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HARMON CRAIG 1926-2003

A Biographical Memoir by
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Harmon Craig

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BY KARL K. TUREKIAN

HARMON BUSHNELL CRAIG (he never used his middle name) was born in the borough of Manhattan in New York City on March 15th, 1926. He died on March 14th, 2003, a day short of his seventy-seventh birthday. Craig was the product of two major forces in his life. His father, John Craig Ir., was from a family long in the theater as actors, directors, and producers. Indeed, John Craig's major activities, after his heroic involvement in World War I, were in running theaters in the northeastern United States. Young Harmon was surrounded by a theatrical crowd during his early childhood. His mother came from a long line of activist Quakers, who, starting before the Civil War, established schools for freed slaves. This activity moved the family from its initial homestead in Virginia westward, finally to Kansas. The influence of his mother's ethos permeated young Harmon as his mother fed his inquisitive mind with books on a wide range of subjects, especially those heroic and exploratory in nature. It was the blending of the thespian and the Quaker ethos that shaped young Harmon in his early years and set the behavior pattern of his later life.

Harmon's youthful love of adventure, adventurers, and science blossomed into a career in the earth sciences when he discovered fossils in a rock on a family outing. He went off to the University of Chicago as a freshman with a clear idea of pursuing studies in geology. World War II interrupted his education. He went off to an officer's training program in the navy and eventually joined the fleet in Norfolk, Virginia. He returned to the University of Chicago after demobilization, and his future scientific life was shaped there.

After World War II the faculty of the University of Chicago, weary of their part in the development of the atomic bomb, turned to research in areas of the most esoteric sorts. With mass spectrometers in place, Harold C. Urey and his students, postdocs, and research collaborators delved into the arcane worlds of determining the warmth of an ocean 100 million years ago, determining the ages of rocks and the Solar System, and exploring the chemistry of the Universe. It was in this hotbed of national-defense-irrelevant research that Harmon Craig found himself. An undergraduate geology major at the University of Chicago, he was propelled into this world of geochemistry and cosmochemistry without waiting to get his undergraduate degree.

The measurement of ancient sea temperature depended on analyzing carbon dioxide released from calcium carbonate fossils and measuring the relative masses of carbon dioxide composed of ¹⁸O and ¹⁶O. The constancy of the carbon isotope loading on the carbon dioxide was tacitly assumed. Craig, for his thesis, measured the natural variability of ¹³C/¹²C to establish the baseline for all future studies involving the carbon system.

The independent discovery of natural radioactive ¹⁴C by W. F. Libby at the University of Chicago immediately led to the application of ¹⁴C to dating in archaeology and Pleistocene geology. The stable carbon isotope study from Craig's thesis allowed for corrections due to mass fractionation and permitted the proper determination of radiocarbon ages

(later to be corrected to calendar years by accommodating the variations in the initial $\rm ^{14}C/^{12}C$).

Craig's thesis is still today the primary citation for all studies involving variations in ¹³C/¹²C in natural materials, Cited in studies ranging from the establishment of food chains to identifying sources of ancient marbles for statues, this remarkable thesis was the harbinger of the impact Craig would have in various areas of geochemistry and cosmochemistry.

In the quest for the best measure, using meteorites, of the composition of the Solar System, the common assumption was that there was a uniform composition of the most likely nonvolatile raw material of the Solar System, chondrites. There was a need for criteria by which bad analyses of chondrites could be systematically identified and separated from the worthy ones; that is, meteorites modified by weathering had to be rejected so that the true makeup of meteorites, in particular the chondrites, could be ascertained. With his mentor Harold Urey, Craig discovered that once the veil of quality certification had been rent, the chondrites fell into at least two major groups. The Solar System was not so uniform after all. This discovery, later affirmed in several additional ways by others, gave us a totally new view of how and from what materials planets formed.

In 1955 the eastern universities were not yet ready to accept the strange new world of geochemistry heralded primarily by Harold Urey and his friends, but California was not afraid to go where no man had gone before. Caltech, through the wisdom of Robert Sharp and Harrison Brown, hired a bevy of University of Chicago geochemists. The Scripps Institution of Oceanography—mainly through the foresight of Roger Revelle, its director—brought in Craig from Chicago.

Back then, instruments were not built in a day. As Craig was tooling up, he solved the fundamental problem of the fate of carbon dioxide in the atmosphere and the oceans. His theoretical solutions are valid to this day. Indeed, they anticipated the program for atmospheric CO_2 measurements begun at Scripps by C. D. Keeling in 1957 at the instigation of Roger Revelle.

Craig decided that somebody had better figure out all the controls on oxygen and deuterium isotopes in the hydrologic cycle, especially if these isotopes were going to be used for paleoenvironmental reconstructions. In two elegant papers that resulted from his meticulous treatment of the problem for an appreciative Italian audience at Spoleto, he laid out the entire framework for discussing the role of kinetics and equilibrium in determining the isotopic composition of the hydrosphere, including the oceans. (These papers are not generally available in the common literature; neither are J. Willard Gibbs's classic thermodynamics papers, which were published in an obscure Connecticut journal.) These Spoleto papers are the fundamental documents that all atmospheric geochemists as well as hydrologists and oceanographers turn to for guidance in many aspects of light isotope geochemistry.

He established the meteoric water line, which defines the unique linear relationship between hydrogen and oxygen isotope ratios in natural terrestrial waters. He also discovered the oxygen isotope shift in geothermal and volcanic fluids, which showed (contrary to prevailing ideas) that the water in these fluids is overwhelmingly meteoric in origin. This work provided the basis for studies of water-rock interactions in geothermal systems and in hydrothermal vents.

