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1892—1973

A Biographical Memoir by

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BY GEORGE R. TILTON

CHARLES SNOWDEN PIGGOT WAS one of the founding fathers of ocean-bottom marine research. He was, in fact, a pioneer in the study of geologic phenomena in the ocean basins, whose work has revolutionized the way earth scientists view earth evolution. There were no ocean drilling projects when he began his research into radium activity in sediments. Consequently, he not only had to develop laboratory facilities for the measurements, but also had to design and build a coring device capable of obtaining undisturbed cores up to three meters in length. Techniques up to that time had been capable of collecting small surface samples of sediments with a grappling device, but the stratigraphic record was destroyed even in those limited samples. As Piggot stated, such samples give information about present conditions only and divulge nothing of past events. It is these past events that reveal valuable information about geologic processes at work on land as well as in the ocean basins.

His investigations finally produced reliable dates on sediments and sedimentation rates from the North Atlantic Ocean and the Caribbean Basin over a time span of some 300,000 years. Foraminiferal data from the cores, provided mainly by J. A. Cushman, showed that variations in the abundances of species marked changes in water temperatures that could be correlated with glacial and interglacial stages on the continents. This approach established a new and important method for working out the chronology of the glacial epochs. In addition, experiments with W. D. Urry established that the sedimentation rate in a Pacific Ocean red clay was lower by about a factor of five to ten compared to the Caribbean Basin. Although the techniques for measuring the water temperatures and ages of oceanic sediments by radioactive disequilibria have improved greatly since those early experiments, the pioneering work of Piggot and his colleagues placed the science on a firm footing for future studies.

Although Piggot is best known for his ocean sediment work, his scientific career was characterized by remarkable breadth. His doctoral dissertation was in the area of inorganic chemistry and involved oxidation of ammonia, a process for which he later received a U.S. patent. His first position after graduation was in organic chemistry at the United States Industrial Chemical Company, where he worked on processes for large-scale production of ethyl alcohol and acetic acid and their derivative products. Part of the work of his group there led to an improvement in the anesthetic quality of ethyl ether by addition of small quantities of ethylene.

Later in his career he entered the fields of geochemistry and geophysics at the Carnegie Institution of Washington. He provided the lead samples that Aston used to produce the first isotopic composition data for "common" lead and radiogenic lead from uranium decay. He received the Order of the British Empire for work on mine disposal in World War II and the Bronze Star from the U.S. Navy for service at the Bikini atomic test site in the postwar era.

Charles Snowden Piggot was descended from two generations of educators. His grandfather, Aaron Snowden Piggot, was a professor of natural philosophy in Baltimore and taught chemistry, geology, and mining engineering. He also taught chemistry in the School of Dentistry, which later merged with the Maryland Medical School. Finding no suitable text book he wrote *Piggot's Dental Chemistry,* a standard text book for many years. His son, Cameron Piggot, M.D. (1856-1911), studied medicine in Baltimore and was Professor Ira Remsen's scientific assistant at the Johns Hopkins University. He became a professor of chemistry at the University of the South in Sewanee, Tennessee, in 1889, and was later dean of the Academic Department.

Charles was born in Sewanee in 1892. His mother was Anne Olivia Cockey of a Maryland family. The Piggot family was always short of money because his father's salary was small and the university sometimes failed to pay it altogether. It never occurred to him that their financial plight was unusual because all the rest of the university staff was in the same condition. He found the Sewanee environment ideal for a growing boy. In many ways it seems to have been a classical mountain environment of the Old South. Their house was surrounded by forest with unlimited space for hiking, riding, hunting, and camping. Charles was required to carry drinking water from a nearby spring. It was also his duty to cut kindling, which, as the first one up each morning, he used to start a fire in the kitchen stove.

He remembered considerable feuding among the local mountaineers. The only person in the hills with medical training, his father treated all clans who sought aid and did not charge for his services; he considered them too poor to pay. As a result, both Charles and his father were treated kindly by all the local people.

There were also moonshiners as well as hunters and farmers among the population. He recalled that the killing of a federal revenue agent "seemed quite understandable and reasonable at the time!"

