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SAMUEL EPSTEIN
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A Biographical Memoir by

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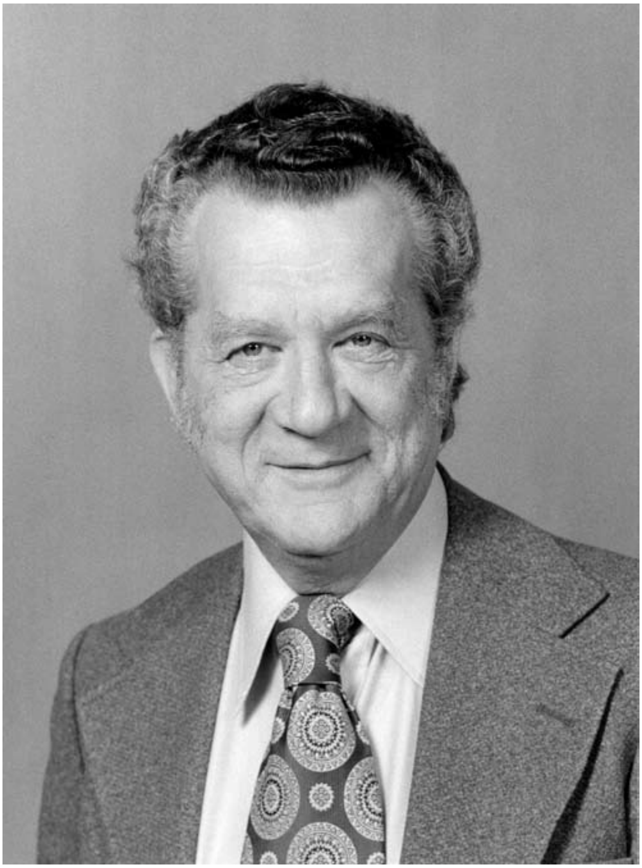

SAMUEL EPSTEIN

December 9, 1919–September 17, 2001

BY HUGH P. TAYLOR JR. AND ROBERT N. CLAYTON

SAMUEL EPSTEIN WAS ONE of the principal geochemists responsible for pioneering discoveries regarding variations of the stable isotopes of oxygen, hydrogen, carbon, nitrogen, silicon, and calcium on Earth, the Moon, and in meteorites. He was fortunate to have been at the forefront of great advances in the physics and chemistry of isotopes that were an outgrowth of atomic energy investigations in the wake of World War II. Although several scientists in the 1950s and 1960s recognized the power of stable isotope measurements to solve scientific problems, it was Sam more than anyone else who had the energy and insight to carry this out in such fundamental ways and in so many diverse fields, including: paleothermometry of carbonate fossils; geothermometry of minerals and rocks; origins of natural waters, including fluid inclusions in minerals; paleoclimatology records in glaciers, continental ice sheets, and tree rings; biological processes including living plants and animals, fossil plants and animals, and paleodiets; petroleum and natural gas; hydrothermal ore deposits; water and rock interactions; oceanography; meteorology; gases in Earth's atmosphere; weathering and soil formation; studies of meteorites, lunar rocks, and tektites; and studies of igneous, metamorphic, and sedimentary rocks and their constituent minerals. Born in eastern Europe, he grew up and was educated in Canada, but lived most of his professional life in the United States. For most of his career he was a professor of geochemistry at the California Institute of Technology in Pasadena, California.

Sam was born in a village near Kobryn, Poland (now Belarus), on December 9, 1919. His parents, Zelig and Rifke, were also born in this region (the "Pale of Settlement," where Jews were allowed to live in Czarist Russia). Around the turn of the 20th century Zelig Epstein traveled to New York City, where he lived for about four years. In 1904 he returned to Russia (Poland was a part of Russia at the time) for required military service, and in 1907 he and Rifke were married. They had planned to emigrate to the United States, but when World War I began this became problematical. They attempted to travel eastward through Siberia but were apprehended and turned back in Harbin, near the Russian-Chinese border. After many difficulties associated with living in the midst of battlegrounds, during which time Sam's two sisters Dora and Bella were born, the family settled on the tiny farm where Sam was born. Soon thereafter, Zelig moved into Kobryn and started a small business, buying and reselling produce from the local farmers. Some years he did fairly well, but other years were quite bad, so he resolved to leave Poland. Sam began school at a one-room *cheder*, where he mainly learned Hebrew and Polish and studied arithmetic and the Bible, but spoke Yiddish at home and White Russian to many of his neighbors. At this time Canada was one of the few countries accepting immigrants from Poland on a nonquota basis, so Zelig Epstein decided to move there instead of undergoing the long wait required by the United States. As related by Sam himself (1997):

> My father left Kobryn in 1926, and in 1927 my mother, my two older sisters, and I joined him. My gentle and very caring mother was already very ill at that time as a consequence of the horrors she and my father and sisters

suffered during World War I. She passed away in 1928. This tragic loss was somehow made easier by the love and care that my older sisters and my father gave me during the next important and formative years of my life. It goes without saying that the decision my parents made to immigrate to Canada saved us from certain destruction during World War II, a fate that overcame the rest of our family who remained in Kobryn.

