Selected annotated bibliography of the geology and occurrence of uranium-bearing marine black shales in the United States

By Carolyn E. Fix

Trace Elements Investigations Report 535

UNITED STATES DEPARTMENT OF THE INTERIOR
GEOLOGICAL SURVEY

Geology and Mineralogy

UNITED STATES DEPARTMENT OF THE INTERIOR

GEOLOGICAL SURVEY

SELECTED ANNOTATED BIBLIOGRAPHY OF THE GEOLOGY AND OCCURRENCE OF URANIUM-BEARING MARINE BLACK SHALES IN THE UNITED STATES*

By

Carolyn E. Fix

November 1956

Trace Elements Investigations Report 535

This preliminary report is distributed without editorial and technical review for conformity with official standards and nomenclature. It is not for public inspection or quotation.

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

USGS - TEI-535

GEOLOGY AND MINERALOGY

Distribution	No. of copies
Atomic Energy Commission, Washington	2
Division of Raw Materials, Albuquerque	1
Division of Raw Materials, Austin	1
Division of Raw Materials, Casper	1
Division of Raw Materials, Denver	1
Division of Raw Materials, Ishpeming	1
Division of Raw Materials, Phoenix	1
Division of Raw Materials, Rapid City	1
Division of Raw Materials, Spokane	1
Division of Raw Materials, Washington	3
Division of Raw Materials, Salt Lake City	1
Exploration Division, Grand Junction Operations Office	1
Grand Junction Operations Office	1
Pennsylvania State University (Spackman)	1
Technical Information Service Extension, Oak Ridge	6
U. S. Geological Survey:	
Fuels Branch, Washington	2
Geochemistry and Petrology Branch, Washington	1
Geophysics Branch, Washington	1
Mineral Deposits Branch, Washington	1
P. C. Bateman, Menlo Park	1
A. L. Brokaw, Grand Junction	1
N. M. Denson, Denver	1
V. L. Freeman, College	1
R. L. Griggs, Albuquerque	1
W. R. Keefer, Laramie	1
R. A. Laurence, Knoxville	1
L. R. Page, Washington	1
P. K. Sims, Denver	1
Q. D. Singewald, Beltsville	1
V. E. Swanson, Denver	1
A. E. Weissenborn, Spokane	1
TEPCO, Denver	4
TEPCO, RPS, Washington, (including master)	3
	47

CONTENTS

ILLUSTRATION

SELECTED ANNOTATED BIBLIOGRAPHY OF THE GEOLOGY AND OCCURRENCE OF URANIUM-BEARING MARINE BLACK SHALES IN THE UNITED STATES

By Carolyn E. Fix

INTRODUCTION

The occurrence of uranium in marine black shales was first discovered in 1893 by Nordenskiold[1] while analyzing samples of alum shale of Late Cambrian age in southern Sweden. In 1909 this shale was mined for its radium content[2] and in 1948 uranium was being recovered from this shale[3]. In recent years this shale also has been mined and processed for its oil content.

[1] Nordenskiold, A. E., 1893, Rémarques sur le fer natif d'Ovifak et sur le bitumen des roches cristallines de Suède: Acad. Sci. Paris Comptes Rendus, v. 116, p. 677-678.

[2] Nininger, R. D., 1954, Minerals for atomic energy: New York, D. Van Nostrand Co., Inc., 367 p. See p. 76.

[3] Anonymous, 1953, Swedish uranium production: S. African Mining and Eng., v. 64, no. 3146, p. 521.

Since the first discovery was made, uranium has been found in many black shales in many parts of the world. In the United States numerous marine black shales, chiefly of Paleozoic age, have been examined in the recent search for uranium deposits. Although many of these marine black shales contain slightly more uranium than marine sedimentary rocks in general, only a few of these units contain more than 0.005 percent uranium. Because of their wide areal distribution and uniform uranium content, marine black shales are considered to represent vast but low-grade resources of uranium. None of these uraniferous shales, however, is considered to be of commercial significance at the present time.

Some of the domestic black shales have been studied in considerable detail in order to determine the distribution, and the geologic factors controlling the distribution, of their uranium. These include the Chattanooga shale of Devonian and Mississippian age in southeastern United States; the Woodford shale and its correlatives of Devonian and Mississippian age in Oklahoma and adjacent states; the Middle and Lower Pennsylvanian shales of Kansas and Oklahoma; the Cretaceous Eagle Ford shale of Texas; and the Sharon Springs member of the Pierre shale of Cretaceous age in South Dakota and Nebraska. Other black shales that have been examined include: the Devonian and Mississippian Antrim shale of Michigan; shale in the Mississippian Spergen limestone of Missouri; the subsurface Hartville formation of Mississippian(?), Pennsylvanian, and Permian age in western Nebraska and eastern Wyoming; and black shale beds in the Pennsylvanian cyclothems of the Midcontinent and the Appalachian and Eastern Interior Basins.

In general, most of the uraniferous marine black shales have the following in common: (1) they are relatively thin, widespread deposits, probably representing extremely slow deposition over a long period of time; (2) they are carbonaceous, containing abundant plant matter; (3) sulfides and some phosphate are characteristically present; (4) they contain little or no calcium carbonate; and (5) almost all are Paleozoic or older in age.

Two main problems that have arisen in connection with the investigations of uranium-bearing shales are the mineralogical association of the uranium and the mode of origin of the uranium. Intensive laboratory studies have been made, but no uranium-bearing minerals have been identified to date, and the physical separation of a fraction where the uranium is concentrated has been only partially successful. The uranium seems to be finely disseminated and is associated with the organic matter in some shales and possibly with the clay fraction in others.

All investigators seem to agree that the uranium was deposited at or about the same time as the enclosing sediments in an organic- and sulfide-rich reducing environment, but the paleo-geochemistry of precipitation and/or adsorption of the uranium, and the relationship between paleogeographic conditions and uranium concentration are still in question.

EXPLANATION OF THE BIBLIOGRAPHY

The bibliography consists of annotations or abstracts of selected reports that pertain to the geology and occurrence of uranium in marine black shales and their metamorphic equivalents in the United States. Only those reports that were available to the public prior to June 30, 1956, are included. Most of the reports may be consulted in the larger public, university, or scientific libraries. A few reports that have been released to the public in open file may be consulted at designated offices of the Geological Survey.

An effort has been made to include only those references to shales whose uranium is believed to be of syngenetic origin and whose major source of radioactivity is uranium. Many general papers on the geology of uranium deposits refer to marine black shales, and some of these general papers have been included.

The ideas and opinions expressed in the annotations are those of the original authors. In places additional information is included by the annotator for the sake of clarity and is enclosed in brackets when placed within an author's quotation. An asterisk (*) preceding a geologic name indicates that the name has not been adopted for use in classification in publications of the U. S. Geological Survey.

The annotations are arranged in alphabetical order by author and are numbered consecutively for indexing purposes. Reports by the same author or authors are arranged first in chronological order and second in alphabetical order by title. The annotations are indexed at the end of the bibliography by: (1) author, (2) geographic area, (3) stratigraphic names and geologic ages, and (4) subject.

This bibliography was prepared by the U. S. Geological Survey on behalf of the Division of Raw Materials, U. S. Atomic Energy Commission.

Shown on the index map, figure 1, are: (1) the general location of marine black shales containing more than 0.001 percent uranium; (2) the uranium content based on chemical analysis, or, where indicated by subscript "e", equivalent uranium content; (3) one or more references for each location, indicated by numbers corresponding to the number of the reference in the text of the bibliography.

BIBLIOGRAPHIES ON URANIUM

Additional references to uranium deposits may be found in the following general bibliographies:

Allen, R. E., 1953, Uranium and its compounds, a bibliography of unclassified literature: U. S. Atomic Energy Comm. TID-3041, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Cooper, Margaret, 1951, Preliminary bibliography on uranium and thorium and radioactive carbonaceous deposits: U. S. Atomic Energy Comm. RMO-835, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

_____, 1953, Selected bibliography on uranium exploration and the geology of uranium deposits: U. S. Atomic Energy Comm. RME-4007, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

_____, 1953, Bibliography and index of literature on uranium and thorium and radioactive occurrences in the United States, Part I: Arizona, Nevada, and New Mexico: Geol. Soc. America Bull., v. 64, no. 2, p. 197-234.

_____, 1953, Bibliography and index of literature on uranium and thorium and radioactive occurrences in the United States, Part 2: California, Idaho, Montana, Oregon, Washington, and Wyoming: Geol. Soc. America Bull., v. 64, no. 10, p. 1103-1172.

_____, 1954, Bibliography and index of literature on uranium and thorium and radioactive occurrences in the United States, Part 3: Colorado and Utah: Geol. Soc. America Bull., v. 65, no. 6, p. 467-590.

_____, 1955, Bibliography and index of literature on uranium and thorium and radioactive occurrences in the United States, Part 4: Arkansas, Iowa, Kansas, Louisiana, Minnesota, Missouri, Nebraska, North Dakota, Oklahoma, South Dakota, and Texas: Geol. Soc. America Bull., v. 66, no. 3, p. 257-326.

Croxton, F. E., 1951, Uranium and its compounds - a bibliography of unclassified literature: Carbide and Carbon Chemicals Co., K-25 Plant, Oak Ridge, Report K-295, Part 2.

Postell, P. E., and Voress, H. E., 1953, Unclassified bibliographies of interest to the atomic energy program: U. S. Atomic Energy Comm. TID-3043, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Schwind, S. B., and Croxton, F. E., 1950, Radium. A bibliography of unclassified literature: U. S. Atomic Energy Comm. TID-363, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Wallace, J. H., and Smith, H. B., 1955, Bibliography of U. S. Geological Survey trace elements and related reports to June 1, 1954: U. S. Geol. Survey Bull. 1019-B, p. 63-144.

REFERENCES

1. Alexander, F. M., 1953, The Chattanooga black shale - A possible future source of uranium: Tennessee, Dept. of Conservation, Div. of Geology, Inf. Circ. no. 1, 3 p.; Reprinted from the Tennessee Conservationist, June, 1953.

This small pamphlet was written to present information to the public on the federal government's investigations of the Chattanooga shale in southeastern United States as a possibe future source of uranium. A general description of the lithology, thickness, distribution, and correlative formations of the Chattanooga shale is given.

In 1921 and 1922 the Tennessee Division of Geology sponsored an investigation of the oil potential of the kerogen-rich Chattanooga shale. It was found that as much as 10 to 15 gallons of oil per ton could be extracted from the shale, although this figure varied widely from place to place.

"The fact that marine black shales contain very small amounts of radioactive substances has been known to geologists for many years" but before the "atomic age" was ushered in, this knowledge was only of academic interest. The uranium content of the Chattanooga shale has been reported to range from 0.001 to 0.03 percent but "much of the formation probably contains less than 0.2 pound of uranium per ton of shale." Studies indicate, however, that the marine black shales of eastern and central United States are promising as large potential uranium reserves.

Major problems to be solved before this widespread, low-grade source of uranium can be economically exploited are the development of low-cost methods of mining the shale and extracting the uranium.

2. Bachman, G. O., and Read, C. B., 1952, Trace elements reconnaissance investigations in New Mexico and adjoining states in 1951: U. S. Geol. Survey TEM-443-A, 22 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

In 1951 a reconnaissance was made in New Mexico, Arizona, Colorado and Utah for uranium in coal and carbonaceous shale, chiefly of Mesozoic age; and in marine black shale of Paleozoic age. "Tertiary volcanic rocks, considered to be a possible source for uranium in the coal and associated rocks, were examined where the volcanic rocks were near coal-bearing strata." Except for a coal unit in the Mesaverde formation, on La Ventana Mesa, Sandoval County, N. Mex., which contains as much as 0.62 percent uranium, all other coals and associated rocks tested contain 0.01 percent or less uranium. The equivalent uranium content of tested marine black shales did not exceed 0.004 percent.

The results of the tests for uranium in the marine black shales are as follows:

Martin limestone (Devonian), Gila County, Ariz.:
Four samples from the upper shaly portion contain from 0.003 percent to 0.004 percent equivalent uranium but less than 0.001 percent uranium in sample and in ash (90 percent average ash).

Percha shale (Upper Devonian), Otero County, N. Mex.:
Nine samples contain from 0.001 percent to 0.004 percent equivalent uranium; 2 of these samples were from a calcareous zone and contain 0.004 percent equivalent uranium and 0.002 percent uranium.

Lake Valley limestone (Mississippian), Socorro County, N. Mex.: One sample of calcareous shale contains 0.002 percent equivalent uranium.

Madera formation (Pennsylvanian), Mora County, N. Mex.: One sample of bituminous shale contains 0.002 percent equivalent uranium and 0.001 percent uranium.

3. Bain, G. W., 1950, Geology of the fissionable materials: Econ. Geology, v. 45, p. 273-323. See p. 284, 291-92, 308 and 311.

This report is a general summary of the geological associations, types of deposits, and world resources of uranium. Uranium deposits are classified as either (1) primary or hypogene, (2) sedimentary or bedded and (3) oxidized bodies. Maps showing the locations of the principal uranium deposits of the world are included.

"The uraniferous bituminous source rocks for petroleum are marine deposits that accumulated very slowly. Generally only marine shales yielding over 10 percent of oil by distillation have significant uranium." However, a few slates and schists of shale derivation still retain their uranium, but they have been so metamorphosed as to lose most of their volatile hydrocarbons. "The uranium content of 0.02 percent U_3O_8 or more has been confirmed rarely." Uraniferous shales are from 1 to 17 meters in thickness and underlie hundreds of square kilometers. "Each square kilometer has about 225 metric tons of U_3O_8 for each meter of thickness of 0.01 percent content."

Some typical shales are the Antrim and Chattanooga in the United States, with about 0.01 percent U_3O_8; the alum shale of Narke, Sweden, with 0.023 percent U_3O_8; the Cambrian and Ordovician shales of the Leningrad district, Russia, with from 0.008 to 0.03 percent U_3O_8; and the black siliceous schist(?) of Cambrian to Silurian age of the Ferghana region, Russia, with from 0.03 to 0.08 percent U_3O_8. The high values of the latter seem to be the result of surface enrichment.

The uranium in marine black shales was "a part of the original organic-rich mud laid in brackish or salt water and adsorbed from it at pH 6 to 7.5. The uranium content is generally greater in strata that accumulated very slowly.... Uranium was adsorbed on the clay as phosphate and vanadate crystallites along with other metals." The thin black shales of Pennsylvanian age of the Midcontinent, Illinois-Indiana and western Appalachian regions are also potential large, low-grade sources of uranium.

The geology and reserve data of the uraniferous shales and slates of Russia are discussed in some detail. By analogy to the reserve data of the foreign uranium-bearing black shales, "300 square miles may be expected to be underlain by such strata in the Chattanooga shale area [southeastern United States], and at the average content of 1,125 metric tons per square kilometer, it is not unreasonable to expect up to 810,000 tons of U_3O_8 in this sort of occurrence...."

4. Bates, T. F., Strahl, E. O., Short, N. M., Silverman, E. N., and Camilli, Elena, 1954, Mineralogy and petrography of the Chattanooga shale [abs.]: Geol. Soc. America Bull., v. 65, p. 1230.

"Detailed mineralogical and petrographic analysis of a drill core of the Chattanooga formation has resulted in a clearer picture of the relationship of uranium with the composition and texture of the black shale from the Highland Rim area of Tennessee.

"Thin-section point counts, alpha-track counts, and quantitative mineralogical and chemical analyses of 60 samples from 35 feet of core have yielded statistically significant correlations between uranium and some of the shale constituents. The correlation is positive with respect to 'blackness' of the sample, total carbon, pyrite, and total sulfur, and negative with quartz, feldspar, and clay and with Fe_2O_3. The absence of high correlation coefficients indicates that the uranium is not specific to any single constituent measured. On the other hand certain relationships between uranium content and composition are apparent.

"The upper member [Gassaway] of the formation is darkest, shows the least mineralogical and petrographic variation, and contains the highest concentration of uranium. The four lower members are more heterogeneous and consist of alternating gray and black beds. The gray beds are finer-grained and contain more quartz, feldspar, and clay and less pyrite and organic matter than the black layers.

"Autoradiographic studies demonstrate that the alpha-particle source material is localized about concentrations of organic matter and pyrite." (author's abstract)

5. Beers, R. F., 1945, Radioactivity and organic content of some Paleozoic shales: Am. Assoc. Petroleum Geologists Bull., v. 29, p. 1-22.

Detailed measurements were made of the total alpha activity, the total beta activity and the radium content of some Paleozoic sedimentary rocks taken from oil wells in Osceola County, Mich. and in Oklahoma. A method was developed for the rapid determination of the three radioactive constituents in ancient sediments (uranium, thorium and potassium 40) with the aid of a beta counter. In addition quantitative analyses were made of the potassium, carbon, and hydrogen content of selected samples. A detailed description of the apparatus and methods of measurement used are given, as are several figures and tables of results.

Samples of cuttings of shale from oil and gas wells penetrating the Coldwater, Sunbury, Ellsworth and Antrim shales and Traverse formation of Michigan and the Chattanooga, Woodford and Sylvan shales in Oklahoma were studied. The radioactive elements in these rocks are in three principal loci: 1) in heavy minerals (e.g., zircon), 2) in potassium 40 and 3) uranium and thorium in clays and organic materials. The studies indicate a positive correlation between the uranium and carbon content of the Antrim and Sunbury shales. Positive correlations were found between radioactivity and fineness of particle size; radioactivity and thorium-uranium ratio; and radioactivity and potassium content.

Methods cited as to how the uranium and thorium may have become associated with the sediments making up black shales are: 1) adsorption to clay minerals during sedimentation; 2) inclusion in a stabilized colloidal form in the existing organic colloid; and 3) formation as a compound on, or adsorbed to organic particles.

The high thorium-uranium ratio (8.0) observed in the samples of Antrim shale may have been due to a pH value in excess of 3.7 in the shallow sea that caused the thorium to be precipitated in excess of the uranium. "If all of these elements inhabit a colloidal dispersion at one time, then we have a mechanism which may account for the widespread and uniform distribution of radioactive substances and the correlative organic matter in these black shales." The selective action of the colloidal environment on the distribution of radioactivity and organic matter is illustrated by the fact that the radioactivity increases with the fineness of texture of the Antrim shale.

A hypothesis of the environmental conditions that may account for the distribution of the Sunbury and Antrim shales with their high content of uranium, thorium, potassium and organic matter is set forth as follows. The complete cycle of sedimentary rock types from the Traverse formation through the Coldwater shale indicates a tectonic cycle during their formation. First there was a stable period during which the limestone of the Traverse formation was deposited in a clear, deep epicontinental sea. During this time the land was

gradually weathered and eroded and a deep residual mantle of fine clay and resistant heavy minerals, some of which were radioactive, was accumulated.

The sea became oscillatory as the period of unstability began and transgressed the adjacent low-lying lands. The residual debris was carried into the sea, the heavy and coarse fragments settling first and the fine particles remaining in colloidal suspension until an increased salinity was met in deeper water. Then the fine clay particles with their adsorbed uranium, thorium and potassium and the organic matter were precipitated. The gentle oscillations of the sea may have been responsible for the further lateral spread of the fine sediments. At the close of Antrim time, the eastern edge of the basin became the strand line and the clastic Berea sandstone was deposited. This evidently marked the period of maximum crustal upwarp. The Sunbury shale was then deposited at the beginning of a cycle of downwarping under conditions similar to those in Antrim time. The maximum downwarping came as the predominantly calcareous Coldwater shale was being deposited. Thus, the full cycle had been completed.

