# Uranium in Sharon Springs Member of Pierre Shale South Dakota and Northeastern Nebraska

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GEOLOGICAL SURVEY BULLETIN 1046-R

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# Uranium in Sharon Springs Member of Pierre Shale South Dakota and Northeastern Nebraska

*By* ROY C. KEPFERLE

CONTRIBUTIONS TO THE GEOLOGY OF URANIUM

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CONTRIBUTIONS TO THE GEOLOGY OF URANIUM

# URANIUM IN SHARON SPRINGS MEMBER OF PIERRE SHALE, SOUTH DAKOTA AND NORTHEASTERN NEBRASKA

By Roy C. Kepferle

ABSTRACT

A study of the uranium content of the Sharon Springs member of the Pierre shale in South Dakota and northeastern Nebraska was made in 1954. More than 300 samples of this black organic marine shale of Late Cretaceous age from 30 exposures and 2 core holes show that the uranium content averages about 0.0015 percent throughout the region studied. The most uraniferous parts are about 3 inches thick and contain as much as 0.025 percent uranium. The richest sample comes from an exposure along the Missouri River in Nebraska and is thought to be secondarily enriched by weathering. Water from the Sharon Springs member generally has a pH of less than 4, and 7 samples from seeps and springs contained from 7 to 780 parts uranium per billion.

The thickness of the Sharon Springs member ranges from 1 to 42 feet in exposures along the Missouri River and is as much as 106 feet on the eastern flank of the Black Hills. Gamma-ray logs and published lithologic logs from holes drilled for oil and gas indicate that rocks equivalent to the member extend northward from central South Dakota into North Dakota and that the radioactivity decreases northward.

Along the Missouri River in eastern South Dakota, the noncalcareous Sharon Springs member lies on the calcareous Niobrara formation. At some places the two units appear conformable; at others a disconformity separates them. Along the southeast flank of the Black Hills and northward to North Dakota, the Sharon Springs rests conformably on rocks equivalent to the Gammon member of the Pierre shale.

The upper boundary of the Sharon Springs member is poorly defined except at a few exposures along the Missouri River where a marly zone marks the base of the overlying Gregory member of the Pierre. Elsewhere, the upper boundary is placed at the change from the darker color and steeper slopes of the Sharon Springs member to the lighter color and gentler slopes of the overlying Gregory member.

Semiquantitative spectrographic analyses of 82 samples of black shale from the Sharon Springs member indicate that the molybdenum, iron, and phosphate contents tend to vary directly with the uranium content, whereas the aluminum, gallium, boron, titanium, magnesium, and sodium contents vary inversely with the uranium content.

The uranium in the Sharon Springs member is thought to have been emplaced during or shortly after the deposition of the shale.

## INTRODUCTION

A reconnaissance search for uranium in the Sharon Springs member of the Pierre shale was made by the author during the summer of 1954 in southern South Dakota and northeastern Nebraska in continuation of the investigation begun in 1953 by H. A. Tourtelot (1956). The main objective of the search was to determine the areal and stratigraphic distribution of uranium in the black shale of the member as a means of evaluating it as a source of uranium. Secondary objectives were to determine controls that influenced the occurrence of uranium in the shale that could be used as a guide in future prospecting and to examine the overlying and underlying rocks for uranium.

The investigation by Tourtelot (1956), using surface exposures and gamma-ray logs of holes drilled for oil, showed that the Sharon Springs member is radioactive in the subsurface over an area extending through western Kansas and Nebraska, northeastern Colorado, and southern South Dakota. Gamma-ray logs examined during the present study show that the radioactive shale extends through most of South Dakota into northeastern Nebraska and most of North Dakota, as well.

Simultaneously with the present study, other investigations of the Sharon Springs member were made in the Chadron area of Nebraska and South Dakota by R. J. Dunham (1955, p. 164) and in eastern Colorado and western Kansas by E. R. Landis (1959). The search was made by the U. S. Geological Survey on behalf of the Division of Raw Materials of the U. S. Atomic Energy Commission.

The cooperation of Dr. E. P. Rothrock, State geologist of South Dakota at the time this investigation was made, and L. B. Underwood and R. E. Fisher of the U. S. Corps of Engineers, as well as the many farmers and ranchers of the area, is greatly appreciated. The writer also wishes to thank L. B. Riley and J. D. Vine of the Geological Survey for their assistance in interpreting the spectrographic data. Geological Survey analysts, who made chemical and spectrographic analyses and to whom the writer is grateful, include W. Tucker, J. Budinsky, A. Sweeney, S. Furman, R. Cox, J. Schuch, M. Finch, H. Lipp, Mona Frank, Roosevelt Moore, G. Burrow, N. M. Conklin, R. Dufour, and T. Miller.

## METHOD OF INVESTIGATION

In order to determine the areal and stratigraphic distribution of uranium within the Sharon Springs member of the Pierre shale, stratigraphic sections were measured in detail at 15 localities along the Missouri River valley in South Dakota and Nebraska and at 15 localities along the eastern and southern flanks of the Black Hills in

South Dakota. The surface was scanned with a scintillation counter, and anomalously radioactive zones were sampled. Most of the route traveled between outcrops was scanned with a jeep-mounted scintillation counter. At a few localities channel samples were taken throughout the complete thickness of the Sharon Springs member; at other localities, complete channel samples were taken only through the most radioactive zones. A few grab samples of concretions, wood fragments, and other material anomalous to the general lithologic character of the member were taken from the overlying and underlying rocks. In all, more than 300 samples were collected. Most of the samples showing radioactivity of 0.003 percent or less equivalent uranium (eU)[1] were not analyzed chemically for uranium content. Many of the samples were analyzed semiquantitatively by spectrographic methods in the laboratories of the U. S. Geological Survey. A statistical study of the analyses of 82 samples of shale was made to determine factors which may affect the distribution and concentration of the uranium.

## SHARON SPRINGS MEMBER OF THE PIERRE SHALE

### STRATIGRAPHIC SETTING

The Sharon Springs member was first described by Elias (1931) as the basal member of the Pierre shale in western Kansas. Searight (1938) applied the name to the black organic shale at the base of the Pierre shale of South Dakota but included overlying gray shales since named the Gregory member by Gries and Rothrock (1941), who restricted the Sharon Springs member to the fossil fish bearing shale that overlies the Niobrara formation and underlies the Gregory member in central South Dakota. The better exposures along the Missouri River are referred to by Searight (1937), Gries and Rothrock (1941), Gries (1942), Petsch (1946), and Simpson (1955). Many of the exposures have since been inundated behind Fort Randall Dam. On the southeastern flank of the Black Hills the Sharon Springs member does not lie on the Niobrara formation but apparently on a wedge edge of rocks lithologically similar to the Gammon ferruginous member of the Pierre shale described by Rubey (1930) in northeastern Wyoming. On the northeastern flank of the Black Hills the Sharon Springs member cannot be distinguished from the Gammon ferruginous member.

In the present report the Gammon ferruginous member is referred to as the Gammon member, inasmuch as equivalent rocks on the eastern and southern flanks of the Black Hills lack the abundant ferrugi-

[1] Percent equivalent uranium is an expression of the radioactivity in terms of the amount of uranium which, in equilibrium with its daughter products, would produce that amount of radioactivity.

nous concretions found in the type area described by Rubey. Spivey (1940) and Gries (1942) discuss many of the better exposures along the southeastern flank of the Black Hills. The generalized stratigraphic relationships of the Sharon Springs member to other rocks in the region are shown in table 1; the detailed relationships to overlying and underlying rocks are shown by four measured sections (fig. 85).

