STATE OF ARKANSAS

ARKANSAS GEOLOGICAL AND CONSERVATION COMMISSION

Norman F. Williams, Geologist-Director

Information Circular No. 22

GEOLOGY OF A URANIUM-BEARING BLACK SHALE OF LATE DEVONIAN AGE IN NORTH-CENTRAL ARKANSAS

by
Vernon E. Swanson and Edwin R. Landis
U. S. GEOLOGICAL SURVEY

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ABSTRACT

Selected samples of black, semilustrous organic material collected from a small lens of marine black shale in the basal sandstone of the Boone formation in north-central Marion County, Ark., contain as much as 0.71 percent uranium. This lens and several other lenses of black shale are in what is believed to be a northward extension of a time-transgressing sandstone called the Sylamore sandstone member of the Chattanooga shale in other areas of Arkansas and Oklahoma. The ages of the shale lenses and the enclosing sandstones, based mainly on conodont assemblages, are either Late Devonian or Early Mississippian (Kinderhook), or both.

Three types of organic matter can be differentiated in the black shale at the Marion County locality: a) coaly plant remains, probably fragments of driftwood *Callixylon;* b) minute spore-like bodies of the genus *Tasmanites;* and c) black colloidal humic material. Relatively pure samples of collodial humic material contain the most uranium, and are comparable in general appearance, physical characteristics, and range in uranium content to the kolm in the shales of Cambrian age of Sweden.

INTRODUCTION

At an exposure of black shale in Marion County, Ark., samples were collected that have the highest uranium content yet reported from black shale in the United States. The most uraniferous samples collected by the authors contained 0.55 percent uranium, but a shale sample from the same locality with a uranium content of 0.71 percent has been reported. Less than 1 ton of highly uraniferous shale is present at the locality, but the geologic relations are significant because they suggest specific conditions under which uranium may be syngenetically concentrated in marine black shale. The study of this and other nearby deposits in the Chattanooga shale in north Arkansas was made as part of an investigation on behalf of the U. S. Atomic Energy Commission of uranium-bearing black shale in the Midcontinent region.

The locality was first visited in May 1955 by Swanson in the company of Mr. C. E. West of Bentonville, Ark., who had detected the anomalously radioactive shale while prospecting in the area. Representative samples were collected at this time; and the locality was studied and sampled in more detail during subsequent visits by the authors in June 1955, by Landis in November 1955, and by Swanson in June 1958. The type section of the Sylamore sandstone member and many other outcrops of the Chattanooga shale in Arkansas have also been studied.

A suite of shale samples was submitted to James M. Schopf, U. S. Geological Survey, for petrographic study of the organic matter, and another collection was submitted to the late Wilbert H. Hass, U. S. Geological Survey, for fossil determinations. The authors gratefully wish to acknowledge the descriptive material and data determined by Schopf and Hass and used in the preparation of this paper.

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U. S. Geological Survey

GEOLOGIC SETTING

Location

The outcrop of black shale is located in the NE¼SW¼ sec. 11, T. 19 N., R. 17 W., Marion County, Ark. (fig. 1), near the head of a small, south-trending ravine that opens onto Georges Creek, which is about 1 mile south of the outcrop. The outcrop is reached by following an old logging road for about 200 yards south-southeast of Arkansas Highway 14; this logging road, which can barely be detected from the highway, enters onto Arkansas Highway 14 5.2 miles southeast of the junction of Arkansas Highways 14 and 125. The locality may also be reached by travelling 11.0 miles on Arkansas Highway 14 from the intersection of this highway with U.S. Highway 62 at the northwest corner of the square in Yellville, the county seat of Marion County.

About 150 yards south and down the ravine from the outcrop of black shale are the ruins and dumps of the old Bear Hill zinc mine, last operative in 1917. The zinc ore was mined at depths between 100 and 200 feet stratigraphically below the black shale, from the lower part of the Everton formation and the upper part of the Powell dolomite, both of Ordovician age.

Stratigraphy

Marion County lies on the highly dissected southwest flank of the Ozark uplift where rocks of Ordovician age are widely exposed. Only on the high ridges are younger rocks of Paleozoic age present, and then generally of only one formation, the Boone formation of Mississippian age. The rock unit of special interest in this report is found at the contact between these Ordovician and Mississippian rocks. McKnight (1935) presented a comprehensive report on the zinc and lead deposits of northern Arkansas, which included a geologic map of the Yellville quadrangle that covers most of Marion County, Ark.; his report has served as the basis for much of the general geologic description given here.

The highly uraniferous material was collected from a shale lens in a sandstone that was called the "basal sandstone of the Boone" formation by McKnight (1935, p. 68). This sandstone is a persistent unit at the base of the Boone formation in Marion County and parts of adjacent counties of north-central Arkansas, and rests upon a broad, plane surface that unconformably cuts across several formations of Ordovician and Silurian ages. The sandstone is generally a few inches to 5 feet thick, but in a few areas it is between 5 and 12 feet thick. In the Yellville quadrangle, this sandstone is everywhere overlain by the distinctive crinoidal, reddish St. Joe limestone member of the Boone formation.

The sandstone at the base of the Boone formation is known to contain black shale at two other places in the Yellville quadrangle. Both places were noted by McKnight (1935, p. 69), and are located near Everton, Ark., about 14 miles airline southwest of the locality described in this report (fig. 1). Maher and Lantz (1952, p. 10; 1953) observed three similar occurrences of black shale in this sandstone about 25 miles to the south, two in central Searcy County and one in eastern Newton County (fig. 1). Thus, 6 localities are known, and there is little doubt that local lenses or pockets of black shale in the sandstone are present at many other places in the area to the north and east of the zero-isopach line of black shale as shown in figure 1.

Age and Stratigraphic Correlation

The age and stratigraphic relations of this sandstone at the base of the Boone formation in northern Arkansas are still not definitely known, largely because of the uncertainty (for example, see Croneis, 1930, p. 38-40) of the age and distribution of the Sylamore sandstone member, whose type locality is about 30 miles southeast of the uraniferous shale locality (fig. 1). An interpretation of the stratigraphic relations of Upper Devonian and Lower Mississippian rocks of northern Arkansas based on additional evidence is suggested here, but the new information is not adequate to nullify other interpretations.

Rocks of Late Devonian age, mostly marine black shale referable to the Chattanooga shale, are present flanking the Ozark dome in a large area of central and northwestern Arkansas, northeastern Oklahoma, and southwestern Missouri. Over most of this area the Chattanooga is composed of two lithologic units: an upper black to dark-gray fissile shale, underlain by a