The interaction of the organic matter and radioactive elements in shales may be favorable for petroleum genesis. There is "the possibility that a significant amount of organic matter may have been altered by alpha-particle bombardment." The original organic endowment and the products of this alteration are not known, however.

6. Beers, R. F., and Goodman, Clark, 1944, Distribution of radioactivity in ancient sediments: Geol. Soc. America Bull., v. 55, p. 1229-1254.

Over 300 samples of sedimentary rocks including limestone, sandstone, shale, arkose, and dolomite were tested for their radioactive constituents (potassium 40, thorium, and uranium) for the purpose of evaluating the role of radioactivity in the genesis of petroleum. Quantitative analyses of the potassium, carbon, and hydrogen content of selected samples were made. A detailed description of the apparatus and methods of measurements, particularly the beta counter, is given.

Samples of shale from oil and gas wells penetrating the Sylvan and Woodford shales of Oklahoma; the Traverse formation and Antrim shale of Michigan; the Chattanooga shale of Oklahoma; and the Ellsworth, Sunbury and Coldwater shales of Michigan were studied. The highest concentration of uranium, thorium, and potassium-40 was found in black shale samples of the Antrim from a well in Osceola County, Mich. The highest total beta activity was observed in well cuttings of the Woodford chert (a correlative of the Chattanooga shale) from a well in the Seminole district, Okla. Samples of organic black shale from the Antrim shale of Late Devonian age, Michigan, were selected for intensive study. A nearly 50 percent increase in beta activity was observed in the minus 200 mesh fraction as compared with the coarser fractions.

Conclusions resulting from this study are as follows: "In ... organic black shales, potassium and uranium content increase directly with one another. Organic black shales show excellent correlation of uranium content, organic matter, abundance of colloidal size grades and a high thorium/uranium ratio.... All these correlations indicate possible genetic relationships in petroleum source beds and may have a bearing on one possible origin of oil."

7. Bell, K. G., 1954, Uranium and thorium in sedimentary rocks; in Faul, Henry, editor, Nuclear geology: p. 98-114, New York, John Wiley and Sons, Inc.

The uranium cycle is briefly reviewed. The uranium present in sediments may (1) be indigenous to the clastic sediments (mainly uraniferous resistates), (2) have been deposited syngenetically with the sediments or (3) have been deposited epigenetically after sedimentation was completed. Some types of uraniferous sediments undoubtedly belong to more than one of these categories. The uranium in fine-grained clastics is predominantly indigenous while that of the chemical and carbonaceous sediments is commonly of syngenetic origin.

Syngenetic deposition of uranium

The chemical reactions that bring about the extraction and deposition of uranium from aqueous solutions are not fully understood. The uranium may be: (1) directly precipitated as insoluble compounds; (2) adsorbed by clays and carbonaceous materials; or (3) deposited in lesser amounts by gel precipitates of iron, aluminum, manganese and silica. In addition, the uranium may have been an original constituent of some organic compounds or it may have entered some minerals as an isomorphic substitution for calcium.

The distribution of uranium in sedimentary rocks indicates that a reducing environment containing organic matter and sulfides and little or no dissolved oxygen can precipitate uranium from solution. The deposition of uranium in

carbonaceous shales may have taken place by both direct precipitation of insoluble compounds and by adsorption onto clays and carbonaceous material.

There are two main groups of carbonaceous sediments: (1) the humic group with a low hydrogen and high carbon and oxygen content and (2) the sapropelic group with high hydrogen and carbon contents and little, or no, oxygen content. Some marine sapropelic black shales have been found to contain the greatest concentration of uranium of syngenetic origin of the sedimentary rocks. The uranium exists in an acid-soluble form in these uraniferous black shale formations, all of which have the following features in common: (1) they are usually less than 100 feet thick, but represent long periods of very slow deposition, (2) contain abundant organic matter and sulfides, and are poor in carbonates, (3) are bituminous and probable source beds of petroleum, (4) are all pre-Mesozoic in age, and (5) the greatest amount of uranium is found in nodules and layers of almost pure bitumen. Many marine black shales are non-uraniferous and non-marine black shales, oil shales, asphaltites and coals are among the least radioactive of all sedimentary rocks.

Some examples of uraniferous marine black shales are the Chattanooga and Antrim shales of the United States which contain about 0.01 percent U_3O_8; the alum shale of Sweden with as much as 0.023 percent U_3O_8; the shales of Cambrian and Ordovician age of the Leningrad district of Russia, which contain from about 0.008 to 0.03 percent U_3O_8 and the metamorphosed black

siliceous schist of Cambrian to Silurian age of the Ferghana region of Russia with amounts ranging from about 0.03 to 0.08 percent U_3O_8. The latter may possibly be surface-enriched.

All marine phosphate formations that have been tested contain significant amounts of uranium. The phosphates resemble the black shales in that they are relatively thin and represent long periods of very slow deposition. They were probably formed along the edges of continental shelves. The uranium seems to be combined with the phosphate minerals probably as an isomorphous substitution for the calcium. A reducing environment in the presence of organic matter may have aided in the concentration of uranium in the phosphatic sediments. Phosphatic nodules in some black shales are uraniferous, as at the top of Checkerboard and the Fort Scott limestones of Pennsylvanian age in Oklahoma, but some phosphatic black shales in Kansas and Oklahoma have uraniferous layers of non-phosphatic shale. The phosphatic nodules, however, generally contain more uranium than the enclosing shale.

Note: The book has an extensive bibliography of references dealing with geology and radioactive materials.

8. Bell, K. G., Goodman, Clark, and Whitehead, W. L., 1940, Radioactivity of sedimentary rocks and associated petroleum: Am. Assoc. Petroleum Geologists Bull., v. 24, p. 1529-1547.

Determinations of the radium and radon content of 21 sedimentary rocks and 7 associated crude oils have been made by the precision method developed by R. D. Evans. "The object of the investigation was to fix, if possible, the source of the radium emanation in the crude oils, to correlate the physical properties of the oil and its radioactivity and to determine the radioactivity of some sedimentary rocks...."

The radioactive elements in sedimentary deposits "are laid down as components of some mineral in the beds or are precipitated with the sediment subject to existing physical, colloidal and chemical conditions." The chemical effects of radioactivity in the generation of petroleum is suggested.

Radium determinations were made on six drill-core samples from oil and gas wells in the Fitts Pool, Pontotoc County, Okla. and in Texas, of shale ranging in age from Ordovician through Oligocene. The results of these tests, as well as tests upon other sediments, are given in tabular form. The radioactivity of five shale samples from the Frio clay of Starr County, Tex., and the Woodbine formation of Navarro County, Tex., was found to be relatively high and fairly constant, averaging 1.08 x 10^{-12} gm. Ra/gm. rock. "If uniform radioactivity in contemporaneous shales is real and is explicable by adsorption of uranium by colloidal constituents, then variation of radioactivity in shales of widely different ages may suggest variation of uranium in the sea during geologic time."

9. Beroni, E. P., and McKeown, F. A., 1952, Reconnaissance for uraniferous rocks in northwestern Colorado, southwestern Wyoming and northeastern Utah: U. S. Geol. Survey TEI-308A, 41 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

In 1950 a reconnaissance examination was made to determine the radioactivity of coal and lignite of Tertiary age in the Green River Basin in Wyoming and the Uinta Basin in Utah. Several areas of copper-uranium mineralization and a few miscellaneous prospects also were examined. Only those deposits that contain 0.010 or more percent uranium or that represent large potential low-grade reserves, such as lignite, are discussed in the report.

Samples of phosphatic shale of the Phosphoria formation near Cokeville, Lincoln County, Wyo., contain as much as 0.027 percent uranium. Near Rawlins, Carbon County, Wyo., thin layers of green micaceous shale of Devonian(?) age are interbedded with pinkish-gray quartzite. The green shale and the chlorite coating of the quartzite contain about 0.003 percent equivalent uranium. One grab sample of carbonaceous shale from the Mancos shale of Late Cretaceous age near Steamboat Springs, Routt County, Colo., contains 0.002 percent equivalent uranium, both in the sample and in 93.31 percent ash. Five beds of oil shale, totaling six feet in thickness, from the Laney shale member of the Green River formation near Rock Springs, Sweetwater County, Wyo., contain an average of 0.003 percent equivalent uranium.

10. Bradley, W. H., 1950, Occurrence of uranium deposits: Bull. Atomic Scientists, v. 6, no. 5, p. 149-152.

This article summarizes the work of the U. S. Geological Survey in the exploration for uranium deposits in the United States.

Uranium deposits are classified into three general types: (1) vein deposits, (2) deposits in sedimentary rocks and (3) pegmatite deposits. The geology of the important uranium deposits of the world are briefly described. Under the heading "Deposits in Sedimentary Rocks," uranium-bearing marine rocks, including black shales and phosphorites are discussed.

Most of the known uranium-bearing marine rocks are either black shales or phosphorites. Large, low-grade deposits of uranium-bearing marine black shales have been found in many areas of the world. Uranium is being extracted from some of these deposits, the oil shales of Sweden, for example, which contain an average of 0.01 to 0.02 percent uranium. In the future other similar deposits may well prove to be important sources of uranium.

Examples of uranium-bearing marine black shales include: shales in Estonia and Russia, which are similar to the oil shales of Sweden; the Chattanooga shale of east-central United States; the Woodford chert of Oklahoma; the Nonesuch shale of Michigan; and the Calico Bluff formation of east-central Alaska. All marine bituminous or carbonaceous rocks, however, are not appreciably uraniferous, and the nonmarine oil shales, coals and associated rocks as a group are among the least radioactive of all rocks.

In the uraniferous marine shales, however, the most uranium generally is found in those rocks containing the most organic matter. The uranium content increases in direct proportion to the carbon content in the Sunbury and Antrim shales of Michigan. The highest concentrations of uranium are found in nodules and lenses of nearly pure bitumen in the shale of Sweden and in very thin layers of almost pure bitumen in the Chattanooga shale.

Significant deposits of uranium in phosphorite rocks are known in several regions of the world. In the United States the Phosphoria formation of northwestern United States and the Bone Valley formation of Florida contain important low-grade concentrations of uranium. Uranium is concentrated in the phosphatic nodules of many black shales, such as those at the top of the Checkerboard limestone and in the Fort Scott limestone of Oklahoma; and at the top of the *Bourbon formation and in the *Hushpuckney and *Stark shales of Kansas.

All marine phosphate formations that have been tested contain significant amounts of uranium. In general, the uranium content of the phosphorites increases in direct proportion to the phosphate content, although there are many exceptions to this rule when applied to the phosphorites of the United States.

11. Branson, C. C., Burwell, A. L., and Chase, G. S., 1955, Uranium in Oklahoma, 1955: Okla. Geol. Survey Mineral rept. 27, 22 p. See p. 19.

This booklet has been prepared for the use of prospectors searching for uranium in Oklahoma. Several known uranium occurrences in the state are discussed. Maps of the major surface rock types and radioactive occurrences and a geologic map of a portion of southwestern Oklahoma are included. No radioactive deposits of proven commercial value have been found in Oklahoma to date.

Under the heading "Phosphatic black shales" several shales that are notably radioactive are described. These are the Woodford and Chattanooga shales of Devonian and Mississippian age and shales of Pennsylvanian age from the Senora formation, the Marmaton group and the Missouri series (includes the Checkerboard limestone). No figures are given to indicate the uranium content of these shales.

12. Breger, I. A., 1955, Radioactive equilibrium in ancient marine sediments: Geochim. Cosmochim. Acta, v. 8, p. 63-73.

The radioactive equilibrium of eight marine sedimentary formations was studied by means of direct determinations of the content of uranium, radium, and thorium and by alpha-particle counting techniques. "The maximum deviation from radioactive equilibrium that has been noted is 11 percent--indicating that there is probably equilibrium in all the formations analyzed."

Of the eight formations studied, six are of Cretaceous age from the Gulf Coastal Plain of eastern Texas and southern Mississippi. The other two are the Miocene nodular shale of the Los Angeles Basin, Calif., and the Antrim shale of Devonian age from Osceola County, Mich., which contain 14.4 and 17.5 parts per million uranium (0.0014 percent and 0.0017 percent), and 3.6 and 2.8 parts per million thorium (0.0003 percent and 0.0002 percent) respectively.

"The uranium content of marine sediments may be appreciably higher than the average values that have been reported for sedimentary rocks. Data show that there is up to fourteen times the percentage of uranium as of thorium in the formations studied, and that the percentage of thorium never exceeds that of uranium. While the proximity of a depositional environment to a land mass may influence the concentration of uranium in a marine sediment, this is not true with thorium."

13. Breger, I. A., and Deul, Maurice, 1956, The organic chemistry of uranium: Proc. Internat. Conf. on peaceful uses of atomic energy, Geneva, 1955, v. 6, p. 418-421, United Nations, New York; U. S. Geol. Survey Prof. Paper 300, p. 505-510.

"It has become clear within recent years that coal or materials derived from plants are avid receptors for uranium." Following a brief review of previous studies, the concentrations of uranium associated with carbonaceous materials and their probable genesis are considered for the following: living plants, marine carbonaceous shales, carbonaceous limestones, coal, coalified logs, petroleum, asphaltic pellets, and unclassified materials.

Some marine carbonaceous shales have a uranium content of from 20 to 50 ppm, few contain over 50 ppm. Early studies on the alum shale and associated kolm lenses of Sweden led to the conclusion that the uranium had been precipitated in the shale by means of organisms. However, the very old organic complexes, such as the Babrosco "carbon" of Precambrian age in South Africa or the kolm of Cambrian age in Sweden "are probably of algal origin."

"Recent experimental studies ... indicate physical rather than direct chemical association of uranium with the organic components of certain shales." Probably "... the decomposition of the organic matter under reducing conditions leads to the formation of hydrogen sulfide, which is known to reduce the uranyl ion to the insoluble uranium dioxide." However, the organic matter may have served as a primary precipitant by forming a complex organo-uranium compound from the sea water. If this was the case, then the chemical bonds between the uranium and the organic compounds have since been broken.

14. Breger, I. A., and Schopf, J. M., 1955, Germanium and uranium in coalified wood from Upper Devonian black shale: Geochim. Cosmochim. Acta, v. 7, p. 287-293.

"Microscopic study of black, vitreous, carbonaceous material occurring in the Chattanooga shale in [Davidson County,] Tennessee and in the Cleveland member of the Ohio shale in [Cuyahoga County,] Ohio has revealed coalified woody plant tissue. Some samples have shown sufficient detail to be identified with the genus Callixylon. Similar material has been reported in the literature as 'bituminous' or 'asphaltic' stringers.

"Spectrographic analyses of the ash from the coalified wood have shown unusually high percentages of germanium, uranium, vanadium, and nickel." A small sample of coalified wood from the Chattanooga shale contains 1.27 percent ash, and this ash contains 2.58 percent uranium and about 4.0 percent germanium. A sample of coal from the Cleveland member of the Ohio shale contains 1.7 percent ash, and this ash contains 0.50 percent uranium and 2.0 percent germanium. "The inverse relationship between uranium and germanium in the ash and the ash content of various samples shows an association of these elements with the organic constituents of the coal.

"On the basis of geochemical considerations, it seems most probable that the wood or coalified wood was germanium-bearing at the time logs or woody fragments were floated into the basins of deposition of the Chattanooga shale and the Cleveland

member of the Ohio shale. Once within the marine environment, the material probably adsorbed uranium with the formation of organo-uranium compounds such as exist in coals.

"It is suggested that a more systematic search for germaniferous coals in the vicinity of the Chattanooga shale and the Cleveland member of the Ohio shale might be rewarding." (authors' abstract)

15. Brown, Andrew, 1956, Uranium in the Chattanooga shale of eastern Tennessee: Proc. Internat. Conf. on peaceful uses of atomic energy, Geneva, 1955, v. 6, p. 439-444, United Nations, New York; U. S. Geol. Survey Prof. Paper 300, p. 457-462.

The uraniferous Chattanooga shale of Alabama, Tennessee and adjoining states in southeastern United States is part of a widespread blanket of bituminous shales of Late Devonian and Early Mississippian age which were deposited over much of central United States from New York to Texas.

This report concerns the most uraniferous part of the Chattanooga shale in the Eastern Highland Rim area of the Nashville Dome and the Walden Ridge area, both northwest of Chattanooga, Tenn. The Chattanooga shale dips eastward from the axis of the Nashville Dome at the rate of 30 feet per mile to the Sequatchie Valley where it is broken by the Sequatchie overthrust fault in the vicinity of Walden Ridge. Here the average dip is 8 degrees to the southeast. The Chattanooga shale in this area is of Late Devonian age and consists of two members, the Dowelltown and the overlying Gassaway. The Dowelltown member, which includes a basal sandstone, a black shale unit, and a gray shale unit, attains a maximum thickness of about 18 feet; the Gassaway member, which includes two black shale units separated by a siltstone unit, ranges in thickness from 7 feet to about 21 feet. The top black shale unit is the most widespread and most uraniferous unit of the Chattanooga shale, and ranges in thickness from 4 to 10 feet in Tennessee.

Analyses of two cores from drill holes near Smithville, Tenn. reveal that all units of the Chattanooga shale are composed chiefly of silicate minerals of silt and clay sizes;

pyrite and carbon are next in abundance. The organic matter in the shale is composed largely of spores, and most of the uranium-rich stringers, formerly identified as asphaltite, are now known to be woody fragments.

Semiquantitative spectrographic analyses of samples from three cores of the total thickness of the Chattanooga shale and from 15 cores of the Gassaway member are summarized and show that the chemical composition is fairly constant, both vertically and laterally. No correlation between the uranium and the minor metal content could be made.

While all units of rock in the Chattanooga shale contain more uranium than most marine shales, the uranium content varies from unit to unit. The top gray shale unit of the Dowelltown member contains the least (0.001 to 0.002 percent uranium) and the top black shale unit of the Gassaway member contains the most uranium (0.006 to 0.0086 percent). Only the Gassaway member can be considered a potential source of uranium under present conditions.

"No uranium minerals have been identified in the shale. The element is present as a separate colloidal phase disseminated through the organic matrix and is not chemically combined with the organic matter. Autoradiographic studies ... demonstrate that the alpha-particle source material is localized around concentrations of organic matter and pyrite."

Observations in the field and reflectance studies in the laboratory have established a positive correlation between the "blackness" of the shale, which is a function of sulfide and

carbon content, and its uranium content. Analyses of Chattanooga shale show a positive relationship between uranium content and: (1) total sulfur content, (2) carbon content and (3) pyrite content.