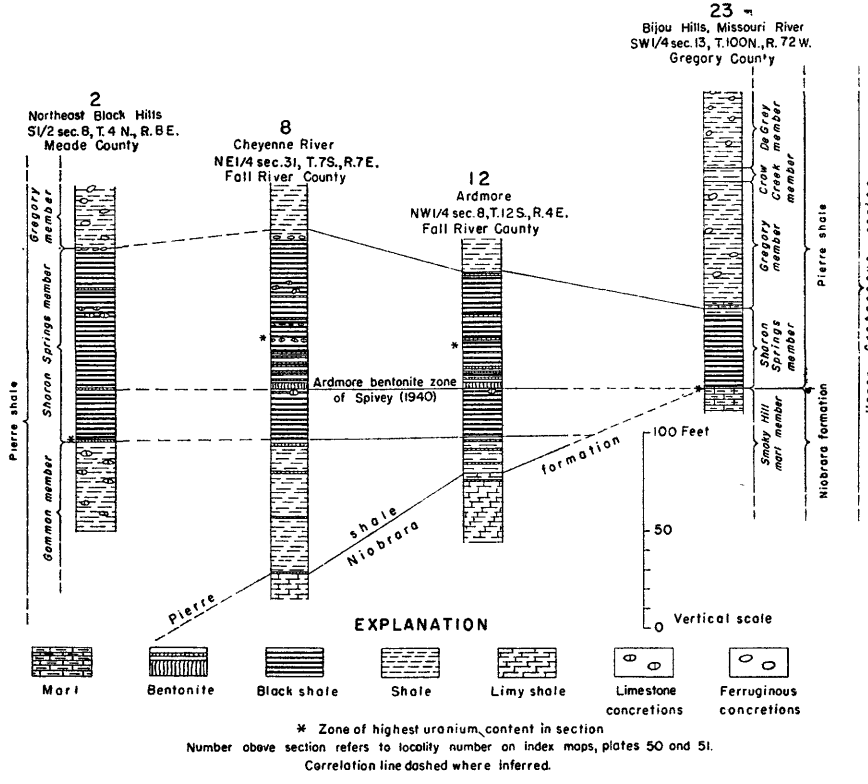


**FIGURE 85.—Stratigraphic sections showing correlation of Sharon Springs member of the Pierre shale and associated rocks in South Dakota.**

### LOWER BOUNDARY OF THE MEMBER

At most places along the Missouri River the noncalcareous Sharon Springs member appears to lie conformably on the calcareous Niobrara formation. The two can be readily differentiated by their calcium carbonate content. Test holes at the Oahe Dam site near Pierre show an interfingering of the calcareous and noncalcareous lithologies. At other exposures along the Missouri River, however, a disconformity between the two is indicated by a "trashy" zone consisting of a lag concentrate of fish bones, scales and scutes, plant debris, and some silt.

The Gammon member of the Pierre shale, which lies beneath the basal Sharon Springs member in southwestern South Dakota, pinches out between the Black Hills and the Missouri River. At Ardmore (loc. 12, fig. 85 and pls. 51 and 52) 15 feet of shale and bentonite, thought to be equivalent to the Gammon member, lie conformably between the Sharon Springs member above and the Niobrara formation below. Southeast of Hot Springs at the Cheyenne River (loc. 8, fig. 85 and pls. 51 and 52) this shale is 67 feet thick. The Gammon member is 800 feet thick at the north end of the Black Hills (Cobban, 1952).

### UPPER BOUNDARY OF THE MEMBER

The upper boundary of the Sharon Springs member is locally clearly defined by the basal marly zone of the overlying Gregory member of the Pierre shale. However, where the marly zone is absent, the two members can be differentiated by the change from the darker color and steeper slopes of the Sharon Springs to the lighter color and gentler slopes of the overlying Gregory member. The upper contact of the Sharon Springs is marked at some places by a thin bentonite bed or a layer of concretions, most of which are gypsiferous claystone. A fairly continuous 3-inch bed of ankerite, goethite, and limonite, containing more than 10 percent manganese, occurs at the upper contact at locality 2 (fig. 85 and pls. 51 and 52).

### ORGANIC CONTENT

An analysis made by W. Tucker, J. Budinsky, and A. Sweeney of a sample of the shale near the Fort Randall Dam (loc. 25, pl. 50) shows 10.3 percent organic matter, from which about 4 gallons of oil per ton of shale may be distilled. A sample from locality 21 (pl. 50) yields 8 gallons of oil per ton. The organic matter and sulfur in the shale are sufficient locally to ignite spontaneously, baking the shale to a brick-red clinker (loc. 9, pl. 51, and locs. 20 and 23, pl. 50). Much of the organic matter is macerated plant material, although two large coalified wood fragments were found (loc. 14, pls. 51 and 52, and loc. 25, pl. 50).

### FOSSILS

Scales belonging to a chirocentrid fish, *Portheus* or *Icthyodectes*, and teeth and scales of the teleost fish, *Enchodus*, from locality 23 (pl. 50) were identified by D. H. Dunkle of the U.S. National Museum. Mr. Dunkle also referred parts of a lower left jaw from locality 14 (pl. 51) to the mosasaur genus *Clidastes*, because of their similarity to specimens of *Clidastes westii* described from the Sharon Springs member of the Pierre shale in western Kansas.

Marine mollusks from the limestone concretions directly below the Ardmore bentonite zone of Spivey (1940) at locality 9 (pls. 51 and 52)

TABLE 1.—*Generalized section of Late Cretaceous and younger formations between the east side of the Black Hills and the Missouri River, S. Dak.*

| System | Series | Group, formation, member | | Thickness (feet) | | General characteristics |
|---|---|---|---|---|---|---|
| Quaternary | Recent and Pleistocene | | | 0–100 | | Loess and dune sand; terrace deposits and alluvium along stream channels. |
| Tertiary | Pliocene | Ogallala formation | | 0–100+ | | Siltstone and sandstone, tuffaceous, and local beds of volcanic ash and conglomerate; fluviatile and eolian. |
| | —Unconformity— | | | | | |
| | Miocene | Arikaree formation | | 0–1150 | 0–500 | Siltstone and sandstone, tuffaceous, whitish- to greenish-gray or pinkish-gray, and a few thin beds of conglomerate and volcanic ash; fluviatile. |
| | —Unconformity— | | | | | |
| | Oligocene | White River group: Brule formation | Whitney member of Schultz and Stout, 1938 | | 0–200 | Siltstone and sandstone, tuffaceous, greenish- to whitish-gray, and local volcanic ash beds; fluviatile. |
| | | | Orella member of Schultz and Stout, 1938 | | 0–300 | Siltstone and fine-grained sandstone interbedded with some claystone, yellowish-gray to pinkish-gray; member is calcareous in part; fluviatile. |
| | | Chadron formation | | | 0–190 | Claystone, montmorillonitic, greenish-gray to dark-gray, massive in part; interbedded with sandstone and siltstone and containing local conglomerate channels. Fresh-water limestone beds occur near the top; sandstone is common near the base; fluviatile. A basal conglomerate is widespread near the Black Hills. |
| | —Unconformity— | | | | | |
| Cretaceous | Upper Cretaceous | Fox Hills sandstone | | 1200–2500 | 25–200 | Sandstone, soft, massive, some sandy shale, and near the top a few thin coal beds; mainly marine. |
| | | Pierre shale | Elk Butte member | | 60–310 | Shale, medium-gray; forms gumbo; sandy near the top; marine. |
| | | | Mobridge member | | 90–240 | Shale, calcareous, and marl; weathers yellowish gray; marine. |
| | | | Virgin Creek member | | 85–285 | Shale; lower part weathers silver gray; upper part forms gumbo; marine. |
| | | | Verendrye member | | 100–200 | Shale, light olive-gray to dark-gray; forms gumbo; marine. |
| | | | DeGrey member | | 40–160 | Shale, claystone, and bentonite, siliceous in part, dark-gray; upper part has many Fe-Mn carbonate concretions; contains marine vertebrate remains. |
| | | | Crow Creek member | | 0–15 | Shale and sandstone, calcareous, light-gray; weathers to yellowish gray; marine. Developed mainly in eastern South Dakota. |
| | | | Gregory member | | 35–125 | Shale, gray; contains calcareous and ferruginous layers and concretions; marine basal part is locally calcareous along Missouri River. |
| | | | Sharon Springs member | | 9–175 | Shale, organic-rich, dark-gray to black; fish remains are common; marine bentonite beds are prominent along the southern and eastern Black Hills. |
| | | | Gammon member | | 0–67 | Shale and mudstone, light-gray; marine. A few ferruginous concretions 1 to 2 feet in diameter are present northeast and north of the Black Hills but are absent southeast and south. Member pinches out to the east. |

| | | | | | | |
|---|---|---|---|---|---|---|
| | | | Niobrara formation[1] | Smoky Hill marl member | 100–220 | 60–120 | Marl, calcareous shale, and bentonite, medium- to dark-gray where fresh; weathers to light yellow; marine. Fish scales are abundant. |
| | | | | Ft. Hays limestone member | | 40–100 | Limestone, calcareous shale, and bentonite; more resistant and weathers to a lighter color than overlying member; marine. |
| | | | Carlile shale | | | 270–800 | Shale, some interbedded siltstone, and in upper part as many as 3 thin sandstone beds, dark-gray; contains large limestone concretions; marine. |
| | | | Greenhorn formation | | | 25–800 | Limestone and calcareous shale; weathers yellowish gray; thin bedded and ridge forming; marine. |
| | | | Belle Fourche shale | | | 500–850 | Shale, soft, dark-gray; marine. |
| | Lower Cretaceous | | Mowry shale | | | 150–250 | Shale, siliceous, and some bentonite, light-gray; marine. |
| | | | Newcastle sandstone | | | 0–60 | Sandstone and dark-gray shale interbedded with bentonite and a few thin coal beds; generally marine. Pinches out to the east. |
| | | | Skull Creek shale | | | 150–300 | Shale, dark-gray, marine. |

