irregularly shaped masses of unaltered shale are completely surrounded by altered shale in the lower part of the zone. Figure 8 shows the general relation of the altered zone to the unaltered shale beneath, and to the White River group above.

Radioactivity and uranium content

At most places, the lower several feet of the altered zone is more radioactive than the top of the zone or than the unaltered shale beneath. In general, the difference in radioactivity is very small, about 0.01 milliroentgens per hour. The altered zone developed on different parts of the Pierre shale was examined at many places in Nebraska and South Dakota. Altered shale of the Niobrara formation was examined only at two places, but at both places the difference in radioactivity was much higher than where the zone was developed on Pierre shale, being as high as 0.4 milliroentgens per hour. Since these localities (locs. 3 and 6, fig. 1) are about 50 miles apart, it seems unlikely that the occurrence of high radioactivity contrasts in the altered zone developed on the Niobrara are merely coincidental. The radioactivity and uranium are concentrated in tongues of unaltered shale in the lower part of the altered zone and in masses of unaltered shale isolated in the lower part of the altered zone. Figure 9 shows the distribution of radioactivity and uranium in the altered zone developed on the Niobrara.

The highest radioactivity was detected at a locality approximately in sec. 2, T. 35 N., R. 46 W., Shannon County, South Dakota, where the tip of an isolated mass of unaltered shale of the Niobrara formation gave scintillation detector readings of 0.44 milliroentgens per hour. A sample from

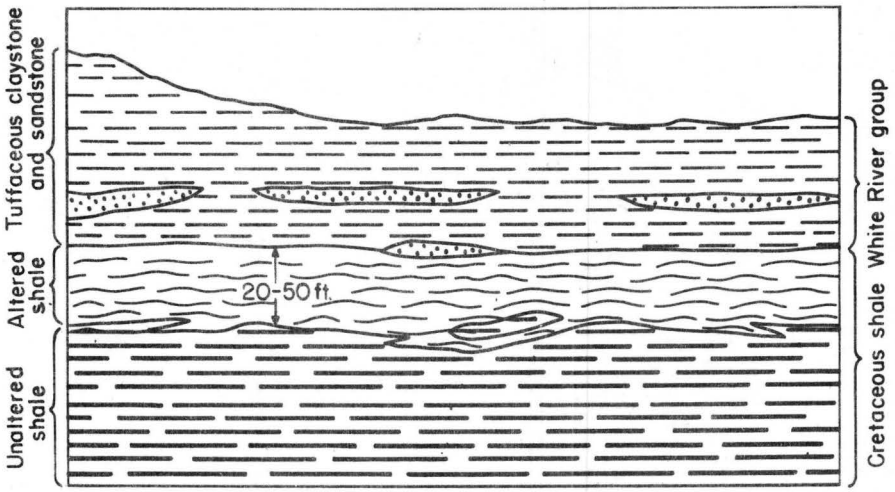
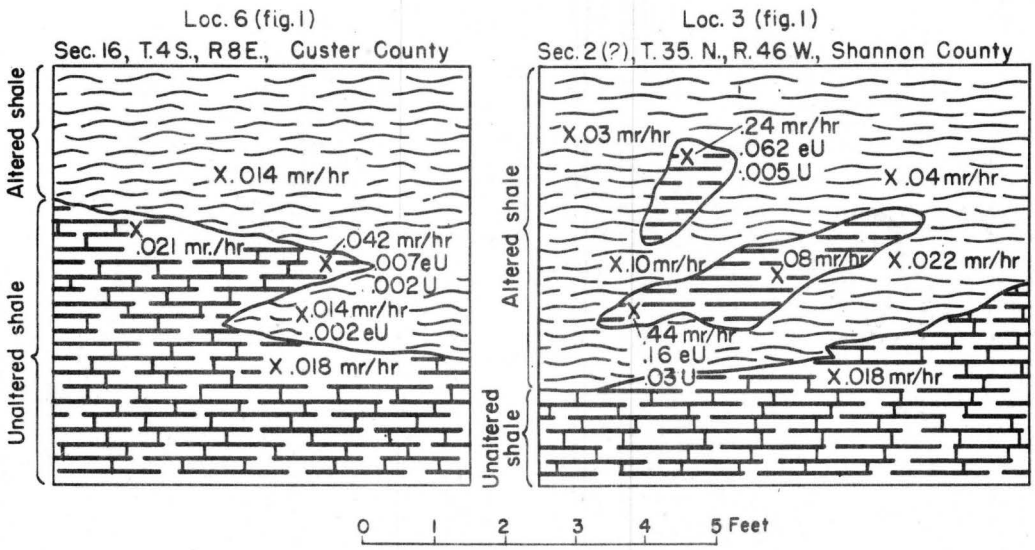