"...The preponderance of evidence points to deposition of the /Chattanooga/ shale in a sea that was for the most part landlocked and comparatively shallow. The uraniferous Eastern Highland Rim-Walden Ridge area was apparently in the central and deepest part of this sea. ... Lack of circulation on the bottom of such a sea would provide anaerobic conditions and permit carbonaceous material, which has a strong affinity for uranium, to accumulate. The suggestion that the high uranium content of certain black shales represents concentration of only normal quantities of the element in sea water over tremendously long periods of geologic time, and that no particularly uraniferous source material is necessary, may not apply to the Chattanooga shale." The uranium in the Chattanooga shale could have been concentrated in part from the small quantity of volcanic ash that probably fell and was transported into the sea. The landmass of crystalline rocks to the east also may have been a possible source of uranium.

Note: For recent stratigraphic descriptions and nomenclature of the Chattanooga shale in southeastern United States see: Hass, W. H., 1956, Age and correlation of the Chattanooga shale and Maury formation: U. S. Geol. Survey Prof. Paper 286.

16. Brown, J. H., Jr., and Keller, W. D., 1952, Uranium in the clay of a black radioactive shale: Science, v. 116, no. 3023, p. 632-633.

A thin, black, radioactive shale layer in the Spergen limestone near Sainte Genevieve, Sainte Genevieve County, eastern Missouri was studied in an attempt to determine the chemical association of the contained uranium. The presence of uranium and vanadium was determined spectrographically.

The uranium content of three size fractions of the shale was tested by the use of the direct fluorimetric method using a sodium fluoride flux. The shale fraction containing the greatest amount of carbon emitted the weakest fluorescence. Therefore, it was concluded that the uranium was associated with the clay fraction and not with the organic fraction.

Leaching experiments were conducted and it was found that hydrochloric, nitric and sulfuric acids, aqua regia and dilute sodium carbonate were effective solvents of the uranium in the shale. "Normal propyl alcohol dissolved a small amount which fluoresced weakly. Hot and cold water, ethyl alcohol, carbon disulfide and carbon tetrachloride, however, did not extract enough uranium to produce fluorescence."

An attempt was made to exchange the uranium in the shale cationically by using barium chloride and potassium chloride. It was concluded that the uranium was held in a condition not easily exchangeable. No information on uranium or vanadium content is given in this article.

Note: An equivalent uranium content of 0.37 percent for this shale was reported in: Muilenberg, G. A., 1949, Notes on uranium: Missouri Geol. Survey Inf. Circ. 5, 18 p. See p. 9, 17.

17. Burbank, W. S., and Pierson, C. T., 1953, Preliminary results of radiometric reconnaissance of parts of the northwestern San Juan Mountains, Colorado: U. S. Geol. Survey Circ. 236, 11 p.

In 1951 a reconnaissance was made of the radioactivity of the rocks of a portion of the northwestern San Juan Mountain region in parts of Gunnison, Ouray, San Juan, San Miguel and Dolores Counties in southeastern Colorado. The localities examined included the uranium-bearing base-metal and precious-metal sulfide-type deposits in the Uncompahgre district of Ouray County and the Red Mountain district of Ouray and San Juan Counties, and uranium-bearing Precambrian slates and black bituminous shales of Jurassic age.

A one-foot channel sample of altered bituminous shale from the Pony Express limestone member of the Wanakah formation of Jurassic age near the Pony Express mine, lower Uncompahgre district, contains 0.010 percent uranium. Nearby unaltered shale from the same horizon was essentially nonradioactive. A chip sample of limonite-stained black slate from the Uncompahgre formation of Precambrian age near Bear Creek Falls, upper Uncompahgre district, contains 0.050 percent uranium. A spectroscopic analysis of this slate is included.

The uranium in the altered bituminous shale from the Pony Express limestone member of the Wanakah formation is thought to have been introduced by mineralizing solutions. The uranium in the black slate from the Uncompahgre formation is believed to have had a dual origin: "The widespread radioactivity of the slate and the structural environment of local uranium concentrations are suggestive of an initial Precambrian low-grade concentration, which has been enriched locally by early Tertiary processes of mineralization."

18. Burton, V. L., and Sullivan, G. R., 1951, Carbon content and radioactivity of marine rocks: Am. Geophys. Union Trans., v. 32, p. 881-884; also in Report of progress -- Fundamental research on occurrence and recovery of petroleum, 1950-1951: Am. Petroleum Inst., p. 221-224, (n. d.).

This is a report of studies of the radioactivity of sedimentary rocks done at the Massachusetts Institute of Technology in connection with fundamental research into the origin of oil sponsored by the American Petroleum Institute. The net beta count and organic carbon content of 315 samples from two sandstone units, two limestone units, and four shale units were determined.

Shales, which make up 82 percent of all sedimentary rocks, "have by far the largest concentration of radioactivity." Organic matter, estimated at 1.6 times the percentage of organic carbon, is also more concentrated in the shales than in the limestones and sandstones. Determinations for organic carbon, potassium, and beta emission were made on samples of oil well cuttings and cores of four shale formations: the Miocene nodular shale from the Los Angeles Basin, the Cherokee shale (former usage) of Pennsylvanian age from Oklahoma, the Eagle Ford shale of Cretaceous age from eastern Texas, and the Eutaw formation of Cretaceous age from southern Mississippi. A more detailed study was made of the Miocene nodular shale (130 determinations) and Cherokee shale (105 determinations) because they exhibited the greatest radioactivity.

When the net beta counts (the beta activity due to potassium was excluded) were plotted against the average organic carbon content, an increasing and consistent linear relationship was observed. These results suggest a possible genetic relationship between the uranium, thorium and carbon contents of the formations studied.

19. Butler, A. P., Jr., 1952, The Geological Survey's work on the geology of uranium and thorium deposits: U. S. Geol. Survey TEI-207, 26 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge. See p. 11, 19-20, 24.

This is a summary of the work done by the U. S. Geological Survey on the geology of uranium and thorium from 1939 to November 1951, and records the many hundreds of occurrences of uranium, or of materials that might possibly be uraniferous, that were examined during that period.

Other than sandstones, marine phosphorites and black shales are the principal uraniferous sedimentary rocks and their uranium content ranges from a few thousandths of a percent to about 0.02 percent. In general, the uranium in these marine sedimentary rocks is distributed rather uniformly in relatively thin beds of wide lateral extent ranging in size from a few to several thousand square miles.

Brief statements are made about the studies of the Chattanooga and other Devonian shales, some shales of Pennsylvanian age, and the Phosphoria formation of Permian age; plate 3 of the report indicates areas where the radioactivity of these units has been measured. Other plates indicate areas in the United States and Alaska that have been examined by the U. S. Geological Survey during the course of reconnaissance investigations for radioactivity.

Under the heading "Studies of Alaskan deposits" the results of reconnaissance investigations, mainly in east- and south-central Alaska, are discussed. Of the sediments examined, only the phosphate rocks in the Lisburne group of Mississippian age

cropping out on the north flank of the Brooks Range of northern Alaska and the black shale beds at the base of the Calico Bluff formation, also of Mississippian age in the upper Yukon River region of east-central Alaska are appreciably radioactive. The uraniferous phosphate rock in northern Alaska is similar in lithology and uranium content to that of the Phosphoria formation of Permian age in northwestern United States. The radioactivity of the black shales of the Calico Bluff formation "is comparable to that of the Chattanooga shale" of Devonian and Mississippian age in southeastern United States. Most of the marine black shales examined in Alaska were only weakly radioactive.

20. Butler, A. P., Jr., and Schnabel, R. W., 1956, Distribution of uranium occurrences in the United States: Proc. Internat. Conf. on peaceful uses of atomic energy, Geneva, 1955, v. 6, p. 224-230, United Nations, New York.

Butler, A. P., Jr., and Schnabel, R. W., 1956, Distribution and general features of uranium occurrences in the United States: U. S. Geol. Survey Prof. Paper 300, p. 27-40.

This report is a compilation of data regarding the general geology and distribution of the major types of uranium deposits in the United States and includes index maps of the United States showing the locations and types of the major uranium occurrences. Under the heading "Uraniferous Marine Sedimentary Rocks," uraniferous marine black shales and phosphate rocks are discussed.

In general, the more uraniferous marine black shales contain abundant plant remains, are relatively rich in iron sulfides, are noncalcareous, and some contain phosphate. Identifiable uranium minerals in these rocks are extremely rare.

"Most of the appreciably uraniferous carbonaceous black shales in the United States are distributed from the western limit of the Appalachian Mountains in Virginia, Kentucky, Tennessee and Alabama westward to the eastern front of the Rocky Mountains in Wyoming. All are of Paleozoic age and contain 0.005 to 0.02 percent U_3O_8. These include the Chattanooga shale in Tennessee and its stratigraphic equivalents in adjacent states westward to Oklahoma, and some thinner, less persistent black shales of Pennsylvanian age in eastern Oklahoma and eastern Kansas and in southeastern Wyoming." Weakly uraniferous shales are known west of the Rocky Mountains, but in only a few places are these as rich in uranium as the Chattanooga shale.

21. Conant, L. C., 1956, Environment of accumulation of the Chattanooga shale: Proc. Internat. Conf. on peaceful uses of atomic energy, Geneva, 1955, v. 6, p. 435-438, United Nations, New York; U. S. Geol. Survey Prof. Paper 300, p. 463-467.

The Chattanooga shale that underlies central Tennessee and adjacent parts of Kentucky, Alabama and Georgia consists largely of interbedded fine-grained black shale and gray claystone overlying a remarkable smooth peneplain of truncated, gently-dipping sediments of Middle Ordovician to Early Devonian age. The formation, which averages 30 to 35 feet thick, represents most of Late Devonian time and is divided into the Dowelltown member and the overlying Gassaway member. Thin sandstone beds are present at the base of each member, a thin, persistent bentonite bed is present near the top of the formation and scattered phosphatic nodules are present in the top-most beds further to the northeast. The denser and darker parts of the shale, which have proved to be the most uraniferous, are made up of about 25 to 30 percent clay and mica, 20 to 25 percent quartz, 15 to 20 percent organic matter, 10 to 15 percent pyrite, 10 percent feldspar, 5 percent chlorite, ferric oxides and miscellaneous minerals. The fossil assemblage includes algae, marine and land plant spores, tree logs, radiolaria, brachiopods, conodonts, and fish bones and teeth.

The uranium content is highest in the Gassaway member, which contains slightly more than 0.005 percent uranium. The uranium probably was brought into the depositional basin in normal quantities by streams from the surrounding land areas. The uranium is believed to have been extracted from sea water by organic matter in some unknown manner. Much of the uranium is "probably associated with the plant matter in an ionic form." The higher uranium content of the driftwood logs of Callixylon "probably results from the absence of mineral matter, and perhaps from longer exposure to water."

The environment of accumulation of the Chattanooga shale, as inferred from lithologic data and the regional, stratigraphic and structural relationships, is as follows: (1) A broad shallow sea, at the most only a few tens of feet in depth, encroached slowly from the northeast across a very smoothly peneplained land surface. (2) The limestone peneplain contributed very fine mud, and probably aeolian dust, at a very slow rate (most of the coarse sediments from the highland areas were trapped in the deeper Appalachian geosyncline to the northeast), where it was deposited on a well-graded sea floor. (3) The water of the sea was stratified, with a strongly reducing bottom layer and an overlying layer of more aerated water where life could be sustained. (4) Conditions prevented strong wave or tidal action so that the bottom muds were subjected to only mild movement and sorting. (5) At the

close of the Late Devonian, the sea had spread over a large area, inundating the peneplain and the low islands that had existed in the Chattanooga sea.

Note: Similar material is included in: Conant, L. C., 1953, Shallow water origin of the Chattanooga shale (abs.): Geol. Soc. America Bull., v. 64, p. 1529-1530.

22. Deul, Maurice, 1955, Mode of occurrence of uranium in the Chattanooga shale [abs.]: Geol. Soc. America Bull., v. 66, p. 1549; see abs., Econ. Geology, v. 50, p. 772.

"The Chattanooga shale, an abnormally uraniferous marine carbonaceous shale of Late Devonian age, has been studied to determine the relationship of uranium to gross organic and mineral constituents. Mineral-rich, organic-rich, and uranium-rich fractions have been obtained mechanically from a shale sample containing 0.009 percent uranium and 13.7 percent carbon.

"Separation of shale components after ball-mill grinding in a mixture of water and kerosene for several hundred hours yielded a mineral concentrate containing 0.0035 percent uranium and 4.7 percent carbon, an organic concentrate containing 0.0038 percent uranium and 39.8 percent carbon, and a middlings fraction, consisting of the finest particles, assaying 0.019 percent uranium and 9.1 percent carbon.

"With increased grinding time more uranium has been liberated from the shale components. The middlings fraction from a sample milled for 1361 hours contained 0.031 percent uranium and 26.7 percent carbon. This fraction, consisting of less than 8 percent of the total material, contained more than 24 percent of the uranium.

"These experimental data indicate that uranium in the Chattanooga shale exists largely as a colloidal phase dispersed through the organic matrix and that most of the uranium is not now combined with the organic material or with the minerals.

These conclusions are confirmed by other experiments where colloidal fractions markedly enriched in uranium were obtained by air elutriation of air-jet pulverized shale and by dialysis of a hydrosol of shale." (author's abstract)

Note: The technique for the separation of components in shales of the Chattanooga, Phosphoria, Dakota, Chinle and Lockatong formations and the resulting analytical data (not including that on uranium) are given in Deul, Maurice, 1956, Colloidal method for concentration of carbonaceous matter from rocks: Am. Assoc. Petroleum Geologists Bull., v. 40, p. 909-917.

23. Deul, Maurice, and Breger, I. A., 1954, Uranium in carbonaceous rocks, Geochemistry and petrology, Geochemistry of uranium-bearing shales, in Geologic investigations of radioactive deposits, Semiannual progress report, June 1 to November 30, 1954: U. S. Geol. Survey TEI-490, p. 178 and 180-182, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Samples of Chattanooga shale, of vanadium shale from the Phosphoria formation in Coal Canyon, Lincoln County, Wyo., and of shale from the Dakota sandstone near Gallup, McKinley County, N. Mex., were fractioned by ball-mill grinding in mixed media. "These shales were chosen because they represent different geologic environments and contain different percentages of uranium. All the shales have about the same percentage of organic material. Preliminary results ... indicate that uranium is present in the Chattanooga shale [and the vanadium shale of the Phosphoria formation] as a separate colloidal phase disseminated through the organic matrix but not combined with the organic matter." ... Complete data (the percentages of ash, uranium, carbon and hydrogen in various unrefined and refined separates) are listed in tabular form.

"Analysis of fractions from the Dakota shale [of non-marine origin] show that the uranium is retained by and associated with the carbonaceous constituents and is not present as a separate phase." This contrasts sharply with the uranium association found in the Chattanooga and Phosphoria shale fractions.

"The organic concentrate from the Chattanooga shale exhibits no unusual trace element enrichments. In contrast, the organic fraction from the Phosphoria formation contains striking

enrichments of nickel, vanadium, copper, silver, and molybdenum; and the organic concentrate from the shale of the Dakota shale contains notable enrichments of copper, lead, cerium, and lanthanum." ... "The data obtained have clarified a number of points with regard to the mechanism responsible for the precipitation of uranium in carbonaceous shales."

Note: This is essentially the author's entire report with the exception of analytical data given in tabular form.

24. Duncan, D. C., 1953, Reconnaissance investigations for uranium in black shale deposits of the Western States during 1951 and 1952: U. S. Geol. Survey TEI-381, 89 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

"Reconnaissance examinations for uranium in 80 formations containing black shale were conducted in parts of Arizona, Colorado, Idaho, Montana, Nebraska, Nevada, New Mexico, Utah, and Wyoming...." The search was directed primarily toward tests of thin black marine shale zones (about 100 feet or less thick) of wide areal extent similar to some of the larger known deposits of uraniferous black shales. "Most of the uranium-bearing black shales that were found...occur in rocks of Carboniferous and Permian ages."

The majority of the black shales examined contain very little uranium. Samples from thirteen of these shales, however, contain from 0.003 to 0.006 percent uranium and were collected from the following rocks: an unidentified black shale of Ordovician or Silurian age from central Nevada; an unnamed shale of Mississippian and Pennsylvanian age, Manning Canyon shale, the Oquirrh formation and the Gardner dolomite - all from central Utah; the Brazer limestone and the Park City formation - both from northern Utah; the Hermosa formation of southern Colorado; the Paradox formation and Belden shale - both from central Colorado; the Heath shale from central Montana; the Madison limestone from southern Montana and the Lakota(?) sandstone of northeastern Wyoming.

Four of the shales locally contain from 0.011 to 0.026 percent uranium and are as follows: a black shale bed, one foot in thickness in the Weber(?) quartzite of Pennsylvanian

age, Lake County, Colo., (0.019 percent uranium, possibly of hydrothermal origin); a shale bed, one foot in thickness, in the Pony Express limestone member of the Wanakah formation of Jurassic age, Ouray County, Colo., (0.011 percent uranium, possibly of hydrothermal origin); several thin beds of black shale from the subsurface Hartville formation of Mississippian(?), Pennsylvanian, and Permian age in eastern Wyoming and western Nebraska (from a few thousandths to 0.019 percent uranium); and phosphate-rich beds from the Phosphoria formation of Permian age in Lincoln and Teton Counties, Wyo., (as much as 0.026 percent uranium). Of the shales examined "only the phosphatic black shales of the Phosphoria formation appear to be sufficiently thick and extensive to be of possible economic interest."

25. Dunham, R. J., 1954, Black shale, Chadron area, Nebraska and South Dakota, in Geologic investigations of radioactive deposits, Semiannual progress report, June 1 to November 30, 1954: U. S. Geol. Survey TEI-490, p. 157-158, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

About 250 square miles in northeastern Dawes County and northwestern Sheridan County, Nebr., and southern Shannon County, S. Dak., were mapped on aerial photographs at scales of 1:20,000 and 1:31,680. Concentrations of 0.003 to 0.013 percent uranium occur in two kinds of deposits.

"Uranium is concentrated sporadically in the Carlile shale, Niobrara formation, and the Pierre shale, all of Cretaceous age in gray shales and marls immediately below the base of an altered zone formed by weathering of the Cretaceous strata. The maximum known uranium content in shale of the altered zone is 0.01 percent, and in most places the content is less than 0.003 percent uranium. Greatest uranium content is generally restricted to the highest part of the highest and thinnest remnant or pinnacle of unaltered shale."

About 20 feet of the middle part of the Sharon Springs member of the Pierre shale exhibits a persistent radioactivity of from two to four times the normal background count. Two samples from the richest part of the interval show 0.003 percent equivalent uranium and 0.001 percent uranium, and 0.007 percent equivalent uranium and 0.001 percent uranium. The Sharon Springs member is a hard black marine shale that bears abundant fish remains and pyrite.

Note: This is the complete text of the author's report concerning uranium in marine black shales with the exception of a map showing exposures of the Sharon Springs member of the Pierre shale.

26. Eargle, D. H., 1954, Uranium in carbonaceous rocks, Black shales in Texas, in Geologic investigations of radioactive rocks, Semiannual progress report, December 1, 1953 to May 31, 1954: U. S. Geol. Survey TEI-440, p. 132-136, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

The radioactivity of the Eagle Ford clay of Late Cretaceous age was studied from Dallas in northeast Texas to the Eagle Mountains in Hudspeth County, southwest Texas. The work included sampling from outcrops and well cores. In addition, a partial study was made of the radioactivity of several adjacent Cretaceous formations, and a few Paleozoic rocks were spot-sampled in several localities. A table showing the average radioactivity and uranium content of the rocks examined is included.