[1] Members not clearly separable around Black Hills.

have been identified by W. A. Cobban of the U.S. Geological Survey (written communication, 1954) as *Baculites aquilaensis* Reeside and *Baculites aquilaensis* var. *separatus* Reeside. Similar fossils were found at the same horizon at localities 8 and 10 (pl. 51).

### LITHOLOGY

The areas where the Sharon Springs member crops out along the Missouri River in South Dakota and Nebraska and along the southern and eastern flanks of the Black Hills are shown in plates 50 and 51. Graphic sections of the shale in these two general areas are shown in plates 50 and 52. The member ranges from 96 to 106 feet in thickness along the eastern and southern flanks of the Black Hills (fig. 85), where it is characterized by three distinct units: a lower shale unit, a bentonitic unit, and an upper shale unit. The upper and lower shale units are characterized by tough, dark-gray to black shale containing numerous scales, spines, and bone fragments of fish. On the outcrop the shale weathers to silver-gray flakes, and the more resistant parts form rounded buttresses surrounded by subdued topography. The bentonitic unit includes as many as 20 bentonite beds, ranging from an inch to slightly more than 2 feet in thickness, distributed through an interval of about 30 feet. This unit generally has a thick bentonite bed near its base that may range from 2½ to 3½ feet in thickness. This bentonite zone was named the Ardmore bentonite bed from outcrops near Ardmore, S. Dak. (Spivey, 1940; Cobban and Reeside, 1952). Some of the beds within this bentonite zone can be traced with fair certainty from Rapid City southward to Ardmore, and thence northwestward to the South Dakota-Wyoming line.

Along the Missouri River, the thickness of the Sharon Springs member ranges erratically from 7 to 42 feet. Simpson (1955) reports that the Sharon Springs is only 1 foot thick in sec. 23, T. 33 N., R. 4 W., Knox County, Nebr. Similarly, 16 bentonite beds may be present at one outcrop, and only 2 may appear at another. The variation in thickness and number of beds suggests local interruptions in the deposition of the member caused by periods of local erosion. The bentonite beds along the Missouri River are much thinner and less persistent than those in the western part of South Dakota; the thickest bentonite is about 4 feet thick along the Black Hills and is slightly less than 2 feet thick along the Missouri River. Slumping of overlying rocks partially obscures the Sharon Springs at several localities.

Ellipsoidal concretions of limestone ranging from a few inches to 3 feet in thickness and from 6 inches to 6 feet in diameter occur at as many as 15 horizons along the eastern flank of the Black Hills. Some contain calcite and barite as septarian vein fillings. Along the Mis-

souri River, limestone concretions, where present, occur at only 1 or 2 horizons. A few ferruginous concretions generally are present near the top of the member.

Fresh samples of the shale of the Sharon Springs member contain shotlike accretions of pyrite which range from a fraction of a millimeter to 3 millimeters in diameter. The decomposition of this pyrite probably forms much of the iron oxides and sulfates which stain outcrops a light yellow.

Selenite commonly occurs as fracture coatings, isolated crystals ranging from less than 1 millimeter to about an inch in length, or cone-in-cone concretions as much as 1½ feet in diameter. The cone-in-cone concretions, as well as crystals up to 5 inches in length showing multiple twinning, are common in the thicker bentonite beds.

A standard rock analysis of a sample from the Sharon Springs member of the Pierre shale is reported by Simpson (1955) from the NE¼NE¼ sec. 17, T. 93 N., R. 56 W., near locality 28, plate 50:

| | *Percent* |
|---|---|
| $SiO_2$ | 60.98 |
| $Fe_2O_3$ | 3.20 |
| $Al_2O_3$ | 16.84 |
| CaO | 1.84 |
| MgO | 1.69 |
| $SO_2$ | None |
| Volatiles | 7.53 |
| Moisture | 5.14 |

This analysis, compared with the average analysis for shales in general (Pettijohn, 1949, p. 82) indicates that the Sharon Springs contains somewhat more silica, alumina, and moisture, and somewhat less iron, lime, and magnesium. The volatiles include carbonaceous matter and probably the sulfur from iron sulfide.

## RADIOACTIVITY AND URANIUM CONTENT

The maximum uranium content of samples of the Sharon Springs member in the report area is 0.025 percent, and the average uranium content for the entire member is about 0.0015 percent. This average is about 12 times higher than that given for sedimentary rocks in general (Evans and Goodman, 1941) and is on the order of the average uranium content reported by Breger (1955) for the Antrim shale of Late Devonian age in Michigan and a nodular shale of Miocene age from California. At three localities, zones having a uranium content of at least 0.005 percent aggregate more than 1.5 feet in thickness (locs. 7, 10, and 22, pls. 50 and 52).

Disequilibrium between uranium content and radioactivity expressed in percent generally favors radioactivity by about 0.002; for

example, samples showing radioactivity of 0.004 percent equivalent uranium generally have a uranium content of 0.002 percent. The shale is consistently out of balance, as indicated by analyses of core samples from the Oahe Dam site and from an oil test well drilled in the SE¼ sec. 17, T. 15 N., R. 20 E., Ziebach County, S. Dak. Water from seeps in the Sharon Springs member contains 7 to 780 parts per billion uranium (table 2), thus indicating that some of the disequilibrium may be caused by leaching.

TABLE 2.—*Uranium content of water in seeps issuing from the Sharon Springs member of the Pierre shale along the eastern and southern flanks of the Black Hills, South Dakota*

[Analysts: J. Schuch, G. Burrow, and N. M. Conklin]

| Laboratory No | Location (pl. 51) | Uranium content (ppb) | pH | Remarks |
|---|---|---|---|---|
| 220340 | SE¼ sec. 8, T. 4 N., R. 8 E., Meade County. | 25 | 7. 8 | Water is from base of member. |
| 231104 | SE¼ sec. 8, T. 7 S., R. 7 E., Fall River County. | 215 | 3. 0 | Water is from shale below Ardmore bentonite. |
| 223376 | NW¼ sec. 33, T. 9 S., R. 5 E., Fall River County. | 20 | 2. 6 | Water is from shale between bentonite beds above Ardmore bentonite. |
| 219172 | NW¼ sec. 30, T. 10 S., R. 5 E., Fall River County. | 7 | 7. 9 | Water is from shale above bentonite beds. |
| 219174 | NW¼ sec. 28, T. 11 S., R. 1 E., Fall River County. | 57 | 3. 5 | Water is from shale between bentonite beds above Ardmore bentonite. |
| 231105 | NW¼ sec. 8, T. 12 S., R. 4 E., Fall River County. | 300 | 2. 9 | Do. |
| 231106 | SW¼ sec. 16, T. 35 N., R. 47 W., Shannon County. | 780 | 2. 6 | Water is from shale above bentonite beds. |

Marl of the overlying Gregory member of the Pierre contains less than 0.001 percent uranium, but gamma-ray logs (fig. 86 and pl. 53) indicate that the underlying Niobrara formation is more uraniferous. It contains as much as 0.004 percent uranium directly beneath the Sharon Springs member at locality 19 (pl. 50). Analyses of 8 samples of water from the Niobrara formation show a uranium content ranging from 2 to 90 parts per billion and averaging about 35 parts per billion. Part of this uranium could have been derived from the overlying Sharon Springs.