FIGURE 8—DIAGRAMATIC SKETCH SHOWING RELATION OF ALTERED AND UNALTERED CRETACEOUS SHALE BENEATH WHITE RIVER GROUP OF LOCALITIES IN SOUTH DAKOTA AND NEBRASKA.



E X P L A N A T I O N

X—Location of sample and scintillation detection reading.

.04 mr/hr—Radioactivity in milliroentgens per hour.

.007 eU—Equivalent uranium in percent.

002 U—Uranium content in percent.

FIGURE 9—FIELD SKETCHES SHOWING DISTRIBUTION IN VERTICAL SECTION OF RADIOACTIVITY AND URANIUM AT BASE OF ALTERED MARL OF NIOBRARA FORMATION AT LOCALITIES IN CUSTER AND SHANNON COUNTIES, SOUTH DAKOTA.

this spot contained 0.16 percent equivalent uranium and 0.03 percent uranium. Other samples shown in figure 9 are similarly out of balance. The general insolubility of radium sulfate suggests that uranium has been leached away from the present exposures within the past few thousand years and that sufficient uranium was once present to support the present radioactivity. It is possible, of course, that radium salts were transported to their present positions. A near by source of uranium is indicated, however, since sulfate ions are abundant in many ground waters in the Great Plains region and radium sulfate would be precipitated at the first place that a solution containing excess radium ions was mixed with waters high in sulfate.

The sample of unaltered shale within the altered zone that contained 0.03 percent uranium was analyzed spectrographically and is included in table 1 (sample 95839). The metal content of the unaltered shale within the zone is similar to that of the shale of the Sharon Springs member and a sample of unaltered marl of the Niobrara formation (sample 95856). Copper, silver, and lead, however, are present in conspicuously greater amounts than in the Sharon Springs or in the unaltered marl; gallium, scandium, yttrium, and ytterbium are present in slightly greater amounts than in most of the other samples analyzed. The concentration of these metals in the unaltered shale within or at the base of the altered zone may have resulted from the same processes by which the uranium was concentrated.

Origin of uranium in altered zone

The origin of the uranium and other metals is involved with the origin of the altered zone. Ward (1922) thought the color of the altered zone, which he called the Interior formation, was the normal color of a rock unit

overlying the Pierre shale. Wanless (1923) concluded that Ward's Interior formation was the result of weathering of Cretaceous shales prior to the deposition of the White River group. Russell (1928) inferred desert conditions immediately prior to White River time on the basis of the red colors in the altered zone and the presence at a few places in the zone in central South Dakota of pebbles believed by him to have been faceted by wind. Clark (1937) traced the Red River valley, an ancient channel from which the altered zone had been eroded before being filled with the basal sediments of the White River group. Altered shale has been reworked into the basal White River strata at a few places. Thus it seems clear that the altered zone in large part is pre-Oligocene in origin. The zone has been affected also by ground water in subsequent geologic time, since the altered zone is more porous and permeable than unaltered shale.

The source of the uranium concentrated in the unaltered marl within the altered zone where it is developed on the Niobrara formation may have been the marl that was altered in the formation of the zone. Unaltered marls of the Niobrara formation 5 to 10 feet below the base of the altered zone are not radioactive, however, where tested at the localities shown on figure 9 and the effectiveness of the unaltered marls as a source of the uranium cannot be completely evaluated.