In Winnipeg, Manitoba, Sam's father found work in a factory, and soon remarried. Sam grew up during the Great Depression, and given the family circumstances, he knew that if he wanted to pursue his studies beyond high school he would have to earn some money. He had a paper route for the Winnipeg *Free Press* and found other temporary jobs, ranging from working on the boardwalk at the beach on Lake Winnipeg, to spraying pesticides against the mosquitoes that annually plagued Winnipeg. Going through the public schools, Sam developed a strong interest in mathematics and science. He entered the University of Manitoba in 1937, selecting mathematics, physics, chemistry, and geology as optional subjects for the B.Sc., and chemistry and geology for his honors degree. In his senior year there were only three students in the geology honors program, and Sam was unique because he qualified for the B.Sc. with very little field geology experience. Geology majors typically worked for the Manitoba Geological Survey during the summer, but this opportunity was apparently denied to Sam because of his Jewish background. As he recalled (1997):

During my final year before graduation, one of the members of the Manitoba Survey gave me the unsolicited advice that I did not have much of a future in geology. This was a devastating blow, and I was very discouraged. However, it turned out to be a blessing in disguise! That summer I visited one of my chemistry professors, Alan N. Campbell, to discuss an examination I had written for his course. I had apparently done well, because our conversation turned to the possibility of whether I might like to do research with him for a Master's Degree, the only advanced degree in chemistry available in Manitoba at that time.

Sam's M.Sc. thesis was to measure the density of molten selenium at various temperatures; he encountered many problems with the quality and strength of the laboratory pycnometer, but after much hard work he obtained his degree in 1942. As recounted by Epstein (1997):

"Our year of research on this project involved a relatively simple objective, but solving the problem and carrying it through to completion was a thoroughly new and exciting experience . . . my first bona fide publication."

Campbell encouraged Sam to apply to McGill University in Montreal to work toward a Ph.D. in chemistry. Although Canada had entered the war in 1939, it was government policy to have students like Sam continue their university studies, because they might later be important to the war effort. Sam was indeed motivated to do anything he could to defeat Nazi Germany. He sent in his application and received an acknowledgment from McGill that he had the proper qualifications for graduate work. Assuming, therefore, that he had been formally accepted as a graduate student, in September 1942 he went to the Canadian Pacific Railroad station to inquire if there were some way he could earn his way to Montreal. He got a job selling fruit and sandwiches on the train that was to leave that night, so he hurriedly went home and packed, ready to travel away from Winnipeg for the first time since 1927.

Two days later, arriving in Montreal, Sam contacted a friend from Winnipeg, Leo Yaffe, a graduate student in the Chemistry Department at McGill. Yaffe took him to Professor Carl Winkler, but Sam was shocked to find out that Winkler was not expecting him, as there had been a mix-up in his application. Epstein (1997) states:

Dr. Winkler obtained my file from the Dean's Office and found that I had written a perfect paper on an advanced physical chemistry exam. . . In addition, Raymond Boyer, a professor of organic chemistry at McGill, seemed very positive about accepting me for graduate school. Dr. Winkler offered to get me a job in Ottawa, but I expressed my preference for staying as a graduate student . . . within a few weeks I was informed that I could help Dr. Boyer in his organic chemistry laboratory course and earn the regular stipend for graduate students . . . sufficient for me to support myself throughout graduate school. Clearly, Dr. Boyer came through for me, in spite of the fact that my past experience would not have qualified me to do a thesis in his field.

At McGill, Sam had his first experience working closely with a group of other scientists, the 16 Ph.D. students doing research under Winkler. Like Sam they were mainly from provinces other than Quebec, because at that time McGill was one of only two Canadian universities (the other being Toronto) that offered Ph.D. degrees. Winkler and Boyer were doing research on the properties and mechanism of formation of RDX, an important explosive used during World War II, trying to produce a maximum amount of RDX and a minimum of HMX, a less stable by-product. In their crowded laboratories the students worked long hours, including weekends, helping each other and many becoming close friends. At the laboratory bench next to Sam was the future Nobel laureate Rudy Marcus, who later became a colleague at Caltech. Epstein (1997) recalls:

I don't quite remember how I got involved in this problem of analyzing the relative proportions of RDX and HMX, but I do recall an experience I had in Dr. Campbell's class involving the kinetics of homogeneous reactions, which gave excellent results in determining the order of the reaction. I decided to try a base hydrolysis on the intimate mixture of RDX and HMX. . . It worked very well. The RDX was preferentially hydrolyzed.