### SURFACE INFORMATION

The vertical distribution pattern of uranium and radioactivity within the measured outcrops of the Sharon Springs member is shown by graphic sections (pls. 50 and 52). In sections along the eastern and southern flanks of the Black Hills, uranium generally occurs

within 10 feet above the bentonite zone (the Ardmore bentonite of Spivey, 1940). The upper limit of this zone is marked by a bentonite bed about 1 foot thick. Exceptions to this pattern are found at two places: first, at locality 2 (pl. 52) where the most radioactive portion of the shale is a bed less than 4 inches thick just above the base of the member, and second, at locality 4 (pl. 52) where the most radioactive and stratigraphically highest shale sampled is at least 25 feet above the bentonite zone.

The measured sections along the Missouri River appear to be enriched in uranium along the same general pattern as in the Black Hills. Where numerous bentonite beds make up the basal part of the member, the zone richest in uranium lies just above the uppermost thick bentonite bed in the group. Where the bentonite beds are less than 0.2 foot thick or are absent, the enriched zone lies at the base of the member, just above the Niobrara formation. The sections at localities 16 and 17 (pl. 50) are partial sections, being incompletely measured. The upper part of the Sharon Springs member contains higher than average amounts of uranium at localities 23, 25, and 26 (pl. 50). This increase in uranium content appears to be associated with an increase in fish remains and plant debris in a "trashy" zone.

Bentonite beds sampled at localities 19 and 21 (pl. 50) contain 0.0007 and 0.007 percent uranium, respectively. An analysis by S. Furman, R. Cox, and J. Schuch of fish scales, bones, and spines, shows a uranium content of 0.094 percent and a $P_2O_5$ content of 21.6 percent. A 3-inch zone directly overlying the Niobrara formation at locality 29 (pl. 51) contains 0.025 percent uranium. The sample is thought to be secondarily enriched, for it shows radioactivity of only 0.017 percent equivalent uranium, and a fresh sample from the same horizon contains only 0.004 percent uranium.

## SUBSURFACE INFORMATION

### CORES

Eighteen selected core samples of the Sharon Springs member from the Irish Creek well in the SE¼ sec. 17, T. 15 N., R. 20 E., Ziebach County, S. Dak., and 10 core samples from a test hole at the Oahe Dam site in sec. 1, T. 111 N., R. 80 W., Hughes County, S. Dak., contain 0.002 percent or less uranium. These selected samples are representative of the lithology of 140 feet and 98.4 feet of core, respectively (Russell, 1925; L. B. Underwood and R. E. Fisher, written communication, 1954).

### GAMMA-RAY LOGS

The pattern of distribution of radioactivity in rocks of central South Dakota (fig. 86) indicates that the highest radioactivity is in

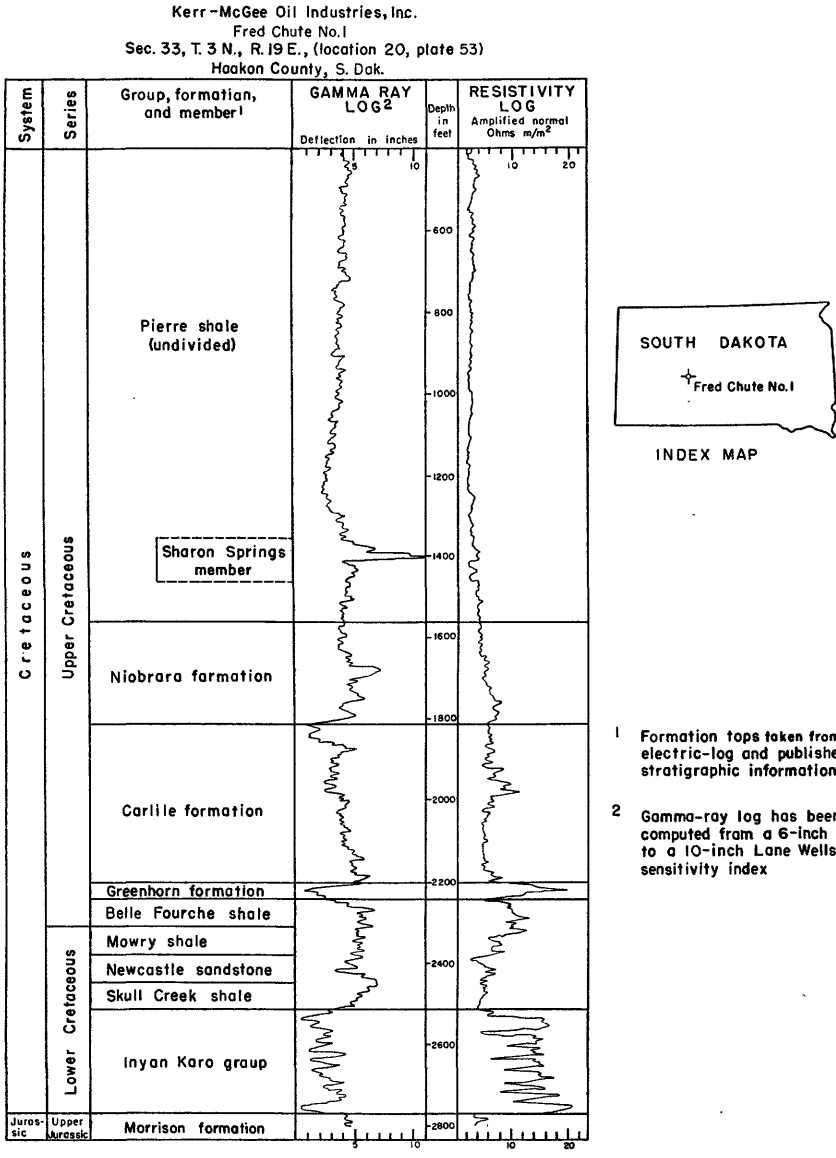


FIGURE 86.—Gamma-ray and resistivity logs of rocks of Cretaceous age in Fred Chute well 1, Haakon County, S. Dak.

the Sharon Springs member of the Pierre shale. The 35 gamma-ray logs examined from holes drilled for oil and gas in this and adjacent regions indicate that this high radioactivity may be correlated from well to well and that the Sharon Springs extends from northern Nebraska northward through North and South Dakota into southern

Canada and westward into eastern Wyoming and northeastern Montana. Twenty gamma-ray logs reproduced at a 10-inch sensitivity index with the top of the Greenhorn formation as datum are shown in plate 53. The rocks between the radioactive zone of the Pierre shale and the top of the Greenhorn formation (table 1 and fig. 86) thicken westward in North and South Dakota (fig. 87 and pl. 53). Although some thickening takes place in each rock unit (Reeside, 1944), much of the thickening takes place in the Gammon member of the Pierre shale.

A study of the gamma-ray logs indicates that the radioactivity of the Sharon Springs ranges from 0.002 to 0.010 percent equivalent uranium. These determinations were based on the premise that, at a 10-inch sensitivity index, a 1-inch deflection is caused by 0.0007 percent equivalent uranium (Gott and Hill, 1953).

A check was made during the present study to determine whether Gott and Hill's calibration factor is applicable to black shales. Chemical analyses [2] for uranium were made of 23 samples of core representing black shale, limy shale, and dolomite of Paleozoic age from the Carter, Sinclair, and Socony-Vacuum Companies' Lockwood well 1 in sec. 5, T. 147 N., R. 93 W., Dunn County, N. Dak. In comparing the gamma-ray log of this interval with the equivalent uranium content of the samples collected, the radioactivity of the samples could be predicted to within 0.001 percent equivalent uranium by using the calibration factor of Gott and Hill (1953).

Gott and Hill (1953, p. 70–71) pointed out, however, that there are many complicating factors that might cause erroneous interpretation of gamma-ray logs. The more important of these factors are the thickness versus the grade of the bed, the fluid content of the well, the shielding effect of casing, differences in individual instruments, and the rate of movement of the gamma-ray detector. Of the parts of the logs shown in figure 86 and plate 53, none of the intervals shown were cased at the time of logging and the rate of movement of the detector in most of the holes was about 35 feet per second. Even though the other factors cannot be critically evaluated, it is assumed their effect does not differ greatly from well to well.

Because of the demonstrated accuracy of predictions using the Gott and Hill calibration factors, the data gleaned from the study of gamma-ray logs of wells drilled for oil and gas in North Dakota, South Dakota, and Montana showing radioactivity of the Sharon Springs member are presented as a fairly reliable guide to the radioactivity of the member. The highest radioactivity shown by each well

[2] Analysts: S. Furman, M. Finch, and H. Lipp.

