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PASSING OF THE LAST OF THE THREE WHO ESTABLISHED ^{14}C DATING: A HISTORICAL FOOTNOTE

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ABSTRACT. Ernest Anderson, the last of the three scientists responsible for the initial research establishing the validity of the radiocarbon dating method at the University of Chicago in the late 1940s, has died. From early 1947 to the end of 1949, Anderson and James Arnold joined with Willard Libby to undertake three critical experiments, employing what one of them called a "procedure developed in hell" (solid carbon decay counting). They laid the foundation of what has developed over more than 6 decades into the "gold standard" for the chronometric dating of the most recent phases of the technological and cultural evolution of anatomically modern human societies, the geology of the terminal Pleistocene and Holocene, critical events in the history of the Earth's climate over the last 50,000 yr, and a range of other scientific and historical applications.

INTRODUCTION

Ernest Anderson, who was, beginning in 1946, Willard Libby's first graduate student at the University of Chicago, died on 20 May 2013, at the age of 92 after a lengthy period of disability. James Arnold, who worked with Libby and Anderson in the development of the radiocarbon method at Chicago from 1948 to 1952, had earlier passed away on 6 January 2012 at the age of 88 after a long illness. Willard Libby, who received the Nobel laureate in Chemistry in 1960 for the development of the ^{14}C method, died unexpectedly in 1980 at the age of 71. These individuals undertook the foundational research that demonstrated that ^{14}C could be used as a chronometric dating isotope. They were the coauthors of the first paper reporting the first two age determinations based on ^{14}C measurements (Libby et al. 1949).[1]

ERNEST CARL ANDERSON (1920–2013)

Anderson's important role in the development of the instrumentation that was used in the initial experiments that established the validity of using ^{14}C as a dating isotope has not been sufficiently stressed in some discussions of the early history of the method. During World War II, he had worked on the Manhattan Project at the Los Alamos Scientific Laboratory. He joined Libby in early 1946, originally as a laboratory assistant, before becoming his first Chicago graduate student (Figure 1). According to Libby, Anderson asked him if he had any interesting problems that could be pursued for a dissertation. Libby reports that he said to Anderson, "Well, I want to measure the natural abundance of radiocarbon." To which Anderson replied "I'm your boy" (Libby 1979:36–7; Marlowe 1999:31).

Anderson joined Libby at the very beginning of a 3-yr period during which the first two of three critical experiments were undertaken that empirically examined the implications of the predictions that Libby had published in a brief 1946 *Physical Review* paper where he outlined the basic idea be-

1. The first two published ^{14}C-based age determinations reported in this paper were on two wood samples from the tombs of two Old Kingdom Egyptian kings, Snefera and Zoser, which had been assigned historic ages of 4575 (±75) and 4650 (±75) yr, respectively, by University of Chicago Egyptologist John Wilson. The first two ^{14}C measurements were combined and expressed in the publication as a weighted average of the measured specific activity of both samples in units of counts per minute of grams of carbon (cpm/gm/c). Based on the assumed historical age, and the then recently measured ^{14}C half-life of 5720 ± 47 yr (Engelkemeir et al. 1949), the expected value was 7.35 ± 0.15 cpm/gm/c and the measured weighted average was 7.15 ± 0.35 cpm/gm/c.

hind ^{14}C dating (Libby 1946).[2] A search of previous literature in a number of journals and reference sources has failed to this date to locate any use of the term "radiocarbon" prior to its appearance in this paper.

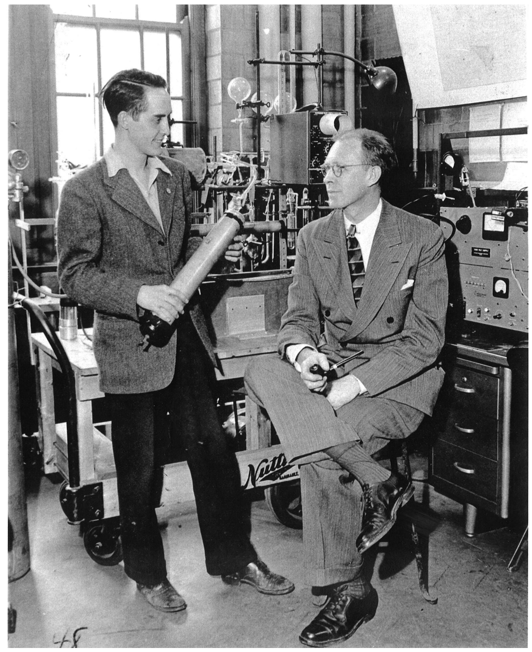

Figure 1 Ernest Anderson (1920–2013) [on left] and Willard Libby (1905–1980) taken in 1948 in Libby's laboratory in Jones Hall, University of Chicago. In the background are parts of the instrumentation used in making ^{14}C measurements. Source: University of Chicago Regenstein Library, Special Collections, Photographic archive.