This research turned out to be Sam's Ph.D. thesis; it contributed to the war effort as well as produced fundamental information about an important reaction. Although the work was classified as secret during the war, it was finally published in 1952 in the *Canadian Journal of Chemistry*. Raymond

Boyer, who had been indispensable in helping Sam remain at McGill, was later convicted of transmitting RDX secrets to the Soviet Union and was sentenced to two years in prison. He was caught up in the famous spy scandal triggered by the 1945 defection of Igor Gouzenko, a cipher clerk at the Soviet Embassy in Ottawa.

After receiving his Ph.D. in 1944, Sam joined the European-Canadian project on nuclear fission at the University of Montreal, under Professor John Cockcroft. Sam was the third member of a team trying to extract and measure the concentrations of the tiny amounts of argon, krypton, and xenon present in uranium metal irradiated with slow neutrons. Much of the needed equipment was not commercially available in Canada; for example, they had to regrind their own glass stopcocks and build their own mercury valves. Therefore, in the first few months Sam spent much of his time at the glass-blowing table, learning a skill that served him very well throughout his later career, as can be attested to by all his subsequent students. In time they succeeded in isolating pure samples of these gases, taking advantage of their different degrees of adsorption on activated charcoal. Although Sam worked diligently on this project, he found that the work was more relaxing than during his intensive two years at McGill. Thus, with a less demanding workload and with his evenings now sometimes free for social activities, Sam started to get out and around in the city of Montreal, which had a large Jewish community. One of the girls he met arranged a blind date for him with her sister, Diane Vool. It turned out that Diane and Sam's friend Rudy Marcus had attended the same high school, Diane being one year behind Rudy. It didn't take long for Sam and Diane to fall in love, and they were married on September 22, 1946.

After the war, when the University of Montreal Laboratory was about to be shut down and its operation moved to Chalk River, Ontario, Sam became acquainted with Professor Harry Thode of McMaster University, who at that time had the only mass spectrometer in Canada. Thode suggested they analyze the mixture of noble gases that Sam's group had extracted, in order to accurately determine the abundances of all the isotopes of krypton and xenon produced by fission under varying conditions of neutron radiation. Thus in 1946, although still an employee of the Canadian Atomic Energy Project, Sam moved with Diane to Hamilton, Ontario, where he began construction of the high-vacuum extraction apparatus needed for their joint project. However, this work depended on the cooperation of the Atomic Energy Commission in the United States, their only source of irradiated uranium. Because of the ongoing Canadian-Soviet spy scandal mentioned above, the U.S. government was reluctant to ship such material to Canada.

With his research at a standstill Sam and Diane strongly considered relocating to the Chalk River facility, where they could live in a comfortable home among a community of people with common interests. At this crucial moment Harry Thode received a telephone call from Professor Harold Urey at the University of Chicago inquiring whether he knew of a young Ph.D. with laboratory skills who would be willing to come to Chicago to work with him on a problem dealing with paleoclimates. Thode immediately thought of Sam, and soon thereafter Sam received an offer of a postdoctoral fellowship at a reasonable salary. Sam decided he should not pass up this opportunity to work with a Nobel laureate in chemistry.

In his classic 1947 paper "The Thermodynamic Properties of Isotopic Substances" Harold Urey had calculated the temperature coefficient of the oxygen isotope exchange reaction between carbonate ion and H_2O. This led to the idea that the oxygen isotope compositions of marine carbonate fossils might provide information on the temperatures of ancient oceans if the recognized and rather formidable analytical problems could be resolved. Urey was fascinated with the mass extinction of the dinosaurs 65 million years ago, and if an abrupt temperature change was the cause, an isotope geothermometer might decipher this. Sam could appreciate the significance of the problem because of his undergraduate experience in geology.

After obtaining a visa that allowed him to work in the United States for one year, Sam moved to Chicago with Diane in October 1947. They rented a tiny, relatively uninviting apartment above Professor Urey's garage. Sam was thus able to have a daily, remarkably close scientific interaction with Urey as they were driven back and forth to the university by Mrs. Frieda Urey. Their first child, Reuben, was born in February 1948, and for the young newlyweds, this continuing proximity to the forceful and dynamic Harold Urey made life interesting, to say the least. Sam frequently recounted his experiences in the often unheated garage apartment, such as when Harold Urey would stand at the back door of his house and summon Sam to the door of his apartment on those occasions when Urey had some thought that he wanted to try out on Sam. Often late at night, even in the subzero temperatures of Chicago winters, the two men would shout scientific ideas across the width of a patio and driveway.

Sam plunged into the carbonate paleotemperature project with immense drive and dedication. Within a year, with Sam at the center of the effort, tremendous strides were made by Urey's talented research team. They had to proceed simultaneously along several avenues of research. First and most important was to construct and then modify the superb gas-source mass spectrometer recently developed at the University of Minnesota by Alfred O. C. Nier. They needed to measure the $^{18}O/^{16}O$ ratio of a sample of carbon dioxide gas to at least 0.2-0.3 per cent in order to have any chance of measuring paleotemperatures to within a few degrees Celsius. This was accomplished with great ingenuity by measuring only the deviations in $^{18}O/^{16}O$ relative to that of a standard CO_2 gas, rather than trying to measure absolute isotope ratios. They employed a dual-inlet system that allowed for rapid switching back and forth between sample gas and standard gas, so that conditions in the mass spectrometer ion source were as uniform as possible during both measurements.