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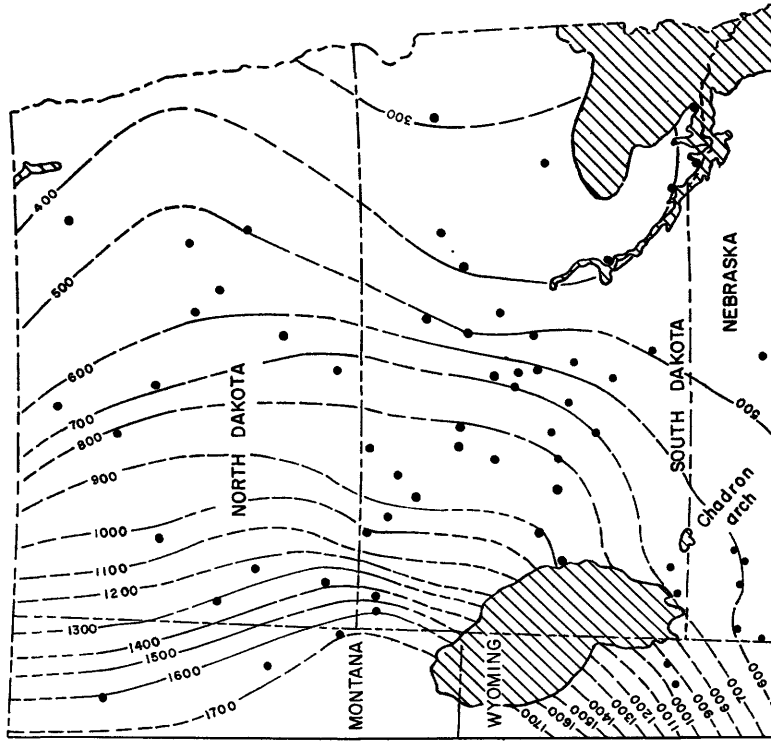


FIGURE 87.—Thickness map of the interval between the zone of high radioactivity in the Pierre shale and the top of the Greenhorn formation, northern Great Plains.

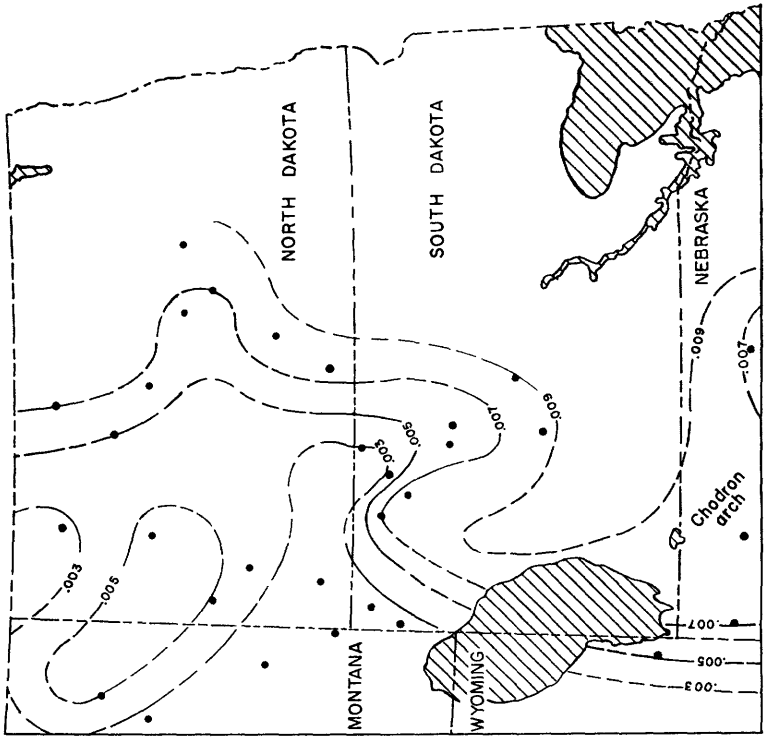


FIGURE 88.—Isoradioactivity map of the Pierre shale, northern Great Plains.

was plotted on a map and lines were drawn through points of equal radioactivity intensity. This map (fig. 88) indicates that the radioactivity is highest in south-central South Dakota and decreases westward and northwestward in western South Dakota and North Dakota to the extent that the Sharon Springs could not be identified with certainty in eastern Montana and Wyoming. This decrease in radioactivity probably reflects a decreasing amount of organic matter for a given volume of shale, a factor that may be explained in part by the increasing nearness to the source of the sediments, which may have been chiefly in the west throughout most of Late Cretaceous time. It is interesting to note that the direction of increasing radioactivity coincides roughly with the eastward thinning of the interval between the Sharon Springs member and the Greenhorn formation even though the thinning is mainly due to the wedging out of the Gammon member.

## CONTENT OF OTHER ELEMENTS

### CHEMICAL ANALYSES

Eighteen channel samples (loc. 19, pl. 50) were analyzed for arsenic and selenium. These analyses show a parallel relationship between the uranium content of the shale and the contents of arsenic and selenium (fig. 89). This relationship appears to be due to the reducing

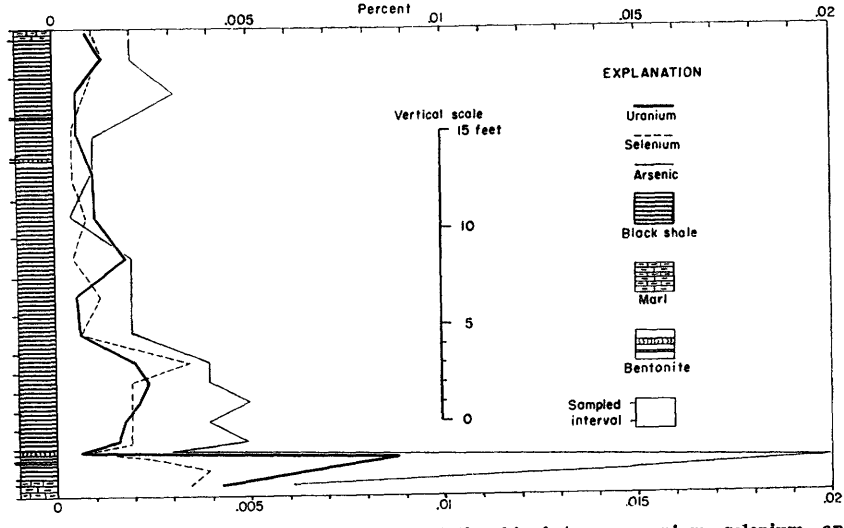


FIGURE 89.—Graphic section showing relationship between uranium, selenium, and arsenic in channel samples of the Sharon Springs member, Lyman County, S. Dak.

environment under which arsenic, selenium, and uranium are deposited (Rankama and Sahama, 1950, p. 742, 749; and Davidson and Ponsford, 1954). Phosphatic fish remains are also known to concentrate arsenic as well as uranium (Goldschmidt and Peters, 1934).