After three years of study he graduated from the University of the South in 1914 with B.A. and B.S. degrees. He received a prize for the best paper in economics. Although he had aspirations towards medicine, a lack of funds caused him to concentrate on chemistry, which promised earlier returns. He won a scholarship to the University of Pennsylvania for postgraduate work in chemistry. He also was awarded a scholarship at Johns Hopkins, his first choice, but it was canceled when the dean learned that he had also applied to Pennsylvania. Piggot stated that at Pennsylvania he was "amazed and shocked" by the lack of ethics and courtesies to which he was accustomed in the south. He studied there for two years and then, under a new scholarship, transferred to Johns Hopkins, where he was "once again among gentlemen." There he came into contact with Professor Remsen, who had guided his father in earlier years.

He spent the summer of 1916 at a civilian military training camp, where he received a commission as first lieutenant in the field artillery. In April 1917 his study at Johns Hopkins was interrupted by service in World War I when he served briefly with a field artillery battery, but was then transferred to a team at Johns Hopkins under J. C. W. Frazer, who was working on a means of protection from carbon monoxide gas. "We worked for over a year without a single positive or encouraging result. While working on a form of very finely divided manganese dioxide I had the luck to add a small amount of silver oxide and had the good fortune to obtain a catalyst which completely oxidized carbon monoxide to carbon dioxide whenever the incoming air mixture was thoroughly dry and there was sufficient oxygen to link up with the carbon monoxide," said Piggot in an autobiographical sketch. That catalyst has continued to be used by fire departments and in coal mine rescue devices.

After the war he returned to his graduate studies at Johns Hopkins where he received a Ph.D. degree in 1920, presenting a thesis titled "The Catalytic Oxidation of Ammonia by Manganese Dioxide." Upon graduation he started to work at the U.S. Industrial Chemical Company in a research laboratory devoted to preparing chemical products from ethyl alcohol and acetic acid as the principal starting substances. They developed processes for production of absolute alcohol in tank car lots; also anhydrous ethyl acetate, absolute ethyl ether, and ethylene gas. They found that 100% pure ethyl ether lost its anesthetic properties and that addition of a small amount of ethylene rendered it extraordinarily effective. He also developed a series of methylbenzenes from monobenzene to hexamethyl-benzene, which were useful as solvents. Thus, after completing a thesis in inorganic chemistry, he became active in the field of organic chemistry.

In 1922 the National Academy of Sciences received an invitation from the Ramsey Memorial Fellowship Trust of England to appoint a young chemist for advanced studies in chemistry as a Ramsey Memorial Fellow. Piggot became the first American to receive that appointment and went to University College, London, to study under the physical chemist, Frederick G. Donnan. Donnan introduced him to many stimulating British scientists. Among them was Sir William Bragg, who used some crystals of hexamethylbenzene that Piggot had prepared while working at the United States Industrial Chemical Company for his determination of the crystal structure of the benzene ring. This led to more work in Bragg's laboratory and with the help of Bragg's assistant, Dr. Mueller, Piggot constructed a small X-ray machine, which was used to determine the diameter of the CH_2 chain.

From England he traveled around Europe on vacations. While spending one of them at a winter resort in Switzerland he met a young woman from South Africa, Ruth Blaine, whom he married in 1927 after his return to the United States.

In 1925 he joined the staff of the Geophysical Laboratory of the Carnegie Institution of Washington to conduct research into the significance of radioactivity in geophysical phenomena. This work involved studies of the radium content of the various layers of the earth's crust with resultant heat production and efforts to improve the determination of geologic time by comparing the amounts of lead associated with its radioactive parent, uranium. Much of his early work at Carnegie concerned the geologic time aspect. At that time even such fundamental data as the isotopes of lead and uranium and their abundances were imperfectly known. To answer some of those questions he contacted F. A. Aston, who then operated the most advanced mass spectroscopy laboratory in the world, and arranged to supply lead samples for isotopic analysis. In 1927 he took a sample of pure lead tetramethyl to Cambridge, and, according to Aston, this resulted in the first successful determination of the mass spectrum of ordinary lead "after repeated failures" in earlier experiments.[1] The lead ions, which were collected on photographic plates, indicated isotopes at masses 206, 207, and 208. The isotope at mass 204 was not positively identified due to problems with interference from the isotope of mercury at mass 204. The relative abundances were reported as 206:207:208::4:3:7.

