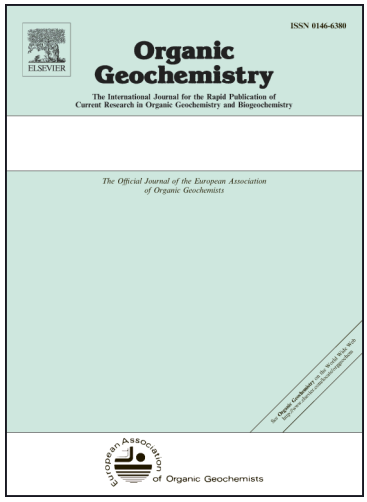

John M. Hayes 1940-2017. Father of isotopes in modern and ancient biogeochemical processes, biosynthetic carbon and hydrogen isotope fractionation and compound specific isotope analytical techniques

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In Memoriam

John M. Hayes 1940-2017. Father of isotopes in modern and ancient biogeochemical processes, biosynthetic carbon and hydrogen isotope fractionation and compound specific isotope analytical techniques.

John Michael Hayes, Professor of chemistry and geology for 26 years at Indiana University (Bloomington) until 1996, then director of the National Ocean Sciences Accelerator Mass Spectrometry facility at Woods Hole Oceanographic Institution and adjunct professor at Harvard University until 2007, died peacefully at his home in Berkeley, California, on February 3rd, 2017.

John was born in Seattle, Washington and grew up in Montana and Iowa. He attended thirteen schools as his family moved along with his father's job for the Chicago, Milwaukee, St. Paul and Pacific Railroad. He graduated from High School in Perry, Iowa and enrolled at Iowa State University, where he graduated with a B.Sc. in Chemistry in 1962. The same year he married Janice Maria (Boeke) Hayes of Hubbard, Iowa, whom he met at Iowa State University. He was awarded a Ph.D. degree from Massachusetts Institute of Technology in 1966 for thesis work (Hayes, 1966) performed on the organic constituents of meteorites (Hayes, 1967; Hayes et al. 1968) under the supervision of Klaus Biemann.

After spending 3 months in 1966 as a Postdoctoral Research Associate at the Enrico Fermi Institute, University of Chicago, John undertook three months of military training to then spend another 21 months of military duty as Captain in the Medical Service Corps of the U.S. Army, specifically in the Chemical Evolution Branch at NASA Ames Research Center in Moffett Field, California. He subsequently went abroad as a NATO-NSF Postdoctoral Fellow to the Organic Geochemistry Unit at The University of Bristol, UK, where he worked with Geoff Eglinton and others on the carbon chemistry of lunar samples brought back to Earth by Apollo 11 and 15 missions (Hayes et al., 1970; Abell et al., 1970).

In 1970 John was one of the attendees of the first Gordon Research Conference on Organic Geochemistry in New Hampshire, which he regularly attended throughout his career. In 2010, celebrating the 40th anniversary of this conference, he and six other first attendees shared their views and scientific career experiences with the audience and specifically the Gordon Research Seminar young graduate attendees in a truly inspirational session.

Also in 1970 he was appointed Assistant Professor at the Department of Geological Sciences, Indiana University, Bloomington, becoming Associate Professor in 1974 and Full Professor in 1997. As of 1984, he held Professorships in the Department of Geological Sciences and in the Department of Chemistry. In 1990 he became Distinguished Professor of Biogeochemistry. While at Indiana University he also was Visiting Research Geochemist at the University of California, Los Angeles in 1979 and 1980 and Visiting Scientist at the Bureau of Mineral Resources, Geology and Geophysics, Canberra, Australia in 1988.

During the 1970s at IU, John continued studying the fate of carbon in lunar samples (DesMarais et al., 1973), advanced studies of carbon isotope fractionation in biogeochemical processes, including bacterially produced acetic acid (Meinschein et al., 1974; Rinaldi et al., 1974 a,b), carbon isotope analysis of aquatic samples, and high resolution mass spectrometric techniques (DesMarais et al., 1976; Hayes and Schoeller, 1977; Hayes et al., 1978; Mathews et al., 1978) and working with NASA (Novotny et al., 1975). During this period of remarkable productivity, his first co-authored book (Peters et al.,1974) was soon followed by a second (Peters et al., 1976). During this time, he also made the first measurements of the distribution of the isotopes of carbon within biolipids (Meinschein et al., (1974a), thereby providing a foundation for new studies on the pathways of carbon in natural environments, both modern and ancient.