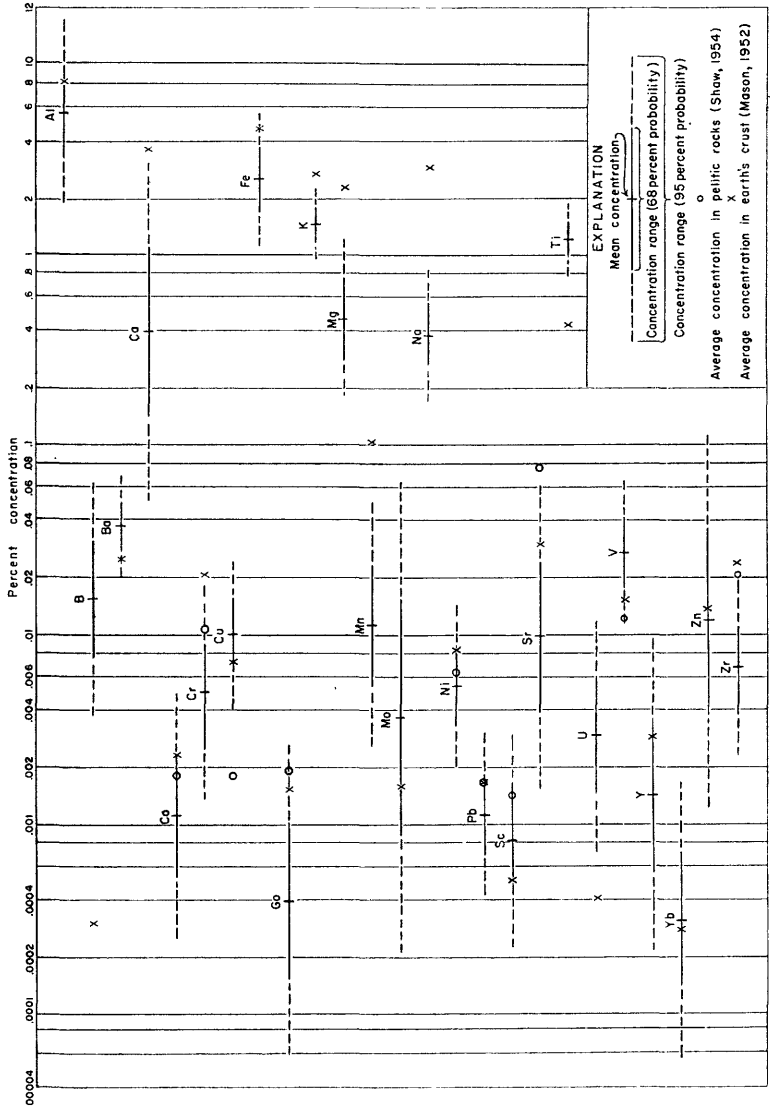


FIGURE 90.—Diagram showing concentration range and mean for 25 elements in 82 samples of the Sharon Springs member of the Pierre Shale, South Dakota and Nebraska.

### SPECTROGRAPHIC ANALYSES

More than 300 samples of the Sharon Springs member and associated rocks were analyzed by the semiquantitative spectrographic method to determine the content of trace elements and their relationship to uranium in the black shale of the Sharon Springs. In this semiquantitative spectrographic method, standard plates for each element are prepared from artificially prepared standard samples. These samples contain amounts of each element that divide each decimal order of magnitude into three equal intervals or groups on a logarithmic scale. (See table 6.) The interval between 1 and 10, for example, is divided into 1–2, 2–5, and 5–10 percent, when rounded off to one significant figure. The spectrogram of the sample is visually compared with that of the standard plates and the values for individual elements are reported within the appropriate group. Comparisons of this type of semiquantitative results with those obtained by quantitative methods, either chemical or spectrographic, show that the assigned group includes the quantitative value at least 60 percent of the time (A. T. Myers, oral communication, 1958).

Trace amounts of 25 elements were detected in nearly all the samples. These elements, except silicon and with the addition of chemically determined uranium, are listed in figure 90. Those elements which occur in amounts greater than uranium, in order of their apparent abundance, are silicon, aluminum, iron, potassium, titanium, magnesium, calcium, sodium, barium, vanadium, boron, zinc, manganese, copper, strontium, zirconium, nickel, chromium, and molybdenum. Those elements which occur in concentration less than uranium, in order of decreasing abundance, are yttrium, lead, cobalt, scandium, gallium, and ytterbium. The mean concentration of copper and vanadium in the Sharon Springs is notably higher than in other shales (pelitic rocks) (Shaw, 1954). Those extremely rare elements which are found in a few of the samples are beryllium, niobium, and silver. Elements which may be present in amounts greater than uranium but which are rarely detected are arsenic, phosphorus, cerium, dysprosium, lithium, neodymium, samarium, lanthanum, and cadmium. (See table 3.)

The germanium content of two samples of coalified wood is particularly noteworthy. Partial analyses of these two samples are given in table 4. A similar association of uranium and germanium in coalified wood from black shale of Upper Devonian age has been reported by Breger and Schopf (1955).

Five-gallon samples of water were collected from the Sharon Springs member at two of the localities listed on table 2 and from an additional locality on the Chadron arch in Shannon County, S. Dak.,

TABLE 3.—*Minimum concentrations of the elements generally detectable by the semiquantitative spectrographic method*

[August 10, 1954]

| Element | Percent [1] | Element | Percent [1] |
|---|---|---|---|
| Ag | 0.00005 | Na | 0.05 |
| Al | .001 | Nb | .001 |
| As | .05 | Nd | .01 |
| Au | .003 | Ni | .0005 |
| B | .005 | Os | .005 |
| Ba | .0001 | P | .1 |
| Be | .0001 | Pb | .001 |
| Bi | .001 | Pd | .0005 |
| Ca | .001 | Pt | .003 |
| Cd | .005 | Re | .005 |
| Ce | .05 | Rh | .005 |
| Co | .0005 | Ru | .005 |
| Cr | .0001 | Sb | .01 |
| Cu | .00005 | Sc | .001 |
| Dy | .05 | Si | .001 |
| Er | .005 | Sm | .01 |
| Fe | .001 | Sn | .001 |
| Ga | .001 | Sr | .0001 |
| Gd | .005 | Ta | .05 |
| Ge | .0005 | Te | .08 |
| Hf | .05 | Th | .05 |
| Hg | 1.0–0.1 | Ti | .0005 |
| In | .001 | U | .05 |
| Ir | .005 | V | .001 |
| K | .5 | W | .01 |
| La | .005 | Y | .001 |
| Li | .01 | Yb | .0001 |
| Mg | .001 | Zn | .02 |
| Mo | .001 | Zr | .001 |
| Mn | .0005 | | |

[1] The limits of detection given are based on the semiquantitative method used in the analytical laboratory at the time of analysis.

TABLE 4.—*Analyses of two samples of coalified wood from Sharon Springs member of the Pierre shale, South Dakota*

[Analysts: Mona Frank, Roosevelt Moore, and Carmen Johnson]

| Item | Percent by weight | |
|---|---|---|
| | NW¼ sec. 28, T. 11 S., R. 1 E., Fall River County (loc. 14, pl. 51) | NE¼ sec. 8, T. 95 N., R. 65 W., Charles Mix County (loc. 25, pl. 50) |
| Uranium | 0.003 | 0.006 |
| Equivalent uranium | .002 | .009 |
| Ash | 19.3 | 58.9 |
| Uranium in ash | .014 | .010 |
| Germanium in ash [1] | .5 –1 | .05 –0.1 |
| Phosphorus in ash [1] | 1.0 –2 | .5 –1 |
| Vanadium in ash [1] | .2 –0.5 | .1 –0.2 |
| Arsenic in ash [1] | .05 –0.1 | .1 –0.2 |

[1] Semiquantitative spectrographic determinations. (See table 3.)

to the southeast. An aliquot from each sample was analyzed for uranium and the remainder was evaporated to dryness. The weight of the residues ranged from 125 grams in the least uraniferous to 232 grams in the most uraniferous. These residues were analyzed spectrographically and the results are presented in table 5.

TABLE 5.—*Elements detected by semiquantitative spectrographic analyses of water residues from the Sharon Springs member of the Pierre shale, South Dakota*

[Analyst: N. M. Conklin]

| Element | Laboratory No. 231104 | | Laboratory No. 231105 | | Laboratory No. 231106 | |
|---|---|---|---|---|---|---|
| | Sec. 8, T. 7 S., R. 7 E., Fall River County | | Sec. 8, T. 12 S., R. 4 E., Fall River County | | Sec. 16, T. 35 N., R. 47 W., Shannon County | |
| | Percent in residue [1] | Parts per million in water [2] | Percent in residue [1] | Parts per million in water [2] | Percent in residue [1] | Parts per million in water [2] |
| Na | 3.2 | 200 | 6.8 | 700 | 6.8 | 850 |
| Mg | 3.2 | 200 | 3.2 | 350 | 3.2 | 400 |
| Ca | 3.2 | 200 | 1.5 | 150 | 3.2 | 400 |
| Al | 1.5 | 100 | 1.5 | 150 | 1.5 | 200 |
| Fe | .15 | 10 | .15 | 15 | 3.2 | 400 |
| Mn | .15 | 10 | .32 | 35 | .15 | 20 |
| Zn [3] | .19 | 12 | .22 | 24 | .18 | 22 |
| Li | .15 | 10 | .068 | 7 | .068 | 8.5 |
| Si | .068 | 4.5 | .068 | 7 | .068 | 8.5 |
| Ni | .015 | 1 | .032 | 3.5 | .015 | 2.0 |
| Sr | .015 | 1 | .015 | 1.5 | .0068 | .85 |
| Cu | .0068 | .45 | .0068 | .7 | .015 | 2.0 |
| Co | .0068 | .45 | .0068 | .7 | .0068 | .85 |
| Y | .0068 | .45 | .0068 | .7 | .0068 | .85 |
| V | (4) | | (4) | | .0032 | .4 |
| P [3] | .0019 | .12 | .0014 | .15 | .092 | 11 |
| B | Trace | <.28 | Trace | <.52 | (4) | |
| Cd | Trace | <.28 | Trace | <.52 | Trace | <.62 |
| Cr | .00068 | .045 | .00032 | .035 | .0015 | .2 |
| Yb | .00068 | .045 | .00068 | .07 | .00068 | .085 |
| Sc | (4) | | (4) | | .00068 | .085 |
| Be | .00015 | .01 | .00015 | .015 | .00015 | .02 |
| Ba | .00015 | .01 | Trace | <.015 | .00015 | .02 |
| U (ppm) [5] | 0.215 (pH 3.0) | | 0.300 (pH 2.9) | | 0.780 (pH 2.6) | |

