UNIVERSITY of PENNSYLVANIA
PHILADELPHIA 19104

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Franklin Building
3451 Walnut Street
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February 4, 1971

Mrs. Mary W. Greene
Associate Program Director
for Anthropology
National Science Foundation
Washington, D. C. 20550

Dear Mrs. Greene:

Submitted herewith are twenty (20) copies of proposal for research support entitled "The Investigation of Archaeological Remains Using Organic Chemical Techniques, under the direction of Dr. John Winter, University Museum.

The proposal has been approved by appropriate University officials and signed on behalf of the University by Dr. John N. Hobstetter, Vice Provost for Research.

Should additional information be required, please do not hesitate to communicate with us.

Sincerely,

Joel E. Kinley
Contracts Administrator

JEK:vm
encl.

cc: Dr. Winter
Dr. Rainey ✓

UNIVERSITY OF PENNSYLVANIA
Philadelphia, Pennsylvania 19104

PROPOSAL FOR RESEARCH PROJECT

Submitted to: Division of Earth Sciences, National Science Foundation

Title of proposal: Empiric Approach to Measurement of Rate of Equilibration of Surface Water with Atmospheric CO_2.

Principal Investigator: Robert F. Giegengack, Assistant Professor
Department of Geology

Faculty Associate: Elizabeth K. Ralph, Associate in Physics
Director, Radiocarbon Laboratories
Associate Director, Museum Applied Science Center for Archeology

School: College   Department: Geology

Starting Date: September 1970   Duration: 2 years

FUNDS REQUESTED

First Year: $ 75,510

Total: $126,425

Corporate name of University: THE TRUSTEES OF THE UNIVERSITY OF PENNSYLVANIA
(a Pennsylvania non-profit corporation)

Contracting Office: OFFICE OF RESEARCH ADMINISTRATION
3451 Walnut Street
Philadelphia, Pa. 19104

Principal Investigator ______________________ Date May 20, 1970

Faculty Associate ______________________ Date May 20, 1970

Approved

Department Chairman Henry Faul   May 20, 1970
Date

Dean of the College ______________________ ________
Date

Vice Provost for Research ______________________ ________
Date

INTRODUCTION

Radiocarbon investigators have long looked with suspicion on dates obtained from analysis of carbonate in fresh-water limestone, particularly spring-deposited travertine (e.g. Broecker and Walton 1959). This is because ground water typically contains a high proportion of old carbon, a result both of the actual C^{14} age of the water that has been moving underground, perhaps for tens of thousands of years, and because carbonate in ground water exchanges readily with "dead" carbonate from limestone bedrock.

The apparent reliability of C^{14} ages of travertine samples from the Great Basin (Broecker and Kaufman, 1965) indicates that there has been sufficient exchange of CO_2 between the atmosphere and surface water to bring about equilibration of the C^{14}/C^{12} ratio in the water to the atmospheric value between the time that ground water emerges at the Earth's surface and the time that $CaCO_3$ is precipitated from it in the surf zone around the margin of a pluvial lake.

It is the goal of the proposed study to measure empirically the distance of surface flow required for emerging ground water to achieve such equilibration. The experimental procedure will be to measure the apparent C^{14} ages of travertine samples collected along the length of Havasu Creek, Arizona, and to date independently the same samples by their close association with dead wood. The extent of possible depletion of C^{14} activity in the wood by incorporation of old carbon will be closely approximated by comparing its carbon-isotope spectrum with that of wood of known age.

The following discussion of available radiocarbon ages of travertine will underline the uncertainties in the data and will demonstrate that the equilibration described above must occur.

AVAILABLE RADIOCARBON DATA

A perusal of the first ten volumes of Radiocarbon demonstrates conclusively that travertine is a suspect material. It is difficult by a statistical manipulation of the data compiled there to arrive at a realistic estimate of the reliability of C^{14} ages of travertine, however, as not all workers who have dealt with the results of such determinations have been aware of the uncertainties. Nor is it simple to categorize the different types of fresh-water carbonate deposits as to material or environment of deposition, as each sample represents a problem unique in itself, and not all descriptions of dated samples are sufficiently comprehensive. Assessments of probable validity of individual ages by those who submitted the samples are not entirely reliable, as the most-often-quoted criterion for validity is consonance with stratigraphy, which in many cases has been itself established on the basis of C^{14} ages of travertine.

In the brief discussion that follows, available C^{14} determinations are considered under the following arbitrary categories:

1. CO_2 in ground water.
2. Carbon in vegetation and $CaCO_3$ in travertine at spring orifices or in wells.
3. $CaCO_3$ in caliche zones in soils.
4. $CaCO_3$ deposited as dripstone in limestone caves.
5. $CaCO_3$ deposited as travertine around the margins of pluvial lakes.

Ground water

Of 265 C^{14} analyses of CO_2 in ground water in the southwestern United States, Germany, Venezuela, and Saudi Arabia, fully 247 had C^{14} activities lower than that of modern water (normalized to prebomb C^{14} activity). Eight samples had activities equivalent to modern water, within limits of experimental error. Anomalously high C^{14} concentrations, presumably the result of contamination by bomb carbon, were reported for six.

Springs and wells

Measurements of C^{14} activities of aquatic vegetation growing in springs indicate that spring vegetation extracts much of its CO_2 from the water and typically yields apparent ages that are anomalously old, but often somewhat less so than the water itself. Only a very few ages have been determined for travertine being precipitated in or near the actual spring orifice, and all of these have yielded apparent ages somewhat older than predicted by the estimated stratigraphic age of the spring. A crude correction of 2,500 years was subtracted from travertine ages at the Tule Springs, Nevada, site, and the counting error was arbitrarily increased to $\pm$ 1,000 years "to allow for the uncertainty in C^{14} activity at zero age" (Radiocarbon, v. 6, p. 319; UCLA-503, UCLA-641, UCLA-642). While it was apparently felt that a correction of such magnitude was reasonable, there was no independent determination of its validity.

Carbonate scraped from a wooden bathtub, patently of historic age, at the orifice of a hot spring at Bridgeport, California, contained only 2.0 $\pm$ 0.8% of the activity of the oxalic-acid standard (ibid. v. 3, L-487P). A sample of travertine-coated twigs from a hot spring in Idaho yielded apparent ages of >39,000 (ibid. v. 6, W-1225) for the travertine and <200 (ibid. v. 6, W-1226) for the wood. Lime crust collected from living aquatic vegetation in Queechy Lake, Connecticut, was found to be anomalously old (ibid. v. 3, Y-754B), as was the pond vegetation itself (ibid. v. 3, Y-754A). Four tufa samples from Panamint Valley, California, were separated into organic and inorganic fractions, and each fraction was dated separately (ibid. v. 8; UCLA-1110-I, UCLA-1110-0, UCLA-1118-I, UCLA-1118-0, UCLA-1121-I, UCLA-1121-0, UCLA-1123-I, UCLA-1123-0). In each case, the age of the inorganic portion was significantly older than that of the organic portion.