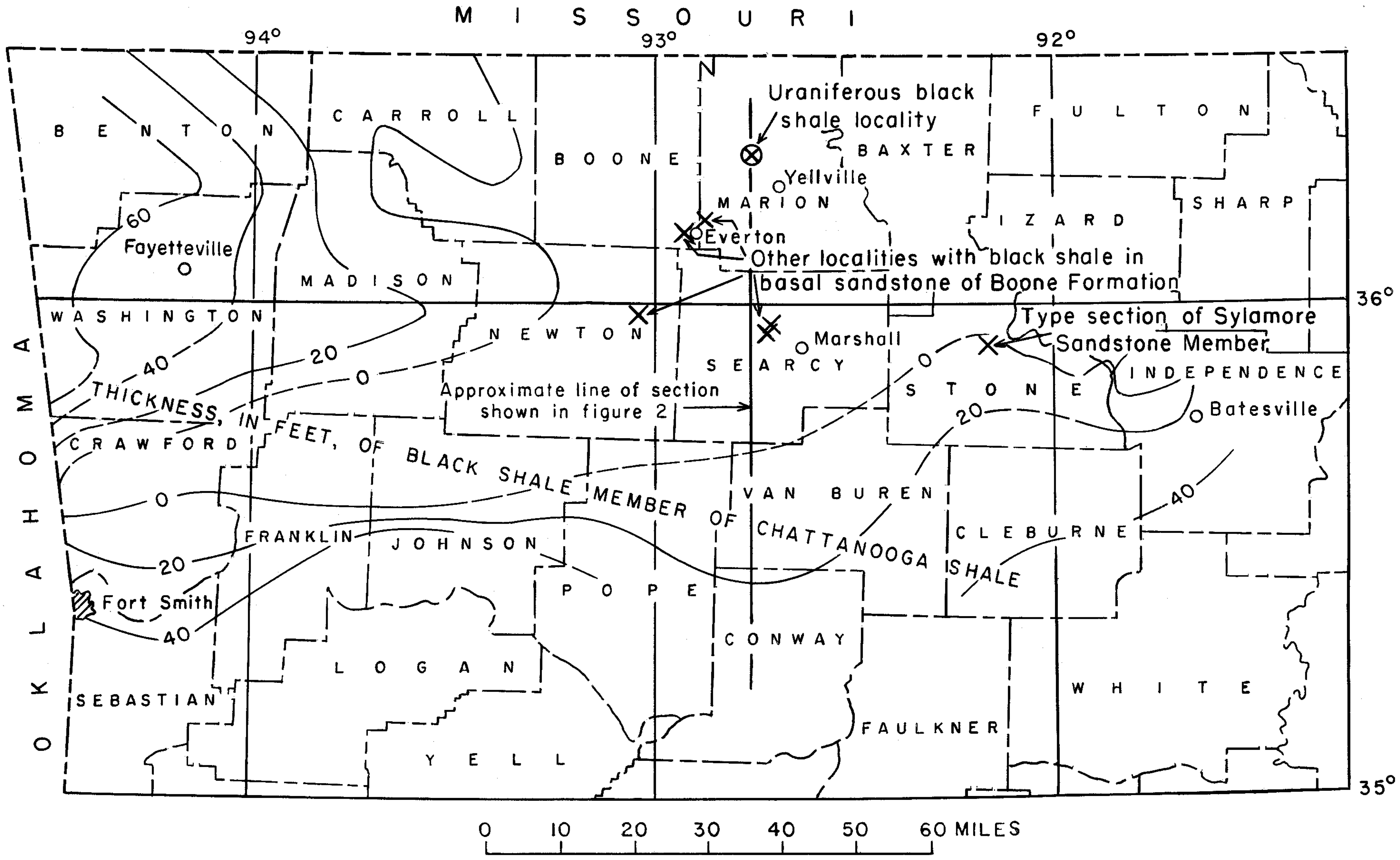


Figure 1. Map showing location of uraniferous black shale in Marion County, and other localities with black shale lenses in the basal sandstone of the Boone formation. Thickness lines of the Chattanooga shale, exclusive of its basal sandstone member, modified from Frezon and Glick (1959, pg. 25).

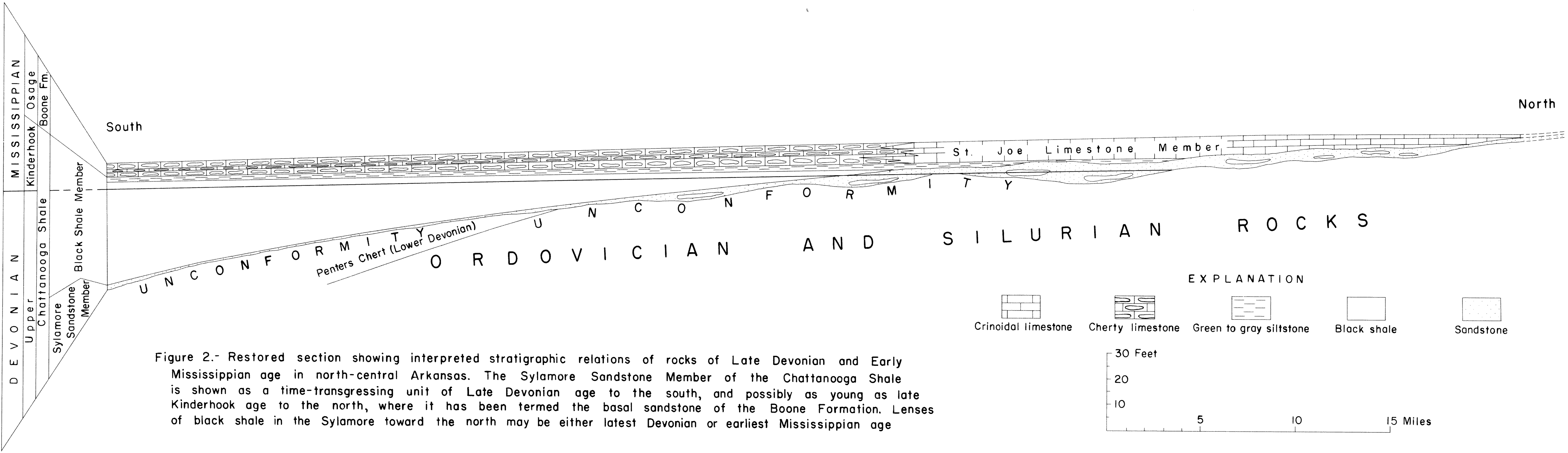


Figure 2.- Restored section showing interpreted stratigraphic relations of rocks of Late Devonian and Early Mississippian age in north-central Arkansas. The Sylamore Sandstone Member of the Chattanooga Shale is shown as a time-transgressing unit of Late Devonian age to the south, and possibly as young as late Kinderhook age to the north, where it has been termed the basal sandstone of the Boone Formation. Lenses of black shale in the Sylamore toward the north may be either latest Devonian or earliest Mississippian age

light-gray brown-weathering sandstone that characteristically contains scattered small phosphatic nodules. The black shale generally thins and disappears on the flanks of the Ozark dome, as indicated by the isopach lines on figure 1, modified from a map by Frezon and Glick (1959, pl. 25). At or just beyond the edge of the black shale unit, the stratigraphic position of the Chattanooga is occupied by a sandstone named the Sylamore sandstone by Branner (Penrose, 1891, p. 112-114; Branner, 1897, p. 580), and assigned a Devonian age by Williams (1900, p. 277). Subsequent workers have correlated this sandstone on the basis of lithology, stratigraphic position, and fossils (for example, see Quigley, 1942) with the basal sandstone of the Chattanooga in areas where the black shale is present, so that now the basal sandstone of the Chattanooga in northwestern Arkansas and northeastern Oklahoma is called the Sylamore sandstone member of the Chattanooga shale, and is thereby considered to be of Late Devonian (?) age.

The age of the sandstone at the base of the Boone formation in the Yellville quadrangle was considered by McKnight (1935, p. 73) to be of Osage, or about middle Mississippian age, based primarily on the age of the abundant megafossils in the conformably overlying limestone units. McKnight (1935, p. 74) did report that conodonts were collected from the black shale in the sandstone near Everton, and he lists the following genera, as identified by R. S. Bassler: *Prioniodus, Bryantodus, Lonchodina, Hindeodella, Polygnathus, Palmatolepis,* and *Panderodella.* Bassler stated that these genera are common in the Chattanooga shale in Tennessee, which at that time he considered to be of Early Mississippian age. McKnight's conclusion (1935, p. 75), considering the proximity and conformable relation of the black shale near Everton with the overlying St. Joe limestone member of the Boone formation, was that the conodont fauna represented a survival of this fauna into Osage time. However, he also recognized that this sandstone was probably of different ages in different areas because it was the basal sandstone of an overlapping series of strata, and probably was a continuation of what is called the Sylamore sandstone in large areas to the south and west.

Maher and Lantz (1953) reported the results of a conodont study by Wilbert H. Hass of 2 samples of shale and 15 samples of sandstone that they collected from the aforementioned localities during their study in Searcy County to the south. The 2 samples of shale contained conodonts questionably of Kinderhook age, and 1 sample of sandstone contained a fauna of definite Kinderhook age; 1 sample of sandstone contained a fauna of either Late Devonian or Early Mississippian age; and 13 samples of sandstone contained a conodont fauna of Late Devonian age. At the locality in eastern Newton County (Maher and Lantz, 1952, p. 10), the sandstone above the shale contained a conodont fauna that was regarded as Kinderhook by Hass; conodonts from the underlying shale and sandstone were not suitable for age determinations. From these age designations based on conodonts, the sandstone at the base of the Boone formation in Searcy and Newton Counties is here concluded to be of latest Devonian or earliest Mississippian age, probably both at many localities.

Two samples containing abundant conodonts were collected from the uraniferous shale lens in Marion County, and were submitted to Hass for identification and age determination. Eleven conodont genera were identified, including all except one of the genera of those identified by Bassler in McKnight's collection. Hass (written communication, 1955) in his report stated: "Specimens of *Palmatolepis glabra* dominate the conodont fauna. This species together with *Palmatodella delicatula* Bassler, *Palmatolepis distorta, Palmatolepis perlobata, Palmatolepis superlobata,* and *Polyophodonta confluens* are characteristic fossils of the lower faunal zone of the Gassaway member of the Chattanooga shale in central Tennessee and adjoining States. . . The two samples are considered to be from rocks of Late Devonian age." Hass further stated that he had studied 42 conodont collections from 14 other localities in the same general area and from the same stratigraphic interval as these samples. All of these collections contained faunas like those found elsewhere in the United States in rocks of Late Devonian and Early Mississippian (Kinderhook) age; none of these collections contained conodonts indicating an age as young as late Kinderhook or early Osage. (Also see Hass, 1951, 1956a, 1956b, 1958).