Second, they had to develop routine laboratory procedures for transforming the oxygen in a $CaCO_3$ shell into a CO_2 gas sample that accurately reflected the $^{18}O/^{16}O$ of the original shell. After many trials and errors, they found this could be done by reacting it with 100 percent phosphoric acid in a constant-temperature bath. The first account of their findings was presented at the Geological Society of America meeting in the autumn of 1948 by H. C. Urey, S. Epstein, C. R. McKinney, and J. M. McCrea. Soon they were able to improve their analytical precision to better than 0.02 per cent (i.e., 0.2 per mil), far exceeding the precision Urey had calculated to be necessary for a viable oxygen isotope paleotemperature scale. With very little modification these pioneering techniques are still being used in the thousands of stable isotope laboratories that have sprung up all over the world during the past 60 years.

At this critical time Sam's visa expired, and the family was forced to return to Canada, remaining for several months with Diane's parents in Montreal. However, Sam had become so essential to the project that Urey personally visited the United States Immigration Bureau on his behalf, finally persuading them to allow Sam, Diane, and Reuben to return to the United States in early 1949. By this time

John McCrea and Charles McKinney had left the project and Urey had moved on to several other research activities. The day-to-day running of the oxygen isotope laboratory was largely turned over to Sam and to Toshiko Mayeda, his newly hired technician.

Among the many remaining problems they had to: (1) demonstrate which organisms laid down their carbonate in equilibrium with sea water; (2) calibrate the $^{18}O/^{16}O$ of the growing shell as a function of temperature in the laboratory; and (3) prove that the original $^{18}O/^{16}O$ of the shell had been preserved during the millions of years since the organism had died. With the help of the biologist Ralph Buchsbaum and Sam's lifelong friend and future Caltech colleague Heinz Lowenstam, then professor of paleontology at the University of Chicago, they were able to resolve most of the remaining difficult problems connected with the chemistry of extracting CO_2 from the biogenic skeletal carbonate shells, as well as refining and testing the carbonate paleotemperature scale.

Ultimately, in the absence of any complications, they were able to measure carbonate paleotemperatures to better than 0.8°C, an astonishing achievement when compared with the state-of-the-art only five years earlier. Urey, Lowenstam, Epstein, and McKinney (1951), for example, analyzed a 100-million-year-old fossil shell and showed that this belemnite lived for four winters and three summers at temperatures ranging from 15 to 21°C, and that the winters grew progressively colder during its lifetime. One of these belemnites from the Cretaceous Pee Dee Formation in South Carolina was particularly well preserved and was chosen by Sam and Heinz to be the comparison standard for all their isotopic analyses. Remarkably, this Chicago PDB standard is still the one being used for carbon isotope analyses in all laboratories throughout the world.

The years 1950 and 1951 were very productive for Sam, as most of the experimental difficulties were solved and he had success in applying these new techniques to geological problems, including his pioneering work with Tosh Mayeda on $^{18}O/^{16}O$ variations of natural waters. Utilizing an elegant method involving equilibration of a small amount of CO_2 gas with each water sample as it was held in a constant-temperature bath for a few days, they examined the spatial isotopic homogeneity of ocean waters, finding a nice correlation between $^{18}O/^{16}O$ and salinity. They also demonstrated that fresh waters were consistently lower in $^{18}O/^{16}O$ than sea water, attributing their $^{18}O/^{16}O$ variations to the known differences in vapor pressure of the different isotopic species of water, which in turn could be related to local atmospheric temperatures.

The isotopic relationships they observed form the basis for almost all present-day $^{18}O/^{16}O$ and D/H paleoclimate studies. Sam also gained a lot by attending the weekly Thursday seminars and associating with such outstanding scientists as Enrico Fermi, Willard Libby, Clyde Hutchison, Tony Turkevich, and Julian Goldsmith, as well as Cesare Emiliani, Gerald Wasserburg, and Harmon Craig, among those just beginning their careers. Sam was always available to help these younger scientists; for example, he showed Jerry Wasserburg how to blow glass. The ease of interactions among all the junior and senior scientists at the University of Chicago made a lasting impression on Sam.