Caliche

Numerous C^{14} ages are available from carbonate in caliche soils in the southwestern United States; some such ages have been interpreted in terms of local Pleistocene history (e.g. Rightmire, 1967), with recognition of the fact that the actual mechanism of formation of caliche is not well understood. Enough is known of the post-formation behavior of caliche to testify to its vulnerability to contamination by older carbon derived from limestone bedrock in rising ground water and by younger carbon carried into the C_{ca} zone by percolating rain water. A number of localities (e.g. Radiocarbon, v. 5, A-248, A-249; A-272, A-273; A-277, A-278, A-279; Rightmire, 1967) indicate that the apparent age of caliche is proportional to depth below the ground surface; this supports the hypothesis that caliche originates by precipitation from descending rain water which derives its carbon-isotope spectrum from CO_2 in the atmosphere and in the soil air. Correction of apparent C^{14} ages after determination of the relative abundance of the stable carbon isotopes (see discussion p. 6) has led to adjusted C^{14} ages of caliche horizons that can be applied with some confidence to the solution of Pleistocene stratigraphic problems (e.g. Rightmire, 1967).

Caves

Dates from travertine (dripstone) deposits in limestone caves are prone to similar uncertainties; two studies of the rate of accretion of annular layers of cave stalactites and stalagmites (Broecker, Olson, and Orr, 1960; Geyh and Schillat, 1966) defined rates of precipitation of cave travertine by measurement of the relative C^{14} activity of successive accretionary layers, but only maximum ages of the cave features were cited, as the authors felt that contamination by old carbonate had been significant. In a similar study of rate of accumulation of dripstone in a cave in France (LaBeyrie, DuPlessy, DeLibrias, and LeTolle, 1967), correction for initial concentration of C^{14} was attempted, and yielded adjusted C^{14} ages consistent with a climatic history deduced from other data. A sample of dripstone carbonate encrusting a piece of lumber carried into Crystal Cave, California, by miners around the year 1880 was found to be anomalously old (Radiocarbon v. 3, L-451C). A C^{14} date of the lumber itself was essentially modern, as expected.

Pluvial lakes

A comprehensive study by Broecker and Kaufman (1965) of the radiocarbon chronology of Lakes Lahontan and Bonneville II, in the Great Basin of the southwestern United States, has confirmed the validity of the radiocarbon-based chronology (Broecker and Orr, 1958) by demonstrating the essential identity of values so obtained with uranium-series isotope ages of materials in equivalent stratigraphic contexts. Measurements of C^{14}/C^{12} ratios in fish, mollusks, and subaqueous plants taken alive from various Great Basin lakes and from their supply rivers (Broecker and Walton, 1959; Broecker and Kaufman, 1965, p. 539; see also Thurber and Broecker, 1969), and comparison of such ratios with that of atmospheric CO_2, led to the conclusion that carbonates formed in Lakes Lahontan and Bonneville had C^{14}/C^{12} ratios of $.94 \pm .06$, representing an apparent age of 500 years for lacustrine carbonate at the time of its precipitation. The chronology presented by Broecker and Kaufman (1965) is based on C^{14} ages so corrected, and as presented affords a wholly reasonable correlation with the chronology of glaciation of the North American mid-continent region.

Summary

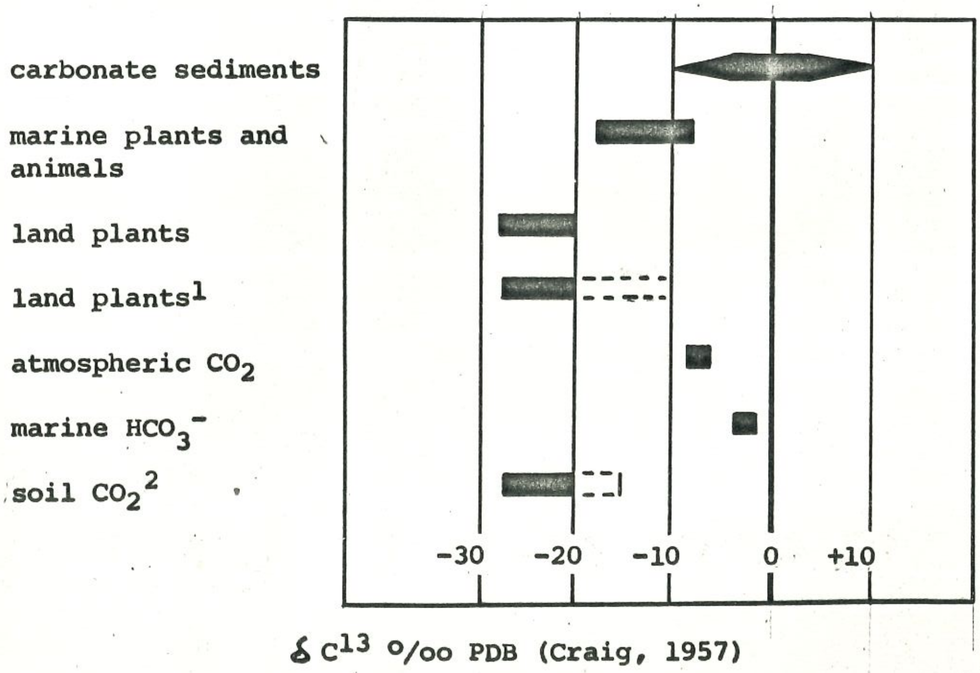
With the exception of the several studies cited above, most dating of travertine that has been undertaken in the past has been of single specimens whose ages, it was hoped, would offer significant documentation to specific sites. It is apparent that the reliability of C^{14} ages of travertine is related to the length of time the water has been in contact with the atmosphere before precipitation takes place. Thus, C^{14} ages of travertine deposited adjacent to spring orifices are typically anomalously old, but ages obtained from travertine deposited along the margins of pluvial lakes can, with minor corrections, be used to document a realistic Pleistocene stratigraphy.

STABLE CARBON ISOTOPES

Numerous measurements of C^{13}/C^{12} ratios in natural materials are now available:

TABLE 1

C^{13} abundances in various natural materials (after Craig, 1953; Ingerson and Pearson, 1964; Friedman, et al, 1970)



[1]Rightmire, C.T., pers. comm. 1970; Craig, 1954.
[2]Rightmire, C.T., 1967; pers. comm. 1970.

The consistency of the measured C^{13}/C^{12} ratios for marine limestone and for land plants has led to the technique used successfully by Pearson (1966 a, b, Pearson and White 1967, Ingerson and Pearson 1964), among others, to approximate the proportion of the carbon in ground water that is derived from exchange with ancient carbonate. When the C^{14} activity is normalized to the equivalent of full concentration of atmospheric CO_2 in the sample, realistic water ages are obtained which, when divided by the distance between recharge area and sampling station, give rates of ground-water movement that are consistent with those obtained by more unwieldy, conventional methods. Direct measurements of C^{13}/C^{12} ratios in soil air (Rightmire, 1967; pers. comm. 1970) indicate that a realistic δC^{13} value for soil CO_2 might be closer to -17 than to -25, particularly in arid or semi-arid environments. The value of -25, used by most workers, is based on C^{13}/C^{12} ratios in land plants grown under normal (i.e. humid) conditions, on the assumption that no fractionation of the carbon isotopes occurs during root respiration or during oxidation of dead vegetable material, and on the δC^{13} value of -23.2 measured by Broecker and Olson (1960) on CO_2 from a humid soil. The correction applied by Pearson (1966) does not correct for the age of the water, of course, since that is the desired information, but the age of the water represents a significant source of contamination of travertine ages.