Craig and his students subsequently studied the isotopic composition of atmospheric and dissolved oxygen and variations in the composition of dissolved gases. This work led to a method for determining biological oxygen production and consumption in the ocean mixed layer, as distinct from physical effects, and thus to a better quantification of biological primary production rates in the oceans.

In 1967 Henry Stommel suggested to a bunch of geochemists at a meeting at Woods Hole that it was about time that some scientists implemented a systematic study of the geochemistry and oceanography of all the oceans. With the new tracers and chronometers available to geochemists, this was the right moment to embark on this daunting enterprise. George Veronis let the group of geochemists get together with his theoreticians meeting at the Geophysical Fluid Dynamic Summer Institute to begin the planning. It became obvious to all who participated in the summer session that the leaders of what ultimately was to be called the Geochemical Ocean Sections Study (GEOSECS) should be Wallace Broecker of the Lamont-Doherty Earth Observatory, Harmon Craig of Scripps, and Derek Spencer of the Woods Hole Oceanographic Institution. With the help of many other geochemists the program did not self-destruct as some people thought (or hoped?), but rather accomplished its main goals.

GEOSECS spawned a number of important projects, many of which continue as follow-ups to this day. Craig was interested in the rate of turnover of the oceans. Fritz Koczy had suggested that ²²⁶Ra with a 1,620-year half-life might be a good tracer of circulation, being introduced at the ocean bottom from sediments and making its way up with the water to the surface, decaying along the way. Edward Goldberg of Scripps had suggested that the daughter of ²²⁶Ra, ²¹⁰Pb, could be measured as a surrogate. When Craig and his colleagues pursued this path, they discovered that ²¹⁰Pb was particle reactive and removed from the ocean by settling. Indeed, all the elements in the ocean that were particle

reactive like ²¹⁰Pb would have similar distributions and the removal from surface to depth and ultimately into the sediments occurred.

Another incorrect assumption was the expectation that ⁴He would be released from the ocean bottom. The expectation was to use atmospheric helium with its ³He dissolved in seawater in an isotope dilution experiment to measure the excess ⁴He putatively released from sediments. When Craig collected an ocean water profile and the talented Brian Clarke of McMaster University—who developed a technique for measuring ³He/⁴He—measured this profile, the astounding result was that it was ³He that was in excess not ⁴He. This discovery of primordial ³He in the oceans was made at the same time that I. N. Tolstikhin discovered primordial ³He in hot springs in the Kuriles. The consequences of the oceanic discovery impacted not only the tracing of ocean circulation but also the understanding of the way the mantle expresses itself at ocean spreading centers and ocean island basalts. The discovery of excess ³He in the oceans from this productive collaboration was exploited in every way by Craig, his students, and his postdocs with many additional remarkable discoveries resulting.

Craig's interests were not restricted to the oceans and the rocks at their boundaries; he also sought to understand the record of atmospheric changes recorded in cores from the Antarctic and Greenland ice sheets. He was one of the earliest workers to study gases trapped in glacier ice, and he showed that atmospheric methane has roughly doubled due to human activities over the past 300 years. He was also one of the first to study the geochemistry of atmospheric nitrous oxide and to work on the production, rate of increase, and isotopic budget of this natural and anthropogenic modulator of Earth's protective ozone layer. More

recently his work focused on the physics and chemistry of gases in polar ice cores, including pioneering work on the gravitational separation of gases and isotopes within the permeable firn layer, and on the gravitational separation of rare gas isotopes as a measure of firn temperatures and thicknesses. This work is fundamental to the reconstruction of past atmospheric composition and isotopic variations based on measurements of gases in polar ice, and plays an important role in continuing efforts to understand past climatic change.

In one of his last papers Craig made sense of the ³²Si measurements made in the Geochemical Ocean Sections Study. Some scientists saw in the original measurements a hopelessly flawed set of data when tested with a simple model. Craig and his coauthors—including Somayajulu, who initially made the measurements and was rightly indignant that the quality of his measurements was challenged—wrote a paper titled "Paradox Lost: ³²Si and the Global Ocean Silica Cycle," wherein the role of mixing of two sources of silica trapped by the collecting fibers explained the results and justified the measurements made by Somayajulu. So we see the man who's eye for recognizing quality measurements first showed up in the paper on meteorites was active in deciphering a major marine geochemical problem.

Craig influenced many areas as a result of his brilliance as a field observer, his skill and meticulousness as a measurer, and his genius as a profound theoretical thinker. These qualities, when found in one person, make that person able to improve our understanding of Earth in all its facets with the strength of a whole army. Yet this one-man army was not acting alone. In everything he did he was accompanied and encouraged by his wife, Valerie. Her patience with Craig's perennially searching mind, his friends

with diverse qualities and interests, and the system in which she and her husband ultimately triumphed made the Craig enterprise one of inevitable success.

Craig's success was recognized and rewarded by a number of prestigious awards, including the Balsan Prize, the Vetlesen Prize, the V. M. Goldschmidt Medal of the Geochemical Society, the Arthur L. Day Medal of the Geological Society of America, and the Arthur L. Day Prize and Lectureship of the National Academy of Sciences. He was elected to the American Academy of Arts and Sciences in 1976, and elected to the National Academy of Sciences in 1979. The University of Paris awarded him an honorary degree (an interesting follow-on to his father having received the Croix de Guerre from the French for his bravery in World War I). His alma mater, the University of Chicago, also awarded him an honorary doctorate while denying him an ex post facto bachelor's degree.

I thank John Craig III and Valerie Craig for insights into Harmon Craig's career throughout his productive life. I have borrowed extensively from an obituary that I wrote for *Nature* and one that Ray Weiss wrote for the *Transactions of the American Geophysical Union (EOS)*.

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