On the basis of Hass' statement, the age of the uraniferous shale in sandstone at the base of the Boone formation in central Marion County is considered of Late Devonian age, the shale near Everton is probably of the same age, and the shales in the sandstone reported by Maher and Lantz (1953) in Searcy County are considered as either of latest Devonian or of earliest Mississippian or Kinderhook age. Other similar bodies of shale in the Sylamore sandstone mem-

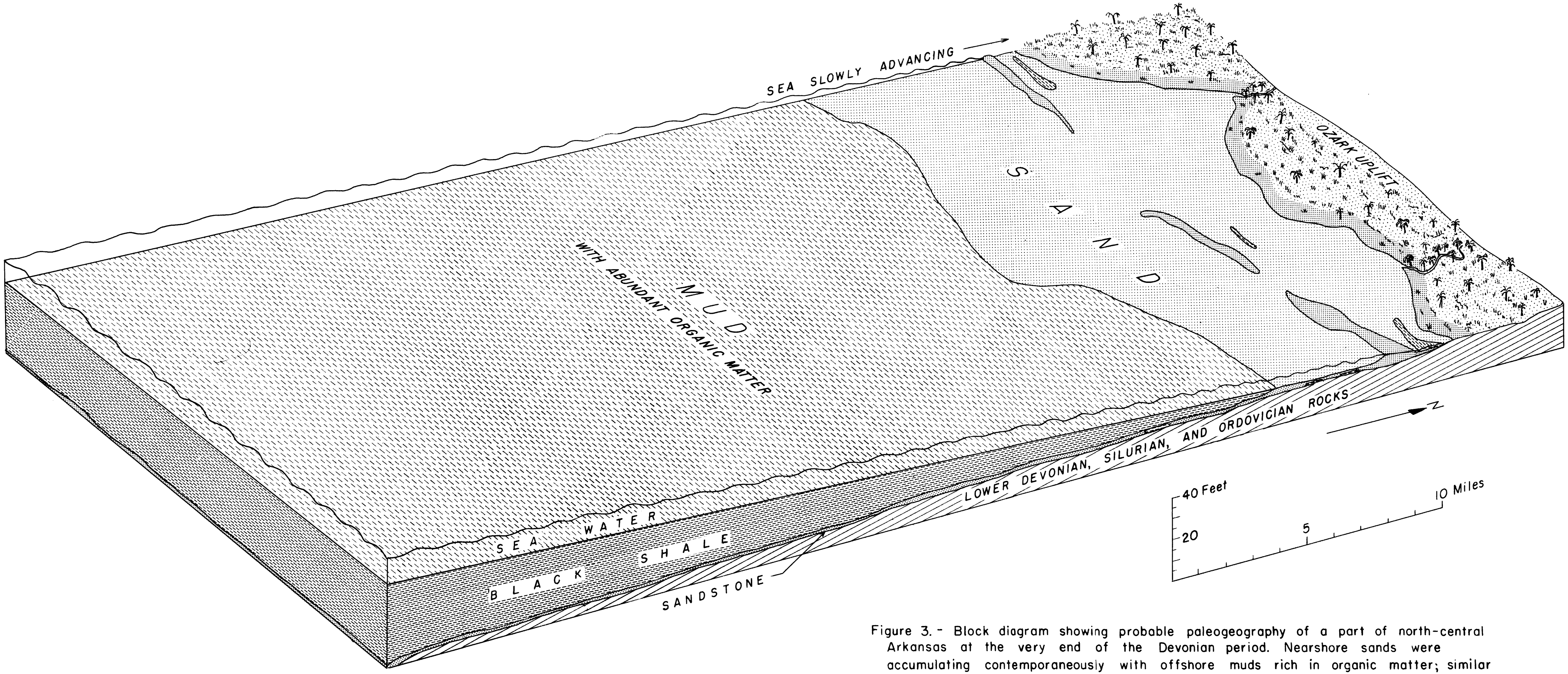


Figure 3. - Block diagram showing probable paleogeography of a part of north-central Arkansas at the very end of the Devonian period. Nearshore sands were accumulating contemporaneously with offshore muds rich in organic matter; similar mud was also accumulating in local depressions of quiet water nearer shore

ber of the Chattanooga shale or in the sandstone at the base of the Boone formation in north-central Arkansas may similarly be either of Late Devonian or Kinderhook age.

Figures 2 and 3 present one possible interpretation, consistent with the available information, of the conditions that may have prevailed in north-central Arkansas during the Late Devonian and Early Mississippian. The diagram on figure 2 represents a generalized restoration of the section extending from the Arkansas-Missouri line southward for about 70 miles. The Sylamore sandstone member of the Chattanooga shale is shown as continuous with the sandstone at the base of the Boone formation. As illustrated, this sandstone is a time-transgressing unit that is of Late Devonian age to the south and of Early Mississippian age to the north. The main body of black shale in the Chattanooga shale thins to zero, but small bodies of shale probably were deposited in small depressions on the sea floor at scattered positions north of the margin of continuous shale deposition. Some of these local deposits might be even slightly younger than the main deposit of black shale.

Paleogeographic relations that could account for the somewhat erratic distribution of black shale lenses in the sandstone are represented on figure 3. The Late Devonian sea is shown at a stage when it was gradually encroaching on the flank of the Ozark upland. The shoreward sand deposits were derived from detrital residuum of the Ozark upland; the shale was derived from the same residuum from clay- and silt-size material that had remained in suspension longer. The latter material probably was supplied in smaller quantity, but was supplemented by organic material from both terrestrial and pelagic organisms. Water circulation in the areas of shale deposition was restricted, and anaerobic bottom conditions prevailed to account for preservation of the carbonaceous matter. Only very locally were the carbonaceous muds deposited with the nearshore sands; these small areas of mud preservation were probably minor depressions on the sea floor, semi-protected from bottom currents and severe wave action, possibly by offshore bars as shown on figure 3. A much greater abundance of vitrainized fragments of the land plant *Callixylon* in the black shale lenses than in the main body of black shale to the south also suggests that these lenses were deposited near to shore.

An almost identical set of conditions has been described as having prevailed at the same time in central Tennessee (Conant and Swanson, 1961, p. 41, fig. 13). There, detailed study showed the Chattanooga shale of Late Devonian age to thin to extinction on an island-area of several hundreds of square miles, known as the Hohenwald platform. The basal sandstone of the Chattanooga shale, which there too rests unconformably mainly on Ordovician limestone and contains phosphatic pellets, is a sandstone that transgresses in age from early Late Devonian where it underlies black shale, to Kinderhook where it is the basal sandstone of the Maury formation of Mississippian age on the Hohenwald platform. It seems reasonable to believe that the same general circumstances of deposition might be interpreted for similar rocks adjacent to the Ozark uplift as on a part of the ancient Nashville dome.

Other interpretations of the origin and intra-age relations of the Upper Devonian and Lower Mississippian rocks in north-central Arkansas are possible using the limited and inconclusive facts now available. For example, Ernest E. Glick (written communication, 1960) has suggested that early in Late Devonian time a shallow sea may have spread over a peneplane that existed in all of northern Arkansas and the Ozark area. For many millions of years this sea reworked the residuum which had formed prior to inundation, resulting in a lag concentrate similar to that described by Rich (1951, p. 2026-2027); numerous pockets of black mud were deposited; most of them were subsequently destroyed, but some were preserved. According to this interpretation, age designations based on conodonts are considered indefinite and the age of the black shale lenses and the enclosing sandstone is taken to range from early Late Devonian to Early Mississippian (Osage) time.

The authors thus interpret the basal sandstone of the Boone formation in north-central Arkansas to be lithologically continuous with the Sylamore sandstone member of the Chattanooga shale; however, until detailed mapping proves that the two sandstones are parts of the same time-transgressing sandstone unit, no revision or extension of existing nomenclature is recommended. In general, the Sylamore contains Late Devonian fossils in the subsurface and outcrop areas of Northern Arkansas, northeastern Oklahoma, and southwestern Missouri where it underlies the black shale of the Chattanooga shale. In general, the basal sandstone of the Boone formation of McKnight (1935) and Maher and Lantz (1952, 1953) in north-central Arkansas, with its lenses of black shale, contains either latest Devonian or earliest Mississippian fossils, or both.

DESCRIPTION OF URANIFEROUS SHALE

Lithology

The uraniferous black shale deposit in north-central Marion County is sandwiched between two beds of fine-grained, fairly well-sorted sandstone. As shown in figure 4, the contacts of the shale with the sandstone, particularly the lower one, are very irregular, and, though the lateral extent of the black shale could not be determined exactly, the shale probably does not extend more than at most a few tens of feet in any direction from its outcrop. The shale thus is probably in the form of an uneven-surfaced lens or pocket in the sandstone. Its maximum thickness is about 2 feet, but the shale thins from 2 feet to as little as 6 inches along the outcrop. Most of the "black" shale is actually medium dark gray to dark gray, with a few thin grayish-black streaks; where the outcrop is wet, the shale appears as glistening black.