The next step was to study the isotopes of radiogenic lead. The first experiment sought to determine the lead isotopes due to the radioactivity of uranium. Since both thorium and uranium were know to decay to Pb, Piggot selected from Norway a Broggerite sample known to contain a high concentration of uranium, but very little thorium. The first vial of lead tetramethyl was sent to Aston in 1928, but broke in transit. A second one, sent a few months later, arrived safely. For this sample Aston reported 206:207:208::86.8:9.3:3.9.[2] There was still no data for the isotope of mass 204. The contrast in the ^{207}Pb/^{208}Pb ratios between the ordinary and radiogenic leads left no doubt that uranium possessed a second radioactive isotope postulated to be either mass 239 or 235. Through these activities Piggot's researches played a significant role in unraveling the mysteries of the radioactive decay systems and initiating the science of geochronology.

Upon arrival at the Geophysical Laboratory Piggot constructed an apparatus for determination of the radium content of igneous rocks by measurement of the activity of radon gas that was released by fusion of samples in carbonate fluxes. The resulting publications on the radium content of granites and their constituent minerals and Hawaiian basalts appeared between 1929 and 1932. He also tested for interstitial radium by leaching samples with hot water, finding that radon activity was typically lowered by 10 percent after the process, but that it grew back into equilibrium with its radium parent again.

The granite experiments paved the way for what was to be his major contribution from the radon work. Around 1933 he expanded his radium abundance survey to include some surficial ocean-bottom sediments. He was struck by the fact that the samples contained several times as much radium as the igneous rocks with which he was then working and wondered whether this high radium abundance was a surface effect or continued below the surface. To answer that question he had to develop a coring technique. The available bottom samples had been obtained with a telegraph snapper, which took a small bite out of the bottom surface, destroying the stratigraphy in the process. By 1936 he had published a description of a coring device capable of obtaining cores up to about three meters in length, with preservation of the stratigraphy. Its main feature was the use of a powder charge to drive a steel tube into the sediments. The charge could be adjusted according to the depth of water and type of sediment. After retrieval the tube was split into halves to reveal the stratigraphy.

Using the newly developed coring device on board a cable repair ship, he collected his first cores from a traverse across the North Atlantic Ocean from Ireland to Newfoundland and obtained information that aroused renewed interest in oceanography and marine sedimentology. The record of glacial epochs (discussed below) could be recognized in the cores, as well as strata characterized by volcanic ash. The lack of thick sediment layers at the Mid-Atlantic Ridge was noted. The cores became the subject of *U.S. Geological Survey Professional Paper 196-A,* published in 1940, in which M. N. Bramlette and W. H. Bradley described the geological and lithological interpretations and J. A. Cushman and L. G. Henbest discussed foraminiferal studies. In addition, colleagues at the Department of Terrestrial Magnetism used the cores to measure changes in orientation of the earth's magnetic field over the past several hundred thousand years.

Piggot realized that the sediment layers in the ocean bottom would reveal a valuable historical record of the ocean basins, which should provide useful information about processes that have occurred on land. This can best be described in his own words (1938):

> These sediments, lying layer upon layer in the bottom, have become the repository of the historical record of the oceans. This record includes the contributions of rivers, reflecting the changing conditions on the continents, as well as those of ice and wind and the myriad of life in the water

above. The record of what happened in this water above is filed away in the mud and clay and ooze below. The rocks and pebbles and sand brought by ice, the clay and mud brought by rivers and ocean currents, the skeletons of marine organisms which lived and died and evolved into various forms throughout the ages constitute this record. . . . In addition to these records of life and its many changes there exists a chemical and a physical record. Oxidation and reduction and the nature of the dissolved matter in the water have all left the record of their changes in the bottom, and the nature and size of the minerals and rock fragments bear evidence of the direction and strength of former ocean currents, the movements of ice and the depths of the ocean in the past.

Heretofore, the samples obtained from the deep ocean bottom have been a mere handful of material taken from the very surface of the bottom. These samples give information of present condition only and reveal nothing of past events, so that although the historical record has been known to exist we have been able to see only the top page.

On land the geologist studies the exposed rock strata, but a study of material lying beneath miles of water is enormously more difficult. If, however, we could bring up a vertical section of several feet of this bottom, in its original, undisturbed condition, we might read the history of oceanic events as the geologist deciphers the record in the rocks.

Those remarks sum up quite well the advances that research in marine geology has brought to our knowledge of geological processes upon the earth.