The first experiment addressed the question of whether there was a measurable difference in ^{14}C content between living and fossil organics (Anderson et al. 1947a,b). The second tested whether the ^{14}C content of living terrestrial organics was essentially (±10%) similar no matter their geographic location on Earth and to establish the specific ^{14}C activity of contemporary terrestrial living materials. That study became the basis of Anderson's Chicago dissertation carrying the simple title "Natural Radiocarbon" (Anderson 1949). The third examined the relationship between the assumed historically documented known age and the ^{14}C-deduced age of historic and tree-ring-dated wood samples to produce the first ^{14}C "Curve of Knowns" (Arnold and Libby 1949).

To undertake the second and third experiments, it was necessary to develop a counting technology that allowed natural ^{14}C concentrations to be measured without recourse to very costly isotopic enrichment as had been done in the first experiment, which demonstrated that fossil and modern carbon exhibited the predicted differences in their ^{14}C content. The most important element in making possible the decay counting of natural ^{14}C without recourse to enrichment was the application of the

2. Where, when, and under what conditions Libby first conceived or encountered the basic idea behind ^{14}C dating has been the subject of some discussion. In an interview undertaken in 1979, Libby states that he conceived the idea of ^{14}C dating after reading a 1939 article that reported the presence of neutrons in the atmosphere caused by cosmic rays. The principal author of that article was Serge Alexander Korff [1906–1983], who, at this time, was studying the physics of cosmic rays. As Libby expressed it: "As soon as I read Korff's [1939] paper, where he'd found neutrons in cosmic rays, that's carbon dating" (Libby 1979:33, 40). The article was Korff and Danforth (1939). This understanding of the origin of the idea behind ^{14}C dating may be contrasted with a comment made in an autobiography of Emilio Segrè [1905–1989], an Italian physicist resident at the University Chicago immediately following World War II. Segrè had previously been a doctoral student in Italy of Enrico Fermi [1901–1954] who was also at Chicago during this period. Segrè notes that he had "been told that the suggestion [for ^{14}C dating] came from Fermi in a Chicago seminar" (Segrè 1991:150). Segrè's earlier biography of Fermi made no mention of this rumor (Segrè 1970).

anticoincidence principle to reduce background count rates in the counter used to obtain the measurements. Without the use of this strategy to reduce background counts, the decay-counting-based measurements of ^{14}C in samples would have required isotope enrichment[3] (Figure 2).

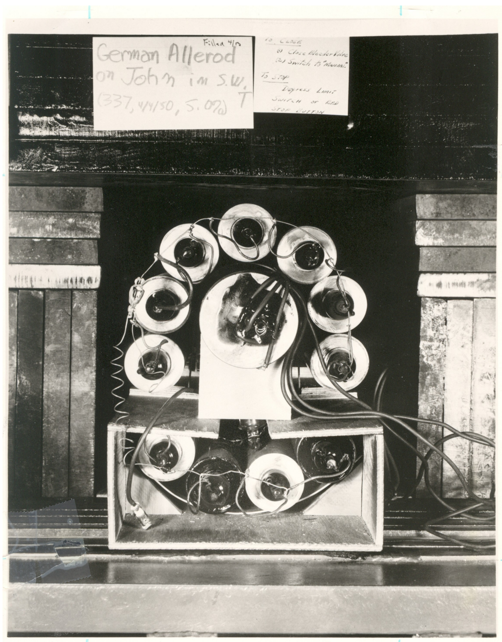

Figure 2 Chicago solid carbon counting instrumentation. Screen-wall counter used for ^{14}C measurements (center counter) surrounded by a ring of GM tubes. Source: R Berger and LM Libby, University of California, Los Angeles (USA) and Isotope Foundation.

