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Strontium-90 Content of the Stratosphere

A low concentration of strontium-90 in the stratosphere indicates a short stratospheric residence time.

Herbert W. Feely

Prediction of the future hazard to the human population from radioactive fallout from past and future tests of nuclear weapons requires an estimate of the amount of debris held in the stratosphere and its rate of release to the troposphere and thence to the ground. Estimates of the stratospheric burden and residence time of strontium-90, generally considered to be the most hazardous fission product, have been made in the past (*1*, *2*) by combining assumed stratospheric bomb yields with measurements of fallout in rain and soil. However, each of these numbers is known only approximately. Some data are available from Project Ashcan (*3*), a project in which filter sampling of air is carried out by means of balloon-borne samplers. Unfortunately, uncertainties in the sampling efficiency and in the radiochemical analyses have limited the usefulness of these data (*2*, *4*, *5*).

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Research Program

Since August 1957, long-range, high-altitude Lockheed U-2 aircraft of the U.S. Air Force have been systematically sampling the radioactive debris in the stratosphere as part of the High Altitude Sampling Program sponsored by the Defense Atomic Support Agency. The scientific direction of the sampling program, the radiochemical analysis of samples, and the meteorological interpretation of the data are the responsibility of Isotopes Inc., Westwood, N.J. (*6*).

After an initial training and testing phase, from August to October 1957, at Laughlin Air Force Base, Texas, the sampling aircraft were based at Plattsburg Air Force Base, New York, and Ramey Air Force Base, Puerto Rico. From these two bases a north-south sampling corridor along the 70th meridian (W) extending from latitude 66° N to 6° S, was monitored systematically from November 1957 through May 1958. During June and July 1958, sampling was conducted only in the vicinity of Puerto Rico. From September 1958 through August 1959 the aircraft operated from Ramey Air Force Base and from Ezeiza Airfield, near Buenos Aires, Argentina, in order to sample the atmosphere in the South-

ern Hemisphere more completely. A corridor along the 63rd meridian (W), extending from latitude 38° N to 57° S, was monitored. The aircraft from Ramey made occasional deployments to Plattsburg to check the northern reaches of the sampling corridor. It is believed that the air sampled along a meridional corridor is representative of the entire atmosphere if sampling is carried out over a sufficiently long period of time. Eventually the whole atmosphere is carried through the corridor by normal zonal circulation.

Each of the six aircraft used for carrying out the high altitude sampling program has a sampler, capable of exposing four filter papers, consecutively, installed in the nose position. A single sample usually consists of the particulate matter filtered from 5000 to 20,000 standard cubic feet of air. Two missions, each of which collects 16 samples from the corridor, are scheduled per week.

As a result of wind-tunnel and in-flight calibrations of the sampler, carried out by the Air Force under the direction of E. G. Reid, the volume of air represented by a sample may be calculated to within about 15 percent. Special filter paper with a high permeability, which permits operation at a high face velocity, is used. This paper (I.P.C. filter paper number 1478) consists of cotton fibers impregnated with dibutoxylethyl phthalate. Wind-tunnel measurements of filter-paper retention, carried out under the direction of J. A. Van den Akker at the Institute of Paper Chemistry, have indicated that this paper has a collection efficiency of approximately 100 percent for particles as small as 0.005 micron in diameter, and they have suggested that its efficiency in collecting smaller particles is equally high.