Thirty-six samples of clay and siltstone from the Eagle Ford clay (or shale) contain, with one exception (0.002 percent uranium in Brewster County), less than 0.001 percent uranium. One sample of brown shale from the Maravillas chert of Late Ordovician age in Brewster County contains 0.003 percent uranium. Two samples of black shale from well cores of the Woodford chert of Devonian age in Terry County contain 0.0045 percent equivalent uranium but less than 0.001 percent uranium. Clay from the Escondido formation of Cretaceous age in Medina County contains 0.002 percent equivalent uranium, and gray clays from the Del Rio clay (Grayson formation) and Pepper shale member of the Woodbine formation, both of Cretaceous age contain less than 0.001 percent uranium.

27. Evans, R. D., and Goodman, Clark, 1941, Radioactivity of rocks: Geol. Soc. America Bull., v. 52, p. 459-90. See p. 459-460, 468, 475 and 487.

After an evaluation of the methods and data resulting from previous determinations of the radioactivity of terrestrial rocks, many of which are discarded as unreliable, several hundred new measurements of the radioactivity of rocks are given. A description of the analytical methods used in obtaining these newer and more reliable data is included.

Most of the new determinations are of igneous rocks, but 28 are of sedimentary rocks, including five shales from unspecified rock units. Most of the sedimentary rocks tested were core samples obtained from oil and gas wells. Of the five shale samples, two are Cretaceous and three are Oligocene in age and all of these shales are from core samples from oil and gas wells in Starr and Navarro Counties, Tex. These shales contain about 0.0003 percent uranium, using the given radium determinations and the given conversion factor. By similar calculation, the average uranium content of the 28 sedimentary rock samples is less than 0.0002 percent.

28. Everhart, D. L., 1951, Geology of uranium deposits - a condensed version, with mineral tables by Muriel Mathez: U. S. Atomic Energy Comm. RMO-732, 33 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge. See p. 20-22.

This report summarizes the geologic features of the major types of uranium deposits. Deposits in igneous and metamorphic rocks are discussed. Deposits discussed under sedimentary rocks include uranium-bearing black shales, carnotite deposits, copper-uranium deposits, uranium-bearing asphalts, limestones, phosphorites, alluvial and placer deposits, and lacustrine-saline deposits.

"Comparatively low concentrations of uranium in vast tonnages of carbonaceous and bituminous shales have been known in many parts of the world for several decades." Since the discovery in 1893 of uranium in the black alum shale of Cambrian age in Sweden, uranium also has been found in certain black shales of Russia, Estonia, Alaska and central United States.

The significantly uraniferous marine black shales are all very dark in color, have a high content of sulfide and organic matter, are low in carbonate, are in relatively thin units which are thinly-bedded, and, so far as is known, are mostly pre-Mesozoic in age. The radioactive black shales of the United States are characteristically uniform in thickness and composition, and contain small abnormally radioactive lenses. The shales are commonly composed of silica, organic matter or carbon, and clay minerals with associated pyrite, marcasite, and phosphate.

The Chattanooga shale of Devonian and Mississippian age is cited as an example of the radioactive black shales in the United States. It ranges in thickness from 5 to 100 feet throughout most of its extent. The most uraniferous parts of the Chattanooga shale "are in central Tennessee and southern Kentucky where thousands of square miles of the uppermost part of the formation contain from 0.002 to 0.013 percent U_3O_8 and average 0.008 percent U_3O_8. Formations correlative to the Chattanooga, extending through Indiana, Kentucky, Ohio, Michigan, and New York are less radioactive, but the uranium content of the roughly equivalent Woodford formation of Oklahoma and Kansas is comparable /to that of the Chattanooga shale/. Certain black shales of Pennsylvanian age in Alabama, Kansas, and Oklahoma also range from 0.003 to 0.013 percent in U_3O_8 content, but the average is somewhat less than that of the Chattanooga shale and they are not so widely exposed nor so mineable."

"The mineralogy of the uraniferous black shales has not been completely solved as yet, and no definite uranium minerals are identifiable in most cases." The uranium is in the finer-grained portions of the shale and is soluble in acid. "It may be incorporated into the crystal lattices of clay minerals and organic compounds.... All thin black shale formations of Paleozoic and, probably, pre-Cambrian ages, representing periods of stable, slow sedimentation in shallow seas, are worthy of investigation if they afford large tonnages and may be mined at low costs."

29. Ferm, J. C., 1955, Radioactivity of coals and associated rocks in Beaver, Clearfield, and Jefferson Counties, Pennsylvania: U. S. Geol. Survey TEI-468, 52 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

In 1953 a reconnaissance was made of the radioactivity of coal and associated rocks in the coal fields of west-central Pennsylvania. Selected beds were examined in the Pottsville, Allegheny and Conemaugh formations of Pennsylvanian age. The greatest radioactivity of any rock group (0.016 percent uranium) was found in the uppermost section of the underclay underlying the Lower Freeport coal in Beaver County. The coal beds were generally the least radioactive of the rocks examined. A sample of the Lower Freeport coal in Beaver County, however, contains as much as 0.010 percent uranium.

Dark marine shale beds associated with the Lower and Middle Kittanning, and the Lower Freeport coals of the Allegheny formation and the Brush Creek coal of the Conemaugh formation contain from 0.002 to 0.004 percent equivalent uranium at a number of localities in the three counties studied.

The structural attitude and the degree of weathering of the beds appears to have little effect on the amount of radioactivity contained in the rocks examined. No commercially significant uranium deposits were discovered during the course of this study.

30. Fredrickson, A. F., 1948, Some mechanisms for the fixation of uranium in certain sediments: Science, v. 108, no. 2799, p. 184-185.

It is suggested that the wide distribution of uranium in the sedimentary crust of the earth is due principally to its tendency to form large UO_2^{++} ions. "Much of the uranium in shales may be in the form of adsorbed ions and not of discrete minerals." Studies indicate that "if the source rocks containing uranium are weathered to clays with a high base exchange capacity, these clays should show high radioactivity."

"A possible method of concentration of uranium in sedimentary deposits containing carbon may then be as follows:

"Uranium ions are released, on weathering, from the granitic, rhyolitic, etc. rocks in which they occur in relatively large amount and then are adsorbed by base exchange phenomena on layer-lattice minerals such as certain clays." By aging of the clays or chemical or thermal change, the uranium might be displaced and again put into solution. The carbonaceous material in the path of the displaced ions would then concentrate and fix the uranium ions.

"For the concentration of uranium ... it is probably important that an oxidizing environment prevail." Alakaline solutions would attack the carbonaceous material and release the uranium into a colloidal suspension. The uranium would then be adsorbed again on clays and similar minerals.

"Could such a process as this account for the high radioactivity of certain carbonaceous shales or have something to do with the formation of oil...?"

31. Glover, Lynn, 1955, Uranium in carbonaceous rocks, black shale investigations, Chattanooga shale in Alabama, Georgia and Tennessee, in Geologic investigations of radioactive deposits, Semiannual progress report, December 1, 1954 to May 31, 1955: U. S. Geol. Survey TEI-540, p. 170-174, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

The Chattanooga shale was studied for radioactivity and sampled in the southern Appalachian region between Birmingham, Alabama and Knoxville, Tennessee. In the vicinity of Birmingham the shale is absent, but farther to the northeast it is generally 10 to 40 feet thick. Phosphate nodules and chert beds are common in the black shale of this region. The phosphate and uranium content of the shale units is inversely proportional.

A sample of highly weathered coalified material from a bed about one inch in thickness in the top of the Maury formation in northwestern Georgia, contains from 1 to 5 percent yttrium in about 10 percent ash.

32. Gott, G. B., and Hill, J. W., 1953, Radioactivity in some oil fields of southeastern Kansas: U. S. Geol. Survey Bull. 988-E, p. 69-122.

"Abnormally high radioactivity in oil and gas wells in southeastern Kansas was noted in 1948 during an investigation to determine the value of commercial gamma-ray well logs in the search for radioactive ore deposits." Studies showed that many of the large deflections on the gamma-ray logs of these Kansas wells are caused by a radium-bearing precipitate, probably derived by solution and redeposition from adjacent rocks since the drilling of the well. These precipitates, generally associated with limestones in the Arbuckle group of Cambrian and Ordovician age, and the Kansas City group of Pennsylvanian age, contain as much as 0.26 percent equivalent uranium, and were found in more than 60 oil and gas fields in Cowley, Butler, Marion, Sedgwick, and Greenwood Counties, Kans. Most of the other rocks in this area are not abnormally radioactive, though some large deflections on the logs are associated with marine black shales.

Gamma-ray logs and a few radiometric analyses of drill cuttings of Pennsylvanian black fissile shales indicate an equivalent uranium content of from 0.004 to 0.01 percent. A gamma-ray log of the Dilworth no. 2 Fee well (sec. 8, T. 33 S., R. 7 E., Cowley County) reveals a marked anomaly at the level of the Cherokee shale that exceeds the limits of the sensitive logging instruments and could not be recorded. A gamma-ray log of the upper gray shales of the Wabaunsee group shows an

equivalent uranium oxide content of 0.02 percent. It is noted that A. L. Slaughter_/ investigated the radioactivity of the

_/Slaughter, A. L., written communication, 1945.

exposed Pennsylvanian black shales and coals of Kansas and Oklahoma, and found phosphatic nodules disseminated in the black shales to contain as much as 0.095 percent uranium. The uranium content of the black shales alone, however, was much lower.

Only meager information was obtained on the Chattanooga shale of Devonian and Mississippian age because most of it has been removed in the region by pre-Pennsylvanian erosion. A few gamma-ray logs and radiometric analyses, however, indicate the equivalent uranium content of the Chattanooga shale ranges from about 0.002 to 0.007 percent.

Drill cuttings of a green shale and glauconitic sandstone from the Simpson group of Ordovician age contain as much as 0.006 percent equivalent uranium.

33. Gott, G. B., Wyant, D. G., and Beroni, E. P., 1952, Uranium in black shales, lignites, and limestones in the United States, in Selected papers on uranium deposits in the United States: U. S. Geol. Survey Circ. 220, p. 31-35.

Small quantities of uranium occur in carbonaceous deposits at a great many localities in the United States, but the amount rarely exceeds 0.1 percent. The most common uranium-bearing carbonaceous deposits are in the black marine shales, principally Paleozoic in age, that are exposed in the eastern and central parts of the United States. The uranium content of most of these shales ranges from a few thousandths to a little more than 0.01 percent. Uranium-bearing lignites and limestones also are discussed.

The uranium content of the Chattanooga shale ranges from 0.001 to 0.03 percent, and data suggest a general correspondence between the uranium content and the: (1) organic content, (2) iron sulfide content, and (3) vanadium content of the shale. The uranium content of the black shales of Pennsylvanian age in Kansas and Oklahoma is similar to that of the Chattanooga shale except that the Pennsylvanian shales contain phosphatic nodules that have a much higher uranium content. Samples of a carbonaceous marine shale of Mississippian(?), Pennsylvanian, and Permian age [the Hartville formation] from an oil well in east-central Wyoming contain from 0.005 to 0.02 percent uranium. Thin beds of black shale in the Spergen limestone of Mississippian age in Ste. Genevieve County, Missouri contain as much as 0.6 percent uranium.

34. Holland, H. D., and Kulp, J. L., 1954, The transport and deposition of uranium, ionium and radium in rivers, oceans and ocean sediments: Geochim. Cosmochim. Acta, v. 5, p. 197-213.

Data that have accumulated over the past two decades on the concentration of uranium, ionium and radium in ocean water and in ocean-bottom sediments, are evaluated and rate equations descriptive of the economy and history of the elements are presented. "The important factors controlling the concentration of the radioelements in the oceans are the influx, the rate of radioactive decay and the rate at which the radioelements are removed by sedimentation."

The uranium content of marine sediments is probably attributable to several factors, including the original content of the constituent grains derived from the land, uranium removed from the transporting streams and from the ocean by the sediments, and the uranium removed by the marine organisms incorporated in the sediments.

The average uranium content of shales is $3.1 \pm 0.3 \times 10^{-6}$ gm/gm and the fraction of the earth's land surface covered by shales is on the order of 0.33 ± 0.03. The estimated average uranium content of constituent grains that make up the shales on entering the oceans via streams is $1.8 \pm 0.4 \times 10^{-6}$ gm/gm and $1.0 \pm 0.5 \times 10^{-6}$ gm/gm uranium is added during sedimentation. "The geochemistry of uranium influx and deposition in shallow water are of major importance...."

35. Huddle, J. W., and Patterson, E. D., 1954, Uranium in carbonaceous rocks - Coal and lignite in the eastern United States, in Geologic investigations of radioactive deposits, Semiannual progress report, December 1, 1953 to May 31, 1954: U. S. Geol. Survey TEI-440, p. 100-101, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Coal, lignite and associated rocks in southwestern Pennsylvania, western Indiana and southern Illinois were examined and sampled for radioactivity. No coal examined contains much more than 0.001 percent equivalent uranium.

Nine samples of a grayish-black marine shale of Pennsylvanian age, from 4 to 6 feet in thickness and overlying the No. V coal, contain from 0.006 to 0.016 percent equivalent uranium at several localities in Warrick, Pike, Vigo and Vermillion Counties in western Indiana. Grayish-black shale beds directly overlying the No. 5 and No. 6 coal units in southeastern Illinois are also radioactive. The No. 5 coal in Illinois is a correlative of the No. V coal in Indiana.

36. Kepferle, R. C., 1955, Uranium in carbonaceous rocks, black shale investigations, South Dakota and northeastern Nebraska, in Geologic investigations of radioactive deposits, Semiannual progress report, December 1, 1954 to May 31, 1955: U. S. Geol. Survey TEI-540, p. 163-165, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

About 300 samples of black shale from 32 localities in South Dakota and northeastern Nebraska indicate that the Sharon Springs member of the Pierre shale in that region contains 0.002 percent or less uranium. "The most uraniferous parts of the shale contain 0.005 percent or more uranium in zones averaging about 1 foot in thickness. At three localities these zones reach thicknesses of 3 to 6 feet; elsewhere they are less than 1 1/2 feet thick. The most uraniferous parts of the zones are generally about 3 inches thick and contain as much as 0.017 percent uranium.

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

"Chemical analyses of 18 samples of shale from one locality indicate a strong positive relationship between the concentrations of uranium and arsenic, and a lesser positive relationship between the concentrations of uranium and selenium.

"From a study of the relationship between the concentrations of uranium and the elements which vary in concentration with respect to uranium, it appears that the uranium in fresh samples of the Sharon Springs shale was emplaced during the deposition of the shale, in a reducing environment where deposition of allogenic clastic sediments was slow."

Note: This is essentially the author's complete report, with the exception of a diagram showing spectrographic analyses of the shale.

37. Krumbein, W. C., and Slack, H. A., 1956, Statistical analysis of low-level radioactivity of Pennsylvanian black fissile shale in Illinois: Geol. Soc. America Bull., v. 67, p. 739-762.

A sampling experiment utilizing the *Brereton shale of Pennsylvanian age in southwestern Illinois was undertaken in order to: (1) evaluate the average radioactivity of the shale and (2) detect regional radioactivity gradients. The mean equivalent uranium content of this 3-foot shale unit is 0.0075 percent.

An isopach map and a map showing the distribution of radioactivity of the *Brereton shale in southwestern Illinois are included. A restricted shallow-water marine environment of deposition with low oxygen content and almost neutral pH conditions is described for the *Brereton shale. It is believed that the Brereton sea, advancing eastward or northeastward, slowly inundated the essentially flat-lying peat bogs (now the No. 6 coal). The normal open circulation of this sea was restricted at its margin, causing the waters to become deficient in oxygen content and rich in sulfides.

The uranium in the *Brereton shale is thought to be associated with the pyrite-organic clay complex. Living marine organisms probably helped in the extraction and fixation of uranium from the sea water and later the uranium may have become associated with the iron sulfide and clay in the accumulating muck after the death of these organisms.

38. McKelvey, V. E., 1955, Search for uranium in the United States: U. S. Geol. Survey Bull. 1030-A, p. 1-64. See p. 14-16 and p. 37-40.

McKelvey, V. E., 1953, Search for uranium in the Western States [abs.]: Am. Assoc. Petroleum Geologists Bull. v. 37, p. 2614.

"The search for uranium in the United States is the most intensive ever made for any metal during our history.... The purpose of this report is to summarize briefly the information on the common uranium minerals; the kinds of deposits in which uranium is found; the methods used in the search for uranium; the important deposits found in this country thus far; the outlook for future discoveries; and the recent literature on the geology of uranium deposits in the United States."

Uraniferous black shale

Some bituminous marine black shales contain from 0.005 to 0.02 percent uranium and represent one of the world's largest potential sources of uranium. The distinguishing characteristics of the uraniferous shales are not fully known. As a group, however, they are from older formations; are sapropelic rather than humic in origin; highly carbonaceous and bituminous, high in sulfide content, generally somewhat phosphatic; but are noncalcareous. Their sediments accumulated very slowly, probably at the rate of only 1 foot per 200,000 to 1,000,000 years net accumulation as compared to 1 foot per 700 years for other shales, and probably under anaerobic conditions.

The alum shale of Late Cambrian age in Sweden and the Chattanooga shale of Devonian and Mississippian age of southeastern United States are important examples of these

uraniferous shales. "The uranium in these shales is in acid-soluble form, but it probably does not occur in a distinct uranium mineral. In the Chattanooga shale it is concentrated in pyrite-organic-matter complexes" and in the alum shale, the largest amounts of uranium (up to 0.5 percent) are in nodules and lenses of dark bitumen, known as kolm. Microscopic particles of kolm, also present in the alum shale, "may be the uranium carriers in the shale itself." "The uranium may occur both in phosphate and carbonaceous matter" in the Miocene nodular shale of southern California and the Cherokee shale (former usage) of Pennsylvanian age in Oklahoma. The uranium in a black shale in the Spergen limestone of Mississippian age in Sainte Genevieve County, eastern Missouri is in the fine clay fraction and not in the organic matter.

The most uraniferous parts of the alum shale of Sweden and the subsurface Hartville formation of Mississippian(?), Pennsylvanian, and Permian age in eastern Wyoming seem to have been deposited near the margins of deposition of those shales, and, in the case of the alum shale, in restricted areas bordering granitic terranes. "Very likely the uranium in black shales is extracted from the sea by plankton, either alive or dead. The reason uranium is more concentrated in some black shales than in others, however, is unknown." It seems that the controls for uranium concentration in these shales "have to do with the pH, redox potential and other physico-chemical factors that affect the precipitation of uranium in the sea." Although the concentration of uranium in the widespread,

evenly-distributed uraniferous marine black shales probably occurred as the sediments were being accumulated, there is evidence that the uranium in some of the more irregularly distributed nonmarine carbonaceous deposits was adsorbed from percolating waters long after their deposition.