Libby later stated that he was not able to recall who first suggested the anticoincidence approach to reduce background counts (Libby 1970:5, 1979:39). There had been at various times vague hints in oral communications from various individuals that Anderson may have initially suggested the use of this strategy. However, upon directly questioning him about this on a number of occasions many decades later, he indicated he could not confirm that he was the source of the idea, but would only state that he recalled that the idea was kind of "in the air" at the time (EC Anderson, personal communications, 1986, 1990, 2001). He did recall that the principle of *coincidence* counting had been a standard method employed in cosmic-ray studies since the 1930s (Anderson 1953:75). Arnold later expressed his understanding that the anticoincidence concept had been "borrowed by Libby from the cosmic-ray physicists and put together in the first crude form by Anderson" (Arnold 1992:6). Anderson later discovered that Danish scientists in the 1930s had employed the anticoincidence principle for low-level counting applications, but this technology was not known to Libby or anyone in his group while ^{14}C dating was being developed at Chicago because the 1930s application had been published in a Danish language journal (EC Anderson, personal communication, 1986, 2001; Libby LM, n.d.).

3. An anticoincidence arrangement involved surrounding a Geiger-Muller (GM) detector containing the sample with a "guard" ring composed of multiple GM detectors as illustrated in Figure 2. Pulses coming from the sample counter are electronically compared as to coincidence or noncoincidence (anticoincidence) with pulses coming from one or more of the guard counters within a defined time interval measured in milliseconds. The circuitry in the instrument was arranged in order to count pulses from the central counter *only* if these pulses were *not* accompanied by an essentially simultaneous pulse from the surrounding guard counters. By these means, signals caused by the decay of an isotope, in this case ^{14}C, in the central counter could be distinguished from signals in the central counter containing the sample that were due to ionizing radiation coming from the environment external to the detector.

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Upon completion of his PhD at Chicago in 1949, Anderson returned to the Los Alamos Scientific Laboratory in Los Alamos, New Mexico, where he spent most of the remainder of his productive scientific career. He continued to be involved in the development of low-level counting and assisted in the development of the ^{14}C laboratory at the University of Copenhagen in the early 1950s (Anderson et al. 1953). In 1966, he received the Ernest O. Lawrence Award from the United States Department of Energy, successor to the Atomic Energy Agency, for his contributions to nuclear technology in the life sciences. The award citation noted that it had been given to him "for outstanding contributions to nuclear medicine, to biological research, to archaeological dating, and for the development of liquid scintillation counting which made possible early neutrino experiments and the liquid scintillator whole body counter." He retired from Los Alamos in 1995.

JAMES RICHARD ARNOLD (1923–2012)

James Arnold initially had worked with Libby and two other professors at the University of Chicago beginning in February 1946 as a postdoctoral fellow (Figure 3). It was during this time that he later reported that he became cognizant of Libby's serious intention to use ^{14}C as a means of dating. He recalls that he gained this information while attending one of Libby's legendary parties. Arnold had entered Princeton at the age of 16, receiving his bachelor's (1943), Master's (1945), and PhD degrees (1946) there. He had been able to undertake his dissertation research as part of the World War II Manhattan Project.

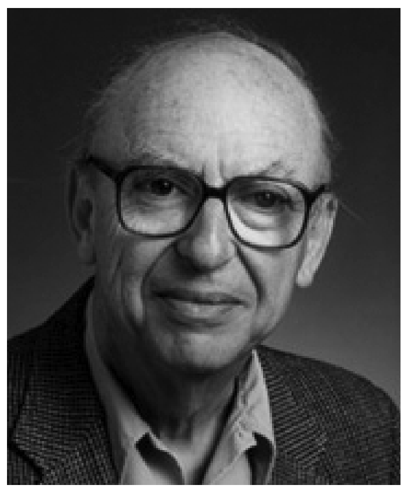

Figure 3 James Richard Arnold (1923–2012) taken in 2005. Source: Mandeville Special Collections, Geisel Library, University of California, San Diego.

For a short period in the spring of 1946, his association with Libby involved working with reactor-produced ^{14}C (Arnold and Libby 1946). Libby had a sample of barium carbonate inserted into a reactor at the Oak Ridge National Laboratory in Tennessee that was producing a high flux of neutrons. This bombardment yielded about a millicurie of artificial ^{14}C. This amount of ^{14}C was many orders of magnitude in excess of what had ever been, to that point, produced. This was sufficient to undertake the experiment (Engelkemeir et al. 1949) that yielded the first ^{14}C half-life of 5720 ± 47 yr that Libby used in his early publications (Marlowe 1980:1005, 1999:11; JR Arnold, personal communication, 2001).