Sam was now 32 years old, and he realized that he needed to find a more permanent position. Again fate intervened when Tony Turkevich suggested he speak to Harrison Brown, who was in the process of leaving Chicago to start a geochemistry program at the California Institute of Technology. Soon thereafter Sam happened to bump into Harrison, and he casually asked him whether he knew of any positions that might be available for someone like himself who was proficient in stable isotope geochemistry. Although this was one of the few times the two had really conversed, Harrison Brown immediately replied, "How would you like to come to Caltech with me?" Sam was taken aback inasmuch as he wasn't quite ready to leave on such short notice. Nevertheless he accepted Harrison's invitation to give a seminar at Caltech in February 1952, and on the train trip he was joined by Harrison Brown's graduate student Clair Patterson, then working with uranium and lead isotopes to measure the age of the earth, and by Charles McKinney, the engineer who had designed the mass spectrometer that Sam had been using for the past four years. As it turned out, all three of them were offered research faculty positions in the Division of Geological Sciences at Caltech, and they all decided to accept. Sam's decision was made easier by the contrast between the gloomy winter weather he left in Chicago and the beautiful sunny days that greeted him in California. Together with the Caltech geologist Leon T. Silver, who met their train in Pasadena, the Brown-Epstein-Patterson-McKinney group would form the nucleus of what turned out to be a remarkably successful geochemistry program at Caltech during the next half-century. Soon they were joined on the Caltech faculty by a couple of Sam's other friends from the University of Chicago, Heinz Lowenstam in 1953 and Jerry Wasserburg in 1955.

Thus began the mutually rewarding relationship between Samuel Epstein and Caltech that was to last a full 50 years. Sam played a major part in the transformation of a good but relatively small geology department whose international reputation had up until 1951 depended mainly upon their faculty in seismology and vertebrate paleontology. During the next 25 years, the number of faculty in the department increased from 13 to more than 30, it had become the Division of Geological and Planetary Sciences, and it consistently ranked at the top of all the earth science departments in the United States, with world-class programs in geology, geophysics, geochemistry, planetary science, and geobiology. The leadership provided by its chairman, Robert P. Sharp, was crucial during the early part of this period, as was the support of the president of Caltech, Lee DuBridge.

Initially Sam was a tenure-track research fellow with his salary paid by Harrison Brown's large research grant from the Atomic Energy Commission. This grant was used to transform the entire basement floor of one of the Caltech geology buildings into a set of state-of-the-art geochemistry laboratories. Sam was quickly promoted to senior research fellow in 1953 and associate professor of geochemistry in 1954. He and Diane became naturalized citizens of the United States, and their second son, Albert, was born in April 1954. Now Sam was free to independently design his own future research program, and he had the funds and the resources he needed to carry it out. The first step was having Chuck McKinney's help in building a gas-source mass spectrometer for CO_2 like the one he had left behind in Chicago. This was done rather quickly with the help of Victor Nenow, an electronics technician, and Curtis Bauman, a machinist. In another couple of years they constructed a similar gas-source mass spectrometer for deuterium and hydrogen analysis. These two homemade instruments with little modification were used for thousands and thousands of isotopic analyses over the next several decades by Sam and his students. Even though these machines were partially constructed with sealing wax and needed fairly constant attention from Sam and from Vic Nenow, they were in heavy use virtually up until Sam's death in 2001.

The principal focus of virtually all Samuel Epstein's research studies was to design high-vacuum extraction lines that could quantitatively extract all the oxygen, carbon, or hydrogen from a given substance, and then to transform these into pure CO_2 or pure H_2 for introduction into the mass spectrometers. The idea was to study the isotopic compositions of as many naturally occurring substances as possible, because Sam now had the instruments and the expertise to measure these variations more accurately than had ever before been possible. Of course, some samples only became available later in Sam's career, such as lunar samples from the Apollo missions and samples from the deep-sea drilling program.

As a chemist Sam was also interested in studying equilibrium phenomena and kinetics as a function of temperature in some of the isotopic exchange reactions that are important in nature. The order in which these various problems were attacked at Caltech was largely dictated by the specific interests of the students and postdoctoral fellows who periodically came along to do research with Sam, but remarkably enough Sam's final list of publications does indeed include multiple papers on almost every kind of natural material containing oxygen, hydrogen, or carbon.

The authors of this memoir were the first two students at Caltech to become closely involved with Sam Epstein. Robert N. Clayton was a newly arrived graduate student in the Division of Chemistry who became intrigued with the idea of doing a thesis on stable isotope geochemistry, and Hugh P. Taylor was an undergraduate geology major who decided to switch into the newly formed geochemistry option. Thus, at Caltech the first Ph.D. in geochemistry, in 1955, and the first B.S. in geochemistry, in 1954, both turned out to be students of Sam Epstein. Taylor went away for a year of graduate work at Harvard, but he would return in 1955 to do his Ph.D. thesis with Sam.