"The most important uraniferous black shale yet discovered in the United States is the Chattanooga shale of Devonian and Mississippian age in Tennessee. The upper part of this formation contains vast quantities of uranium in concentrations of 0.01 percent or slightly less." The highest percentage of uranium in black shale (0.019 percent) thus far discovered in the United States is in the Mississippian(?), Pennsylvanian, and Permian(?) Hartville formation in the subsurface of eastern Wyoming. No uranium has yet been produced from the black shales in the United States.

Uraniferous phosphorite

Marine phosphorites commonly contain from 0.005 to 0.03 percent uranium. Important examples are the Permian Phosphoria formation in western United States, the Upper Cretaceous and lower Eocene deposits of North Africa and the Pliocene Bone Valley formation of Florida.

The uranium in the phosphorites seems to be chiefly in the mineral carbonate-fluorapatite "where it probably substitutes for calcium." "The uranium content of the rock generally increases as the phosphate content increases but deposits or

beds richest in phosphate are not necessarily richest in uranium ...", as is seen in the Phosphoria formation. This may be due to the inverse relationship between uranium and carbonate content.

The weakly phosphatic black shales in the Phosphoria formation contain little or no uranium. Phosphatic nodules in certain black shales, such as those of Pennsylvanian age in Kansas and Oklahoma, are generally more uraniferous than the enclosing black shales. The uranium content of phosphatic nodules on the present sea-bottom off the California coast is about the same as that of older phosphorites. The uranium in most of the marine phosphorite deposits is believed to have been adsorbed from the sea water "at the time of deposition or before burial." An index map showing the location of the more significant deposits of uraniferous black shales and phosphorites is included.

39. McKelvey, V. E., and Carswell, L. D., 1956, Uranium in the Phosphoria formation: Proc. Internat. Conf. on peaceful uses of atomic energy, Geneva, 1955, v. 6, p. 503-506, United Nations, New York; U. S. Geol. Survey Prof. Paper 300, p. 483-487.

Uranium-bearing marine phosphate deposits in the Phosphoria formation of Permian age and its stratigraphic equivalents cover an area of about 135,000 square miles in Montana, Idaho, Wyoming, Utah and Nevada. Deposits of the highest grade, however, are in eastern Idaho and adjacent parts of Montana, Wyoming and Utah. The phosphorites contain from 0.005 to 0.03 percent uranium and locally as much as 0.06 percent uranium in very thin layers.

The Phosphoria formation ranges in thickness from 200 to 1,500 feet and consists of two overlapping "couplets," each composed of a carbonaceous, phosphatic shale member overlain by a cherty member. This represents nearly two cycles of transgression and regression of the Phosphoria sea.

Even though some of the phosphatic shale beds contain layers that resemble the uraniferous black shales of other formations, only the beds high in phosphate content are appreciably uraniferous.

The uranium content of the formation varies areally and with the phosphate content, as shown in maps and tables. The reasons for this areal variation are not well understood. The phosphate content and the uranium content vary in direct proportion, but there are many exceptions seemingly due to other factors such as organic content, carbonate content, textural variations, and effects of weathering. Most of the uranium is in carbonate-fluorapatite, probably substituting

for the calcium. The uranium is thought to enter the carbonate-fluorapatite at the time of precipitation or after burial. Smaller concentrations of uranium may be present also in the organic matter or other constituents. The uranium content and the carbonate content vary inversely with one another. Secondary mineralization has been observed and is believed to have resulted from leaching of uranium by surface weathering and subsequent migration in and precipitation from ground waters.

40. McKelvey, V. E., Everhart, D. L., and Garrels, R. M., 1955, Origin of uranium deposits: Econ. Geology, 50th Anniversary volume, 1905-1955, Part I, p. 464-533. See p. 514-526.

After a brief summary of the geochemistry of uranium and the occurrence of uraniferous deposits, the geology, mode of occurrence and origin of the following types of uranium deposits are discussed: igneous rocks, veins, sandstone-type deposits, coal and associated carbonaceous shale, and marine sedimentary rocks (black shales and phosphorites).

Uraniferous marine black shales such as the alum shale of Sweden, the Chattanooga shale of Tennessee and the Hartville formation of eastern Wyoming contain from 0.005 to 0.02 percent uranium and minor amounts of other metals including vanadium, nickel, molybdenum, and zinc. The uraniferous black shales generally are sapropelic, rather than humic in origin; are rich in sulfides, distillable hydrocarbons and fine carbonaceous matter; are somewhat more phosphatic but less calcareous than other shales; and are relatively thin considering the period of time their sedimentation presumably represents.

Much of the uranium in the Chattanooga shale is in the organic matter-pyrite complexes and studies indicate that the uranium is not present as a discrete mineral. It is possible that the uranium in the alum shale of Sweden is carried mainly in fine-grained kolm pellets, although we are aware of no studies along this line. It is evident from many investigations that uranium may occur in several forms in black shales but the evidence "strongly suggests that the uranium in most black shales is held by one or another type of organic matter."

"The uranium in marine black shales and phosphorites was derived from sea water. The precipitation of uranium in the shales probably is brought about by chemical adsorption by living or dead plankton, but reduction evidently plays an important role in its precipitation and stabilization in the shale. Uranium in the phosphorites substitutes for calcium in the apatite structure." A list of 192 references, many dealing with uranium in carbonaceous materials, is included at the end of the report.

Note: For information of a similar nature see also: McKelvey, V. E., Everhart, D. E., and Garrels, R. M., 1956, Summary of hypotheses of genesis of uranium deposits: Proc. Internat. Conf. on peaceful uses of atomic energy, Geneva, 1955, v. 6, p. 551-561, United Nations, New York; U. S. Geol. Survey Prof. Paper 300, p. 41-53.

41. McKelvey, V. E., and Nelson, J. M., 1950, Characteristics of marine uranium-bearing sedimentary rocks: Econ. Geology, v. 45, p. 35-53.

Uranium occurs in abnormal concentrates in some marine sediments. The most important and widespread deposits are in black shales and phosphorites, many of which contain from 0.01 to 0.02 percent uranium. The beach placer deposits of Nome, Alaska and possibly the Witwatersrand conglomerates of South Africa constitute the only other known significant occurrences of marine origin.

Uranium-bearing black shale

Uranium in black shale was first discovered in 1893 in the Cambrian alum shale of Sweden. It has subsequently been found in the "black shales of several other formations, including the Dictyonema and Leningrad shales of Estonia and Russia, which are the stratigraphic equivalents of the alum shale of Sweden; the Precambrian Nonesuch shale of Michigan; the Devonian(?) Woodford chert of Oklahoma...; the Devonian and Mississippian Chattanooga shale of east-central United States and the Mississippian Calico Bluff formation of east-central Alaska."

The uranium-bearing marine shales have several features in common. "All are black or dark in color, rich in organic matter and sulfides, low in carbonate, relatively thin, and those known are of pre-Mesozoic age." Most of them are of great lateral extent, contain little clastic material, and represent long periods of depositional stability. The uranium in the marine black shales described is syngenetic and is

believed to have been chemically adsorbed by clay minerals or by organic matter. The strong positive correlation between uranium content and carbonaceous material in marine black shales suggests that the uranium has been adsorbed by the organic matter rather than by the clays. A list of formations containing black shale that may be uraniferous is included.

Uranium-bearing phosphorites

Uranium in phosphorite was first discovered in 1924 in the phosphorite deposits of Cretaceous and Eocene age in Algeria. Since then, uraniferous phosphorites have been discovered in many other parts of the world. In the United States, phosphorite in the Pliocene Bone Valley formation of Florida and in the Permian Phosphoria formation of northwestern United States contain significant concentrations of uranium. In contrast to the marine phosphorites, the residual phosphate deposits so far tested do not contain significant amounts of uranium.

Uraniferous phosphatic nodules have been found in many black shales, such as those of Pennsylvanian age in Kansas and Oklahoma. These phosphatic black shales "are a cross between the typical phosphorite deposits and the uraniferous black shales."

The phosphatic rocks that contain significant amounts of uranium range in age from Paleozoic to Cenozoic, are in relatively thin formations, and many are associated with

unconformities. The uranium content generally increases with the phosphate content but decreases with an increase in the carbonate content.

The uranium content of the Phosphoria formation increases westward from Wyoming into Idaho where an increase in the total phosphate content causes a marked increase in total thickness of the formation. Since the uranium-phosphate ratio is not constant in the Phosphoria formation, it is thought that some of the uranium may be associated with the organic and clay fractions in addition to the usual fluorapatite association.

Origin

It is believed that the uranium in these various marine sediments had a syngenetic origin and was removed from the sea water: (1) by direct chemical precipitation, (2) by its selective removal by certain marine organisms, or (3) by adsorption to clay or organic matter in an environment unfavorable for the accumulation of large amounts of clastic or carbonate materials.

42. McKeown, F. A., 1951, Reconnaissance of radioactive rocks of Vermont, New Hampshire, Connecticut, Rhode Island, and southeastern New York: U. S. Geol. Survey TEI-67, 46 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge. See p. 25-33 and 43-46.

In 1948, a reconnaissance using a car-mounted Geiger-Mueller counter was made in Vermont, New Hampshire, Connecticut, Rhode Island and southeastern New York to test the radioactivity of roadside rocks and soils. The author observed abnormal radioactivity in the rocks and soils of three areas which he called "radioactive provinces". These are: (1) the Hudson Highlands of southeastern New York and the Housatonic Highlands of western Connecticut, with an estimated maximum equivalent uranium content of 0.240 percent; (2) central New England, with an estimated maximum equivalent uranium content of 0.006 percent; and (3) the Milton area, Chittenden County, northwestern Vermont, with an estimated maximum equivalent uranium content of 0.029 percent.

Abnormal radioactivity was found in the Hudson and Housatonic Highlands region in gneiss, schist, and pegmatite. The radioactivity of some of the gneiss is believed to be of sedimentary syngenetic origin. The radioactivity in the other gneisses, schists, and pegmatites, however, is of hydrothermal origin.

Abnormal radioactivity in central New England was found in gneiss, schist, phyllite, slate, granite and pegmatite. Phyllite in the Orfordville formation of Middle Ordovician(?) age from two localities near Brattleboro in southeastern Vermont contains a field estimate of from 0.003 to 0.005

percent equivalent uranium. Micaceous slate of early Paleozoic age in the Dixville Notch in northern New Hampshire contains a field estimate of 0.003 percent equivalent uranium. No laboratory analyses were made of these rocks. Black, petroliferous, phosphatic, calcareous clay galls in intraformational breccias of the Milton dolomite of Late Cambrian age near Milton, Vt., contain an average of 0.006 percent uranium and as much as 8.06 percent P_2O_5 and 0.04 percent V_2O_5. A range in grade of 0.002 to 0.018 percent uranium was noted. The radioactivity of the rocks in the Milton area is believed to originate from uranium. Thorium, however, is believed to be the cause of most of the radioactivity of the other two "provinces" since chemical analyses on igneous and metamorphic rock samples to date have revealed a high thorium content and a low uranium content.

"The manner in which the radioactive elements are combined with other elements in the rock is not known. Alpha radiographs of thin sections ... show ... no apparent centers of radioactive concentrations." Phosphate analyses indicate a direct relationship between the uranium and phosphate content. "The fact that the radioactive material apparently has a wide areal and narrow stratigraphic distribution and is disseminated in ... the rock suggests that the radioactivity is a feature of the original sediments."

43. Mapel, W. J., 1956, Uraniferous black shales in the northern Rocky Mountains and Great Plains Regions: Proc. Internat. Conf. on peaceful uses of atomic energy, Geneva, 1955, v. 6, p. 445-451, United Nations, New York; U. S. Geol. Survey Prof. Paper 300, p. 469-476.

In 1953 a reconnaissance search for uranium in black shale was conducted in the northern Rocky Mountain and Great Plains regions. The black shales in these regions range in age from Precambrian to Late Cretaceous. Outcrops, drill cuttings, cores, and gamma-ray well logs were examined. The location and name of the formations examined, and the maximum uranium content of the shale are shown in tabular form and on index maps.

A black shale sequence, ranging in thickness from a few inches to 100 feet, at the base of the Madison limestone of Mississippian age was sampled at 17 localities in Cache County, northwestern Utah and in southwestern and central Montana, and contains as much as 0.005 percent uranium in Gallatin County, Mont.

Core chip samples of shale from oil and gas wells penetrating the *Bakken formation (a correlative of the basal shale of the Madison limestone) were sampled in northeastern Montana and northwestern North Dakota. Some of this shale contains as much as 0.007 percent uranium. Samples from a well in Daniels County, Mont., contain an equivalent uranium content of as high as 0.010 percent but only 0.003 percent uranium. The disequilibrium was found to be caused chiefly by the presence of thorium 230 and radium, both daughter products of uranium. The radioactivity in the *Bakken formation seems

to differ "appreciably and with a definite pattern from place to place. Recognition of the pattern doubtless will help in outlying areas in which maximum concentrations of uranium in the formation might be expected."

A black phosphatic shale sequence, ranging in thickness from a few inches to 75 feet, at the base of the Brazer limestone of Mississippian age was sampled at two localities in southeastern Idaho and one locality in Rich County, northern Utah. The shale at the Idaho localities was essentially non-uraniferous, but an upper shale unit, 14 feet thick, and a lower shale unit, 8 feet thick, at the Utah locality contain as much as 0.006 percent and 0.009 percent uranium, respectively.

The Heath shale, a marine black shale of Mississippian age, contains a reported maximum of 0.006 percent uranium in Fergus County, central Montana, but less than 0.003 percent equivalent uranium in Roosevelt County, northeastern Montana. "Gamma-ray log data from oil and gas wells indicate that a bed of Heath shale, 10 feet in thickness, may contain more than 0.01 percent equivalent uranium in McCone County, northeastern Montana and that parts of the formation in Musselshell County, Mont., may be nearly as radioactive."

Samples of black shale beds as much as 6 feet in thickness, in the Minnelusa sandstone of Pennsylvanian age, were taken from 2 oil and gas wells in southeastern Wyoming. The uranium content of the shale is reported to be from 0.005 to 0.019 percent. Data from gamma-ray well logs of Colorado shale of Cretaceous age in central Montana indicate as much as 0.008 percent

equivalent uranium in Musselshell County and a range of 0.002 to 0.005 percent equivalent uranium for other parts of central Montana. Gamma-ray log data further indicate that black shale from the lower part of the Pierre shale of Late Cretaceous age in northeastern South Dakota may contain 0.008 percent equivalent uranium.

44. Moxham, R. M., and Nelson, A. E., 1952, Reconnaissance for radioactive deposits in south-central Alaska, 1947-49: U. S. Geol. Survey Circ. 184, 14 p. See p. 1-6.

In 1947 a reconnaissance for radioactivity was undertaken in areas adjacent to highways. Known pegmatites and beach placers in south-central Alaska also were examined.

Shale beds from the Kennicott formation of Early Cretaceous age in the Wrangell Mountains contain from an "insignificant amount" to 0.002 percent equivalent uranium. Shale beds of the Matanuska formation of Late Cretaceous age in the Matanuska Valley that were tested were found to be essentially nonradioactive. An argillite of Mesozoic age along the Richardson highway, is also essentially nonradioactive.

45. Narten, P. F., and McKeown, F. A., 1952, Reconnaissance of radioactive rock of the Hudson Valley and Adirondack Mountains, New York: U. S. Geol. Survey TEI-70, 54 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge. See p. 7-10, 25-28.

In 1949 a reconnaissance for radioactivity, using a car-mounted Geiger-Mueller counter, was made of the sedimentary and metamorphic rocks of Paleozoic age in the Hudson and Champlain Valleys of eastern New York and western Vermont and the metasedimentary and igneous rocks of Precambrian age in the Adirondack Mountains in eastern and central New York. The average equivalent uranium content of the most radioactive Paleozoic sedimentary rocks is about 0.003 percent. The highest equivalent uranium content of these rocks is estimated to be 0.005 percent in some slates of Early Cambrian age near Fairhaven, Rutland County, western Vermont. Precambrian pegmatites were the most radioactive rocks encountered, containing as much as 0.043 percent uranium and 0.62 percent thorium.

One sample of each of the following argillaceous rocks of Paleozoic age in New York was submitted for analysis: a dark gray shale from the Deepkill shale near Stuyvesant, Columbia County; a black shale from the Bakoven shale near Medway, Greene County; gray and black shale from the Esopus shale near Ravena, Albany County; black shale from the Canajoharie shale near Ballston Spa, Saratoga County and a slate from the Hudson River group near Chatham, Columbia County. All of these samples contain 0.002 percent equivalent uranium and 0.001 percent uranium.

A reconnaissance examination for radioactivity was made of the following argillaceous rocks in New York: slaty shale, phyllitic slate and dark slate from the Hudson River group; phyllite and slate from the Berkshire schist; shale from the Rensselaer graywacke; slate from the Normanskill shale; shale from the Snake Hill formation and black shale from the Marcellus shale. A slate from the *Bomoseen grit of Early Cambrian age from western Vermont also was examined for radioactivity. The average estimated equivalent uranium content for all of these rocks is about 0.003 percent.

46. Nelson, J. M., and Narten, P. F., 1951, Reconnaissance of radioactive rocks of Maine: U. S. Geol. Survey TEI-68, 43 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

In 1948 a reconnaissance for radioactivity, using a car-mounted Geiger-Mueller counter, was made of roadside rocks, soils and glacial materials in the state of Maine. Abnormal radioactivity was found in granite, pegmatite, diorite, gneiss, schist, phyllite, quartzite, greenstone in tuff matrix, road metal and glacial till. A sample of black phyllite of Silurian age contains as much as 0.003 percent uranium. No uranium deposits of economic importance were found during the course of this reconnaissance examination.

Samples of the abnormally radioactive rocks were collected and analyzed for radioactivity in the field and later in the laboratory. A sample of gray phyllite of Ordovician and Silurian age from the Seboomook area, Somerset County in west-central Maine contains 0.002 percent equivalent uranium and 0.001 percent uranium. Three samples of black phyllite of Silurian age were collected from the Monson, Harrington Lake and Kokadijo areas, Piscataquis County in central Maine. These three samples contain from 0.002 percent to 0.003 percent equivalent uranium. No uranium was found in the samples from the Monson and Kokadijo areas. The sample from the Harrington Lake area was not analyzed for uranium.

47. Nininger, R. D., 1954, Minerals for atomic energy: New York, D. Van Nostrand Co., Inc., 367 p. See p. 75-76 and 155.

This book is a comprehensive guide to exploration for uranium, thorium, and beryllium and includes data on the major producing and potential areas of the world.

Deposits in phosphate rock

Uranium is present in the phosphatic Bone Valley formation of Florida and Phosphoria formation of northwestern United States and also in phosphatic nodules occurring in certain shales of Kansas and Oklahoma. Uranium-bearing phosphorites are also known from Russia, Egypt, Algeria, Tunisia, Morocco and Nigeria. In Devon, England uranium occurs in phosphatic nodules in shaly limestones. Non-marine phosphates, such as those of Tennessee and parts of Florida contain little uranium.

The uranium-bearing marine phosphorites range in age from Paleozoic to Pliocene and all seem equally favorable. In general, there is a good correlation between uranium and phosphate content. The uranium is found to be associated primarily with the calcium phosphate mineral, fluorapatite, although in places uranium may be associated with clay minerals and organic matter.