By that time Piggot had become intrigued by the observation that there was not sufficient radium in ocean water in proportion to the uranium content, and that there was too much radium in ocean sediments in comparison with their uranium content. This was the insight that led to the ocean sediment dating experiments in a series of papers that appeared between 1939 and 1942 dealing with the ocean-bottom studies. They were written in collaboration with W. D. Urry, who had constructed an improved apparatus for Rn determination. For their first detailed study they selected a 2.85-meter core from east of Nova Scotia off the

Newfoundland Banks that was a member of the series of cores Piggot had taken in the North Atlantic traverse described above. They chose that particular core because of its very uniform chemical and lithologic composition. The results must have been something of a disappointment because they found only a small variation in radium content with depth that did not permit determination of a sedimentation rate or dating of the core. They concluded that deposition was too rapid to cover enough of an interval to allow detectable radium decay at this site. Piggot speculated that the Labrador Current caused a high sedimentation rate in the area. More positive results were to follow in future studies.

The results of the papers Piggot and Urry published between 1939 and 1941 were summarized and evaluated in 1942 in a landmark paper in *Bulletin of the Geological Society of America*, which reported radium results on cores from the North Atlantic Ocean, the Caribbean Basin, and Pacific Ocean red clay. Depending upon core length and sedimentation rates the cores spanned periods of time reaching back 10,000 to 300,000 years.

A substantial portion of the paper dealt with correlating their Th disequilibrium dates with the geological and foraminiferal observations of Bramlette, Bradley, Cushman, and Henbest on the North Atlantic cores in order to date glacial epochs as known on the continents. They used the foraminifera data of Cushman and Henbest, which gave qualitative measures of water temperatures, or zones of glacial marine deposits from Bramlette and Bradley to define epochs of glaciation. The best results from the North Atlantic Ocean were obtained from a three-meter core collected about halfway between the Mid-Atlantic Ridge and Newfoundland, which contained a record of the past 73,000 years. Progressing down the core, foraminifera abundances indicated cooling of the water occurred 12,800 years ago, with glacial debris deposits occurring at about the same depth. (Piggot and Urry used a value of 82,000 years for the half-life of ^{230}Th, whereas we now use 75,400 years, which reduces the age to about 11,800 years.[3] This value correlates quite well with the ending of the Wisconsin (Wurm) glacial epoch as presently known. The core next recorded a rise in water temperature between about 61,000 and 70,000 years, which correlates approximately with the beginning of the Wisconsin glacial epoch. Two other cores recording only 12,000 and 24,000 years of history, respectively, yielded results generally consistent with the 72,000-year core over the periods of time they spanned. Piggot and Urry showed that the ocean core results compared reasonably well with data from the land masses as then known (e.g., advance and retreat of Alpine glaciers and studies of soils in the midwestern United States), fulfilling the expectations Piggot had expressed when he first started work on ocean sediments.

They further showed that dates from a core in the Cayman Trough of the Caribbean Sea yielded results in close agreement with the North Atlantic data. The core consisted of Globigerina ooze and covered a time span of 300,000 years. The record of the Wisconsin glacial epoch could be recognized in that core with dates that closely matched those found in the North Atlantic core. Their temperature scale, provided by Cushman, for some reason did not serve to distinguish between the Illinois (Riss) and Kansan (Mindel) glacial epochs, but indicated continuous glaciation before about 110,000 years ago.

Given the amount of data and state of knowledge available then, Piggot and Urry could not be very dogmatic about interpretation of those results, but their work outlined the principles and methodology for ocean sediment dating that opened up the field for future investigation. Although we now have much better information due to the ability to measure water temperatures from ^{18}O data and improved techniques for disequilibrium dating, their results still fit quite well with present knowledge of sedimentation rates in the ocean basins and the dating of glacial epochs from those data.

Finally the paper reviewed sedimentation rates for various environments. The rates in the North Atlantic Ocean varied by nearly a factor of ten, depending upon the location of the cores. Off the Newfoundland Bank the sedimentation rate varied between 10 and 60 cm per 1,000 years over the past 12,000 years; in the basin between Newfoundland and the Mid-Atlantic Ridge it varied between 1 and 6 cm per 1,000 years according to their results. The Pacific Ocean red clay yielded lower rates between 0.2 and 1 cm per 1,000 years. They also found that the surficial clay core surface contained ^{226}Ra in excess of the amount required for secular equilibrium with the ^{230}Th parent, whereas calcareous sediments and oozes from the Atlantic Ocean normally contained less ^{226}Ra than required for secular equilibrium.