In the late 1970s and early 1980s, Hayes continued to push forward the use of stable isotopes as a tracer of biochemistry, working on the carbon isotopic composition of synthesized carboxylic acids, biosynthesized fatty acid fractions and decarboxylation pathways and their isotopic fractionation (Games et al., 1978; Vogler et al., 1979). With David Monson, he produced the first evidence of isotopic ordering in fatty acids, a pattern now widely recognized as characteristic of terrestrial life, and simultaneously proved that the distribution of carbon isotopes in organic matter was not an equilibrium phenomenon (Monson et al., 1980, 1982). Continuing his longstanding interests in early Earth biogeochemistry, he was also an active member of the Paleoproterozoic Research Group (PPRG) organized by Bill Schopf at UCLA. This group produced groundbreaking research into the *Earth's Earliest Biosphere* (Schopf, 1983) and laid the groundwork for future isotopic studies of ancient life. During this same time, he was also beginning to explore what would become one of the seminal contributions of his career: the development of methods for measuring the isotopic compositions of individual organic molecules, a technique he initially called isotope-ratio-monitoring GC/MS (Mathews and Hayes et al., 1978) and which is now more widely known as compound-specific isotope analysis (CSIA). As part of this work he forged a strong collaboration with Finnigan MAT (now part of ThermoFisher Scientific) to develop the first generations of mass spectrometers for the new compound-specific analyses (Hayes at al., 1978). That collaboration on isotope instrumentation was to continue for the rest of his career.

Throughout the 1980s, Hayes – together with his skilled technician Steve Studley, programmer Margaret Ricci (on loan from Finnigan) and numerous graduate students and postdocs, particularly Dawn Merritt and Kate Freeman – continued to refine and perfect methods for compound-specific carbon isotope analysis. These analytical developments would not be recognized in print for almost a decade (Hayes et al., 1989; Merrit and Hayes, 1994; Merrit et al., 1994; Ricci et al., 1994), but in the meantime their application led to a series of groundbreaking papers (Hayes, 1984; Knoll et al., 1986; Hayes et al., 1987; Freeman et al., 1990; Jasper and Hayes, 1990; Hayes, 1991; DesMarais et al., 1992; Kohnen et al., 1992; Freeman and Hayes, 1992; Zaback et al., 1993) on subjects ranging from the biogeochemistry and oxygenation of the Archaean Earth, to isotopic records of P_{CO2} in the Cenozoic, to isotopic characteristics of phytoplankton lipids and pigments, and others. During this period John was also an active member of the Paleoproterozoic Research Group organized by Bill Schopf at UCLA, and which produced groundbreaking research into the Earth's Earliest Biosphere (Schopf,

By the early 1990s John's compound-specific approach had expanded to include nitrogen isotopes (Merritt and Hayes, 1994) and, by the late 1990s hydrogen isotopes (Burgoyne et al, 1998; Sessions et al., 1999). His research interests in Precambrian biogeochemistry continued, including the role of active methane cycling in early climates (Hayes and Bengston, 1993), and previously unnoticed shifts in the isotopic

ordering of *n*-alkyl and isoprenoid carbon skeletons during the Precambrian (Logan et al., 1995). It also expanded to include the origins and history of hydrocarbon biomarkers, with relevance both to the history of life and to the energy industry (Sinninghe Damsté et al., 1993; Jasper and Hayes, 1993; Hayes, 1993; Hartgers et al., 1994; Hayes 1996). With Kai Hinrichs, he helped to discover the long-south sought? organisms responsible for anaerobic oxidation of methane (Hinrichs et al., 1999).

In 1996 he moved to the Woods Hole Oceanographic Institution (WHOI) for the purpose of – as he put it in his typically modest way– "advancing and disseminating some experience to the next generation." There, he undertook a new position as Director of the National Ocean Sciences Accelerator Mass Spectrometry facility, a national facility dedicated to radiocarbon dating for all of science, but for oceanographic research in particular. He also maintained a separate lab where research continued on hydrogen isotopes in lipids (Sessions et al., 2002; 2004) and plant leaf wax (Sauer et al., 2001), and on anaerobic methanotrophy (Hinrichs et al., 2000; Orphan et al., 2001). With his new colleague Tim Eglinton and graduate student Ann Pearson, they began to employ compound-specific radiocarbon as an isotopic tool for distinguishing the origins of organic molecules (Pearson et al., 2002; 2004). Using similar techniques, John, Tim, and postdocs Nao Okhouchi and Gesine Mollenhauer first showed us that organic biomarkers like alkenones can be thousands of years older than the sediments in which they are buried (Okhouchi et al., 2002; Mollenhauer et al, 2005), a finding whose implications continue to ripple today.