Once it appears at the Earth's surface as a flowing stream, ground water, presumably of low C^{14} activity relative to the atmosphere and of C^{13}/C^{12} ratio intermediate between that of marine limestone and soil CO_2, begins to mix with the atmosphere and to exchange its CO_2 with atmospheric CO_2, with a δC^{13} value of $\sim$-7. A travertine sample precipitated from such a solution, then, would have a δC^{13} somewhere between 0 and -25, but a value of -7 would be no assurance that total equilibration had taken place. Furthermore, the demonstration (Craig, 1953) that $C^{12}O_2$ is preferentially extracted from the atmosphere at the water surface adds yet a fourth unknown to a situation describable, at best, in terms of one vague expression. Additional, possibly biogenic, fractionation might occur in the process of precipitation.

Thus it seems unlikely that a study of the stable isotopes of carbon in a travertine sample will, in itself, determine the extent to which the spring-derived surface water from which the travertine was precipitated had equilibrated with the atmosphere, and even when supported by measured C^{14} activity can not be used to demonstrate conclusively that the carbon-isotope spectrum in the water was the same as that of the atmosphere at the time that a given specimen of $CaCO_3$ was precipitated.

MODERN ANALOGS

No systematic attempt has been made to measure C^{14} activity in travertine being deposited in streams under modern conditions, except to establish that apparent C^{14} ages are considerably older than ages of the samples established from other data (see p. 3). Any such study would, of course, be enormously complicated by the anomalously high and locally variable levels of C^{14} activity added to the contemporary atmospheric carbon reservoir as a result of neutron radiation from atomic bomb explosions made during the last 20 years.

An appropriate setting in which to examine the equilibration of surface water with atmospheric CO_2 would be one in which travertine precipitated prior to 1950 could be independently dated by some means other than analysis of the isotope spectrum of the carbonate. The difference between the independently determined date and the C^{14} age of the travertine, then, would be a measure of the disequilibrium at any point. Such an independent calibration of travertine ages can be achieved in Havasu Canyon, Arizona.

HAVASU CANYON, ARIZONA

Geologic setting

Havasu Creek, a tributary of the Colorado River on the South Rim of the Grand Canyon (map, fig. 1) rises from springs near the base of the Supai Formation of Permian age. The creek is 15-30 feet wide, and falls 1,450 feet in the course of the 10 miles it flows to the Colorado (long profile, fig. 2).

Precipitation of travertine begins in Havasu Creek $\sim 2\frac{1}{2}$ miles downstream from the springs, as a result of turbulence which drives off CO_2 and renders $CaCO_3$ less soluble. Travertine first appears on boulders lying on the stream bed, along the shores of the stream where turbulent eddies form, and on twigs, leaves, and roots of streamside trees that disturb the water's surface. Precipitation