Early in 1947, Arnold left Chicago for Harvard as a National Research Fellow. In early 1948, he returned to Chicago as a research associate to work with Libby and Anderson on the ^{14}C project supported by a grant from the Viking Fund, now known as the Wenner-Gren Foundation for An-

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thropological Research.[4] His principal role involved the preparation and counting of the samples that would constituted the first series of ^{14}C measurements on "known age" materials, the oldest of which were assigned ages based on their association with ancient Egyptian archaeological contexts. Although a chemist, Arnold was very familiar with ancient Egyptian archaeology and chronology because he had grown up in a household where ancient Egypt was the topic of many conversations. This was because Arnold's father, while an attorney by profession, was characterized by his son as a "serious amateur archaeologist." The elder Arnold was the secretary of the Egyptian Exploration Fund in the United States, with responsibility for fundraising for this organization in North America (Arnold 1992:4).

The technology that was employed by the Chicago group in measuring natural levels of ^{14}C employed solid carbon ("carbon black"), which was prepared by the reduction of CO_2 on magnesium. The counter into which the solid carbon was introduced employed a screen wall design of the same general type that Libby had employed in his dissertation research at the University of California, Berkeley, in the early 1930s. To place the solid carbon into the counter, it was necessary to create a slurry of the carbon black in a liquid medium and coat the inside surface of a metal sleeve with this mixture. In theory, the evaporation of the liquid should leave the carbon black adhering to the inside surface of the sleeve. Unfortunately, even the slightest movement of the sleeve was often sufficient to dislodge parts of the carbon coating. Arnold's later reflections concerning solid carbon counting was that it was a "procedure developed in hell" (JR Arnold, personal communication, 2001). Interestingly, it appears that Anderson was one of the few early ^{14}C investigators who worked with solid carbon ^{14}C counting that did not share Arnold's view (EN Anderson, personal communication, 2001).

Arnold was directly involved in measuring the first reported ^{14}C "date." It was obtained on a piece of cypress wood taken from the tomb of the Third Dynasty Egyptian king Zoser (Djoser). Arnold later recalled that he calculated that first result of the count rate of that sample in Libby's laboratory on the hot Chicago afternoon of 12 July 1948. If one wished to identify a specific day as the "birthday" of ^{14}C dating, that date could appropriately be considered the natal day of ^{14}C dating. Arnold later stated that for "a couple of heady hours, I was the only person in the world who knew that ^{14}C dating worked. One lives for such moments" (Arnold 1992:8). Arnold was responsible for supervising the measurement of the majority of samples that comprised the first ^{14}C "date list" (Chicago I), which was published in the journal *Science* in February 1951 (Arnold and Libby 1951).

Arnold left Libby's group in 1955 to serve for 2 years on the faculty of his *alma mater*, Princeton University. Prior to leaving Chicago, he had collaborated with a group at the Los Alamos Scientific Laboratory in the development of liquid scintillation technology for low-level decay counting (Arnold 1954). He also discovered a short-lived isotope of beryllium (^{7}Be) in nature, an isotope that is used in meteorological research (Arnold and Al-Salih 1955). Also while he was at Princeton, he discovered the natural occurrence of long-lived ^{10}Be (Arnold 1956). In 1958, he was asked to be the founding chairman of the Department of Chemistry, University of California, San Diego (UCSD).

Arnold spent the remainder of his illustrious scientific career at UCSD, a campus overlooking the

4. A significant portion of the funds provided by the Viking Fund to Libby (actually the grant was awarded to both Libby and Harold Urey) was used to construct and test a thermal diffusion column that was intended to be used to undertake the isotopic enrichment of ^{14}C in samples. Initially, Libby was not sure that decay counting, even with an anticoincidence arrangement, could be developed to the point that acceptable natural level measurements could be obtained. The column was never used because of the success of the anticoincidence feature of the decay counting system that Anderson constructed. Libby also considered moving the counting equipment into a mine as a means of reducing the background (Libby 1973:xxxiv, 1979:38–9; JR Arnold, personal communication, 1986).