In general, the black shale at this locality is lithologically very similar to most of the black shale in the Chattanooga shale in other parts of Arkansas. On close inspection, however, several differences are noted. The sorting of the shale within the bed is poor, with one or more layers near the base having a silty or even a sandy texture, as do several discontinuous streaks scattered throughout the shale. On the west side of the outcrop a thin lens of quartz sandstone in the middle of the shale has a maximum thickness of 1½ inches and a lateral extent of 3 feet. Similar lithologic variations were also observed in the black shale at the locality northeast of Everton.

Some of the silty or sandy streaks and layers are "coquinas" of black to gray conodonts, concentrations of dark reddish-brown to black spore-like orbs and discs, aggregates of pyrite crystals and blebs, or mixtures of all three types of material. Commonly associated with these, but also irregularly scattered through much of the shale, are flattened fragments, or their impressions, of coalified plant stems. These fragments are as much as 4 inches in length and are probably of the genus *Callixylon,* which is commonly found in the Chattanooga shale in many other areas. The conodont collections studied by W. H. Hass were taken from "coquina" layers at the points indicated on figure 4.

The shale at this outcrop also contains many fractures, and slickensided surfaces are not uncommon. For this reason, and because of the poor sorting described, the weathered shale is mainly a mass of small, irregularly shaped chips, rather than the thin sheets of shale typical of the weathered Chattanooga shale elsewhere. Where fresh, however, the shale is just as massive and difficult to break loose as in other outcrops of unweathered Chattanooga shale.

The relatively small size and lenticularity of the shale body, its position in and its uneven contacts with sandstone, the poor sorting and irregular bedding within the shale, and the scattered concentrations and relative abundance of conodonts and carbonaceous debris indicate that this shale accumulated near the shore of a sea in a shallow (about 50 feet deep or less), semiprotected depression on the sea floor. The lower layer of sand, the first deposit of an advancing Devonian sea, was undoubtedly laid down in very shallow waters where wave action and currents existed; this water movement temporarily was greatly lessened, possibly by the buildup of an offshore bar on the seaward side, so that the finer silts and muds, with much organic debris, were swept into the depression where they were not disturbed and destroyed but were deposited, buried, and preserved. Subsequently, the protected position no longer existed, probably because of the northward migration of the protecting bar or the filling up of the minor depression, or both, so that the muds were covered and buried by sands being carried across and deposited on the sea floor. A stage in this interpreted sequence is shown on figure 3.

Petrography

Clastic minerals

Most of the black shale at the uraniferous shale locality in Marion County is estimated to contain 75 to 80 percent of detrital minerals; the remainder consists mainly of organic matter and pyrite. Some thin layers, however, are almost 100 percent detrital minerals, and others, described under organic matter, contain less than 50 percent of these minerals.

Viewed in thin section, the shale has a somewhat similar appearance to most black shales. Organic matter is disseminated throughout the rock and partly masks the clay- and silt-sized mineral matter; the organic matter is estimated to make up from 20 to 60 percent of the shale in 5 thin sections studied. The following quantitative listing of the materials in a thin section of shale bordering a layer of abundant spore-like

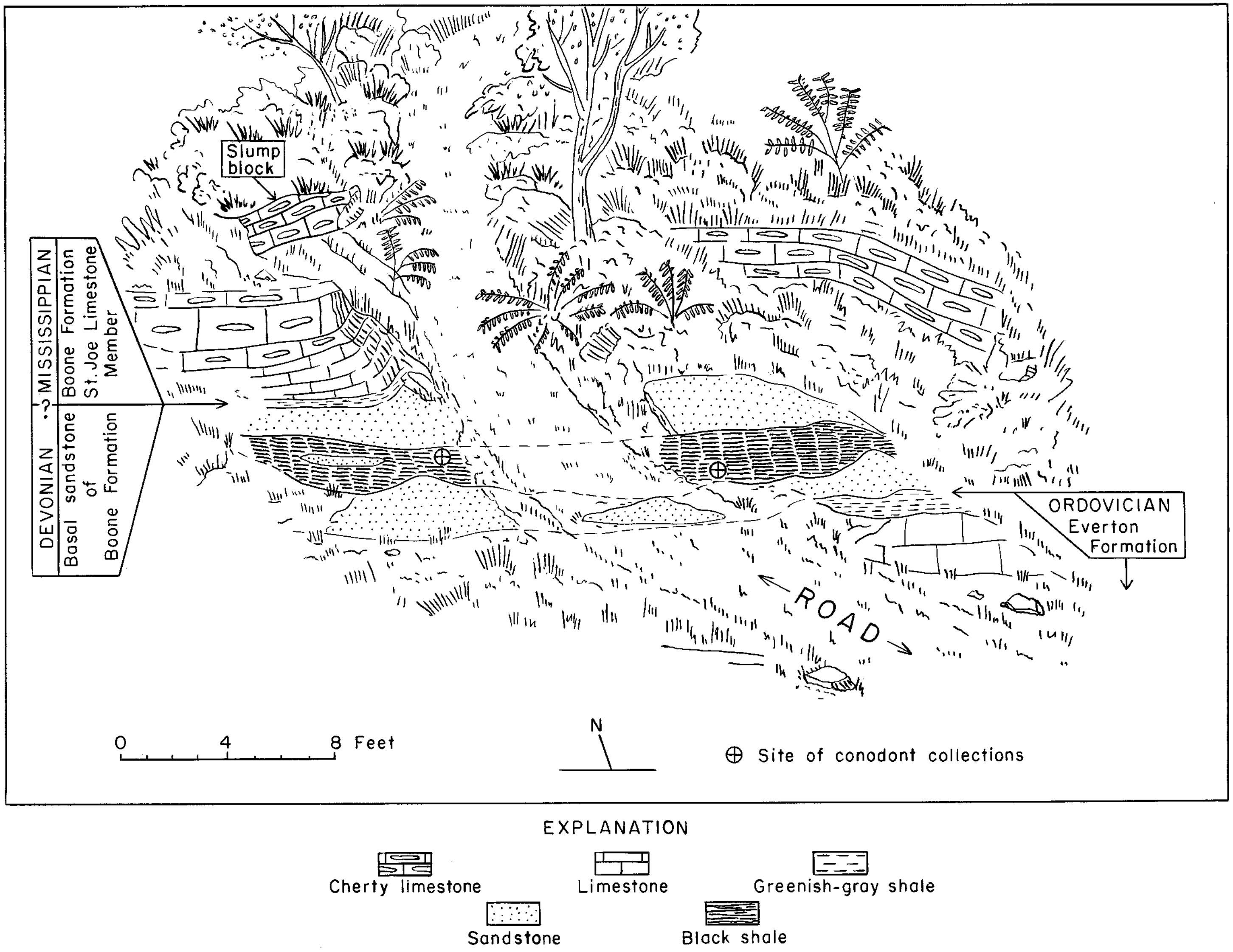


Figure 4. **Diagrammatic sketch of outcrop in north-central Marion County showing lens-like body of black shale in the basal sandstone of the Boone formation of McKnight (1935).**

bodies was prepared by Charles Felix, U.S. Geological Survey:

	Percent
Pyrite	5.7
Platy minerals (mica?)	1.4
Clayey minerals	27.6
Other minerals, largely quartz	7.4
Humic matter	35.2
Brown organic matter	19.5
Sub-anthraxylon	1.5
Walls of spore-like *Tasmanites*	.7
Red, resinous plant remains	.5
Yellow, wax-like plant remains	.2
Conodont(?) fragments	.1
Total	99.8

The major difference between the shale at the central Marion County locality and most other marine shales similar to the Chattanooga shale is the poorer sorting of the materials making up the rock. The lamination of mineral-rich layers and layers containing abundant organic matter, which is characteristic of some other black shales, is very poorly developed in much of the Marion County shale; silt-sized quartz grains in some zones are almost randomly scattered. The initial orientation of the platy minerals and some of the fragmented organic material on deposition, and the subsequent orientation of some clayey minerals and flattening of organic material on compaction, however, partly disguises what originally must have been relatively poor sorting of the fine-grained particles.

A feature of the shale in the thin sections studied that is not very common in other shales similar to the Chattanooga is the presence of discrete masses of black colloidal humic material. Most of these masses are elongated parallel to the bedding and have slightly irregular outlines; they appear to have once been sub-spherical gelatinous bodies which were greatly flattened during compaction of the rock.

No uranium minerals were observed in the thin sections studied. An autoradiograph of a polished section of chips from a sample containing spore-like material, colloidal humic material, pyrite, and black shale showed uranium to be largely concentrated in the colloidal humic material, with much smaller amounts in the spore-like material and in the bulk shale, and none in the pyrite.