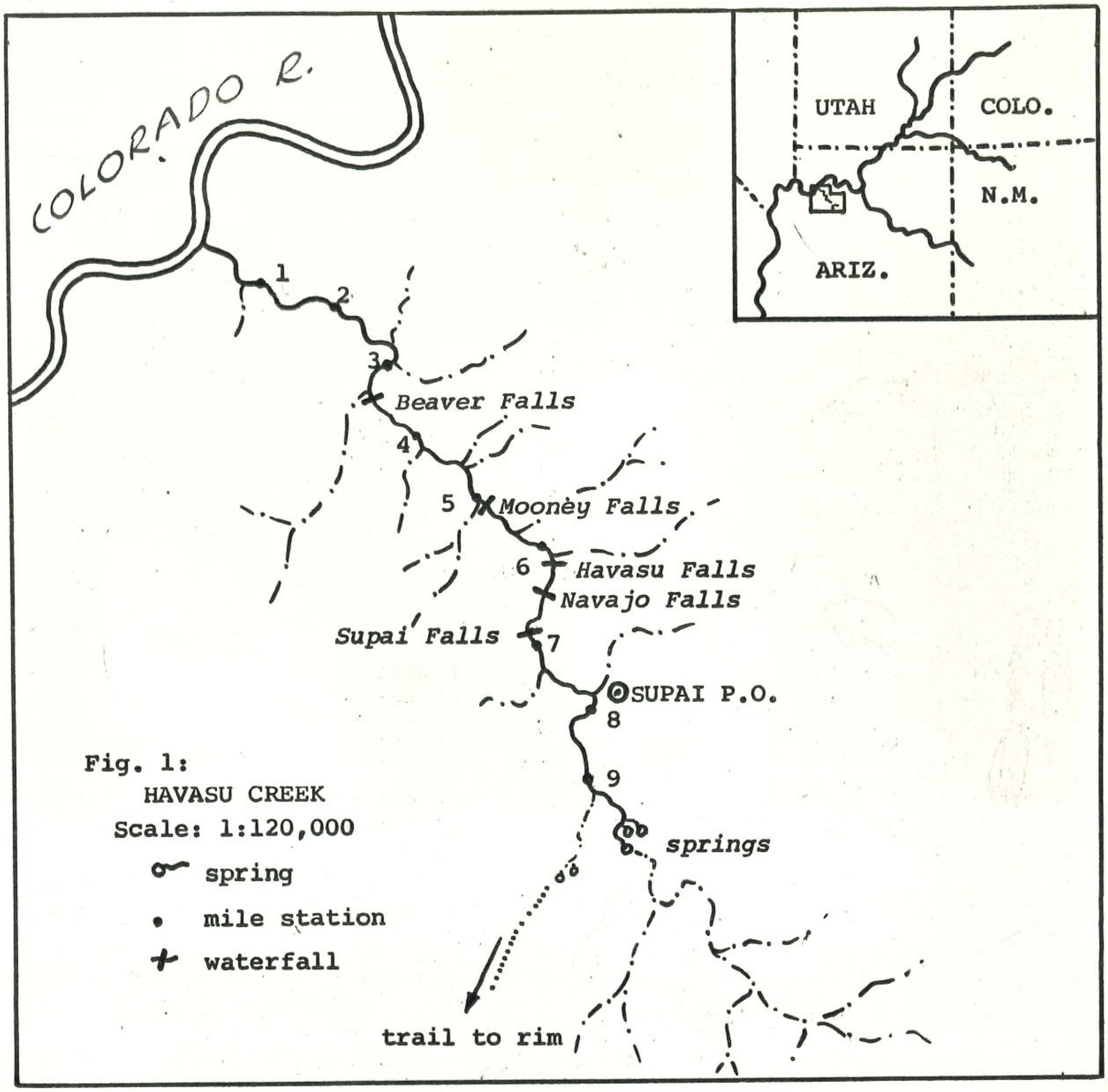


Fig. 1:
HAVASU CREEK
Scale: 1:120,000

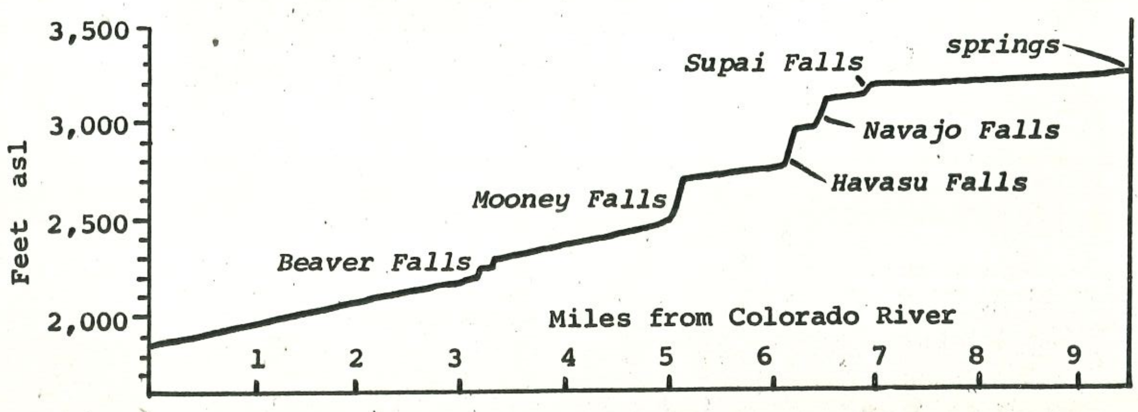


Fig. 2: Long Profile, Havasu Creek

of travertine as a result of the turbulence created by such protrusions typically results in increased turbulence which enhances further precipitation. Massive deposits of travertine thus accumulate which typically assume the form of arcuate dams which may come to span the width of the stream in a festooning, step-like configuration. The creek consists of a series of pools behind dams which may grow to 20 feet in height, and are remarkably resistant to the stress imposed by the water rushing over them.

The growth of a travertine dam may divert Havasu Creek to a new course, in the process flooding part of the bank. Continued upward growth of a dam may eventually render it architecturally unstable and it may be undermined, perhaps during a time of flood runoff when the water is briefly undersaturated with respect to $CaCO_3$. By whatever mechanism, it is apparent that Havasu Creek is continually altering its course within the confines of its narrow, steep-walled valley, and is continually inundating sections of its banks. Trees growing on the banks are thus drowned, are coated with travertine around submerged parts of their trunks, and eventually fall, often into the creek, where they may become coated with travertine and sink to the floor of one of the pools, or may float downstream to become lodged in one of the dams, to which they are rapidly cemented by the continuing process of precipitation. Many pools are floored with a dense mat of tightly intertwined, travertine-coated branches, and some dams fairly bristle with wood fragments, securely locked in the travertine, and crudely identifiable as to relative age by their stratigraphic position in the wall of the dam.

At least two generations of older travertine, presumably precipitated under similar conditions in the past, have been identified; contemporary wood has been observed only in the younger of the two.

RESEARCH PROGRAM

This study will include the following procedures:

1. Samples of travertine-coated trees, killed before the beginning of atmospheric bomb tests, will be collected at frequent intervals along the length of the creek. Both the outermost layer of wood and the adjacent layer of travertine will be dated by C^{14}, thereby establishing at each station the extent of contamination of the $CaCO_3$ by old carbon. This will be done for the modern generation of travertine, and for the older material, if possible.
2. If it can be shown that trees in the canyon respond to seasonal or yearly variation in precipitation, despite the abundant ground water, an attempt will be made independently to fix the age of the wood samples by dendrochronology. This study will be used to approximate the extent of alteration of wood ages as a result of incorporation of carbon from ground water in tree metabolism.
3. A measure of the possible depletion of C^{14} activity in the wood by incorporation of old carbon will be established by dating wood of known age collected from full sections cut from living trees. This information will complement the data obtained from procedure

2, and will represent the only empiric measure of total wood contamination if procedure 2 is unsuccessful.

4. Selected samples of travertine will be dated by thermoluminescence; it is anticipated that modern travertine may be too low in radioactive substances to have stored much thermoluminescence, but the technique may be effective for the older travertine, particularly for the oldest generation in which no wood has been observed.

5. C^{13}/C^{12} analyses of dated samples will be conducted routinely to correct C^{14} ages for fractionation. The carbon-isotope spectrum of selected specimens will be considered critically in an attempt to establish 1) the extent of contribution of carbon from the several sources, and 2) the nature and extent of fractionation during equilibration and/or precipitation.

6. A detailed stratigraphic study of the travertine sequence in Havasu Canyon and in nearby tributaries of the Grand Canyon will be undertaken. Samples for C^{14} dating will be collected outside Havasu Canyon from localities judged likely to yield meaningful ages.

Discussion

It is anticipated that the discrepancy between the age of the wood and the age of the travertine will decrease systematically with distance from the source of the creek, as a function of exchange of CO_2 between the water and the atmosphere. The distance from the springs at which the two values are first identical within limits of experimental error, if it ever occurs, thus empirically defines the distance water must flow in Havasu Creek before equilibration with the atmosphere is achieved. If total equilibration doesn't occur before the Colorado River is reached, an extrapolation of the observed convergence will be made to approximate the distance from the source at which equilibration should be complete. It will be instructive to compare such a convergence with a similar equilibration curve for the older travertine, if such a curve can be established.

PRELIMINARY RESULTS

A brief reconnaissance of Havasu Creek was carried out by the principal investigator in May and June of 1969, that was supported in part by Sub-Grant NSF-1G-69-10 of an NSF Institutional Grant awarded to the University of Pennsylvania. At that time the mechanism of precipitation of travertine was examined, a tentative sequence of earlier (Pleistocene?) travertine events was recognized, and 80 samples of wood coated with travertine were collected from both modern and ancient travertine features at frequent intervals between mile 7.2, the start of precipitation of travertine, and mile 0.47, the site of the most downstream sample of wood observed (because of the difficulty of establishing the point of origin of Havasu Creek, samples were identified by mile numbers measured from the Colorado River in an upstream direction).

An attempt was made independently to fix the wood ages by dendrochronology, but it developed that the cottonwood trees, which make up the great majority of wood samples in the creek, show no systematic variation in width of rings as a function of climate. Mesquite seems to be more responsive to yearly variation in available moisture, but so far only two of the samples collected are mesquite.

Full sections of both a living cottonwood and a living mesquite were collected and will be dated.

<u>Preliminary C^{14} results</u>

C^{14} activity measured in two samples collected in 1969 is presented in Table 2, along with measured C^{13}/C^{12} ratios. It is fortunate that the wood is too old to have been affected by bomb carbon, and too young to fall within the range of uncertain values around 240-350 B.P. (Michael and Ralph, 1969, Stuiver and Suess, 1966). The travertine ages indicate that equilibration is proceeding with distance downstream, but too slowly to achieve total equilibration in the creek. Until more dates are available, however, these two samples simply indicate that 1) the wood is old enough to be reliable, 2) the water is anomalously old, and 3) the convergence "curve" is in the right direction.