Bob Clayton, who like Sam was a Canadian, wanted to see whether the $^{18}O/^{16}O$ ratios of coexisting minerals formed at high temperatures could be used as geothermometers. Compared with the crude geothermometers then available to earth scientists, these would have the advantage of being independent of pressure, and in an assemblage of *N* minerals one might obtain *N – 1* independent temperatures of formation, providing a test for whether equilibrium had been obtained during crystallization. Sam was familiar with the fluorine reaction procedure pioneered by Peter Baertschi and Sol Silverman in Urey's laboratory at Chicago. For safety the dangerous fluorine gas cylinder had to be kept on the roof of the geology building, so Sam and Bob had a penthouse laboratory constructed to do these experiments. However, Bob Clayton also wanted to try another method: to see whether he could extract oxygen from silicates and oxides by reacting them with powdered graphite in a high-temperature reduction furnace. This technique worked well for quartz and magnetite but turned out to be unsuitable for most silicates. Nonetheless, Clayton and Epstein (1958) did demonstrate a close approach to equilibrium for many quartz-calcite-magnetite assemblages in nature, and together with A. E. J. Engel they were the first to apply these techniques to a hydrothermal mineral deposit: the Pb-Zn ore body at Leadville, Colorado. Bob then left Caltech and after a short stay at Penn State, he was offered a faculty position at the University of Chicago, where he inherited Urey's isotope laboratory and where he formed a long and very productive association with Sam's former technician, Toshiko Mayeda.

Sam's second Ph.D. candidate was Rod Park from the Division of Biology at Caltech. Park and Epstein (1960) performed pioneering experiments on the mechanisms of carbon isotope fractionation during photosynthesis in plants.

Rod went on to become vice chancellor of the University of California at Berkeley. Sam's third Ph.D. student was Hugh Taylor, who utilized the penthouse fluorine laboratory to carry out the first systematic study of $^{18}O/^{16}O$ in coexisting minerals of igneous rocks. Sam and Hugh demonstrated that these data gave information about igneous parent materials, as well as proving that the assemblages closely approached equilibrium, as mirrored by their uniform $^{18}O/^{16}O$ fractionations and a consistent sequence of decreasing $^{18}O/^{16}O$ that correlated with the strengths of the oxygen bonds in their crystal structures: quartz-Kfeldspar-plagioclase-pyroxene-hornblende-biotite-magnetite. It was also in this penthouse laboratory that C. D. ("Dave") Keeling was encouraged by Sam Epstein and Harrison Brown to begin his famous atmospheric CO_2 measurements (i.e., the Keeling curve) that in 1957 he continued at Scripps Institution of Oceanography.

In 1959 Hugh Taylor accepted a temporary faculty appointment at Caltech in order to continue working with Sam, before leaving for a short stay on the faculty at Penn State. He returned to the Caltech faculty in 1962, after which he and Sam continued to work together harmoniously for the next 40 years, sharing research grants, laboratory facilities, postdoctoral fellows, and graduate students, as well as traveling together to countless scientific meetings and conferences. Summing up, note that Sam's first three Ph.D. students came from three different departments at Caltech: chemistry, geology, and biology. This kind of scientific breadth was a hallmark of Sam's entire career in research.

Samuel Epstein's unique research style is illustrated by a study he did soon after arriving at Caltech. This involved analyses of $^{18}O/^{16}O$ in waters collected from all the storms that passed over Pasadena during the single rainy season of 1953-1954. The samples were collected, with Diane's help, from the water runoff from the roof of their new home in northeastern Pasadena. As Sam predicted, the $^{18}O/^{16}O$ of rain from a cold-front storm decreased with time, but as the heavier cold air penetrated the area it uplifted the warm moist air, causing adiabatic cooling and increasing the intensity and the ^{18}O content of the rainfall. A reversed isotopic pattern was observed in rain from a warm-front storm. Rapid and dramatic changes in isotopic composition were attributed to stratified clouds, with low ^{18}O rain coming from colder clouds at higher elevations. Sam also analyzed $^{18}O/^{16}O$ of rain from a single storm collected in five separate locations: Santa Barbara, Pasadena, and different elevations in the San Gabriel mountains. As predicted, $^{18}O/^{16}O$ correlated nicely with local atmospheric temperatures (1956).

Sam was not particularly polished as a public speaker or in the classroom, but he was famous among many of his associates and colleagues for his remarkably original insights into scientific problems. The education of his students might come about as they finally understood the deep significance of an idea that he was trying to put into words. Sam left indelible marks on all members of his scientific family, particularly through his knack for solving seemingly intractable problems with a little clever laboratory work, his refusal to become bogged down in unimportant details, and his intuitive feel for the intrinsic accuracy required for a given measurement to be decisive. He understood that in doing science it was crucial to expend one's efforts only on really important problems, a credo that had been instilled in him by Harold Urey. Often his first reaction in discussing some new idea was, "Let's think of a way to make those measurements in the laboratory."

In December 1989 most of Sam's former students and associates gathered at Caltech to celebrate Sam's 70th birthday. The Epstein Symposium was held in a lecture hall festooned with large poster-size photographs of Sam at various stages of his career, including his childhood. The published record of the symposium is contained in Geochemical Society Special Publication No. 3: Stable Isotope Geochemistry, a Tribute to Samuel Epstein, edited by H. P. Taylor, J. R. O'Neil, and I. R. Kaplan, published in 1991. Summarizing the preface of that book, the following paragraphs list some of Sam's other research accomplishments at Caltech.