Pacific Ocean at La Jolla in southern California. Most of his subsequent research was focused on various aspects of space and planetary science including cosmogenic geochemistry. He was one of the scientists participating in the United States National Aeronautics and Space Administration's (NASA) Apollo fights to the moon, conducting geochemical studies of lunar samples returned from NASA missions. He founded the California Space Institute in 1979, served as its Director for a decade, and in the context of that service, had an asteroid named for him. With several colleagues, Arnold demonstrated the approximate constancy of the cosmic-ray flux over a period of several million years. He was elected to the US National Academy of Sciences in 1964 and was appointed as the first holder of UCSD Harold C. Urey Professorship in Chemistry. Arnold retired to emeritus status from UCSD in 1993 (Woo 2012).

WILLARD FRANK LIBBY (1905–1980)

Biographical materials, which include discussions of the scientific career of Libby, are available in a number of publications, as is his own much later recollections of the early years of the development of the ^{14}C method (see discussion and references in Taylor and Bar-Yosef 2014: Chapter 8).

As an undergraduate, Libby attended the University of California, Berkeley, first as a petroleum engineering student, but then transferred to chemistry in his sophomore year, He states that he changed his major primarily because the Berkeley rooming house in which he lived was occupied mainly by chemistry majors. He was sometimes referred to as "Wild Bill Libby" initially based on a reputation gained in high school associated with how he performed during football games. The term followed him in various contexts for the remainder of his life (Arnold 1981, 1992; Taylor and Bar-Yosef 2014: Chapter 8).

In his senior year at Berkeley, for his senior honors project, he had built with what was almost certainly the first Geiger or Geiger-Müller (GM) type ionizing radiation detector assembled in the United States, the design of which was based on a 1928 publication in German by Hans Geiger and Walter Müller (Geiger and Müller 1928). He stayed on at Berkeley for his PhD and, as part of his dissertation, constructed a screen-wall type GM counter to measure weak beta radiation from samarium and other rare earth elements (Libby 1933). With very little modification, the screen-wall counter design used in his dissertation research would be used in his ^{14}C research at the University of Chicago.

As was the case with Anderson and Arnold, during World War II Libby was affiliated with one of the research groups involved in the Manhattan Project efforts to develop an atomic weapon for the United States. His group, directed by Nobel laureate Harold Urey at Columbia University, was focused on the development of a method of enriching ^{235}U by gaseous diffusion. Surprisingly, Libby later expressed the view that his World War II chemical research to develop a thermal diffusion barrier was "better than my carbon 14 dating" (Libby 1978:37).

Following World War II, Libby and Urey along with a number of others in Urey's Manhattan Project group at Columbia, were recruited in late 1945 to the University of Chicago to establish a new Institute for Nuclear Studies, now known as the Enrico Fermi Institute for Nuclear Studies. Libby, who at the time of his appointment was the youngest full Professor on the University of Chicago faculty, was at Chicago until 1952. He then was nominated by President Eisenhower to be a member of the Atomic Energy Commission (AEC), later known as the United States Department of Energy (DOE). At that time, he was the only scientist serving as an AEC commissioner.

During the period of his tenure at the AEC, Libby became a controversial advocate of both military and civilian uses of atomic energy, appearing on the cover of the 15 August 1955 issue of *Time* magazine in his role as an AEC commissioner. He also acquired a public reputation as a political conservative. For example, he openly opposed the reappointment of J Robert Oppenheimer as a member of the General Advisory Committee of the AEC (Hewlett and Holl 1989:46) and wrote an open letter to the Nobel Peace Prize laureate Albert Schweitzer opposing Schweitzer's call for the end of further experiments with atom bombs (News of Science 1957).

In 1959, he returned to the University of California, becoming Professor of Chemistry at the University of California, Los Angeles (UCLA), and later Director of the UCLA Institute of Geophysics and Planetary Physics (IGPP). The following year, he received the Nobel Prize in Chemistry for the development of the ^{14}C method. At UCLA, he established a ^{14}C laboratory in the IGPP that operated from 1959 to 1995 (Berger 1992). It is estimated that approximately 60% of the samples dated at the UCLA laboratory were obtained on archaeological materials.

Libby once indicated that he considered himself something of an "amateur archaeologist," being very supportive of efforts to demonstrate the presence of humans in the New World much older than generally accepted by a majority in the archaeological community (Libby in Olsson 1970:107). For example, in pursuit of this goal, he was very supportive of excavations undertaken at Tule Springs, Nevada, in the early 1960s, serving as a member of the scientific advisory group (Figure 4) for the project and, through his Isotope Foundation, contributed funds to support the research (Shutler 1967). It appears that Libby's interest in supporting a large-scale excavation at Tule Springs was stimulated in large part by a single ^{14}C determination of >23,000 yr (C-914), which had been obtained at his Chicago laboratory in the early 1950s on material excavated from this site.