It is difficult to interpret the significance of the C^{13}/C^{12} ratios without additional data. Each of the travertine samples falls within the range of values expected for marine carbonate, suggesting that no equilibration has occurred, but the difference in the C^{14} ages indicates that equilibration is taking place. Minor fractionation of the carbon isotopes during either equilibration or precipitation could alter C^{13} values by as much as 25 $^{o}/oo$, but would not significantly alter the C^{14} ages, as the corrected values in column 6 of Table 2 indicate.

TABLE 2: C^{14} Dates for Havasu Canyon, Arizona

(5,568 half-life; B.P. 1950)

Column No. 1	2	3	4	5	6
Univ. of Penn. Lab. No.	Sample No. and material	C^{14} date (years B.P.)	C^{13} Oak standard (mils)	C^{13} Lime standard (mils)	C^{14} Date Corrected for C^{13} (Oak St'd.)
P-1631	69-HC-0.47-W1 Cottonwood	196 ± 48	-0.9		181
P-1641	69-HC-0.47-T1 Travertine	13,034 ± 296	+25.2	-1.8	13,443
P-1632	69-HC-5.97-W1 Mesquite	137 ± 42	+6.5		242
P-1642	69-HC-5.97-T1 Travertine	17,006 ± 569	+25.9	-0.9	17,414

GOALS OF THE PROJECT

The goals of the proposed study are the following:

1. To characterize the nature and extent of carbon-isotope fractionation in the exchange of CO_2 between the atmosphere and surface water, and in the precipitation of $CaCO_3$ from such water.
2. To approximate the rate of equilibration of surface water with atmospheric CO_2, at least for Havasu Creek, and possibly for other localities.
3. To place limits on the reliability of C^{14} ages of travertine precipitated from flowing surface water.
4. To apply guidelines so established to a program of dating of travertine within Havasu Canyon, and to contribute thereby to the growing understanding of the history of the Grand Canyon by presenting a dated sequence of travertine-precipitating events during late-Plesitocene time.

Guidelines established in the course of this project may be of value in dating ancient travertine deposits, sediments indicative of greater ground-water flux than is generated by the present hydrologic cycle, that are significant features of desert and semi-desert environments throughout the world.

FACILITIES

RADIOCARBON LABORATORY

The Radiocarbon Laboratory at the University of Pennsylvania was established in 1951 in the University Museum. In 1956 it was moved to the David Rittenhouse Laboratory (Department of Physics) and two gas proportional CO_2 counters were installed (in place of the original solid-carbon screen wall counter). The Laboratory has been supported mostly by the University of Pennsylvania, but the second counter (since 1958) has been supported by NSF grants. The chief function of the Laboratory has been the dating of archaeological samples that have been excavated by, or are of interest to, the curators of the University Museum. It now has the reputation of being one of the most precise and careful C-14 laboratories in the world.

The NSF supports the second counter for the dating of tree-ring-dated sequoia and bristlecone pine samples - for the determination of the causes and magnitudes of fluctuations in the atmospheric inventory of C-14 in past times (Ralph & Michael, 1967; Ralph & Michael, 1969; Michael & Ralph, in press).

These two projects occupy the counting facilities full-time and the Laboratory usually has more than a year's backlog of samples waiting to be dated. The chemical processing train, however, is adequate to supply three counters. Therefore, with the addition of a third counter, 33 1/3% of the counting time could be devoted to geological samples.

THERMOLUMINESCENCE (TL)

Thermoluminescence in pottery is due to the fact that radiations from the traces of radioactive elements in pottery bombard the other constituents of the clays and raise electrons to metastable levels. When the pottery is heated, such as in firing, enough additional energy is supplied to enable each electron to fall back to its stable position and to emit a photon of light. On being reheated, the amount of thermoluminescence observed (as a glow curve) is, therefore, representative of the number of metastable excited electrons and hence of the time elapsed since the original firing of the pottery. The amount of metastable electron accumulation is also dependent upon the rate of radioactive bombardment and upon the susceptibility of the pottery to irradiation, as determined by an artificial dose. Three measurements are, therefore, involved - the glow curves of natural TL, of artificial TL (after being given a fixed dose of X-rays to account for varying susceptibilities), and of the inherent radioactivity.

Initial experiments directed toward the dating of pottery by thermoluminescence were started by Elizabeth Ralph at the University of Pennsylvania in 1959. The program has been pursued more actively by Mark Han since 1962, during which time it has been supported by NSF grants. It has now been demonstrated that most types of fired pottery can be dated by TL within a precision of $\pm 10\%$.

Our glow curve apparatus, even though designed for the measurement of pottery, is extremely sensitive and flexible, and can readily be adapted for use with other thermoluminescent materials. One of the best features of our linearly programmed heating-control system is the Data-Trak Model 5300 Programmer (made by R. I. Controls). By means of inexpensive changeable charts (made of a conducting material) any program can be drawn. Our present program for pottery dictates that heating is first at the rate of 6.5^{o}C/sec up to 170^{o}C, then followed by a faster rate of 25^{o}C/sec up to 500^{o}C. The flexibility of this programming unit makes it much more suitable for these measurements than other types currently in use by other workers. Another unit - the Model 417 Picoammeter (Keithley Instruments) - with a dark current suppresor has enabled us to use much greater sensitivity when required.

Comparisons were made recently with our assembly of units and a "packaged" commercial thermoluminescence reader - namely, the Harshaw Model 2000A TL Detector with the Model 2000B Automatic Integrating Picoammeter. Even though this latter apparatus is rated to have a maximum sensitivity of 10^{-12} amperes, its actual response was only equivalent to 10^{-8}A on our own apparatus. Since our assembly has a usable sensitivity of 10^{-9}A for lower light intensity measurements, it appears to be some ten times more sensitive than this highly-regarded commercial instrument,

It is known that limestones emit 100 times more light (upon heating) than potsherds. However, the extra sensitivity and flexibility of our system may be useful for experiments with travertines, especially when applied to very young samples in which not much alpha decay has occurred.

The glow curve aparatus is available for part-time use, but the three alpha counters (used to measure the inherent radio-activity due to uranium and thorium) are used continuously due to the long counting times required. Therefore, it would be necessary to purchase an additional alpha counter and associated components for the experiments with travertines.

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Michael, H.N., and Ralph, E.K., Correction factors applied to Egyptian radiocarbon dates from the B.C. era: Proceedings of Nobel Symposium XII, Uppsala, August 1969, in press.

Pearson, F.J., Jr., 1966, Use of C^{13}/C^{12} ratios to correct radiocarbon ages of materials initially diluted by limestone: Proc. Sixth Int'l. Conference, Radiocarbon and Tritium Dating, p. 357-366.

__________________, 1966, Ground-water ages and flow rates by the carbon-14 method: Ph.D. diss., Univ. of Texas, Austin, Texas.

Pearson, F.J., Jr., and White, D.E., 1967, Carbon 14 ages and flow rates in Carrizo Sand, Atascosa County, Texas: Water Resources Research, v. 3, p. 251-261.

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Ralph, E.K., and Michael, H.N., 1967, Problems of the radiocarbon calendar: Archaeometry, v. 10, p. 3-11.