Organic matter

Two sets of samples, each containing 3 specimens, were submitted to the Geological Survey's laboratory at Columbus, Ohio, for detailed study. The samples were representative respectively of the main mass of shale, and of shale consisting mostly of three main types of organic material, namely spore-like bodies, coaly plant stem fragments, and black colloidal humic material. The following descriptions of these materials are taken primarily from the reports prepared by James M. Schopf, Charles Felix, and Marcia Winslow; chemical analyses of some of the carbonaceous substances are given in tables 1 and 2.

Coalified plant stems are fairly abundant, but randomly scattered in the shale, and generally are oriented parallel to bedding planes. These stems are longitudinal fragments that are 1 to 4 mm in thickness, 2 to 6 cm wide, and as much as 15 cm in length. The coalified stems probably are driftwood, and closely resemble the coalified wood from the Ohio and Chattanooga shales that is identified as *Callixylon.* The coalified plant substance itself is a black, vitreous, brittle vitrain. A very thin pyritic layer that shows a woody cellular structure commonly coats the surface of the fragments at the vitrain-shale contact.

One of the most distinctive characters of the black shale on close observation with a hand lens is the abundance of ill-defined, discontinuous layers made up predominantly of minute spore-like microfossils known as *Tasmanites.* Some of these layers or stringers have the appearance of a fine-grained oolite. The thickness of these layers ranges from a few millimeters to an observed maximum of about 15 mm. Rounded, slightly frosted grains of quartz of fine-sand size, and bar- and plate-like conodonts are commonly disseminated among the *Tasmanites* in these layers. Elsewhere in the shale, the *Tasmanites* are sparsely scattered, are somewhat flattened to a disc-like form, and can be observed with the naked eye as minute black specks in the dark gray shale.

The *Tasmanites* forms are made up of reddish-brown translucent walls or coats 6 to 20 microns in thickness which originally were spherical in shape, but are generally somewhat flattened by the weight of the overlying sediments. They generally are filled with fine quartz and clay silt, abundant colloidal humic material, and abundant disseminated pyrite, and are enclosed in a matrix of the same materials. Some of these *Tasmanites* specimens are shown on plate 1.

According to Marcia Winslow, the spore-like microfossils represent two rather well defined species, both of which are present in about equal abundance in the samples submitted. The larger form, which is about 0.5 mm in diameter, is

Tasmanites cf. *T. huronensis,* a species that is common in the lower part of the Ohio shale and is generally confined to rocks of Late Devonian age. The second species, which is about 0.2 mm in diameter, is *Tasmanites* sp. nov. (to be described by Winslow), which has a range in Ohio from Middle Devonian to Early Mississippian. These age designations are compatible with those given by Hass based on conodonts. The *Tasmanites* and the conodonts suggest principally a marine site of deposition.

Although the major part of some selected specimens of the shale consist of the *Callixylon* and *Tasmanites* types of organic matter, a black, amorphous type of organic material is by far the most abundant carbonaceous matter in the deposit as a whole. This material is difficult to describe because it lacks definable morphologic features, and it probably contains several different kinds of organic substances. Varieties can be distinguished petrographically, however, by degree of translucency and color. Terms such as humic matter, colloidal humic material, opaque organic material, and opaque attritus all seem appropriate; but, whatever this carbonaceous material is called, it represents an indurated, diagenetically altered mixture of submicroscopic plant shreds, flocculated humic acids, which are a common and abundant decompositional product of decaying plants, and possibly other degraded plant substances. Here, this type of carbonaceous matter is called colloidal humic material.

Colloidal humic material is present throughout the black shale, and makes up much of the matrix in which other identifiable minerals and organic types are embedded. Megascopically it is very dense, hard, and relatively homogeneous in appearance, comparable to fusain in coal. In thin section it generally is black and opaque or very dark reddish brown and only slightly translucent by transmitted light. This humic material has no definite organized structure, though in places it can be described as having a flocculent or ropy texture. Most of the gray to black areas in the photographs on plate 1 are colloidal humic material.

It is this colloidal humic material, particularly the dense opaque humic matter, that contains much or all of the uranium in the black shale in Marion County. This material was apparently deposited fast enough in a few places to form small, discontinuous seams or flattened nodules that contain relatively minor amounts of detrital sediment and other organic matter. The seams or nodules are rarely more than a few centimeters in greatest length, and range from 1 to 20 mm in thickness; their boundaries are not line-sharp but are somewhat gradational through a millimeter or two into the enclosing sediment. The seams are generally a semi-lustrous coalblack substance having a subconchoidal fracture; the nodules are generally larger and have a mixture of very fine detrital minerals and other organic material scattered through them. Both are in normal sedimentary relation with the enclosing shale, with their longest dimensions parallel to bedding planes. The seams or nodules are fairly easy to find in the shale as they are darker than the rest of the shale and glisten in the sunlight; about 15 specimens of shale containing them were collected at the Marion County locality. These seams differ from the *Callixylon* fragments in that the seams of abundant colloidal humic material do not have sharp flat boundaries, they are less glossy, and they have varying amounts of disseminated mineral matter and *Tasmanites.*

CHEMICAL DATA

Uranium

Attention was first called to the black shale of central Marion County by Mr. C. E. West of Bentonville, Ark., who had observed the abnormally high radioactivity of the shale while prospecting. Using a Geiger counter, Mr. West collected and submitted 2 samples of the most radioactive parts of the shale through the U. S. Atomic Energy Commission to the U. S. Geological Survey laboratories for uranium analyses. One of these samples contained 0.71 and the other 0.12 percent uranium. On being informed of black-shale samples that contained about 100 times more uranium than black shales commonly contain, Swanson made arrangements with Mr. West to visit the locality with him. The exact location and stratigraphic position of Mr. West's samples were thus determined and the study which led to this report was initiated.

Shale, as the most abundant type of sedimentary rock, contains an average of 0.0003 or 0.0004 percent uranium. Black shales probably have an average of 0.0008 percent uranium; and marine black shales, excluding the thick marine black shales deposited in geosynclinal areas, have an average uranium content of about 0.0020 percent and a general range of 0.0008 to 0.0250 percent. Prior to the discovery of the marine black shale in Marion County, the highest uranium content of samples from a marine black shale in the United States was found in coaly *Callixylon* remains from the Gassaway member of the Chattanooga shale near Nashville, Tenn. This coaly material contained 0.025 to 0.035 percent uranium, and has been described by

Breger and Schopf (1955). Greater concentrations of uranium in a marine black shale have been known for many years, however, from descriptions and analyses of kolm lenses (0.1 to 0.7 percent uranium) in the alum shales of southern Sweden.

Samples of the Chattanooga shale from 18 localities in Arkansas, Oklahoma, and Missouri have an average uranium content of 0.0020 or 0.0030 percent, with a range of less than 0.0010 percent to 0.013 percent (Landis, 1958, p. 202-203). Bulk shale at the localities northeast of Everton and in central Marion County contains an average of about 0.0060 percent, with a range of 0.0020 to 0.0120 percent.

All of the uranium determinations on samples from the central Marion locality are given in table 1 in which uranium analyses are listed for

Table 1. Uranium analyses of samples from the black shale in central Marion County.

[Analysts: C. G. Angelo, G. Daniels, R. Daywitt, Mary Finch, Irving Frost, C. Johnson, T. Miller, R. Moore, J. P. Schuch, W. Tucker, and James Wahlberg, U. S. Geological Survey]

Laboratory No.	Sample description	Ash (percent)	Uranium in ash (percent)	Uranium in whole sample (percent)
144440	Handpicked coaly material, probably *Callixylon* fragments.	25.4	0.0160	0.0041
231713	Selected coaly material, probably *Callixylon* fragments.	35.81	.025	.008
144441	Selected from zone with abundant *Tasmanites*	63.2	.0050	.0032
147198	Selected sample, black shale with abundant *Tasmanites*.			.003
147368	do.			.0071
231718	*Tasmanites* "oolite" layer; very little colloidal humic material; very little black shale.			.008
228907	Selected sample from talus of black shale containing some colloidal humic material.	83.5		.011
231719	*Tasmanites* "oolite" layer with about 15 percent colloidal humic material and a small amount of black shale.			.043
226627	Selected sample containing abundant colloidal humic material; C. E. West, collector.	78.1		0.12
232436	Seam of predominantly colloidal humic material.	40.0		.42
228906	Selected sample of layer slightly less than 0.1 foot thick; largely black colloidal humic material.	57.98	.92	.55
226624	Selected sample containing abundant colloidal humic material; C. E. West, collector.	42.8		.71
231721	Representative sample of black shale.	88.64	.005	.004
228904	Channel sample of entire black shale unit where 0.9 ft. thick.			.005
144442	Black shale adjacent to *Tasmanites* zone of sample 144441.	86.0	.0060	.0052
228905	Channel sample of entire black shale unit where 1.3 ft. thick.			.006
147381	Selected sample of black shale in lower 0.5 ft. of shale [1]			.007
147380	Selected sample, black shale with no *Tasmanites* or humic material [2]			.008
147197	Black shale from uppermost 0.1 ft. of shale; few scattered *Tasmanites*.			.009
231720	Channel sample of entire black shale unit where 0.9 ft. thick; includes seam of *Tasmanites* and humic material 0.1 ft. thick.	86.18	.013	.012

[1] 6.3 percent organic carbon, 0.08 carbonate CO_2.