With Heinz Lowenstam and Cesare Emiliani, Sam continued to refine the carbonate paleotemperature method and to better understand the temperature-shell growth relations of $CaCO_3$-secreting organisms, extending these studies to Pleistocene and Recent fauna, and to the origin of sedimentary aragonite needles. With Sol Silverman he investigated the origin of petroleum and organic matter in sedimentary rocks. With Robert P. Sharp he made pioneering studies of the D/H and $^{18}O/^{16}O$ climate records in the annual layers of snow accumulation in the Malaspina, Saskatchewan,and Blue glaciers, and in Antarctic snow, firn, and ice. Later joined by Anthony Gow, Epstein and Sharp extended these studies to include a remarkably complete six-year record in firn at the South Pole, and still later to the record in some deep ice cores from Byrd Station, Antarctica.

With Donald Graf and Egon Degens he carried out the first major isotopic studies of the origin of dolomites and cherts, and of the preservation of the $^{18}O/^{16}O$ record in ancient sedimentary rocks. With Hugh Taylor and Arden Albee he carried out the first detailed study of coexisting minerals in high-grade pelitic schists, demonstrating that these assemblages crystallized in equilibrium. With Hugh Taylor he carried out $^{18}O/^{16}O$ studies of all the tektite fields of the world, comparing these data with sedimentary rocks and other terrestrial impact glasses, and thereby helping to confirm that tektites had a terrestrial impact origin. With Jim O'Neil, who had been Bob Clayton's first Ph.D. student at

Chicago and who then continued his career as a postdoctoral fellow at Caltech, Sam pioneered a method for direct isotopic analysis of milligram quantities of water. O'Neil and Epstein also studied the equilibrium $^{18}O/^{16}O$ relationships in the system dolomite-calcite-carbon dioxide, and they followed up an earlier study Sam made with Bill Compston, trying to finalize the correct value of the $^{18}O/^{16}O$ fractionation factor for CO_2-H_2O exchange at 25°C.

With Samuel Savin, who later became dean of the College of Arts and Sciences at Case Western Reserve University, Sam Epstein did the first systematic survey of D/H and $^{18}O/^{16}O$ in clay minerals and sedimentary rocks. They showed that in the weathering environment clay minerals typically form in isotopic equilibrium with their local waters, allowing such clays to be used as climate indicators and defining a kaolinite line that is subparallel to Harmon Craig's famous meteoric water line on a plot of D/H versus $^{18}O/^{16}O$. With Donald Garlick he did the first major study of $^{18}O/^{16}O$ variations in coexisting minerals as a function of increasing grade in some classic areas of regional metamorphism, as well as the first isotopic study of the copper ore deposit at Butte, Montana, showing that heated meteoric ground waters were a major constituent of the ore-forming system.

With J. Hoefs he did the first oxygen isotope studies of migmatites and orbicular igneous rocks. With Lynton Land he continued his studies of the processes of dolomitization. With Simon Sheppard he was the first to characterize D/H and $^{18}O/^{16}O$ ratios of minerals brought up from the earth's upper mantle in diamond-bearing kimberlites. With Hugh Taylor, Mike Duke, Helmut Reuter, and Lee Silver, Sam was the first to systematically measure $^{18}O/^{16}O$ ratios of the minerals in meteorites, showing that there were distinct differences between the two major groups of chondrites, and that both of these groups differed significantly from the basaltic achondrites and the carbonaceous chondrites.

With Bruce Smith, Sam continued his pioneering work on carbon isotope applications to botany, monitoring the isotopic changes between lipids and carbohydrates, and showing that the higher plants are divided into two major groups with different $^{13}C/^{12}C$ ratios. They showed that plants using the C_4 photosynthetic pathway (e.g., maize, millet, sugarcane) were systematically less depleted in ^{13}C than the much more abundant C_3 plants, and this is now a standard technique for distinguishing between C_3 and C_4 plants.

Later with his Ph.D. student Mike DeNiro, Sam developed techniques for looking at stable isotope variations in the paleodiets of humans and animals, and this turned out to have many archeological applications. Together they published a series of papers on isotope variations in animal and plant tissues, including fossil bones, pointing out that the $^{13}C/^{12}C$ and $^{15}N/^{14}N$ ratios in animal tissues strongly correlate with the animal's diet. For $^{13}C/^{12}C$ this was summed up by the phrase,"You are what you eat . . . plus one per mil."