At NOSAMs, Hayes continued his longstanding interest in analytical development. Working with physicists Bob Schneider, Karl von Raden and Mark Robers, they built the first routine continuous-flow ion source for accelerator mass spectrometry (AMS; Schneider et al, 2004; Roberts et al, 2007). They also constructed the first continuous-flow sample introduction system for AMS using pyrolysis of organic matter. Now known as 'ramped pyrolysis' (Rosenheim et al., 2008), it was affectionately known as the 'dirt burner' in the lab, a term coined by John's longstanding technician Sean Sylva. Recognizing the shift in analytical organic chemistry away from GC and hydrocarbons and towards HPLC and biolipids, John also worked again with Finnigan to help develop the prototype moving-wire combustion system for measuring ^{13}C in nonvolatile molecules, including DNA (Pearson et al., 2004; Sessions et al., 2004).

During his time at WHOI, John was appointed as an adjunct faculty at Harvard, where he taught his biogeochemistry class and continued to collaborate with colleagues at Harvard and MIT. Notable among the output from this time was the hypothesis that the huge Neoproterozoic ^{13}C excursions that had puzzled biogeochemists for so long were in fact the signature of a disappearing dissolved organic carbon pool (Rothman et al, 2003).

After his retirement in 2006, John and Janice moved to Berkeley, California, which was conveniently near the geographic center for their children and grandchildren. In total, John published more than 200 research papers, two textbooks and four book chapters. He is widely credited with inventing compound-specific isotope analysis, which utterly transformed the use of stable isotopes as tracers in organic molecules. He performed field work around the globe, including on the R/V Atlantis and in the submersible Alvin, and in Western Australia, South Africa, and the Canadian Arctic. He was a member of the American Geophysical Union, the American Society for Mass Spectrometry, and the European Association of Organic Geochemists. He was elected to the US National Academy of Sciences in 1998 and The Royal Society in 2016. He was a recipient of the Treibs, Goldschmidt, and Urey medals, collectively the three highest honors in his field. He received an exceptional number of honors and awards, a fact that both honored and humbled him, served as editor, associate editor and editorial board member and served the geochemical discipline in multiple honorary roles:

2016	The Royal Society, elected Foreign Member
2003	Geochemistry Division Medal, American Chemical Society
2002	Goldschmidt Medal, The Geochemical Society
2001	Fellow, American Geophysical Union
1998	National Academy of Sciences, elected Member
1998	American Academy of Arts and Science, elected Fellow
1998	Treibs Medal, The Geochemical Society
1997	Urey Medal (with G. Eglinton), European Association for Geochemistry
1996	Geochemical Fellow, Geochemical Society and European Association for Geochemistry
1996-1998	Associated Director, Canadian Institute for Advanced Research
1995	H. Burr Steinbach Scholar, WHOI
1995	Krumbein Lecturer, University of Chicago, Northwestern University, Field Museum
1994	Ingerson Lecturer, The Geochemical Society
1987-1989	Chair, Organic Geochem. Division, Geochemical Society
1987-1988	Fellow, John Simon Guggenheim Memorial Foundation
1986-1988	Chairman, Gordon Conference on Organic Geochemistry
1986	Fellow, American Association for the Advancement of Science
1984-1986	Chairman, Publications Committee, The Geochemical Society,
1983-1989	Member, Publications Committee, The Geochemical Society
1981-1983	Chairman, Gordon Conference on Chemistry and Physics of Isotopes
1977-1998	Member, Editorial Board, Precambrian Research
1975-1983	Member, Editorial Board, Biomedical Mass Spectrometry
1971-1975	Associate Editor, Geochimica et Cosmochimica Acta
1964-1966	NSF Cooperative Graduate Fellow, MIT.
1962	Eastman Prize for outstanding graduate student in chemistry, MIT.

In his personal life John was a determined and skilled photographer (a hobby that began with his use of photographic plates as MS detectors during his PhD thesis), an accomplished flutist and lover of classical music. He never minded lending his car to his postgraduate students, as long as they were fine with him having a selection of classical CDs in the player and in the glovebox compartment. Remembered will be in-depth discussions we had about Brahms Requiem performed by Kurt Masur and the New York Philharmonics vs. the recording of Karajan with the Berlin Philharmonics. Unforgettable remain the evenings at his houses in Bloomington and Woods Hole, enjoying spectacular home-cooked meals, fine wine, and listening to classical music.

After retirement, John continued to regularly attend conferences in the geochemical domain and the Royal Society elected him Foreign Member. He was the John we all got to know and appreciate so much, the same modest, sometimes withholding and gentle person. Those of us, who had the privilege to work with him and enjoy his leadership,

will always remember him and he has become part of our own thought processes and nature. We all sadly heard in 2013 about his wife, Janice, passing away, and who we also always remember as John's inspiration - a gentle, welcoming and truly warm partner of his. When John joined her, dying of idiopathic pulmonary fibrosis, or as his daughter Anne Hayes Hartman called it, "rejoined the carbon cycle", his children, James T. Hayes of Honolulu, Hawaii, Anne Hayes Hartman of Oakland, California, and Rachel M. Hayes of Nashville, Tennessee, and his grandchildren were with him.

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