[2] 6.9 percent organic carbon, 0.03 carbonate CO_2.

coaly *Callixylon*-like fragments, *Tasmanites*-rich samples, samples largely of colloidal humic material, and samples composed largely or entirely of black shale of normal organic content (15 to 20 percent organic matter), in that order.

Samples of the black brittle coaly material that makes up the *Callixylon*-like plant stems can be readily flaked off of the shale, which results in relatively uncontaminated material, as indicated by the relatively low ash of these samples (table 1). As shown by the two analyses, this coaly material contains about the same amount of uranium as the enclosing shale; the uranium content of the ash is, of course, considerably higher than that of the shale. By way of comparison, the *Callixylon* fragments from the Chattanooga shale near Nashville, Tenn., contained as much as 0.035 percent uranium, and six samples of similar fragments from western White County in central Tennessee contained 0.006 to 0.026 percent.

Samples containing abundant *Tasmanites* are not difficult to find and separate, but even the *Tasmanites* "oolite" probably contains less than 50 percent of the waxy coats or walls that make up these fossils. Other organic matter, detrital minerals, and pyrite occupy the spaces between the *Tasmanites*, and completely or partly fill the interiors of the sphere-shaped fossils. For this reason, an analysis could not be made of pure *Tasmanites* material, but only of a mixture of these spore-like fossils and other materials. The uranium content of sample 144441 (table 1), which is 0.0032 percent, is the uranium content of this type of mixture. The shale bordering the *Tasmanites-rich* zone (sample 144442) has 0.0052 percent uranium. If this is taken as representative of the matrix material in the mixture, and the *Tasmanites* coats are estimated to make up 40 percent of this mixture, the *Tasmanites* themselves contain only 0.0002 percent uranium. Whether this estimate of 40 percent is correct or not, it must be concluded that pure *Tasmanites* material probably contains very little uranium.

What might be considered a third group of analyses in table 1 includes the samples containing abundant colloidal humic material. The analyses are listed in the order of increasing uranium content, from 0.011 percent to 0.71 percent. As can be seen in the sample descriptions the uranium content increases with increasing proportion of colloidal humic material and with decreasing ash content. The relation is shown graphically by figure 5 in which the content of colloidal humic material is determined indirectly and imperfectly from the ash determinations, verified by visual estimates of the relative amounts of colloidal humic material. These data confirm that the uranium is concentrated mostly in the colloidal humic material.

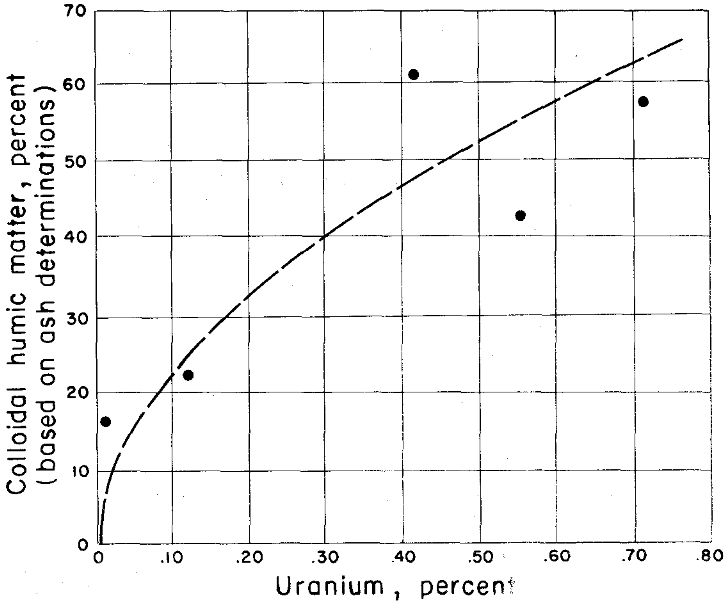


Figure 5. Diagram suggesting a general relation between uranium and colloidal humic material.

Semiquantitative spectrographic analyses

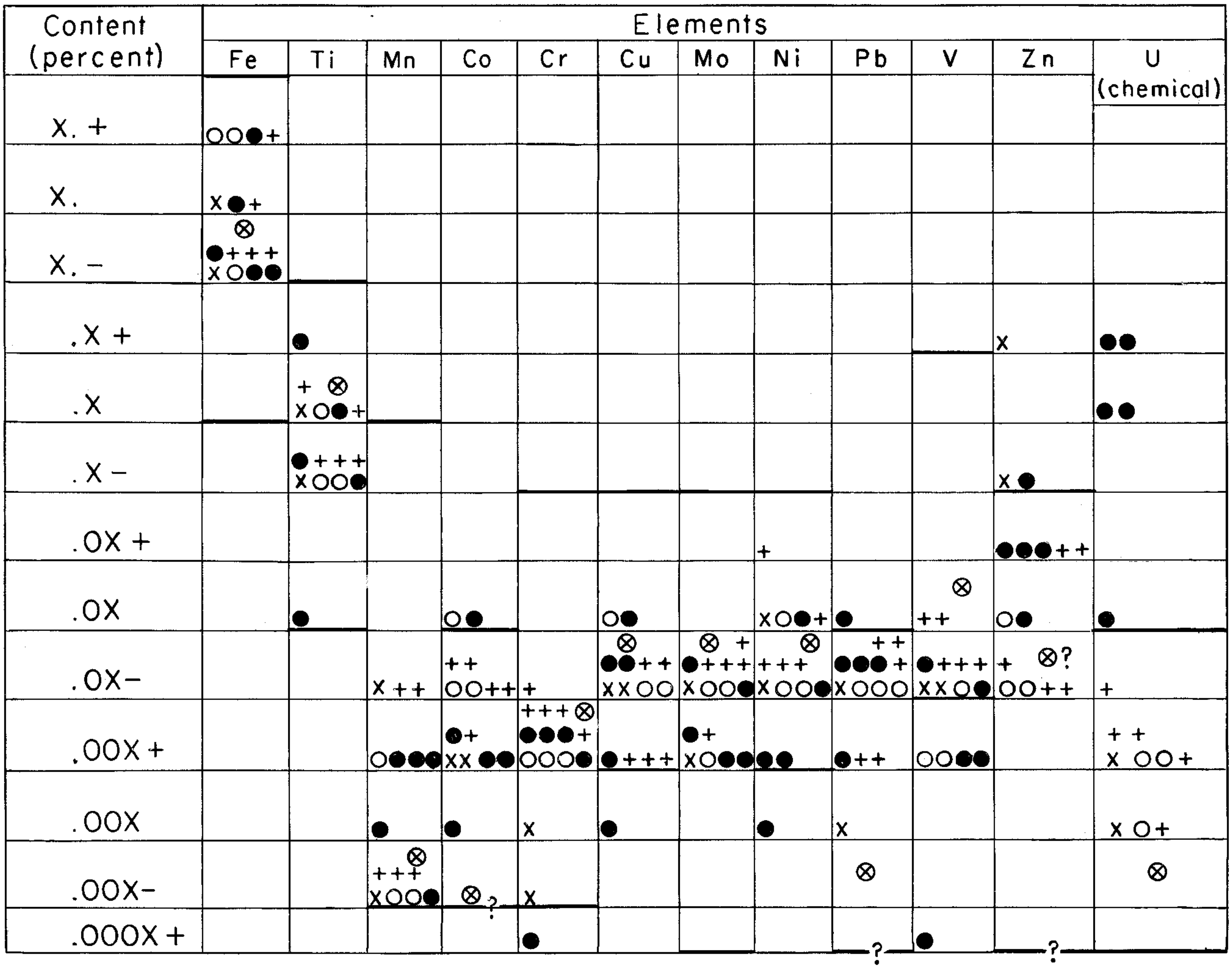
Semiquantitative spectrographic analyses were made of a suite of samples to determine if any metals other than uranium were concentrated in any one of the several constituents of the shale. Table 2 presents the data on 12 metals, including uranium, that are commonly enriched in carbonaceous sediments. Figure 6 shows the distribution of these metals in the samples compared to the modal mean content and range of content of the metals in a suite of 29 samples of the Chattanooga shale and its lateral correlative, the Woodford shale, which were collected in Arkansas and Oklahoma by the authors. The 29 samples include four shale samples plotted on figure 6. Analyses of the other 25 samples are from Landis (1958, p. 205) and from unpublished data.