The Phosphoria formation of Permian age, which is appreciably uraniferous, crops out in Montana, Idaho, Wyoming and Utah. The uranium content varies a great deal from place to place; it increases westward from Wyoming into Idaho and decreases westward in Montana. The formation is from 200 to 500 feet

thick and the beds of phosphate contain much extraneous material. The beds containing more calcium contain little uranium. The Phosphoria formation contains a small amount of many other metals including vanadium which has been mined commercially.

Deposits in black shales

The uranium-bearing black shales have many features in common: (1) they are black and rich in organic matter and iron sulfides, (2) they contain very little lime, (3) they all contain petroleum which, however, appears to exist in inverse proportions to the uranium content, (4) they are all relatively thin formations that were deposited very slowly over a long period of time, and (5) most of them are geologically old formations. Only a few of the numerous black shales contain appreciable uranium. The carbonaceous or coaly type of black shale is essentially barren of uranium.

The uranium in the marine black shales, as well as in the phosphorites, is of syngenetic origin. The uranium in the black shales is probably associated with the clay minerals or the organic matter as no discrete uranium minerals have been found. The uranium content is generally less than one-fifth of a pound per ton and only the alum (kolm) shale of Sweden contains as much as one-half of a pound or more per ton.

The most promising domestic occurrence of uranium in black shales is in the Chattanooga shale in the Sparta-Smithville-McMinnville area midway between Knoxville and Nashville, Tennessee. Other outcrops of the Chattanooga shale and its

equivalents in Alabama, Georgia, Kentucky, Illinois, Indiana, Michigan and Ohio, including the New Albany, Antrim, Sunbury and Utica shales, contain lesser quantities of uranium. The Nonesuch shale of Michigan, the Woodford chert of Oklahoma and the Calico Bluff formation of east-central Alaska and several others in the Great Plains and Rocky Mountain states contain traces of uranium.

Uranium occurs also in the black alum shales of southern Sweden, Estonia, and the Leningrad district of Russia. The Swedish deposits of Cambrian age were first discovered in 1893 and some of these deposits were mined for radium in 1909. These shales have unique nodules and small lenses of almost pure hydrocarbon that often contain as much as 0.5 percent uranium. Experimental extraction of uranium from these shales has been effected only recently.

Uraniferous phosphatic nodules have also been described from black shales of Pennsylvanian age of Kansas and Oklahoma, and from black shales of the English Midlands area.

An index map of the United States showing generalized areas of uranium-bearing shales is included on page 155.

48. Ostrom, M. E., Hopkins, M. E., White, W. A., and McVicker, L. D., 1955, Uranium in Illinois black shales: Ill. Geol. Survey Circ. 203, 15 p.

"One hundred and seventy-five samples of dark gray to black shale [of Ordovician, Devonian, Mississippian, and Pennsylvanian age] were taken from outcrops in 44 Illinois counties. The highest equivalent uranium content, as determined by radiometric assay, of any of the samples was .017 percent. The uranium oxide (U_3O_8) content of 15 of the shale samples having the higher percent equivalent uranium ranged between .001 and .014 percent. The uranium oxide averaged .002 percent lower than the percent equivalent uranium for these fifteen samples..... No obvious regional variations in the equivalent uranium values of the shales were noted.... Phosphatic nodules and bands found in relatively small quantities in some black shales have a higher percent equivalent uranium than the shales in which they occur. The maximum uranium oxide content of the phosphatic materials investigated was .075 percent."

Thirty-nine shale units of Pennsylvanian age, most of them associated with coal beds, were sampled. High values obtained are listed below:

Beds Sampled	Percent Uranium
Shales above Upper McLeansboro coals	0.012
Shale above Cohn coal	0.019*
Shale below *Shoal Creek limestone	0.13
Shale above Macoupin coal	0.014, 0.075*
Shale above No. 6 coal	0.006
Shale above No. 4 coal	0.014
Shale above No. 2 coal	0.008
Shale above Davis coal	0.009
Shale above Stonefort limestone member of the Tradewater formation	0.013
Shale at position of Reynoldsburg coal	0.001

*Analyses of phosphatic nodules

A sample of shale from the Mountain Glen shale of Devonian age in Union County, southern Illinois, contains as much as 0.013 percent uranium.

49. Patterson, E. D., 1954, Radioactive coal and shale of Pennsylvanian and Permian age in northern West Virginia: U. S. Geol. Survey TEI-494, 17 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

During 1953 the commercially important coal beds and associated shales in the Monongahela formation of Pennsylvanian age and coal and shale of the Dunkard group of Permian age were examined and sampled for radioactivity in eight counties in northern West Virginia. The shales of the Monongahela formation average about 0.004 or less percent equivalent uranium. The black and canneloid shales of the Dunkard group average about 0.002 percent equivalent uranium. Most of the rocks examined were nonradioactive. Coal samples of the Dunkard group contain as much as 0.003 percent equivalent uranium and the Monongahela formation, 0.001 or less percent equivalent uranium.

Eighty-eight samples, the majority of which were coal, were collected from 16 localities. Six samples of black shale and gray to black siltstone from the Dunkard group of Permian age were collected in Wetzel and Marion Counties, W. Va. These six samples contain a maximum of 0.002 percent equivalent uranium. A sample of canneloid shale associated with the Washington coal of Permian age in Marion County contains about 0.002 percent equivalent uranium. Black shales, siltstones and mudstones of Pennsylvanian age were not sampled because of the low radioactivity exhibited at their outcrops. These rocks are part of the Waynesburg, Pittsburgh and Redstone coals of the Monongahela formation and the Freeport coal of the Allegheny formation.

50. Patterson, E. D., 1954, Radioactivity of some coals and shales in southern Illinois: U. S. Geol. Survey TEI-466, 23 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

In 1954 a reconnaissance examination was made of the radioactivity of the coal and carbonaceous shales of Pennsylvanian age in central and southern Illinois. Channel samples were collected of the commercially important coal beds and their hard, black, sheety roof shales in the Caseyville, Tradewater, Carbondale and McLeansboro formations. The uranium content of most of the coal and black shale sampled is less than 0.001 percent. However, a sample of bright attrital coal from the Herrin No. 6 coal bed of the Carbondale formation near Harrisburg, Saline County, contains 0.008 percent uranium and 0.125 percent uranium in the ash of the sample, and a sample of black, hard, pyritic shale overlying the Harrisburg No. 5 coal, Equality quadrangle, Saline County, contains 0.017 percent uranium and 0.027 percent uranium in the 61.4 percent ash of the sample.

Of the marine black shale beds sampled, only three contained more than 0.001 percent equivalent uranium. These are as follows: (1) the shale directly overlying the Davis(?) coal of the Tradewater formation in Saline County contains from 0.002 to 0.006 percent equivalent uranium; (2) the shale directly overlying the Harrisburg No. 5 coal of the Carbondale formation in Saline, Williamson, Jefferson, and Gallatin Counties contains from 0.002 to 0.018 percent equivalent uranium; and (3) the shale directly overlying the Herrin No. 6

coal of the Carbondale formation in Williamson, Perry, Randolph and St. Clair Counties contains from 0.005 to 0.007 percent equivalent uranium.

"It is evident that these black shales are radioactive over a wide area." The radioactivity seems to be higher in the thinner black shale beds that contain pyrite.

51. Pepper, J. F., 1953, Uranium in carbonaceous rocks, Reconnaissance, black shale in Eastern States, in Geologic investigations of radioactive deposits, Semiannual progress report, June 1 to November 30, 1953: U. S. Geol. Survey TEI-390, p. 125, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

"Preliminary survey [of the literature] shows that there are approximately 75 named black or partly black shales east of the Mississippi River besides a large number of unnamed black shales that are referred to in geologic sections in some areas."

Measurements of the radioactivity of the Dunkirk shale of Late Devonian age were made at 28 localities in west-central and western New York state. This shale was found to contain on the order of 0.003 to 0.004 percent equivalent uranium. "The radioactivity appears to decrease from west to east, perhaps related to an increase in the silt content of the shale."

Note: This is essentially the author's complete report.

52. Rankama, Kalervo, and Sahama, T. G., 1950, Geochemistry, Univ. of Chicago Press, Chicago, 912 p. See p. 636-638.

This comprehensive review of geochemistry is divided into two main parts: (I) "general geochemistry" and (II) "manner of occurrence of the elements." In Part II, one chapter each on uranium and thorium is included. The geology of uranium is discussed under the sub-headings: "Abundance and general geochemical character," "Uranium in igneous rocks," "Cycle of uranium," "Biogeochemistry of uranium," and "Ores of uranium."

Some terrigenous sediments are rich in uranium because they contain uranium-bearing, detrital heavy minerals. From the few determinations of uranium in sediments, it is concluded that the amount of uranium is small in sediments containing little or no organic matter.

The concentration of uranium by biologic processes is considered an important reason for the enrichment of uranium in some sedimentary rocks. Carbonaceous marine shales contain more uranium than other sediments and even more than most granites. It is believed also that the uranium may have been precipitated as a sulfide, along with iron, in the original sapropelic sediments and, in some cases, along with vanadium, molybdenum and nickel. It has been found that the uranium and potassium content of organogenic black shales and igneous rocks increase directly with one another.

Kolm, an ash-rich coal in the alum shales of Sweden, contains as much as 1 percent uranium. The bulk of the alum shales, however, contains an average of from 50 to 100 grams of uranium per ton with some values as high as 200 grams of uranium per ton. An extensive bibliography is included at the end of the chapter, as well as several geochemical tables.

53. Ross, V. F., 1952, Autoradiographic study of marine shales: Econ. Geology, v. 47, p. 783-793.

An autoradiographic study of five marine shales was undertaken as a part of the research sponsored by the American Petroleum Institute into the influence of radioactivity on the natural production of hydrocarbons from organic matter in sediments. The purpose of the present study was to determine the source of the radioactivity by recording on sensitized nuclear emulsions the tracks of the alpha particles in the shales. Distribution curves of alpha-particle tracks and photomicrographs of the tracks are included.

Investigations of kolm from the alum shale of Sweden indicate that the radioactivity is associated largely with the inorganic constituents which are chiefly fine-grained pyrite, phosphate and carbonate with occasional salts of uranium. "The radioactivity is often concentrated at the surfaces of colloidal grains." Some radioactivity, however, also was observed in association with the organic matter.

The radioactivity of the Woodford shale of Texas, with its finely-disseminated pyrite, was found to be mainly associated with certain mineral grains. The seasonal white layers and grains of phosphate in the Woodford shale in Oklahoma exhibit about 3 times the radioactivity of the surrounding organic matter. There may be a relationship between the uranium and the mineral sulfides. Studies of the Cherokee shale (former usage) of Oklahoma revealed that the radioactivity is largely associated with phosphatic nodules

which are randomly distributed throughout the shale. The organic matter which surrounds these nodules exhibited only a small amount of radioactivity. The radioactivity of the Miocene *Playa del Rey shale /Miocene nodular shale(?)/ of southern California is "more extensively localized in the inorganic nodules and layers than in the organic layers and veinlets."

Conclusions are that: (1) "the radioactivity of the shales investigated is associated with both the organic and the inorganic matter, (2) the radioactivity is concentrated to a greater extent in the inorganic matter by a factor of 2.5," and (3) alpha particles originating in inorganic matter close to organic matter "might have been effective in transforming the latter into hydrocarbons."

54. Ruch, J. W., 1954, Status of uranium recovery from low grade sources: Mines Mag. (Denver), v. 44, no. 3, p. 105-106.

Problems encountered in processing several types of uranium ores are reviewed briefly. These include ores from vein deposits, Colorado Plateau deposits and various byproduct sources such as the Rand deposits of South Africa, the phosphate deposits of Florida and northwestern United States, and the Chattanooga shale deposits of southeastern United States. These materials range in grade from perhaps 0.005 percent to about 0.50 percent U_3O_8.

The Chattanooga shale is a very low-grade but uniform and widespread deposit. It is a bituminous shale containing about 60 to 80 parts per million of uranium, but containing only about one-third the oil potential of the oil shale in the Green River formation of Colorado and Wyoming. "If this large potential source is ever to be a uranium producer, the recovery of sulphur and hydrocarbon values appears mandatory." So far, attempts at beneficiating Chattanooga shale have met with little or no success, even though most known physical methods have been tried.

55. Runnels, R. T., Kulstad, R. O., McDuffee, Clinton, and Schleicher, J. A., 1952, Oil shale in Kansas: Kansas Geol. Survey Bull. 96, pt. 3, p. 157-184. See p. 183-184.

A stratigraphic and petrographic description of the oil shales of Pennsylvanian and Cretaceous age in Kansas is presented. Total recoverable oil from the shales that contain over 5 gallons per ton is estimated to be about 3 billion barrels.

Other possible uses for these oil shales include the recovery of phosphate and uranium. These shales contain, chiefly in phosphate nodules, about 136 grams of uranium per ton of shale (0.0136 percent).

56. Runnels, R. T., Schleicher, J, A., and Van Nortwick, H. S., 1953, Composition of some uranium-bearing phosphate nodules from Kansas shales: Kansas Geol. Survey Bull. 102, pt. 3, p. 93-104.

The phosphatic nodules in seven black shales of Pennsylvanian age from 11 localities in eastern Kansas contain an average of 30.2 percent P_2O_5, 0.017 percent U_3O_8, and 3.2 percent fluorine. Nodules from the *Muncie Creek shale member of the Iola limestone from Wyandotte County have the highest phosphate content (37.34 percent) and the highest uranium oxide content (0.03 percent) of any of the nodules analyzed. The mineral in these nodules is tentatively identified as a carbonate-bearing fluorapatite though chemically the component percentages lie between those of fluorapatite and dahllite.

As shown by a series of graphs, the uranium oxide, fluorine, and calcium oxide contents of the nodules generally increase with the increase in phosphate content and as the fluorine content increases, so does the uranium oxide content. Thus, "...if a location where the phosphate concentration in the nodules is 35 percent were selected, then slightly more than 0.02 percent U_3O_8 could be expected..." and if "...the maximum phosphate content of 42 percent were obtained, 0.04 percent or more U_3O_8 would be present."

57. Russell, W. L., 1944, The total gamma ray activity of sedimentary rocks as indicated by Geiger counter determinations: Geophysics, v. 9, p. 180-216.

"Geiger counter determinations of the gamma ray intensity of 510 rock samples have been made to determine the average radioactivity and frequency distribution of radioactivities of the various types of sedimentary rocks. The results, expressed in units of gamma ray intensity, show that limestones, sandstones and dolomites are of relatively low radioactivity, shales much higher, and black bituminous shales highest of all. The new data on the gamma radiation of the sediments may be used to improve the interpretation of radioactivity logs and to determine the values of surface radioactivity surveys." (author's abstract)

A listing of the radioactivity determinations and location, formation, age, and lithologic character of the 510 samples is included. The average radioactivity of 164 shale samples is 16.2 units (each unit is equivalent to 1×10^{-12} grams of radium per gram of rock). The highest radioactivity in the sequence of averages for lithologic types is 26.1 for 40 black and grayish-black shale samples. Generally, the more calcareous or sandy the shale, the less the radioactivity. "Another general rule is that the darker the shade of a rock, the greater its radioactivity .... This increase in radioactivity as the shade grows darker is probably related to the increase in organic matter.

The shale samples studied were from both outcrops and wells in a number of areas in the United States, Venezuela, and Trinidad, and are from formations ranging in age from Ordovician to Pliocene. The highest radioactivity (equivalent to 104 to 220 x 10^{-12} gm. Ra/gm. rock) of any rock measured was from phosphatic concretions in thin black shales associated with the Oologah limestone and the Checkerboard limestone. Both are of Pennsylvanian age and were sampled in a quarry near Tulsa, Oklahoma. The highest radioactivity of any shale studied was from a black shale of Pennsylvanian age overlying No. 2 coal in western Illinois. Other high radioactivity determinations are from the Chattanooga shale of northeastern Oklahoma, eastern Kentucky and Illinois and from several black shales of Pennsylvanian age and their associated phosphatic nodules of Oklahoma.

58. Russell, W. L., 1945, Relation of radioactivity, organic content, and sedimentation: Am. Assoc. Petroleum Geologists Bull., v. 29, p. 1470-1493.

"A comparison between the radioactivity and organic content of 510 samples of sedimentary rocks (see Russell, W. L., 1944) indicates a marked relation between certain types of organic content and radioactivity. Marine oil shales are associated with exceptionally high radioactivity, coals with abnormally low radioactivity, and other types of organic matter with intermediate radioactivities." Calculating from figures given, Russell suggests that the average uranium content of all shales is probably about 0.003 percent. Russell suggests that the uranium in shales may be (1) indigenous in the mineral grains, (2) extracted from solution by organisms, (3) precipitated from solution in a reducing environment, or (4) adsorbed by clay minerals. "Shales which yield oil when distilled are in general much more radioactive than those containing other types of organic matter. An analysis of the material balance between sedimentary and igneous rocks indicates that the sediments should have about the same radioactivity as the igneous rocks from which they were derived; the averages of the writer's tests seem to confirm this conclusion if the igneous source of the sediments resembles a granite." From available sample tests, it is suggested that the Chattanooga shale of Oklahoma is more radioactive where it is thicker, though it is probably the chemical nature of the environment rather than the speed of deposition or relative thickness which

controls the concentration of radioactive matter. "An analysis of the data bearing on the radioactivity of deep-sea deposits and oil shales indicates no evidence of a general increase in radioactivity with slowness of deposition." The fact that shales of Paleozoic age have a higher average radioactivity than younger shales is not because the Paleozoic shales are older, but that certain tectonic and chemical conditions were more prevalent during the Paleozoic era which led to a higher concentration of radioactive materials being deposited.

Determinations of radioactivity, indications of organic content, and lithologic characteristics are presented in tabular form.

59. Schnabel, R. W., 1955, The uranium deposits of the United States: U. S. Geol. Survey Mineral Inv. Resource Map MR 2.

This is a map (scale 1:5,000,000) of the United States showing the locations of the principal types of uranium deposits. The map is keyed to a listing of these deposits and brief descriptions of the principal types of deposits are given below. A short bibliography is included.

The outcrop of the Chattanooga shale, a marine black shale formation containing small percentages of uranium, is shown in green in the southeastern and south-central United States. The Woodford chert and Phosphoria formation are similarly indicated.

60. Schopf, J. M., 1953, Search for and geology of uranium in carbonaceous rocks, black shale investigations, organic matter of the Chattanooga shale, in Search for and geology of radioactive deposits, Semiannual progress report, December 1, 1952 to May 31, 1953: U. S. Geol. Survey TEI-330, p. 146-152, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Studies of portions of two drill cores of the Chattanooga shale were made to determine whether a correlation between laminae could be established over a distance of a half mile. No periodicity in the thick or thin bands was noted and "... variation in the persistence of microlaminae can be demonstrated in individual core specimens."

Identifiable plant materials in the Chattanooga shale include Foerstia, a group of planktonic marine algae, drift wood of Callixylon, and one large algoid plant, Prototaxites. Some "... nearly pure coaly mineral-free streaks that probably represent the bituminous remains of Callixylon ... have proved /to be/ unusually radioactive."