[1] Logarithmic average of concentration range reported by semiquantitative spectrographic analyses (table 6).
[2] Computed from semiquantitative spectrographic analyses (table 6), this figure is accurate to plus or minus one half reported value.
[3] Chemical determinations from residue. Analysts: J. Schuch and R. Dufour.
[4] Not detected.
[5] Chemical determinations from water. Analyst: T. Miller.

A statistical study of the spectrographic analyses from 82 samples of black shale followed a method outlined by A. L. Miesch (written communication, 1954), in which logarithmic averages were assigned to each concentration range detected spectrographically (table 6).

TABLE 6.—*Numbers and symbols used in referring to semiquantitative spectrographic analyses of Sharon Springs member*

| Concentration range (percent) | Logarithmic average (percent) [1] | Symbol used on figure 92 |
|---|---|---|
| More than 10 | | A |
| 5–10 | 6.8 | B |
| 2–5 | 3.2 | C |
| 1–2 | 1.5 | D |
| .5–1.0 | .68 | E |
| .2– .5 | .32 | F |
| .1– .2 | .15 | G |
| .05– .1 | .068 | H |
| .02– .05 | .032 | I |
| .01– .02 | .015 | J |
| .005– .01 | .0068 | K |
| .002– .005 | .0032 | L |
| .001– .002 | .0015 | M |
| .0005– .001 | .00068 | N |
| .0002– .0005 | .00032 | O |
| .0001– .0002 | .00015 | P |
| .00005– .0001 | .000068 | Q |
| .00002– .00005 | .000032 | R |
| .00001– .00002 | .000015 | S |

[1] This figure was used in the statistical treatment of the samples.

These samples were collected from 30 localities to be representative of unweathered black shale containing no appreciable bentonite or lime. Chemical and radiometric determinations indicate that the samples have an average uranium content of 0.003 percent.

The mean concentration and range in concentration of the elements present in the Sharon Springs member are plotted on a semilogarithmic scale (fig. 90). This is to conform with the logarithmic method used by the laboratory in reporting semiquantitative spectrographic data (table 6) and is at least tentatively substantiated by results of experiments by Ahrens (1953), who reports that trace elements are log-normally distributed in the earth's crust.

### CORRELATION WITH URANIUM

The correlation between concentration of uranium and other elements in the Sharon Springs member (fig. 91) was determined statistically by means of correlation coefficients. A correlation coefficient is used to express the relationship of two variables as a single number. This coefficient ranges from a positive 1 to a negative 1, where 1 indicates a complete association of two components and a value of zero indicates that there is no association whatsoever between the two com-

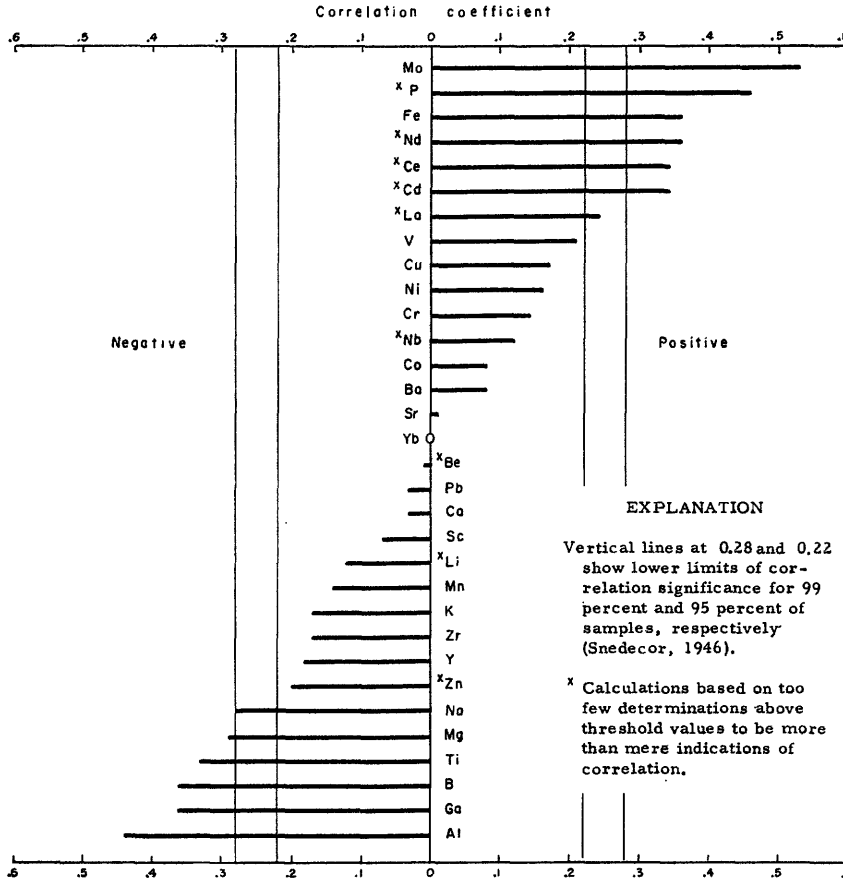


FIGURE 91.—Diagram showing correlation between concentration of uranium and 32 other elements in 82 samples of the Sharon Springs member of the Pierre shale, South Dakota and Nebraska.

ponents. A positive correlation indicates that the two components vary directly, whereas a negative coefficient indicates that they tend to vary inversely in relation to each other.

A good positive correlation was found between the content of uranium and molybdenum and the content of iron (fig. 91). A tentative correlation is indicated between the uranium content and the contents of phosphoros, neodymium, cerium, cadmium, and lanthanum. A strong negative correlation is shown between the content of uranium and the contents of aluminum, gallium, boron, titanium, magnesium, and sodium. A discussion of a few of these correlations follows.

#### POSITIVE CORRELATION

In addition to positive relationships indicated for uranium with arsenic and selenium by chemical analyses, a positive correlation coefficient of 0.53 was found between uranium and molybdenum, and a correlation coefficient of 0.36 was found between uranium and iron (fig. 91). This relationship may be explained in part by the fact that in a reducing environment hydrogen sulfide causes precipitation of all four elements in the original sapropelic sediments (Rankama and Sahama, 1950, p. 637). Abundant pyrite in the shale of the Sharon Springs member, together with the positive correlation between uranium and iron, indicates that the environment in which uranium occurs in black shale is favorable for the occurrence and deposition of pyrite. Recent work by E. D. Strahl and others at Pennsylvania State University (written communication, 1955) has disclosed a very strong correlation between the concentration of uranium and the abundance of pyrite in the Chattanooga black shale, confirming an association pointed out by McKelvey and Nelson (1950), Goldschmidt (1954, p. 566), and others. Autoradiography by Davidson and Ponsford (1954) indicates a tendency toward close association of uranium and pyrite in lignite.

The correlation between uranium and phosphoros results from high phosphate content of fish scales and bones and the corresponding high uranium content of that material (p. 587). Many of the shale zones showing particularly numerous fragments of such material have a correspondingly higher uranium content. This relationship may also hold for the samples that are very low in uranium, but confirmative data are lacking because the phosphorus content of such samples is below the amount detectable by semiquantitative spectrographic methods (table 3). The tentative correlation between uranium and the elements neodymium, cerium, cadmium, and lanthanum is little understood. These rare elements may be associated with phosphatic material.