Comparison of the data shown on figure 6 indicates that the samples from Marion County contain, in addition to uranium, slightly more manganese, cobalt, lead, and zinc, and slightly less vanadium than most of the samples of the Chattanooga and Woodford shales. However, the differences are within or nearly within the range of abundance also shown on figure 6, and it is

Table 2. **Content of a suite of metals in samples from the black shale in central Marion County, as determined by semiquantitative spectrographic analysis.**
[Chemical analyses for uranium shown in parentheses.
[Analysts: Mona Frank, Joseph Haffty, and R. G. Havens, U. S. Geological Survey]

Laboratory No.	Sample description	Elements (percent)											
		Fe	Ti	Mn	Co	Cr	Cu	Mo	Ni	Pb	V	Zn	U
[1] 144440	Coaly fragments, probably *Callixylon*	X.−	0.X−	0.00X−	0.00X+	0.00X−	0.0X−	0.0X−	0.0X−	0.0X−	0.0X−	0.X−	(0.0041)
231713	do.	X.	.X	.0X−	.00X+	.00X	.0X−	.00X+	.0X	.00X	.0X0	.X+	(.008)
[1] 144441	Zone with abundant *Tasmanites*	X.−	.X−	.00X−	.0X−	.00X+	.0X−	.0X−	.0X−	.0X−	.0X−	.0X−	(.0032)
147368	Selected sample, black shale with abundant *Tasmanites*	X.+	.X−	.00X−	.0X−	.00X+	.0X−	.0X−	.0X−	.0X−	.00X+	.0X−	(.0071)
231718	*Tasmanites* "oolite" with no colloidal humic material	X.+	.X	.00X+	.0X	.00X+	.0X	.00X+	.0X	.0X−	.00X+	.0X	(.008)
231719	*Tasmanites* "oolite" with about 15 percent colloidal humic material	X.+	.X	.00X+	.0X	.00X+	.0X	.00X+	.0X	.0X−	.00X+	.0X+	(.043)
232436	Seam of predominantly colloidal humic material	X.	.X+	.00X+	.00X+	.00X+	.0X−	.0X−	.0X−	.0X−	.0X−	.0X+	(.42)
228901	Selected fragments of colloidal humic material (sample 232436)	X.−	.0X	.00X−	.00X	.000X+	.0X−	.00X+	.00X+	.0X−	.000X+	.X−	0.X+
228902	Selected black shale coating fragments of sample 228901	X.−	.X−	.00X	.00X+	.00X+	.00X	.0X−	.00X+	.00X+	.00X+	.0X+	.X
228903	Sample of predominantly colloidal humic material	X.−	.X−	.00X+	.00X+	.00X+	.00X+	.00X+	.00X	.0X	.0X−	.0X	(.71)
231721	Channel sample of black shale; no *Tasmanites;* no humic material	X.	.X	.0X−	.00X+	.00X+	.00X+	.00X+	.0X	.00X+	.0X	.0X+	(.004)
[1] 144442	Black shale adjacent to sample 144441	X.−	.X−	.00X−	.0X−	.0X−	.00X+	.0X−	.0X−	.0X−	.0X−	.0X−	(.0052)
147381	Selected sample of black shale	X.−	.X−	.00X−	.0X−	.00X+	.0X−	.0X−	.0X−	.0X−	.0X−	.0X−	(.007)
147380	Selected sample, black shale with no *Tasmanites*	X.−	.X−	.00X−	.0X−	.00X+	.00X+	.0X−	.0X−	.00X+	.0X−	.0X−	(.008)
231720	Channel sample of black shale; includes some *Tasmanites* and humic material	X.+	.X	.0X−	.0X−	.00X+	.0X−	.0X−	.0X+	.0X−	.0X	.0X+	(.012)

[1] Spectrographic analysis made on ash.



Content (percent)	Elements											
	Fe	Ti	Mn	Co	Cr	Cu	Mo	Ni	Pb	V	Zn	U (chemical)
X. +	○ ○ ● +											
X.	x ● +											
X. −	⊗ ● + + + x ○ ● ●											
.X +		●									x	● ●
.X		+ ⊗ x ○ ● +										● ●
.X −		● + + + x ○ ○ ●									x ●	
.0X +								+			● ● ● + +	
.0X		●		○ ●		○ ●		x ○ ● +	●	⊗ + +	○ ●	●
.0X−			x + +	+ + ○ ○ + +	+	⊗ ● ● + + x x ○ ○	⊗ + ● + + + x ○ ○ ●	⊗ + + + x ○ ○ ●	+ + ● ● ● + x ○ ○ ○	● + + + x x ○ ●	+ ⊗? ○ ○ + +	+
.00X +			○ ● ● ●	● + x x ● ●	+ + + ⊗ ● ● ● + ○ ○ ○ ●	● + + +	● + x ○ ● ●	● ●	● + +	○ ○ ● ●		+ + x ○ ○ +
.00X			●	●	x	●		●	x			x ○ +
.00X−			⊗ + + + x ○ ○ ●	⊗ ?	x				⊗			⊗
.000X +					●				?	●	?	

EXPLANATION

x Analysis of coaly fragments *(Callixylon)*

○ Analysis of *Tasmanites*-bearing material

● Analysis of collidal humic material

\+ Analysis of black shale

⊗ Modal mean content of 29 samples of Chattanooga(11) and Woodford (18) Shales. Includes 4 of the 5 shale samples shown above. Queried where in doubt. (From Landis, 1958, p. 205, and unpublished analyses)

— Range of content in 29 samples of Chattanooga and Woodford Shales. Queried where in doubt or unknown

Figure 6. Distribution of metals in samples from black shale in north-central Marion County (see table 2 for sample descriptions) and from the Chattanooga and Woodford shales in Arkansas and Oklahoma.

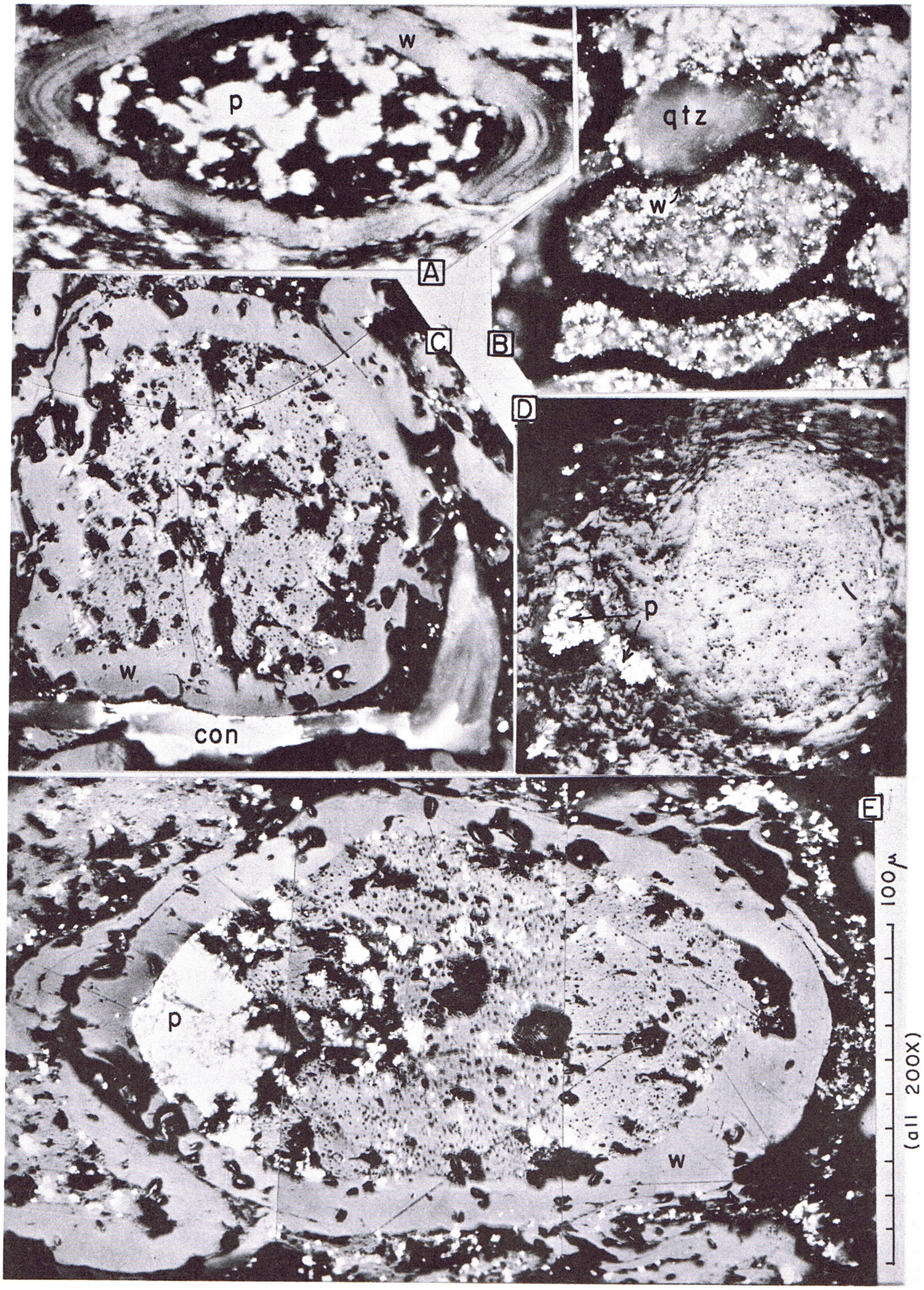


Plate 1. Photomicrographs of organic materials in black shale from central Marion County (photography by R. W. Bouman).