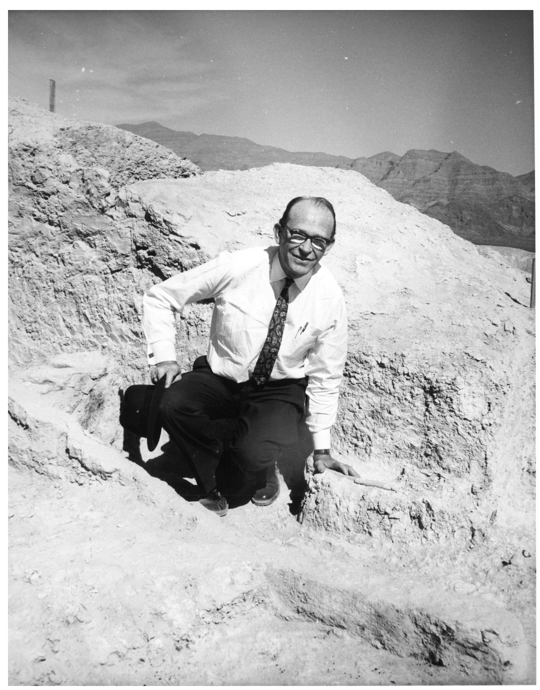

Figure 4 Willard Libby visiting excavations underway at the Tule Springs Site, Nevada (USA) in 1963. Source: Isotope Foundation, Santa Monica, California (USA).

The Tule Springs sample measured at Chicago was characterized as "charcoal" recovered from what had been labeled a "hearth-like feature" by excavations conducted in the 1930s. The excavators associated its occurrence with sediments containing the bones of extinct fauna. Detailed geological

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investigations carried out during the Tule Springs project in the early 1960s by C Vance Haynes, Jr., determined that the features originally labeled as "hearths" containing "charcoal" were in fact concentrations not of charcoal, but of decayed plant remains associated with water channel debris or spring deposits (Worminton and Ellis 1967). One may assume that Libby was disappointed when the extensive excavations and a large suite of ^{14}C determinations from this site obtained at his UCLA laboratory failed to confirm a pre-Clovis status for the Tule Springs site.

Libby retired as emeritus professor at UCLA in 1976, but continued to pursue a number of research projects (Arnold 1981). His widow, a well-known nuclear chemist in her own right, Leona Marshall Libby (1919–1986), later noted, "Until the day of his death, [Libby] was carrying on his researches at top speed as he always did" (LM Libby 1981).

CONCLUSION

More than 6 decades have passed since Ernest Anderson, James Arnold, and Willard Libby published the first paper that contained the first set of ^{14}C "dates." In his retrospective comments concerning the development of the method, Libby noted the "invaluable contributions" of his two collaborators, expressing the view that "certainly nothing would have been done without them" (Libby 1952:v).

ACKNOWLEDGMENTS

On a number of occasions over a number of years, both Ernest Anderson and James Arnold very graciously and patiently related to the author background information focusing on various historical details not contained in any previously published source that provided detailed insights with regard to the scope of the often difficult and frustrating work that resulted in the initial establishment of the ^{14}C method. Many photographs and documents concerning the history of ^{14}C dating were obtained by the author while a graduate student in Libby's Isotope Laboratory at the University of California, Los Angeles (UCLA) in the 1960s. The support of Libby was instrumental in establishing the author's ^{14}C laboratory at the University of California, Riverside. The records of Libby's laboratory at the University of Chicago and UCLA are currently being curated by the author. A more detailed and expanded presentation of some of the information presented in this paper has been included in a discussion of the history of ^{14}C dating in a book length treatment of ^{14}C dating in archaeology (Taylor and Bar-Yosef 2014: Chapter 8). The author wishes to thank the University of Chicago Regenstein Library, Special Collections, Photographic Archive, Mandeville Special Collections, Geisel Library, University of California, San Diego, and the Isotope Foundation, Santa Monica, California, for permission to use the figures used in this article. He also wishes to acknowledge the support of the Gabriel R. Vierra Memorial Fund.

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