Beginning in the late 1960s Sam Epstein and Hugh Taylor prepared their laboratories for analysis of the soon-to-be-returned lunar samples. Their role was to measure isotope ratios of carbon, oxygen, hydrogen, and silicon. Although technicians did a lot of the routine work during their studies of terrestrial samples, Sam and Hugh both felt that they should do all the extractions on the priceless lunar samples themselves, in a newly constructed laboratory on the top floor of the geology building. Among other things they showed that hydrogen in the lunar soil was essentially deuterium free and thus totally derived from solar wind, and they were also the first to discover the enormous ^{18}O and ^{30}Si enrichments of the grain surfaces in the lunar soil, a result they attributed to micrometeorite and particle bombardment. For Hugh this was a memorable opportunity to work side by side with Sam for several years, and to again observe at close hand the unique way he attacked scientific problems. For example, if varying a particular procedure in a sequence of analytical steps in an experiment shouldn't *a priori* make any difference to the final result, Sam would often modify it on purpose. Usually nothing came of this, but occasionally this allowed Sam to gain an unexpected insight into some analytical problem.

Much of Sam's research in the 1970s continued to be focused on the lunar samples. However, with Tetsuro Suzuoki he developed hydrogen isotope geothermometers relating to exchange between hornblendes, micas, chlorites, and water. With Paul Knauth, J. Karhu, and Yehoshua Kolodny he instituted some systematic studies of D/H and $^{18}O/^{16}O$ in cherts throughout the geologic record, including very ancient Precambrian rocks. With Steve Lambert he studied drill-core samples from hydrothermal systems at The Geysers, California, and the Valles Caldera, New Mexico. With Crayton Yapp and later with R. V. Krishnamurthy and X. Feng he developed techniques for determining the D/H ratios of nonexchangeable hydrogen in cellulose of plants, using these to monitor past climatic changes recorded in tree rings. With Francois Robert, Richard Becker, Jon Yang, R. V. Krishnamurthy, and John Stone he started a major program of looking at hydrogen, carbon, silicon, and nitrogen isotope variations in unequilibrated meteorites, including the carbonaceous chondrites. They discovered some truly gigantic isotope fractionation effects, some of which can be traced back to before the formation of the Solar System.

During the last few years before his retirement in 1990, coincident with the arrival at Caltech of a young colleague in igneous petrology named Ed Stolper, Sam started to do experiments on oxygen isotope exchange between volatiles and silicates at high temperature and also on the hydrogen isotope fractionation between coexisting vapor and the different species of H_2O dissolved in silicate melts. Stolper and Epstein studied the $^{18}O/^{16}O$ fractionations between silica glass and CO_2, and later they were joined by Alan Matthews and Mike Palin in an analogous study of albite, albite glass, and CO_2. A whole new younger generation of graduate students and research fellows at Caltech (e.g., Phil Ihinger, Laurie Leshin, Pat Dobson, Jen Blank, and Sally Newman) worked with Ed and Sam on these important problems, including a study of the SNC meteorites, thought by many workers to be derived from the planet Mars. The studies with Stolper and his students continued to flourish during the decade of the 1990s, even after Sam became Professor Emeritus.

Remembering the times that various of his students interacted with Sam, we recall how many of us suffered with him on those days when his ulcer was acting up, or when we happily witnessed him finally abandon smoking, or when we had lunch with him in the Athenaeum at Caltech where he would voice strong opinions on science, world politics, baseball, and many other topics before negotiating with the waiter for a piece of Camembert of just the correct degree of ripeness. We remember his twinkling eyes and smiling countenance, and how we were charmed by his sincere affection for children—anyone's children. Countless times he also indicated how grateful he was to his own family and how important they were to him. His son Reuben Epstein graduated from Caltech in 1970, received his Ph.D. in physics from Stanford in 1976, and spent four years at MIT working with Irwin Shapiro on astrophysical problems involving relativistic gravitation. He is currently senior scientist in the theory group at the Laboratory for Laser Energetics at the University of Rochester working on inertial confinement fusion. Son Albert Epstein received a B.S. in business administration from Cal State, Los Angeles, in 1985. He is presently senior judicial assistant in the Superior Court of Los Angeles and is involved in semiprofessional folk dancing and choral music groups.

Samuel Epstein was widely recognized for his scientific achievements. In 1975 he went to the Hebrew University of Jerusalem as a Lady Davis Fellow, and in 1977 he was elected to both the National Academy of Sciences and the American Academy of Arts and Sciences. He received the Geochemical Society's Goldschmidt Medal in 1977 and the Geological Society of America's Day Medal in 1978. Sam was president of the Geochemical Society in 1978-1979, and in 1980 he received the honorary LL.D. degree from the University of Manitoba. In 1984 Sam was named William E. Leonhard Professor of Geology at Caltech. In 1993 he received the Wollaston Medal of the Geological Society. He shared the 1995 Urey Medal of the European Association for Geochemistry with his former students Bob Clayton and Hugh Taylor. In 1997 he became a foreign fellow of the Royal Society of Canada.

Although he "retired'' in June 1990, he continued to carry out productive full-time work in the laboratory every day, virtually up until his death from cancer on September 17, 2001. Sam's widow, Diane, died on November 4, 2007; they are survived by their sons, Albert and Reuben; Reuben's wife, Jody; and three grandchildren: Aaron, Eric, and Chloe.

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