#### NEGATIVE CORRELATION

Most of the elements showing a strong negative correlation with uranium (fig. 91) are common in clay and other inorganic allogenic detrital material, indicating that the most concentrated accumulations of uranium are in association with organic material. These elements include aluminum, magnesium, and sodium as clay minerals, and gallium associated with aluminum (Rankama and Sahama, 1950, p. 727). Boron and titanium are thought to be mainly present as detrital minerals—boron being one of the constituents of tourmaline, and titanium being found in sphene, ilmenite, rutile, brookite, and anatase.

### RELIABILITY OF THE CORRELATIONS

Because of ever-improving techniques of spectrographic analysis, and because of the ease with which large numbers of samples may be analyzed for a suite of as many as 60 elements, so much data are amassed that they can be interpreted only by statistical methods. Table 6 shows the ranges used in reporting concentrations determined by the semiquantitative spectrographic method, and it also shows the numerical amount assigned to each range for the purpose of statistical study. The histograms (fig. 92) present a condensation of the data on which the statistical study is based by showing the number of samples (of a possible 82) reported by the laboratory to contain a given concentration of the elements studied. In order to treat statistically the semiquantitative spectrographic analyses of all 82 samples for the 25 elements shown on figure 92, concentrations reported as "trace" or "zero" by the laboratory were arbitrarily assigned percent values relative to the lowest amount generally detectable by semiquantitative spectrographic methods (table 3).

### ENVIRONMENT OF DEPOSITION

The overall chemical environment of deposition and diagenesis of the Sharon Springs member of the Pierre shale was reducing, as is indicated by its sapropelic nature. In common with other marine uranium-bearing black shales (McKelvey and Nelson, 1950, p. 43), the Sharon Springs is noncalcareous and highly carbonaceous. It is high in sulfides and somewhat phosphatic (due largely to fish remains), and it probably accumulated slowly, as is evidenced by the high proportion of organic matter (plant and fish remains) to allogenic sediments. That the source of the sediments lay to the west is indicated by the sandstones and light-colored shales which interfinger with the black shales in eastern Montana and Wyoming, by the fact that the thicker bentonites in the Sharon Springs member are also found in the western part of South Dakota, and by the westward thickening of the Sharon Springs member.

The depth at which the Sharon Springs sediments accumulated is inferred from the following evidence to have been less than 250 feet: (1) the excellent state of preservation of the long delicate shells of *Baculites* found along the southeastern flank of the Black Hills indicate that the water may have been quiet and fairly shallow (W. A. Cobban, written communication, 1955); (2) the marl of the Gregory member directly overlying the Sharon Springs in the Missouri River valley contains Foraminifera typical of the neritic zone and probably representing a depth no greater than 250 feet (Steven K. Fox, U. S. National Museum, written communication, 1955); and (3) thin zones

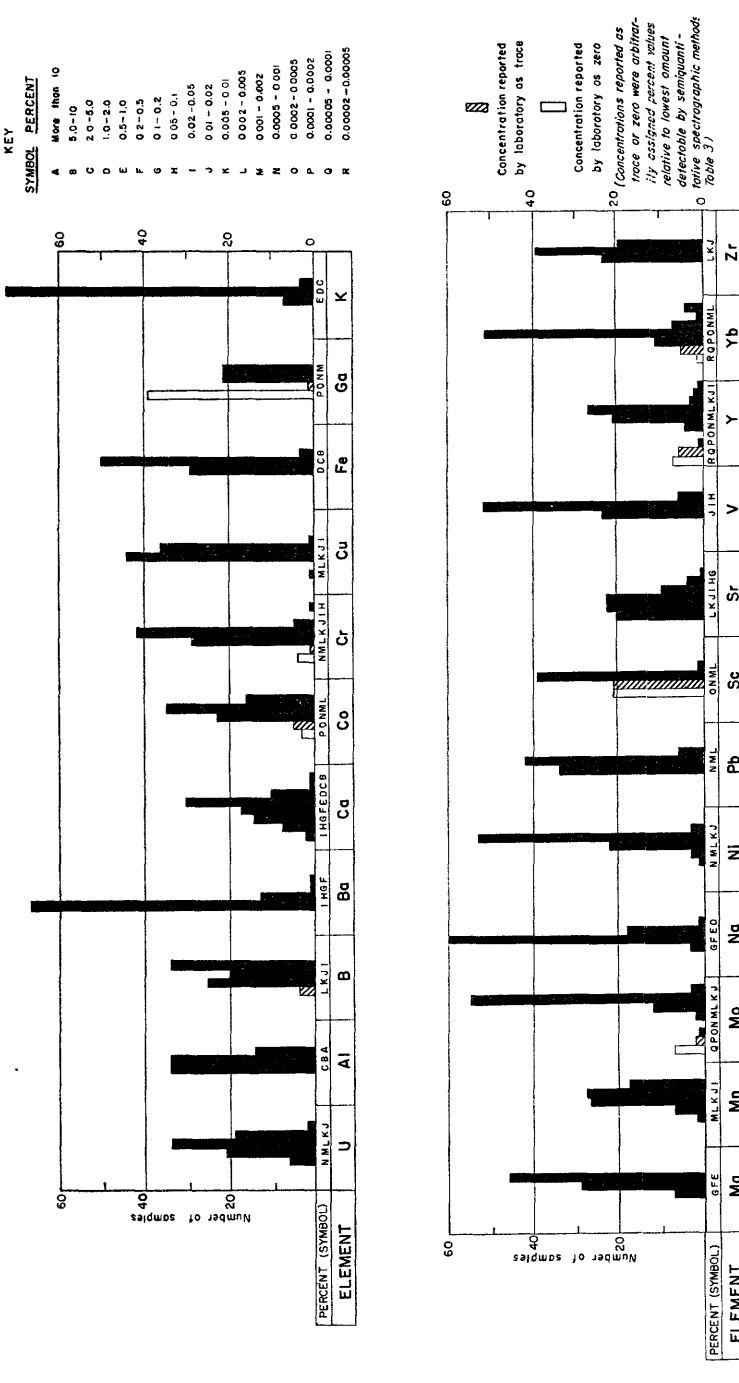


FIGURE 92.—Histograms showing frequency distribution of elements for 82 samples of Sharon Springs member, South Dakota and Nebraska.

of coarse fragments of fish scales and bones found at a few localities are thought to represent a lag concentration from an underwater surface eroded by wave action.

## ORIGIN OF URANIUM

The uranium appears to have been emplaced in a reducing environment during deposition and diagenesis of the shale, the highest concentrations of uranium being found where deposition of allogenic sediments was probably the slowest. Most of the uranium appears to be associated with the organic constituents of the shale.

The fact that the peak in radioactivity lies just above the bentonitic sequence may be due to fortuitous circumstances. Or, as suggested by Tourtelot (1956),

> * * * the volcanic ash deposited in the sea probably would have released much of its metal content, including uranium, in the early stages of its alteration to bentonite and thus would have increased the metal content of the waters of the Sharon Springs area.

This also may explain uranium-rich layers where no bentonites are present, for the absence of bentonites by nondeposition or erosion still permits the distribution throughout the sea of the elements derived from bentonites.

## CONCLUSIONS

The study of the Sharon Springs member in South Dakota and northeastern Nebraska leads to the following conclusions:

1. The Sharon Springs member of the Pierre shale is a lithologic unit which may be traced from exposures and from subsurface information throughout most of South Dakota and North Dakota and adjacent areas to the south.
2. Most of the member accumulated under quiet water, probably less than 250 feet deep, and the source of the sediments was mainly to the west.
3. The uranium is held, at least in part, by phosphatic material such as fossil fish bones and scales and in part by carbonized macerated plant remains.
4. The vertical distribution of uranium content in the member is thought to be a function of the uranium content of the sea water following the deposition of volcanic ash now altered to bentonite.
5. The highest uranium content in the Sharon Springs member is about 0.025 percent, in beds no thicker than 5 inches. The uranium content of the entire member averages about 0.0015 percent—higher than any average hitherto reported for black shales of post-Paleozoic age.

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# CONTENTS

[The letters in parentheses preceding the titles designate separately published chapters]