Description of photomicrographs of Plate I

A. Thin section of large, partly compressed *TASMANITES* with laminate wall (w). Dark filling material is humic and red translucent. Enough reflected light was used to illuminate pyritic aggregates (p) in white areas. Photograph chiefly by transmitted light using Ultropak. X200.

B. Surface section (polished) in a silty low-luster area showing small *TASMANITES* with simple wall. Wall (w), which is highly translucent in thin section, appears dark (light absorbing). White specks in filling and matrix are pyritic; gray filling is chiefly silty detrital quartz. One large rounded grain, probably quartz (qtz), is in upper central area. *TASMANITES* disseminules, mostly with silty fillings, are tight packed in this area. In normal shale occurrence the fillings are lacking and the organic bodies tightly compressed. Very little organic matter except *TASMANITES* coats are present in the silty area. Photographed using oil immersion and vertical illumination. X200.

C. Surface section (polished) across an "oolitic" high-luster area, showing large *TASMANITES* uncompressed, with humic organic filling, and a conodont (con). In thin section the *TASMANITES* wall and humic filling material appears opaque (black); in this photograph (taken by reflected light) it appears gray and shows greater detail. Both pits (induced in polishing) and transparent mineral matter appear dark; white specks are disseminated pyrite. The conodont shows internal reflection and for this reason appears gray or whitish (note cone structure on the right). The white areas of the "shank" are in part pyritic; a very thin pyrite vein runs obliquely across the fossil. Such pyritic veinlets are uncommon. Photographed using oil immersion and vertical illumination. X200.

D. Surface section (polished) in a high-luster area showing an organic spheroid with micropunctation, as in the filling material shown in C, but without any enclosing wall. Dispersed flocculent organic matter (gray) in transparent mineral matrix (black) surrounds the spheroid. White specks and aggregates represent disseminated pyrite (p). Photographed with vertical illumination as in C. X200.

E. Surface section (polished) in a high-luster area similar to that shown in C, showing the larger and more characteristic form of opaque *TASMANITES*, with micropunctate organic filling. Dark elliptical spots represent pits induced in polishing; one internal edge of the wall (w) is outlined by a dense aggregate of pyrite (p). Dark areas outside the wall are chiefly detrital transparent minerals. X200.

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concluded that the metals shown in table 2 and on figure 6 do not have any direct relation to uranium. Rather, the very minor differences reflect slight differences in the chemical environment in which the black shales were deposited, or possibly very small differences in the available supply of the different metals.

× Analysis of coaly fragments *(Callixylon)*
● Analysis of colloidal humic material
× Modal mean content of 29 samples of Chattanooga (11) and Woodford (18) shales. Includes 4 of the 5 shale samples shown above. Queried where in doubt. (From Landis, 1958, p. 205, and unpublished analyses)
o Analysis of *Tasmanites*-bearing material
+ Analysis of black shale
——Range of content in 29 samples of Chattanooga and Woodford shales. Queried where in doubt or unknown

Comparison of the metal distribution shown on figure 6 also indicates that, with the exception of uranium, none of the metals are abnormally concentrated in the samples of organic materials, as compared to the bulk shale samples.

Other analyses

Analyses for germanium were made on 5 samples of the shale from Marion County; all samples, which included 2 samples of the coaly *Callixylon*-type material, contained less than 0.0010 percent germanium, which is within the general range of this metal in many kinds of carbonaceous rocks.

Two samples of the shale were analyzed for phosphate inasmuch as phosphate is known as a common holder of uranium in sedimentary rocks. Both samples contained less than 0.75 percent P_2O_5, which is an amount common in other black shales. Two samples of the shale analyzed for carbonate contained less than 0.1 percent carbonate which confirms the estimate made by petrographic examination of the shale.

ORIGIN OF URANIUM

Stratigraphic, lithologic, and chemical information on the black shale near the base of the Boone formation in central Marion County suggests conditions under which uranium was concentrated in the shale.

It seems reasonable that most or all of the uranium was concentrated in the shale just prior to, during, or shortly after deposition in marine water. As nearly as can be determined, general associations of uranium in this shale and the average uranium content of the shale are the same as in shale of about the same age in much of the eastern and central United States, and in other uraniferous marine black shale elsewhere whatever the age. The major difference is that certain constituents of the shale in Marion County contain considerably more uranium than known elsewhere in marine black shale, with the exception of the kolm lenses in the Cambrian shale of southern Sweden.

Most of the uranium in this shale apparently is in, and was concentrated by, the organic matter. This is borne out by the analyses of selected samples of relatively pure organic material. Of the three main types of organic matter, the colloidal humic material contains the most uranium—on the order of 0.5 percent. Locally the physical appearance and associations of the colloidal humic material suggest that some of it was once a gel of flocculated humic acids.

Szalay (1954, 1958) has shown that humic acids, the colloidal decomposition product of decaying woody types of plants that have dissolved in alkaline water, are capable of sorbing uranium from solution. These uranium-bearing humic acids may then be flocculated or precipitated as a dark-brown gel by lowering the pH of the solution to the acid side or by the addition of divalent cations such as calcium. The flocculated humic acids may contain several percent uranium by dry weight. Manskaya, Drozdova, and Emelianova (1956) have experimentally verified this process, and Vine, Swanson, and Bell (1958) discuss this mechanism of uranium enrichment in sedimentary rocks, including marine black shale.

Very likely the black opaque organic matter observed in the Marion County black shale was deposited at a uranyl humate, or flocculated humic acid, which picked up uranium while in solution enroute to its site of deposition. The presumed paleogeographic position of this shale body near the shore of a sea possibly explains the presence of flocculated humic acids in relatively large masses and pure form, as compared to the dissemination of humate through other shale deposited at greater distances from the shoreline.

The composition and physical properties of the colloidal humic material in the Marion County shale, though not precisely determined, are very similar to the kolm in the Cambrian black shale of Sweden. Kolm, which has the appearance of the seams of colloidal humic material, generally contains 0.1 to 0.7 percent uranium; the samples from Marion County having large concentrations of colloidal humic material show almost the same range in uranium content. The exact origin of the kolm has not been generally agreed on to the writers' knowledge, so further comparison is not warranted at this time.

CONCLUSIONS

The sandstone at the base of the Boone formation in north-central Arkansas is believed to be a part of an areally extensive time-transgressing sandstone that is called the Sylamore sandstone member of the Chattanooga shale in areas to the south and west in Arkansas. The age of this sandstone, based mainly on conodont assemblages, is generally Late Devonian where it underlies the black shale of the Chattanooga, but, as the initial deposit of a northward-advancing sea, it becomes progressively younger toward the north, where it is either Late Devonian or Early Mississippian (Kinderhook), or both.

The stratigraphic relations of the several lenses of marine black shale in the sandstone at the base of the Boone formation indicate that these shale bodies were probably deposited near the shore of the sea as it advanced northward on the southern flank of the Ozark uplift. Fine-grained carbonaceous sediment in which fragments of the land plant *Callixylon* were abundant probably accumulated in minor depressions on the sea floor partly protected from current and wave action, possibly because the depressions were located on the landward side of offshore bars.

One of the lenses of shale in this sandstone in north-central Marion County contains organic material having the largest uranium content (0.1 to 0.7 percent) known from any black shale in the United States. The black opaque organic material is called colloidal humic material and consists in large part of material interpreted to have once been soluble humic acids that sorbed uranium from river and sea water prior to being deposited as a gel on the sea floor. In uranium content and general appearance, the colloidal humic material is similar to the kolm in the Cambrian alum shale of southern Sweden. Fragments of black coaly material, probably driftwood of the plant *Callixylon,* contain only about 0.005 percent uranium; and the spore-like bodies of the genus *Tasmanites,* so abundant in some thin layers as to appear as an oolite, probably contain considerably less than 0.001 percent uranium. None of the three types of organic matter, nor the inorganic constituents of the shale, contains other metals in significantly different quantities than is common to carbonaceous shale elsewhere.

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