______________________________, 1969, University of Pennsylvania radiocarbon dates XII: Radiocarbon, v. 11, no. 2, p. 469-481.

Rightmire, C.T., 1967, A radiocarbon study of the age and origin of caliche deposits: M.S. diss., Univ. of Texas, Austin, Texas.

Stuiver, Minze, and Suess, H.E., 1966, On the relationship between radiocarbon dates and true sample ages: Radiocarbon, v. 8, p. 534-540.

Thurber, D.L., and Broecker, W.S., 1969, The Geochemistry of dissolved carbon in surface waters of the Great Basin: (abs. V100) Trans. American Geophys. Union., v. 50, no. 4, p. 347.

National Science Foundation

BUDGET	Funds Requested 1970-71	1971-72	University Contribution 1970-71	1971-72
A. Salaries and Wages				
1. Faculty:				
Robert F. Giegengack, Asst. Prof., Geology, Co-Principal Investigator. $1,333 per month (First year, full time 2 months summer)	2,666			
(2nd year, full time 1 month summer; 1/9 time 9 months academic year)		2,666		
(1/9 time 9 months academic year)			1,333	1,333
Elizabeth K. Ralph, Faculty Associate, C^{14} and Thermoluminescence, Co-Principal Investigator (20% of full time, 12 months)			2,800	2,800
2. Other Professional and Technical:				
Barbara Lawn, Research Specialist II, C^{14}. (33 1/3% full time, 12 months)			2,700	2,700
Mark C. Han, Research Chemist, Thermo-luminenscence (10% full time, 12 months)			950	950
Chemical Electronics Technician (Full time, 12 months)	7,150	7,150		
2 Graduate Students @ $3,500 9 months academic year; $1,200 summer	9,400	9,400		
2 Undergraduate Student employees $1,200 year, 12 months	2,400	2,400		
Physics Department:				
1. Electronics Shop	500	500		
2. Machine Shop	1,000	200		
3. Glass Blower	300	300		
4. Mass Spectrometer Technician for C^{13}/C^{12} analysis (50 hours/year, $9/hour)	450	450		
Geology Department:				
1. Mass Spectrometer Technician (½ time, 12 months)	4,200	4,200		
3. Non-technicial:				
Secretary, Geology Department (¼ time, 12 months)	1,500	1,500		
TOTAL SALARIES	$29,566	$28,766	$ 7,783	$ 7,783
B. Employee Benefits (10.2% of above)	3,016	2,934	794	794
	$32,582	$31,700	$ 8,577	$ 8,577

Budget, continued - 2

	Funds Requested 1970-71	Funds Requested 1971-72	University Contribution 1970-71	University Contribution 1971-72
C. Permanent Equipment				
1. C^{14} Equipment:				
1 Model GEC-12 Electronic Console Johnston Labs.	$11,975			
1 CO_2 Counter	1,500			
30 G-M Counters - anti-coincidence	1,500			
9 tons steel shield: 1 ton cold-drawn blocks; 8 tons hrs plates + I beam	3,000			
Transformers, circuit breakers, etc.	300			
Valves and Cu tubing - filling line	500			
Temperature Indicator	500			
10 Steel CO_2 storage cycls.	700			
Vacuum pump incl. Hg diff pump and Hg	600			
Pirani gauge	500			
2. Thermoluminescence Equipment:				
Components for additional Alpha Counter				
1 Scaler	1,500			
1 Amplifier and 1 HV Supply	1,000			
1 Preamp and Photomultiplier	500			
TOTAL EQUIPMENT	24,075			
D. Supplies and Expendable Equipment				
1. Research Supplies for C^{14} Lab (Liquid Oxygen, Glassware, Chemicals, Dry Ice, Minor Electronic Components)	5,000	5,000		
2. Office Supplies for C^{14} Lab	300	300		
3. Expendable Field Equipment (Tents, Stoves, Packs, First Aid equipment, etc.)	600			
4. Film and Processing	200	250		
TOTAL SUPPLIES	6,100	5,550		
E. Travel and Field Expenses				
1. Use of private vehicle 2 field seasons @ 10,000 miles/season	1,000	1,000		
2. Support in Field 3 persons for 50 days/season @ $10/day/season	1,500	1,500		
3. Local Air Reconnaissance 20 hours/season @ $20/hour	400	400		
4. Hire of Riding and Pack Animals	260	260		
5. Travel to scientific meetings, to other laboratories (e.g., Tree-Ring Laboratory of the University of Arizona) and to other travertine localities.	750	1,250	500	500
TOTAL TRAVEL	3,910	4,410	500	500

Budget, continued - 3

	Funds Requested 1970-71	Funds Requested 1971-72	University Contribution 1970-71	University Contribution 1971-72
F. Publication Costs (Drafting, Page Charges)		500		500
G. Total Direct Costs	66,667	42,160	9,077	9,577
H. Indirect Costs				
1. On Campus - 34% of salaries excluding secretarial	7,820	8,001	2,646	2,646
2. Off Campus - 20.2% of summer salaries of R. Giegengack and graduate students	1,023	754		
I. Total Project Cost	$75,510	$50,915	$11,723	$12,223

CURRICULUM VITAE

Robert Francis Giegengack, Jr.

BORN: November 27, 1938

MARRIED: May 14, 1967

EDUCATION:

B.A. Yale University 1960; Geology major.
M.S. University of Colorado 1962
Thesis: Recent volcanism near Dotsero, Colorado
Ph.D. Yale University 1968
Thesis: Late-Pleistocene History of the Nile Valley in Egyptian Nubia.

POSITIONS:

Teaching Assistant, University of Colorado 1960-62

Teaching Assistant, Yale University 1962-63, 1965-66

Geologist, Yale University Prehistoric Expedition to Nubia 1963-1967

1963-66: Field work in Africa, including:
1) Study of the Pleistocene history of the Mile Valley in Egypt and the Sudan.
2) Study of coastal morphology and Pleistocene stratigraphy on the Egyptian coast of the Red Sea.
3) Study of modern and fossil soils in Kenya, Tanzania, and Uganda, with particular emphasis on the occurrence, distribution, and modes of transportation of various forms of iron.

Acting Instructor, Yale University 1966-67

Assistant in Research, Peabody Museum of Natural History, Yale University; Research Associate and Field Director, Third Yale North India Expedition. 1967-68

Field investigation of the age and stratigraphic position of specimens of the genera Dryopithecus and Ramapithecus recovered from late-Tertiary sediments in the Siwalik hills of northwest India.

Assistant Professor of Geology, University of Pennsylvania 1968-

PUBLICATIONS:

Giegengack, R.F., 1962, Volcanism near Dotsero, Colorado: Geol. Soc. America, Rocky Mountain Section, Program, 1962 Annual Meeting, p. 22 (abs.)

______________, 1969, Late-Pleistocene history of the Nile Valley in Egyptian Nubia: VIII^e Congres INQUA, Paris 1969, Resume des Communications, p. 202 (abs.).

Veeh, H.H., and Giegengack, Robert, 1970, Uranium-series ages of corals from the Red Sea: Nature, v. 226, p. 155-156

October, 1969

CURRICULUM VITAE

NAME: ELIZABETH K. RALPH

BORN: February 5, 1921. Trenton, New Jersey.