The Chadron formation of the White River group contains secondary uranium minerals in the Big Badlands near Scenic, S. Dak. (Moore and Levish, 1954) and in the Slim Buttes near Buffalo, S. Dak. (Gill and Moore, 1954). In northwestern South Dakota, uranium-bearing lignites are found only where the lignite beds are or have been closely overlain unconformably by the tuffaceous White River group or by tuffaceous strata of the Arikaree formation of Miocene age. This close association has been interpreted to be genetic, the uranium in the lignite having been leached from the tuffaceous

strata and carried to the lignite by ground water, as summarized by Miller and Gill (1954). This moderately widespread occurrence of uranium associated with the White River group suggests that the rocks of the group contain uranium throughout most of their areal extent and that at least some of the uranium in the altered zone developed on marls of the Niobrara formation may have been derived from the White River group.

The Niobrara formation is highly calcareous, but most of the shales of the Pierre formation are conspicuously noncalcareous. The fact that the Niobrara formation seems to show a higher radioactivity at the base of the altered zone than does the Pierre may be related to the more calcareous nature of the Niobrara. Perhaps calcareous shales retain more of the uranium leached from the White River or other source than do noncalcareous shales. The organic material left in the thin tongues and isolated masses of unaltered shale probably played a part in fixing the uranium. An understanding of the geologic factors that controlled the development and subsequent history of the altered zone and the accumulation of uranium in it might lead to the discovery of uranium-bearing shale deposits of commercial interest.

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USGS - TEI Report 298

Part II

POSSIBILITIES FOR URANIUM IN CRETACEOUS SHALES

The Sharon Springs member of the Pierre shale is a uranium-bearing shale comparable to the Chattanooga shale in uranium content and areal extent. Thin beds of the Sharon Springs member contain as much as 0.01 percent uranium in the surface exposures examined so far. Gamma-ray logs from wells drilled for oil and gas in southwestern Nebraska suggest that as much as 20 feet of shale contains 0.01 percent equivalent uranium, although the calibration of the gamma-ray logs has not yet been based on analyzed core samples of Sharon Springs.

The Sharon Springs member contains more uranium in exposures along the Missouri River valley in central South Dakota than it does near the Black Hills. In central South Dakota, the Sharon Springs rests disconformably on the underlying Niobrara formation. The disconformity between the two formations extends south into Nebraska and then southwest into Kansas and northeastern Colorado, and gamma-ray logs indicate a relatively high level of radioactivity throughout most of this large area. Near the Black Hills, and to the north, the disconformity is represented by a wedge of shale called the Gammon ferruginous member of the Pierre shale, and the radioactivity of the Sharon Springs is low although still conspicuous compared to overlying and underlying strata. The Sharon Springs also is thicker near the Black Hills than it is in the Missouri River valley and the area to the south and west. The lithology of the shale appears to be uniform

although exposures near the Black Hills contain more beds of bentonite than elsewhere. The shale contains a large suite of other trace elements among which arsenic, selenium, and vanadium are conspicuous.

Only reconnaissance studies have been made so far, but the thickness of the shale and its relation to the underlying Niobrara formation are geologic features that may be related to the uranium content of the shale. Further study of these features has promise of leading to an understanding of the origin of uranium in black shales and the geologic conditions that controlled the distribution and concentration of the uranium in the shale. This understanding will aid in locating areas where the shale has the highest uranium content. The Sharon Springs member has promise of being equal to or better than the Chattanooga shale as a low-grade source of uranium and thus merits the additional study which is underway.

The zone of altered shale beneath the White River group may contain small deposits of uranium-bearing shale of possible commercial interest where the altered zone is developed on marls of the Niobrara formation. Unaltered marl pods incorporated in the lower part of the altered zone contain as much as 0.03 percent uranium and 0.1 percent equivalent uranium. The equivalent uranium determinations are believed to represent the uranium content of the unaltered marl pods prior to leaching of the present outcrops. The potentialities of the altered zone can be judged only after an understanding of the origin and history of the zone has been gained. Such studies are underway.