"Seven varieties of anomalous spore-like bodies are distinguished among organic constituents of Devonian black shale.... Abundant spores of undoubted land plants have been found, but ... these contrast strongly with the spore-like microfossils..." of uncertain affinity referred to the form genus Tasmanites. "Usually the latter types are dominant /in the Chattanooga shale/ and land plant spores are rarely seen."

Segregations of Foerstia thalli were made from the Huron member of the Ohio shale collected near Columbus, Ohio, and ash, colorimetric oil assay, uranium, and semi-quantitative

spectrographic determinations were made. Cu, Mo, Mn, V, Y, and U are shown to be more abundant in the ash of the Foerstia than in the ash of the shale matrix. The determination of organic elements in the Foerstia concentrate is also shown. These data are preliminary and data from more sample localities "... would aid in showing whether any of the trends of variation between the organic concentrate and matrix are consistent...." These studies may reveal a systematic variation in organic elements as does coal in a different depositional environment.

61. Snider, J. L., 1953, Radioactivity of some coal and shale of Pennsylvanian age in Ohio: U. S. Geol. Survey TEI-404, 22 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Measurements were made of the radioactivity of the commercially important coal beds and associated rocks of the Pottsville, Allegheny and Monongahela formations of Pennsylvanian age in eastern Ohio.

Only five samples contain an estimated 0.001 percent or more equivalent uranium; two of these were shale and three were coal. Two samples of shale, both from just above the Middle Kittanning coal of the Allegheny formation, were analyzed in the laboratory. One of these, from a bed of fossiliferous and pyritiferous marine shale about 6 feet in thickness (from Carroll County) contains 0.0004 percent uranium, 89.2 percent ash and 0.0005 percent uranium in ash. The other sample, from a bed of fossiliferous marine shale about six inches in thickness (from Tuscarawas County) contains 0.003 percent equivalent uranium but only 0.0007 percent uranium, 88.6 percent ash and 0.0008 percent uranium in ash.

None of the rocks sampled contain sufficient uranium to be of economic importance as a source of uranium at the present time.

62. Snider, J. L., 1953, Reconnaissance for uranium in coal and shale in southern West Virginia and southwestern Virginia: U. S. Geol. Survey TEI-409, 28 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

In 1952 a reconnaissance for uranium in coal and associated rocks was made in southern West Virginia and southwestern Virginia. In addition to checking the outcrops with radiation-detection equipment, channel samples of coal, shale and clay of Late Devonian, Mississippian, and Pennsylvanian age were collected for laboratory analysis. Two channel samples of a black, fissile shale of Late Devonian age from Lee County, Va. contain as much as 0.004 percent equivalent uranium. Other sandstone and shale associated with the coal beds contain from 0.001 to 0.003 percent equivalent uranium. All of the coals sampled contain 0.001 or less percent equivalent uranium.

The results of radiometric field examinations of measured stratigraphic sections are as follows:

Wise formation, Pennsylvanian age, Wise County, Va.:

Shale and sandy shale in six beds, ranging from 10 to 65 feet in thickness, contain an estimated 0.001 percent to 0.002 percent equivalent uranium.

Norton formation, Pennsylvanian age, Wise County, Va.:

Shale and sandy shale in fifteen beds, ranging from 4 to 125 feet in thickness, contain an estimated 0.001 percent to 0.002 percent equivalent uranium.

Kanawha formation, Pennsylvanian age, McDowell County, W. Va.:

Fossiliferous marine gray shale and sandy shale in four beds, ranging from 1 to 30 feet in thickness, contain an estimated 0.003 percent equivalent uranium.

Price formation, Mississippian age, Pulaski County, Va.:

Shale and silty shale in nine beds, ranging from 3 to 20 feet in thickness, contain an estimated 0.001 percent to 0.002 percent equivalent uranium.

Siltstone in one bed, measuring 17 feet in thickness, contains an estimated 0.001 percent equivalent uranium.

All other shales and siltstones examined in the field or sampled and analyzed contain less than 0.001 percent equivalent uranium.

63. Snider, J. L., 1954, Reconnaissance for uranium in the Indiana coal field: U. S. Geol. Survey TEM-784, 26 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

A reconnaissance examination for radioactivity in coal and associated rocks was made in the Indiana coal field of southwestern Indiana. This field is a part of the Eastern Interior coal province which also includes areas in southern Illinois and western Kentucky. Sandstone, siltstone, shale, limestone, clay and coal were examined in the following formations: the Mansfield, Brazil, Staunton, Linton, Petersburg, Dugger and Shelburn formations, all of Pennsylvanian age. Most of the coal beds examined in Indiana contain less than 0.001 percent uranium.

A marine black shale bed one foot in thickness, and immediately overlying coal No. V of the Petersburg formation in Pike County, Ind., contains as much as 0.021 percent uranium; other marine black shales generally contain 0.005 or less percent equivalent uranium.

Twenty-eight samples of shales of differing lithology were tested for uranium content in the laboratory. Of these, 20 "shale" samples contain an average of about 0.004 percent and a maximum of 0.01 percent equivalent uranium; 8 "gray" shale samples contain an average of about 0.003 percent and a maximum of 0.004 percent equivalent uranium and 20 "black shale" samples contain an average of about 0.007 percent and a maximum of 0.021 percent equivalent uranium. In addition, ten siltstone samples

and one mudstone sample contain less than 0.001 percent equivalent uranium and a pyrite nodule from a sample of black shale contains about 0.001 percent equivalent uranium.

The shale beds above the No. V coal (Petersburg formation) and Minshall coal (*Minshall formation) contain in places an average of about 0.006 percent equivalent uranium. "The radioactive shale was above the coal at all localities sampled.... There appears to be a relationship between radioactivity and lithologic type of shale...rather than to position in relation to the underlying coal or overlying limestone.... The very dark gray shale containing abundant organic matter is more radioactive than the lighter colored shale containing less organic matter. The shale with the greatest radioactivity is described as "grayish black to black, hard, very fissile when weathered, concoidal fracture when fresh, satiny luster, vitrain bands, few pyritic nodules....

"The fact that the radioactive matter was deposited contemporaneously with the shale is indicated by the wide areal extent of the radioactivity in a stratigraphically limited shale unit. The source of the radioactivity could be uranium in sea water...." The vitrain stringers are indicative of plant material which "...may have absorbed radioactive elements from the sea water.... The black shale above coal V is widespread in Indiana and adjacent states and the reserves of shale are large."

64. Stockdale, P. B., 1955, An investigation of the Chattanooga black shale of Tennessee as a source of uranium, Progress report, January 1, 1955 to June 31, 1955 (Univ. of Tenn.): U. S. Atomic Energy Comm. ORO-143, 11 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

This is a progress report of a continuing project begun in November 1951 at the University of Tennessee. The primary purpose of the project "is to obtain a complete geologic picture of the Chattanooga shale in the outcrop area of the Eastern Highland Rim and Northern Highland Rim of Tennessee and in a small area of southern Kentucky, with special emphasis upon stratigraphic relationships...."

The Chattanooga shale lies beneath 15,000 square miles of the Highland Rim and the Cumberland Plateau in Tennessee. "Assuming an average thickness of 25 feet for the Chattanooga formation, and an average grade of 0.0025 percent, the calculations show approximately 20,000,000 tons of uranium."

"The best average uranium grade in the Chattanooga shale in Tennessee is in the Gassaway member and the most readily accessible shale in large amounts with shallowest overburden is on the Highland Rim." Preliminary reserve estimates of uranium in the Gassaway member of five counties in the northern half of the Eastern Highland Rim indicate an average uranium content of 0.0057 percent, an average thickness of the Gassaway member of 15.8 feet, and total reserves of 1,810,000 tons of uranium.

65. Strahl, E. O., Camilli, Elena, Short, N. M., Silverman, E. N., Weiser, L. A., Wright, H. D., and Bates, T. F., 1954, An investigation of the mineralogy, petrography and paleobotany of uranium-bearing shales and lignites, Scope A - Shales; Third annual report - Period of April 1, 1953 to March 31, 1954 (College of Mineral Industries, Pennsylvania State University): U. S. Atomic Energy Comm. NYO-6060, 70 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Continuing studies were made at Pennsylvania State University, on behalf of the U. S. Atomic Energy Commission, to find the nature of the uranium-bearing compound or compounds and their associations and the controlling factors of uranium deposition in uraniferous shales. Sixty samples selected from a drill core of 35 feet of Chattanooga shale from the Young's Bend area of the eastern Highland Rim, Tennessee were utilized in the studies. The perfection of laboratory methods has resulted in the development of several important techniques including the application of ozonization procedures to remove carbonaceous material from thin section.

Qualitative and quantitative mineralogical studies of the 60 drill-core samples have shown that the major constituents, in addition to organic matter, are quartz, illite, pyrite, kaolin, feldspar, mica and chlorite. Accessory minerals include carbonates, zircon, tourmaline and apatite. "Specific samples, however, show a great variation in mineral content."

A careful study of the 60 core samples "has yielded statistically significant correlations between the amount of uranium and some of the shale constituents." The correlation of the amount of uranium is positive with respect to the

"blackness", or color of the sample; sulfur content; and associated carbon and pyrite content. The correlation of the amount of uranium is negative with respect to the content of feldspar, clay, and Fe_2O_3. Contrary to general belief, the black beds which contain more uranium are coarser grained than the gray beds.

Alpha-track studies of 13 thin sections show that significantly fewer tracks are associated with quartz plus feldspar while significantly more tracks are associated with opaques (mainly organic matter and pyrite).

"The data make it evident that the concentration of uranium in the Chattanooga shale is not due to the presence of any one of the components studied but rather to a combination."

66. Swanson, V. E., 1955, Uranium in carbonaceous rocks, black shale investigations, Midcontinent Devonian shales, in Geologic investigations of radioactive deposits, Semiannual progress report, December 1, 1954 to May 31, 1955: U. S. Geol. Survey TEI-540, p. 169-170, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

"An abnormally high-grade uraniferous black shale was found near Yellville, Marion County, /in north-central/ Ark. Two select samples contain 0.12 and 0.71 percent uranium, respectively. The black shale is confined to a lens less than 3 feet thick in the lower Mississippian basal sandstone member of the Boone formation, and is lithologically similar to the Chattanooga shale of northern Arkansas. The shale having the highest radioactivity contains much coalified plant material. Although analytical data on additional samples collected are not yet available, the average uranium content of the shale in the lens is believed to be less than 0.02 percent uranium."

Note: This is essentially the author's complete report.

67. Swanson, V. E., 1956, Uranium in marine black shales of the United States: Proc. Internat. Conf. on peaceful uses of atomic energy, Geneva, 1955, v. 6, p. 430-434, United Nations, New York; U. S. Geol. Survey Prof. Paper 300, p. 451-456.

Hundreds of black shale units, ranging in age from Precambrian to Tertiary, have been examined in the search for uraniferous marine black shales in the United States. This examination has been undertaken, for the most part, on a reconnaissance basis utilizing radiometric scanning methods with some sampling. The radioactivity of some subsurface shale units penetrated by oil and gas wells has been determined from gamma-ray well logs and cores. A few widespread shales contain on the order of 0.007 percent uranium, but "with few exceptions, the uranium content of known marine black shales of the United States does not exceed 0.02 percent, and the contents of few shale units having a thickness of more than 2 feet exceed 0.005 percent uranium." Although not currently being exploited, these vast deposits of low-grade uraniferous shale represent an extremely large reserve tonnage of uranium.

To date, the uranium mineral or minerals in black shales have not been identified. "The uranium is apparently thoroughly disseminated in the shale but is associated most closely with the contained organic matter ..." and was presumably adsorbed from solution by this organic material.

In general, significantly uraniferous marine black shales contain: (1) abundant organic matter with some oil content; (2) pyrite in the form of lenses, nodules and minute crystals; (3) phosphate as nodules or disseminated particles; and (4)

abnormal amounts of trace elements such as copper, nickel, vanadium, germanium and molybdenum. The uranium content generally decreases sharply with small increases of calcium carbonate. Many marine black shales are essentially non-uraniferous even though they are lithologically similar to the more uraniferous marine black shales.

Chattanooga shale

The Chattanooga shale, of Devonian and Mississippian age, is the largest low-grade reserve of uraniferous black shale in the United States. It crops out in central Tennessee and adjacent parts of Alabama and Kentucky, is generally less than 35 feet thick, and contains from 0.001 to 0.035 percent uranium. It consists mostly of black siliceous shale and gray claystone, with a few thin sandstone beds, calcareous lenses, chert layers, and phosphatic nodules. The black shale units of the Chattanooga are composed of about 35 percent clay, feldspar and mica; about 25 percent silt-sized quartz grains; about 20 percent organic matter; about 15 percent pyrite and minor amounts of chlorite, phosphate, calcite and heavy minerals.

"Correlatives of the Chattanooga shale, all slightly radioactive, are the New Albany shale of Illinois, Indiana, and Kentucky, the Antrim shale of Michigan, the Sunbury and Ohio shales of Ohio and Kentucky; the Genesee group, Dunkirk member of the Perrysburg formation and other thin shales of New York, the Chattanooga shale of Kansas, northern Arkansas and northern Oklahoma and the Woodford chert of Oklahoma and Texas. The

Chattanooga shale and its correlatives accumulated in a widespread, relatively shallow, epicontinental sea ..." where an extremely slow accumulation of mineral detrital material and a relatively high accumulation of carbonaceous debris took place.

Pennsylvanian shales, Midcontinent region

Approximately 2,500 feet of rocks of Pennsylvanian age are exposed in eastern Kansas and nearby parts of Oklahoma and Missouri. Marine black shale beds, generally only a few feet thick, are scattered throughout the sequence. Many of these beds are units of cyclic deposition that include layers of sandstone, coal and limestone. About 20 of the black shale units in this sequence are abnormally uraniferous and contain more than 0.003 percent uranium.

"Many of these black shales of Pennsylvanian age are characterized by irregularly distributed small phosphatic concretions or nodules...." These nodules may contain as much as 0.10 percent uranium with an average of 0.015 to 0.020 percent uranium and are generally more radioactive than the enclosing shales. No correlation has been found between uranium content and: (1) the abundance of these nodules in the shales, (2) the thickness of the shale beds or (3) the lithology of the shales.

These thin black shale units of Pennsylvanian age were apparently deposited in an extremely shallow sea with a bottom reducing environment and the accumulation of abundant organic

debris. The uranium in solution "was adsorbed in this bottom sludge, with a preferential chemical exchange bonding the uranium and the phosphate during the accretionary growth of the nodules."

Phosphoria formation

Two black, pyritic and phosphatic shale members of the Phosphoria formation of Permian age, exposed in southwestern Montana, eastern Idaho and adjacent parts of Wyoming and Utah, contain as much as 30 to 38 percent P_2O_5 and from 0.010 to 0.033 percent uranium. A positive correlation exists between the uranium content and the abundance of phosphatic and carbonaceous matter, and apparently these constituents contain the uranium. "The Phosphoria formation may be roughly divided into a miogeosynclinal facies on the west and a platform facies on the east." A genetic similarity to the other uraniferous black shales described is suggested although it is probable that the sediments accumulated in the Phosphoria sea at a more rapid rate.

Other uraniferous shales

Other uraniferous marine black shales in the United States include: thin black shale beds of Mississippian age in Montana and Utah, phosphatic black shale in the Barnett formation of Mississippian age in central Texas, parts of the Heath shale of Mississippian age in central Montana, black shale units in the Hermosa formation of Pennsylvanian age in western Colorado,

and pyritic and carbonaceous shale in the Sharon Springs member of the Pierre shale of Late Cretaceous age in southern South Dakota, northeastern Colorado, and adjacent parts of Nebraska and Kansas.

Geologic significance

Although marine carbonaceous shales accumulated in miogeosynclinal and eugeosynclinal areas, the general tectonic and paleogeographic category into which most uraniferous shales fall is that of the epicontinental area. This environment was common to much of the interior of the United States during several intervals of the Paleozoic. The long-term inundation of a broad expanse of a relatively flat surface, and the lack of a source of a large amount of clastic sediment made possible the development of specific geochemical conditions that controlled the accumulation and preservation of uraniferous carbonaceous muds.

68. Thompson, M. E., 1953, Distribution of uranium in rich phosphate beds of the Phosphoria formation: U. S. Geol. Survey Bull. 988-D, p. 45-67.

"Five sets of 'close' samples [of phosphate-rich beds from the Phosphoria formation] were analyzed radiometrically for uranium and chemically for P_2O_5, CaO, organic matter, and loss on ignition. A Rosiwal analysis was made of thin sections of one set of samples. The results of the analyses have been plotted on graphs and on scatter diagrams, and coefficients of correlation are given for uranium with CaO, P_2O_5, organic matter, and loss on ignition. Preliminary studies indicate that the concentration of uranium in these samples of phosphate rock is not due wholly to phosphate content, but may depend in part on organic matter or on other components. The correlations of uranium with P_2O_5 are poor in the groups of samples with smaller amounts of uranium but are good in the groups of samples containing more uranium." (author's abstract)

Note: The samples contain from 0.004 to 0.065 percent equivalent uranium, from 1 to 36 percent phosphate and from 1 to 12 percent organic matter.

69. Tourtelot, H. A., 1956, Radioactivity and uranium content of some Cretaceous shales, central Great Plains: Am. Assoc. Petroleum Geologists Bull., v. 40, p. 62-83.

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

tabular form and include analyses of samples of Big Stone Gap shale of Devonian and Mississippian age from Wise County, southwestern Virginia, 0.004 percent uranium; shale of Devonian age from Burnet County, central Texas, 0.01 percent uranium; and Chattanooga shale of Devonian and Mississippian age from Tennessee, 0.003 to 0.008 percent uranium.

In the Great Plains region, the upper 30-50 feet of Cretaceous shales overlain unconformably by the White River group of Oligocene age have been altered by pre-Oligocene weathering and later groundwater conditions to a brightly-colored zone. This altered zone in the Niobrara formation has masses of unaltered shale in its lower part that contain as much as 0.1 percent equivalent uranium and 0.03 percent uranium. This uranium is believed to have been derived from the overlying White River group. (from author's abstract)

70. Twenhofel, W. S., Finch, W. I., Osterwald, F. W., Bell, K. G., and Stead, F. W., 1954, Resource studies, in Geologic investigations of radioactive deposits, Semiannual progress report, June 1 to November 30, 1954: U. S. Geol. Survey TEI-490, p. 298-299, issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

Preliminary analytical data on hand indicate that the natural radioactivity of rocks at the surface of the earth varies widely: from a few parts per million to possibly 50 parts per million of equivalent uranium.

"Further analysis of the data should clarify the expectable 'normal' variation in radioactivity for a few limited environments such as the Mancos shale, which ranges from 6 parts per million to 40 parts per million of equivalent uranium where the total radioactivity of 40 parts per million consists of 20 parts per million caused by the uranium series of elements and 20 parts per million caused by the potassium series of elements.

"It is now apparent that a rather significant variation in the total radioactivity of some shales can be attributed to variation in potassium content with no change in uranium content. These studies are being continued."

71. Weaver, Paul, 1942, A theory of the distribution of radioactivity in marine sedimentary rocks: Geophysics, v. 7, p. 192-198.