RESIDENCE: Box 357, Woosamonsa Road, Pennington, New Jersey.

EDUCATION: Wellesley College, B.A., 1938-1942.
University of Pennsylvania, M.S., 1949-1951.

EXPERIENCE: Junior Electronics Engineer, then Chemist, then Assistant to Chief Radio Engineer, then Project Engineer at Foote, Pierson and Company and Kearfott Manufacturing Company, Newark, New Jersey, 1942-49.

University of Pennsylvania
Research Assistant, Department of Physics, 1951-55.
Associate, Department of Physics, 1955-

University of Pennsylvania, the University Museum
Associate Director, Applied Science Center for Archaeology, 1962-

MEMBERSHIP IN SCIENTIFIC ORGANIZATIONS:

American Association for the Advancement of Science
Sigma Xi, Chapter member, 1956-
Archaeological Institute of America, Phila. Chapter, 1962-
American Geophysical Union, Member, 1964-
Philadelphia Anthropological Society

FIELD WORK: In charge of field tests of instruments for archaeological exploration and the search for the buried city of Sybaris, Italy. 1962-67.

Additional archaeological field surveys with instruments for underground exploration:

1963 - Navan Fort, N. Ireland; Fort Louisbourg, Nova Scotia, Canada; Fort Loudon, Pa.; Caleb Pusey House, Chester, Pa.; Hagley Mills, Wilmington, Del.

1964 - Fort Lennox, Ile-aux-Noix, Quebec Province, Canada; Hagley Mills, Wilmington, Del.

1965 - Gordion, Turkey; Artena and Sele, Italy; Hope Lodge, Whitemarsh, Pa.; Camden, South Carolina; Snaketown, Tucson, and San Xavier, Arizona.

1966 - Gravina, Veii, Sele, Artena, and Metapontum, Italy; Helice and Porto Cheli, Greece; Harvard Forest, Petersham, Mass.

FIELD WORK, CONTINUED:

1967 - Siris, Italy; Thera and Elis, Greece.

1968 - Elis, Greece; Dun Ailinne, Ireland; St. Croix Island, Calais, Maine; Morton Mortonsen House, Norwood, Pa.

1969 - Ciro and Cosa, Italy; Divostin and other Neolithic sites, Yugoslavia.

PUBLICATIONS:

1955a University of Pennsylvania Radiocarbon Dates I. Science, Vol. 121, No. 3136, pp. 149-151, Washington, D.C.

1955b Radiocarbon Dates for Kara Kamar, Afghanistan, University of Pennsylvania II. Science, Vol. 122, No. 3176, pp. 921-922 (with C.S. Coon). Washington, D.C.

1956 C-14 Dating. Pennsylvania Archaeologist, Vol. XXVI, No. 1, pp. 27-31.

1957 Age is No Longer a Secret. Wellesley Alumnae Magazine, Vol. XLI, No. 3, pp. 142-143. Massachusetts.

1959a Radiocarbon Dating in the Arctic. American Antiquity, Vol. 24, No. 4, pp. 365-374 (with F.G. Rainey). Salt Lake City.

1959b Double Trouble. Expedition, Vol. 1, No. 3, pp. 24-25. The University Museum, Philadelphia.

1959c University of Pennsylvania, Radiocarbon Dates III. American Journal of Science, Radiocarbon Supplement, Vol. 1, pp. 45-58. New Haven.

1960a Carbon-14 Measurements of Known Age Samples. Nature, Vol. 188, No. 4746, pp. 185-187 (with R.Stuckenrath, Jr.). London.

1960b New Radiocarbon Dates and the Maya Correlation Problem. American Antiquity, Vol. 26, No. 2, pp. 165-184 (with L. Satterthwaite). Salt Lake City.

1961a University of Pennsylvania, Radiocarbon Dates IV. American Journal of Science, Radiocarbon Supplement, Vol. 3, pp. 4-14 (with R. Ackerman). New Haven.

1961b C-14 Dates for Sites in the Mediterranean Area. American Journal of Archaeology, Vol. 65, pp. 357-367 (with E. Kohler). Princeton.

1961c Radiocarbon "Effective" Half-Life for Maya Calendar Correlations. American Antiquity, Vol. 27, No. 2, pp. 229-230. Salt Lake City.

1962a University of Pennsylvania Radiocarbon Dates V. Radiocarbon (supplement of the American Journal of Science), Vol. 4, pp. 144-159 (with R. Stuckenrath, Jr.). New Haven.

PUBLICATIONS, CONTINUED:

1962b New Instrument Techniques in Archaeology. Proceedings of the Symposium on Detection of Underground Objects, Materials and Properties; 19-20 March, 1962. U.S. Army Engineer Research and Development Laboratories, Fort Belvoir, Va., pp. 151-155 (with F.G. Rainey).

1962c Prospezioni Geofisiche. Relazione Sulla Campagna di Prospezioni, Aprile, Maggio, Giugno 1962, nella Piana del Fiume Crati. Relazione sulla Esplorazione Geofisica (Sibari), pp. 2-32. Fundazione Ing. C. M. Lerici del Politecnico, Milano.

1963 Search for a City Buried 2700 Years. Wellesley Alumnae Magazine, Vol. 47, No. 5, pp. 283-285, 310. Brattleboro, Vt.

1964a Comparison of a Proton and a Rubidium Magnetometer for Archaeological Prospecting. Archaeometry, Vol. 7, pp. 20-27. Oxford.

1964b A Spectrographic Investigation of Jar Handles Bearing the "Royal" Stamp of Judah. Archaeometry, Vol. 7, pp. 67-71 (with A. Millett and J. Pritchard). Oxford.

1965a Review of Radiocarbon Dates for Samples from Tikal Related to the Maya Calendar Correlation Problem. American Antiquity, Vol. 30, No. 4, pp. 421-427. Salt Lake City.

1965b The Electronic Detective and the Case of the Missing City. Expedition, Vol. 7, No. 2, pp. 4-8. The University Museum, Philadelphia.

1965c University of Pennsylvania Dates VII. Radiocarbon (supplement of the American Journal of Science), Vol. 7, pp. 179-185 (with H.N. Michael and J. Gruninger, Jr.). New Haven.

1965d University of Pennsylvania Dates VIII. Radiocarbon (supplement of the American Journal of Science), Vol. 7, pp. 187-199 (with R. Stuckenrath, Jr.). New Haven.

1965e Prospezioni Archeologiche per Sibari. Atti del Convegno di Sibari, 10-11 Ottobre, 1965, pp. 98-99. Ente Studi Economici Per la Calabria, Cosenza.

1965f Carbon-14 Date for the Antikythera Shipwreck. Transactions of the American Philosophical Society, N.S. Vol. 55, Pt. 3 (June), pp. 48ff. Philadelphia.

1966a University of Pennsylvania Dates IX. Radiocarbon (supplement of the American Journal of Science), Vol. 8, pp. 348-385 (with R. Stuckenrath Jr. and W.R. Coe). New Haven.

1966b Dating of Pottery by Thermoluminescence. Nature. Vol. 210, pp. 245-247 (with M.C. Han). London.