According to their data sedimentation rates in the Caribbean Basin increased starting 7,000 years ago compared to rates during the Wisconsin glacial epoch. They postulated that the circa 3,000 year delay after the end of the epoch represented the time required to reestablish organic life after a long period of cold surface waters. The present rate was shown to be five to ten times that during Wisconsin glaciation. No such change was noted in their Pacific Ocean clay or in other Pacific and Antarctic red clay data later reported by Urry.[3] The data as a whole, although revealing variable sedimentation rates, generally indicated that deposition rates for red clays were about five to ten times lower than those for the oozes of the Caribbean Basin.

These results answered the questions Piggot had asked at the start of the experiments—showing that the high concentrations of Ra in ocean sediments were a surface effect and that ^{230}Th abundances in marine sediments were higher than those required for secular equilibrium with ^{238}U. With the aid of the coring device it was possible to use that relationship to date sediment horizons and thereby ascertain sedimentation rates. Furthermore, his belief that information of use in deciphering land-based geologic history could be read in the marine sedimentary record was confirmed.

On a personal note, even though I never met Charles Piggot, my admiration for his radium studies dates back to the days of my doctoral dissertation project around 1949. The goal of that project was to work out methods for accurate analysis of microgram quantities of uranium for application to meteorite studies and geochronology. In reviewing the various available methods, I read the radium papers of Piggot and Urry. Although I finally selected stable isotope dilution (a novel technique at that time!) as the best approach, the sediment disequilibrium dating studies described in their papers made a lasting impression on me.

World War II changed Piggot's activities from radium studies to service-related matters. Shortly before the Pearl Harbor attack the navy bureau of ordnance requested his service from the Carnegie Institution and he was granted a leave of absence. He was assigned to develop procedures for the recovery and disassembly of magnetic and other mines, which were then causing much trouble to Allied shipping. Eventually he worked with 400 trained men who volunteered for that hazardous duty. They worked on the recovery of submarine mines and disarming of bombs, booby traps, and other live ammunition. He established two training schools—one for mine disposal and one for research into methods and procedures for stripping dangerous objects. With help from Van de Graaf at MIT they developed a two-million-volt X-ray machine with which they could examine the interior of dangerous objects and decide how best to dissect them. He was awarded the Order of the British Empire for his work in mine disposal.

In March 1946 he was assigned to the staff of Task Force I for Operation Crossroads at Bikini and witnessed two atomic bomb explosions there. His assignment was to devise means for evaluating damage to ordnance equipment and to provide underwater photography using men and equipment from his mine disposal experience. He received a Bronze Star for that service. Upon returning to the United States he became executive director of the Committee for the Geophysical Sciences of the Research and Development Board of the U.S. Navy, where he oversaw ten panels in various fields.

In 1950 he went to London for two years as the first foreign service reserve officer assigned by the State Department to promote cooperation between the scientists and governments of the two countries. While there he served on the Ramsey Fellowship Board as well.

In 1952 he returned to the United States to work at Yale as a consultant and assistant supervisor on a navy project there. In 1955 he surveyed and appraised eighteen scientific institutions in India for the National Academy of Sciences.

Piggot was elected to the National Academy of Sciences in 1946. He was a fellow of the Geological Society of America and the American Geographical Society and a member of the American Chemical Society, the Washington Academy of Sciences, and the Royal Institution of Great Britain. As mentioned above, the Bronze Star and Order of the British Empire were among his awards.

The Piggots had two children, son Deboorne and daughter Anne Marguerite, now Mrs. Robert W. Black.

AN AUTOBIOGRAPHICAL SKETCH prepared for the National Academy of Sciences provided information on Piggot's family, childhood, education, and career activities. Records from the Geophysical Laboratory and an obituary in *The Washington Post*, dated July 9, 1973, yielded additional information covering some of his professional activities, publications, and awards. I am indebted to Gordon L. Davis, former staff member of the Geophysical Laboratory, for locating those sources of information.

NOTES

1. F. W. Aston. *Nature* 120:224 (1927).
2. F. W. Aston. *Nature* 123:313 (1929).
3. W. D. Urry. *J. Marine Research* 7:618-34 (1944).

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