The relative gamma-ray intensity of muds, clays, and shales is usually the highest of any of the sediments, as determined by gamma-ray logs of wells. According to recent estimates, the rate of sedimentation for marine shales is about 1 foot per 900 years (thickness after compaction). Thus, the gamma-ray measurements of marine sedimentary rocks indicate that in general the amount of radioactivity in marine sediments varies inversely with the rate of sedimentation.

"The most recent measurements of the radioactivity of sea water suggest an increase with depth, which indicates a very slow settling rate of small particles." The possible sources of these mineral particles are: (1) fine river muds, (2) volcanic dust from eruptions, (3) meteor clouds and (4) general dust circulated by large wind movements. Of these sources, only the last is considered to have the necessary attributes to account for a widespread and uniform deposit of relatively high radioactivity.

72. Wedow, Helmuth, Jr., 1954, Reconnaissance for radioactive deposits in the Eagle-Nation area, east-central Alaska, 1948: U. S. Geol. Survey Circ. 316, 9 p.

"Reconnaissance of radioactive deposits in sedimentary rocks of Proterozoic and Paleozoic age, and granite of Mesozoic(?) age together with its Tertiary sedimentary derivatives, was conducted in 1948 in the Eagle-Nation area, east-central Alaska. None of the rocks examined contains more than 0.003 percent equivalent uranium except for black shale beds in the Calico Bluff formation of Late Mississippian age and in granite of Mesozoic(?) age and its sedimentary derivatives. The more radioactive black shale beds in the Calico Bluff formation range in thickness from 1/2 to 7 feet. Two units near the base of the formation appear to be persistent in the area: Radioactive unit A, with an average thickness of 6.6 feet, which contains an average of 0.007 percent equivalent uranium and 0.004 percent uranium and radioactive unit B, with an average thickness of 5.2 feet, which contains an average of 0.006 percent equivalent uranium and 0.003 percent uranium. Phosphatic pellets from unit B at one locality contain 0.022 percent equivalent uranium, 0.019 percent uranium, and 15 percent P_2O_5." The maximum uranium content of the black shale in the two units is 0.006 percent. Though marine black shales of Cambrian, Ordovician(?), Devonian, Early(?) Mississippian, Permian and Triassic age are the most radioactive in the rock sequences that were traversed, they contain 0.003 percent or less equivalent uranium.

73. Wedow, Helmuth, Jr., White, M. G., and Moxham, R. M., 1951, Interim report on an appraisal of the uranium possibilities of Alaska: U. S. Geol. Survey TEM-235, 124 p. (open file). See p. 11, 98, 101, 106 and 113.

Summaries of the geology and mineral deposits, and appraisals of the uranium possibilities of the various regions of Alaska to date are presented in this preliminary report. A short statement of previous knowledge and investigations of radioactive materials in the Territory also is given.

During the reconnaissance investigations, outcroppings of various rocks, including sedimentary formations, were tested for radioactivity. A summary of those shales containing uranium of probable syngenetic origin is given below.

Northern Alaska

The uraniferous oolitic phosphate and phosphatic shales in the Lisburne group of Mississippian age along the north flank of the Brooks Range are the only uraniferous sedimentary rocks known in northern Alaska. Random samples from a phosphatic unit, which is similar in appearance and composition to the bedded phosphates of the Phosphoria formation in northwestern United States, average 31.95 percent P_2O_5 and 0.013 percent uranium.

Porcupine district

Radiometric traverses were made over thick sequences of sedimentary rocks of Precambrian and Paleozoic age and associated intrusive rocks in this district which is in the northeastern part of the Upper Yukon region of east-central Alaska. The black shales of Silurian, Devonian, and Mississippian age, which were among the most radioactive rocks traversed, contain not more than 0.005 percent equivalent uranium.

Tolovana district

In this district, which lies northwest of Fairbanks in east-central Alaska, black shales of Mississippian age were found to contain not more than 0.002 percent equivalent uranium.

Eagle district

Black shale beds near the base of the Calico Bluff formation of Mississippian age along this part of the upper Yukon River contain between 0.005 and 0.01 percent equivalent uranium. Phosphatic pellets and nodules that are scattered through some of these radioactive black shale beds, contain up to 0.02 percent equivalent uranium.

74. Wedow, Helmuth, Jr., White, M. G., Stevens, J. M., and Tolbert, G. E., 1954, Reconnaissance for radioactive deposits in east-central Alaska, 1949: U. S. Geol. Survey Circ. 335, 22 p. See p. 1-3.

Chapter A is concerned with the Fairbanks and Livengood quadrangles in east-central Alaska. In 1949, several mines and prospects in these quadrangles were examined for radioactivity. Metamorphic and sedimentary rocks of Precambrian and Paleozoic age were examined for radioactivity along the Elliott Highway.

The greatest radioactivity found in the rocks along the Elliott Highway was in an iron-stained Precambrian schist and in a carbonaceous(?) shale of Middle Devonian or Mississippian and Pennsylvanian age. Samples of these rocks contain 0.003 and 0.004 percent equivalent uranium, respectively.

Subsequent chapters contain no information on radioactivity of possible syngenetic origin in marine black shales or phyllites.

75. Welch, S. W., 1953, Radioactivity of coal and associated rocks in the anthracite fields of eastern Pennsylvania: U. S. Geol. Survey TEI-348, 31 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

In 1953 a reconnaissance study was made of the radioactivity in the anthracite coal fields of eastern Pennsylvania. Selected coal beds and associated rocks were examined in the Pottsville, Allegheny, and Conemaugh formations of Pennsylvanian age from each of the four major coal fields: (1) the Southern, (2) the Eastern Middle, (3) the Western Middle and (4) the Northern. Forty-six localities were visited and 150 coal and 3 shale samples were collected. The radioactivity of the coal, siltstone, and claystone is estimated to be 0.001 or less percent equivalent uranium, and the shale contains from about 0.001 to 0.003 percent equivalent uranium.

Most of the shale is described as dark gray and carbonaceous, and the shale beds range in thickness from 1 inch to ten feet, the majority being four feet or less in thickness. In Schuylkill County in the Southern field a bed of medium light-gray shale two feet in thickness from the Allegheny(?) formation contains about 0.003 percent equivalent uranium.

The author concludes that the uranium content of the rocks in the anthracite fields of eastern Pennsylvania is "too low to be of economic interest at the present time."

76. Welch, S. W., 1953, Radioactivity of coal and associated rock in the coal fields of eastern Kentucky and southern West Virginia: U. S. Geol. Survey TEI-347A, 38 p., issued by U. S. Atomic Energy Comm. Tech. Inf. Service Extension, Oak Ridge.

In 1952 a reconnaissance was made of the radioactivity of the coals and associated rocks of eastern Kentucky and Logan and Mingo Counties, W. Va. The coal-bearing beds of this region are in the Lee and Breathitt formations of Pennsylvanian age. Most of the radioactivity measurements were made from samples from operating coal mines and consisted chiefly of coal, with some samples of shale, siltstone, limestone, sandstone, underclay and flint fire clay. Two samples of intrusive dike rock also were tested. The radiometric tests showed the coal to have little or no radioactivity: 0.000 to 0.001 percent estimated equivalent uranium. The other rocks sampled were slightly radioactive and contained about 0.002 percent estimated equivalent uranium.

Shales were examined and tested at fifty-nine out of the sixty-one localities visited. Only three out of 132 shale samples tested contain as much as 0.002 percent equivalent uranium. These three shale samples were taken from Breathitt, Knott and Perry Counties in Kentucky. They are predominantly dark gray and carbonaceous and two contain marine fossils. The shale beds range in thickness from one to ten feet at the localities sampled.

Seven siltstone beds were tested at four localities but all contain less than 0.001 percent estimated equivalent uranium.

"No correlation between radioactivity and stratigraphic position nor between radioactivity and structural relationship was detected."

77. White, M. G., 1952, Reconnaissance for radioactive deposits along the upper Porcupine and lower Coleen Rivers, northeastern Alaska: U. S. Geol. Survey Circ. 185, 13 p.

A reconnaissance search for radioactive deposits was undertaken in 1948 in northeastern Alaska.

"The highest equivalent uranium content found in the sedimentary rocks on the upper Porcupine River, northeastern Alaska, is 0.005 percent." Black shale and black slate in the Tindir group of Precambrian and Early Cambrian(?) age contain from 0.001 to 0.004 percent equivalent uranium. Black shale of Silurian age contains the highest amount of radioactivity and included a range of 0.001 to 0.005 percent equivalent uranium. Black shales of Devonian age are essentially nonradioactive and samples contain no more than 0.002 percent equivalent uranium. The black shales of Mississippian and Pennsylvanian age contain from 0.001 to 0.003 percent equivalent uranium.

Though black shales have the highest radioactivity of the rocks tested along the lower Coleen River, none of these rocks (of Devonian or Mississippian and Pennsylvanian ? age) contains more than 0.003 percent equivalent uranium.

78. White, M. G., and Killeen, P. L., 1953, Reconnaissance for radioactive deposits in the lower Yukon-Kuskokwim Highlands region, Alaska, 1947: U. S. Geol. Survey Circ. 255, 18 p. See p. 1 and 9.

Investigations in the area between the lower Yukon and Kuskokwim rivers in western Alaska reveal that the only abnormal radioactivity in the vicinity of Flat is due to uraniferous zircon, an accessory mineral in monzonite.

The sedimentary rock sequence in the Flat area, consisting of gray and black shales interbedded with sandy shale and sandstone of Late Cretaceous age, was tested in the field for radioactivity but results were negative. "Particular emphasis was placed on outcroppings of black shale in an attempt to locate any bed that might contain a significant quantity of radioactive material. Samples from three different localities that gave the highest radiometric readings in the field contain only 0.002 percent equivalent uranium."

79. Whitehead, W. L., 1950, Studies of the effect of radioactivity in the transformation of marine organic materials into petroleum hydrocarbons, Research Project 43c, in Report of progress, Fundamental research on occurrence and recovery of petroleum, 1948-1949: Am. Petroleum Inst., p. 226-229.

This is a biennial report on the progress of continuing research conducted by the Massachusetts Institute of Technology for the American Petroleum Institute concerning the quantitative evaluation of the amount of crude oil formed by the effects of radioactivity on marine organic substances. The studies include research on: (1) the yield factor for reactions producing hydrocarbons, (2) the loss of radioactive energy through absorption in the sediments, and (3) the content and mode of occurrence of radioactive elements in organic marine sediments and sedimentary rocks.

Radioactivity determinations of fractions of powdered organic shales indicate that the finest size organic fractions may be more radioactive than the more argillaceous fractions. Measurements of the potassium content and the radioactivity of ten formations were made by using a flame photometer, and by alpha-particle and net beta counts. The rocks studied are from ten sedimentary formations of marine origin. Of these, there are three from east Texas: the Woodbine formation, the Eagle Ford shale and the Austin chalk; three from southern Mississippi: the Tuscaloosa and the Eutaw formations and the Selma chalk; two from Oklahoma: the Cherokee shale (former usage) and the Chattanooga shale; the Woodford shale from western Texas and the Miocene nodular shale from the Los Angeles Basin of California.

Graphs indicate the frequency distribution of total and net beta counts and the potassium content for the Eagle Ford shale, the Woodbine sand, and the Tuscaloosa and Eutaw formations. These curves indicate a very low variation of radioactivity. A graph shows a slight increase in total beta emission with depth for oil well core samples of the *Playa del Rey shale /Miocene nodular shale?/.

80. Whitehead, W. L., 1952, Studies of the effect of radioactivity in the transformation of marine organic materials into petroleum hydrocarbons, Research Project 43c, in Report of progress, Fundamental research on occurrence and recovery of petroleum, 1950-1951: Am. Petroleum Inst., p. 192-201.

This is a biennial report on the progress of continuing research conducted by the Massachusetts Institute of Technology for the American Petroleum Institute concerning the quantitative evaluation of the amount of crude oil formed by the effects of radioactivity on marine organic substances.

Analyses for potassium and organic carbon were made on several organic marine sedimentary rocks (see preceding annotation, Whitehead, 1950). Tables and graphs that show the results of these determinations plus total and net beta counts are included. It was found that the net beta activity increased with an increase of potassium for all the formations tested except the Miocene nodular shale of southern California. The Antrim shale of Michigan was shown to have the highest median content of potassium (2.77 percent).

Excellent correlation between organic carbon content and radioactivity suggests that a genetic relationship exists between the two. An increase in radioactivity with an increase in the phosphorus content (as nodules of calcium phosphate) is noted for the Miocene nodular shale and the Cherokee shale (former usage) of Oklahoma.

The apparent linear relationship between the carbon and phosphate content of the Miocene nodular shale suggests a common genetic factor in the environment of accumulation. The

phosphorus in ancient seas indirectly may have been responsible for the abundant organic matter as well as the precipitation of uranium. The organic matter, when precipitated as colloids, also may have taken some of the uranium out of solution.

Studies of the "tracks" that alpha-particles make on photographic film were made of the Cherokee, the Miocene nodular shale, the Woodford shale of Oklahoma and the Woodford shale of Texas. The results are given in tabular form and two photomicrographs of "tracks" from the first two named shales are included. A ratio of the number of alpha particles originating in inorganic material to the number originating in organic material in the thin sections of the shales tested is 2.4 to 1.

81. Whitehead, W. L., 1954, Hydrocarbons formed by the effects of radioactivity and their role in the origin of petroleum, in Faul, Henry, editor, Nuclear geology: p. 195-218, New York, John Wiley and Sons, Inc.

Previous studies conducted by the Massachusetts Institute of Technology, for the American Petroleum Institute, into the origin of petroleum through the bombardment of certain organic substances and their related compounds, are reviewed.

The average total radioactivity of 8 composite samples of sedimentary formations (including 5 shales) is about 4×10^{-12} grams of equivalent radium per gram of sediment. It was proved that a negligible amount of hydrocarbons are formed by the effects of radioactive bombardment, but this cannot account for the great bulk of the oil to be found in the sediments.

Tables and figures are given that show the relation of radioactivity to the carbon, potassium, and phosphorus content of several sedimentary formations, including the Antrim shale of Michigan, the Woodford shale of western Texas, the Cherokee shale (former usage) of Oklahoma, the Eagle Ford shale of eastern Texas and the Miocene nodular shale of southern California. An excellent correlation between radioactivity on the one hand, and phosphorus and organic carbon content, on the other, was established for the Miocene nodular shale and Cherokee shale. The radioactivity was shown to increase with an increase of potassium content for all the formations studied, with the exception of the Miocene nodular shale.

INDEXES

Part I - Author index

(Numbers correspond to the reference number, not to page number.)

Part II - Geographic index

(Numbers correspond to the reference number, not to page number.)

Ref. no.

Ref. no.

Part III - Stratigraphic unit index

(Numbers correspond to the reference number, not to page number.)

Part IV - Subject index

(Numbers correspond to reference number, not to page number.)

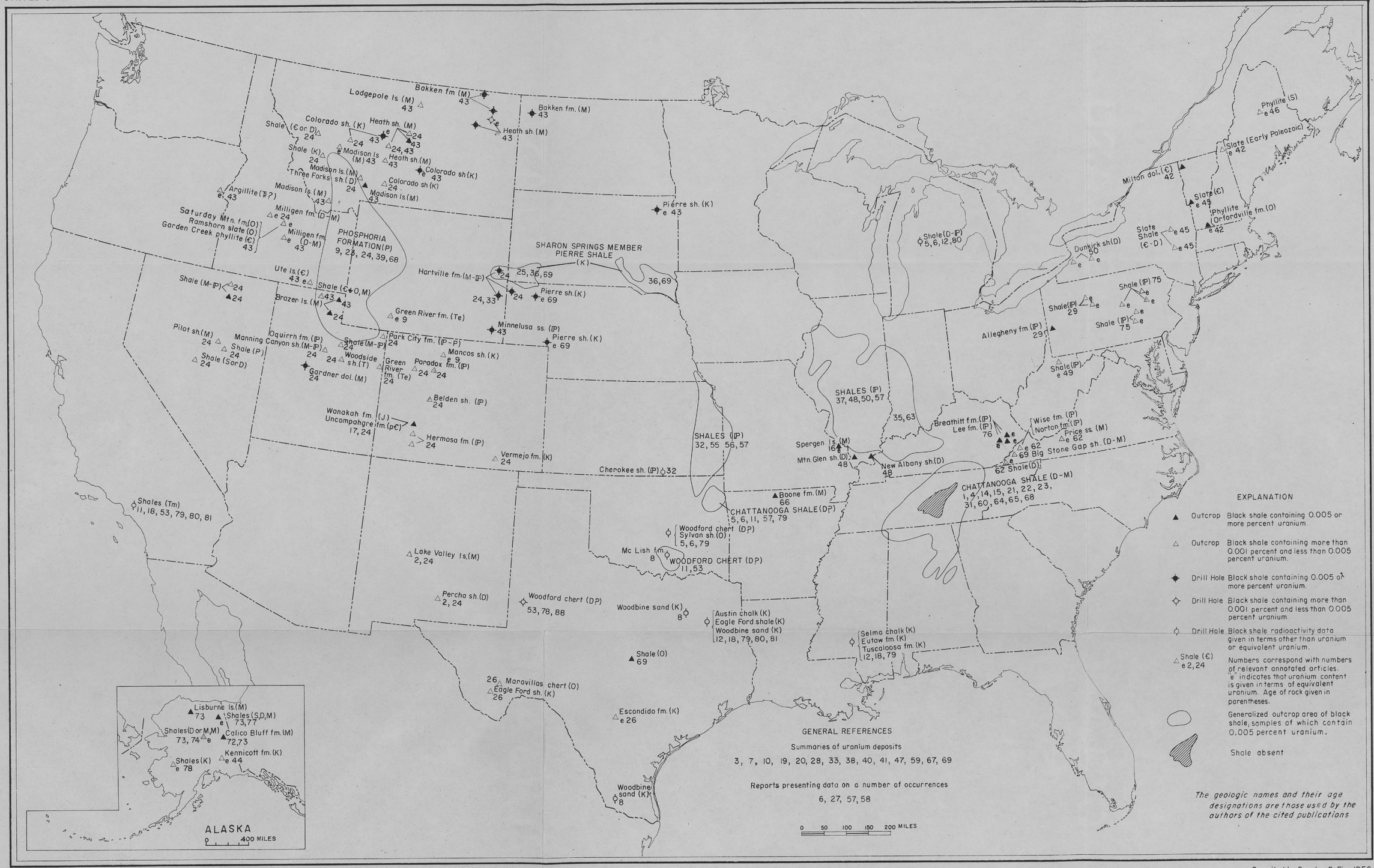
Ref. no.

Relationship studies between uranium content of black shale and:--Continued

Elements

DEPARTMENT OF THE INTERIOR
UNITED STATES GEOLOGICAL SURVEY

TRACE ELEMENTS INVESTIGATIONS
REPORT 535



Compiled by Carolyn E. Fix, 1956

FIGURE 1- MAP OF URANIUM-BEARING MARINE BLACK SHALES IN THE UNITED STATES, INDICATING URANIUM CONTENT AND PERTINENT REFERENCE FOR EACH LOCALITY OR AREA.