1966c Archaeology and Its New Technology. Science, Vol. 153, No. 3743 (September 23), pp. 1481-1491 (with F.G. Rainey). Washington D.C.

PUBLICATIONS, CONTINUED:

1966d Akdeniz Bölgesi Istasyonlarina ait C^{14} Tarihleri. ICOM Turkiye Milli Komitesi Haber Bülteni, No. 7, pp. 74-104 (with E. Kohler). Translation of 1961b (above) into Turkish by Ufuk Esin, published in the Bulletin of the Turkish National Committee of the International Council of Museums.

1967a Instrument Surveys. The Search for Sybaris, 1960-1965, F.G. Rainey and C.M. Lerici et al., pp. 53-124. Lerici Editori, Rome.

1967b Methodological Problems of C^{14} Dating. Archaeological Chemistry. A Symposium, M. Levey, editor, pp. 253-265. Philadelphia.

1967c Problems of the Radiocarbon Calendar. Archaeometry, Vol. 10, pp. 3-11 (with H.N. Michael). Oxford.

1968a Archaeological Surveying Utilizing a High Sensitivity Difference Magnetometer. Geoexploration, Vol. 6, pp. 109-122 (with F. Morrison and D.P. O'Brien). Amsterdam.

1968b Progress in Thermoluminescent Dating of Pottery. Thermoluminescence of Geological Materials, D.J. McDougall, editor, pp. 379-387 (with M.C. Han). New York.

1969a Archaeological Prospecting. Expedition, Vol. 11, No. 2, pp. 14-21. The University Museum, Philadelphia.

1969b University of Pennsylvania Radiocarbon Dates XII. Radiocarbon, Vol. 11, No. 2, pp. 469-481 (with H.N. Michael).

in press Potential of Thermoluminescence in Supplementing Radiocarbon Dating. World Archaeology, September, 1969 (with M.C. Han). England.

in press Potential of Thermoluminescence in Supplementing Radiocarbon for Dating in the Middle Ages. Applications of Science to Medieval Archaeology, International Conference at the University of California at Los Angeles, October 26-28, 1967. UCLA Center for Medieval and Renaissance Studies and Isotope Laboratory, Institute of Geophysics and Planetary Physics, UCLA (with M.C. Han).

in press Potential of Thermoluminescence Dating. Papers of the annual meeting of the American Chemical Society, Atlantic City, New Jersey, September, 1968 (with M.C. Han).

in press Correction Factors Applied to Egyptian Radiocarbon Dates from the B.C. Era. Proceedings of Nobel Symposium XII. Uppsala, August 11-15, 1969 (with H.N. Michael).

STATEMENT CONCERNING FACILITIES FOR PROPOSED RESEARCH PROJECT

This form must accompany all proposals which are to be submitted to the Vice Provost for Research or appropriate Vice-President for approval. The following statements must be completed:

Title: Empiric approach to measurement of rate of equilibration of surface water with atmospheric CO_2.

1. The work to be undertaken will be performed in.

(Building)	(Room No.)	(Size)	(Type i.e., office, lab, etc.)
Hayden Hall	115	12 x 17	Office
Rittenhouse Lab	BW4 BW6	ea. 20 x 20	lab (low-level counting)

2. Alterations required to place the above areas in proper condition for the research work are: (If none, so indicate.) Work described in this proposal can be performed temporarily in space available; more space will be needed eventually in DRL*

3. The estimated cost of the work outlined in No. 2 is $13,000. (This figure must be obtained from Bldgs. & Grds. or the Hospital Administration. The Office of Research Administration will assist the investigator in securing this information.)

* approval to utilize space adjacent to BW6 is in hand; funds are needed to adapt space for low-level counting.

4. The Cost of these alterations will be paid for from:

Contract or Grant Funds	____	Check appropriate block or blocks
Department Funds	____	
University Appropriation	____	
Other (explain)	x	(source unknown)

5. The equipment required to undertake the proposed work is:

Available within the department	____	Check appropriate block or blocks
Will be purchased with contract funds	x	
Will be purchased with dept. funds	____	
Will be secured by other means (explain)	____	

I have presented the above to both the Building Administrator and the Dean or Director concerned. Both approve.

[signature]
(Principal Investigator)

APPROVED

____
(To be signed by administrator of budget to be charged with cost of alterations or equipment)

If the proposed project will require space beyond that currently used by a department approval of the Business Vice-President or his representative must appear below:

APPROVED: ______________________________
Business Vice-President

NATIONAL SCIENCE FOUNDATION
WASHINGTON, D. C. 20550

RESEARCH GRANT BUDGET SUMMARY

INSTITUTION	PRINCIPAL INVESTIGATOR(S)	PROGRAM NAME	DURATION	GRANT NUMBER
Univ. of Pennsylvania	R. Giegengack	Earth Sciences	2 years	

	NSF Funded Man Months			Grantee Man Months			NSF GRANT	GRANTEE SHARE
	Cal	Acad	Sum	Cal	Acad	Sum		
A. SALARIES AND WAGES								
1. Senior Personnel								
a. 1 (Co) Principal Investigator(s)	4	1	3	2	2		$ 5,332	$ 2,666
b. ___ Faculty Associates				4.8	4.8			5,600
Sub-Total							5,332	8,266
2. Other Personnel (Non-Faculty)								
a. ___ Res. Assoc. - Postdoc.								
b. ___ Non-Fac. Professionals - Doc.								
c. 2 Non-Fac. Professionals - Other				10.4	10.4			7,300
d. 2 Grad Students (Res. Asst.)	44	36	8				18,800	
e. ___ Professional School Students								
f. 2 Pre-Baccalaureate Students	44	36	8				4,800	
g. 1 Secretarial-Clerical							3,000	
h. 2 Technical							25,800	
i. ___ Glass Blower							600	
3. Special Allowances								
Total Salaries and Wages							$ 58,332	$ 15,566
B. FRINGE BENEFITS							$ 5,950	$ 1,588
Total Salaries, Wages and Fringe Benefits (A+B)							$ 64,282	$ 17,154
C. PERMANENT EQUIPMENT								
1. Electric Consol and Parts							21,075	
2. Thermoluminescence Equip.							3,000	
Total Permanent Equipment							$ 24,075	$ --
D. EXPENDABLE SUPPLIES AND EQUIPMENT							$ 11,650	$ --
E. TRAVEL								
1. Domestic							8,320	1,000
2. International								
Total Travel							$ 8,320	$ 1,000
F. PUBLICATION COSTS							$ 500	$ 500
G. OTHER COSTS								
1.								
2.								
3.								
Total Other Costs							$ --	$ --
H. TOTAL DIRECT COSTS (A through G)							$ 108,827	$ 18,654
I. INDIRECT COSTS								
1. On Campus 34 % of 46,533 and 15,566							15,821	5,292
2. Off Campus 20.2% of 8,799							1,777	
Total Indirect Costs							$ 17,598	$ 5,292
J. TOTAL COSTS (H and I)							$ 126,425	$ 23,946
K. AMOUNT OF THIS AWARD (ROUNDED)							$ 126,500	

NSF Form 22, September 1967