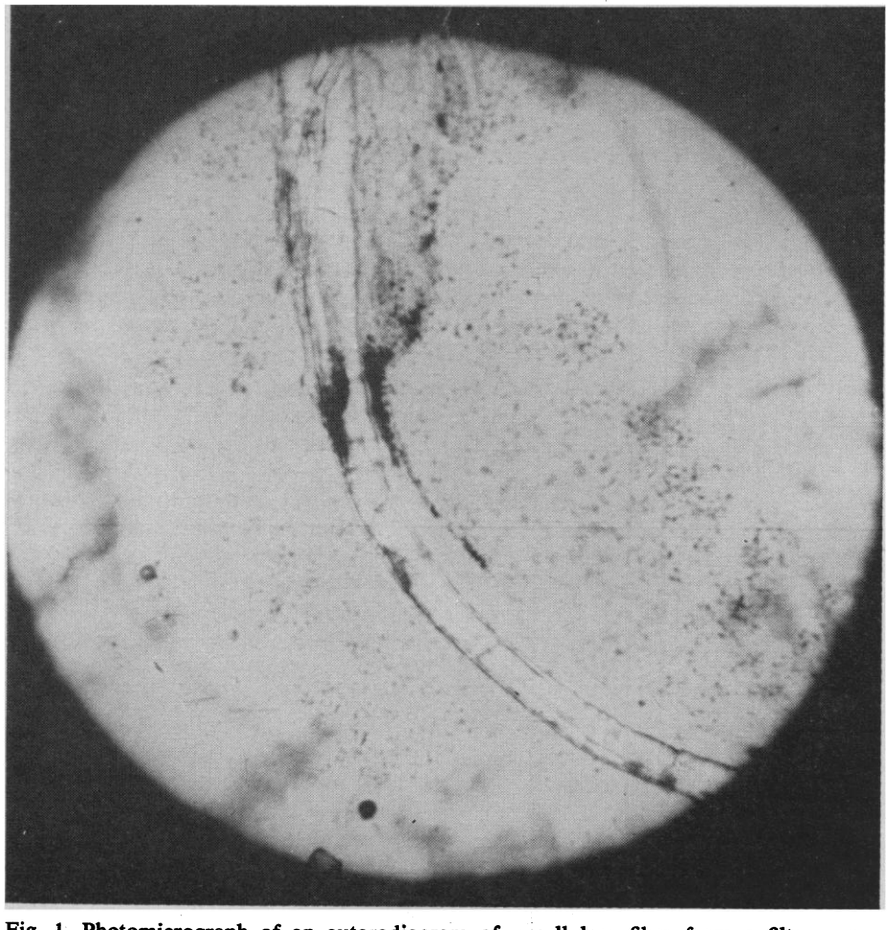

Fig. 1. Photomicrograph of an autoradiogram of a cellulose fiber from a filter paper used to collect stratospheric particles. The blackened silver grains, about 1 micron in diameter, were activated by beta emission from fission products contained in stratospheric particulate matter. The stratospheric particles are too small to be observed in the light microscope, but the autoradiogram discloses their presence in the fiber. The autoradiogram was obtained by coating liquid NTB-3 emulsion on a microtomed section, 5 microns thick, which contained the fiber. The emulsion was exposed for 1 month and developed for 5 minutes in D-19. [Isotopes Inc.]

Analysis

All filter papers, immediately after sample collection, are sent to Isotopes, Inc., where they are analyzed for total beta activity (Fig. 1) and strontium-90 content. Some samples are also analyzed for concentrations of the potentially hazardous nuclides cesium-137 and plutonium. Measurements of barium-140, strontium-89, zirconium-95 and cerium-144 are made to determine the age of the debris. Tungsten-185 and rhodium-102, introduced as tracers during the United States test series of May–August 1958 in the Pacific, are also determined.

An aliquot of each sample is set aside, and beta activity is counted over the course of several weeks. The level of activity and the apparent half-life of the activity are indicative of the age of the debris sampled. Other aliquots are taken from about one-fourth of the samples for analysis of the gamma spectra, which provide measurements of the barium-140 and zirconium-95 concentrations. One half of each paper is ashed at about 450° C. The ash is taken into solution, and separate aliquots are removed for analysis for tungsten-185, rhodium-102, and plutonium and for a sequential analysis for zirconium-95, cerium-144, barium-140, strontium-89, strontium-90, and cesium-137.

From the radiochemical data the disintegration rate per unit mass or unit volume of air is determined, through knowledge of the altitude at which the sample was collected, the air speed of the plane during sampling, the effective cross-sectional area of the filter, and the temperature of the air. A meteorological cross section is constructed along each flight track on the basis of data from radiosonde stations located near the sampling corridor.

Results and Conclusions

By 1 May 1959 more than 1400 samples had been collected. The conclusions given here are based on the radiochemical analyses of over 800 of these samples.

The strontium-90 content of the stratosphere varies markedly with latitude and altitude. Between September 1958

and April 1959, the concentrations of strontium-90 in the Southern Hemisphere were generally between 40 and 80 disintegrations per minute per 1000 standard cubic feet of air. Between November 1957 and April 1959, concentrations of strontium-90 in the stratosphere of the Northern Hemisphere were between 100 and 300 disintegrations per minute per 1000 standard cubic feet. Samples collected in clouds of debris from bomb tests conducted only a month or two before sampling showed much higher activities. The upper troposphere normally has a strontium-90 concentration of less than 3 disintegrations per minute per 1000 standard cubic feet.

The total stratospheric inventory of strontium-90 has been calculated by integrating the specific activities over the entire stratosphere. Because of limitations in the range of the aircraft and in the altitude they could attain, the data must be extrapolated to the upper stratosphere and to the poles. Fortunately, the specific activity of the atmosphere is greatest in the lower layers of the stratosphere, most of which can be sampled by the aircraft. The extrapolation to the top of the atmosphere has been made primarily by using data from Project Ashcan (*3*), which show a maximum in the concentration of strontium-90 per unit mass of air at 65,000 feet, with strong decreases in concentration with altitude up to 90,000 feet (*4*). Actually, only a small percentage of the mass of the atmosphere lies above the limits reached in the sampling program, and no reasonable assumption as to the exact distribution of activity within this portion of the atmosphere will significantly affect the calculated total inventory. The stratosphere above the polar tropopause appears to be fairly well mixed so that extrapolation poleward beyond the range of the sampling program aircraft should introduce only a small error into the calculation. Data from two sampling missions to the North Pole indicate that the concentration of nuclear debris in the polar stratosphere farther north than the limit of normal sampling is roughly the same as the concentration in the part of the northern polar stratosphere regularly sampled. In Table 1 the total strontium-90 concentration of the atmosphere, averaged over the period November 1957 through November 1958, is given as a function of latitude. Strontium-90 data for samples collected during this period give a total concentration of strontium-90 in the stratosphere for that period of about 1.0 megacurie. This is considerably less than the burden of 2.5 to 3.2 megacuries estimated by Libby (*2*) in the past.

Rates of Mixing

The data from the high altitude sampling program have shed some light on the rates of mixing of stratospheric air. The persistence of strong vertical concentration gradients in the specific activity of debris has shown, for example, that vertical mixing through altitudes up to 5000 feet is slow compared with horizontal mixing through several hundred miles above the tropical tropopause. Both the north-south spread of individual "hot clouds" and the poleward diffusion of tungsten-185 introduced during Project Hardtack have demonstrated a fairly rapid lateral spread of debris. Within less than 5 months after the first introduction of large quantities of tungsten-185 into the stratosphere, at about 10° north latitude, stratospheric concentrations at latitude 45°N had increased to about 20 percent of those at 10°. Lockhart found that the transport of debris across the equator in the troposphere is quite slow compared with the rate of mixing within the Northern Hemisphere (*7*). However, by 1 January 1959 the concentrations of tungsten-185 in the northern polar stratosphere were only about three times as high as those in the southern polar stratosphere.

Table 1. Variation in the concentration of strontium-90 in the stratosphere with latitude.

Latitude	Millicuries per square mile	Megacuries
	Northern Hemisphere	
0°–10°N	6.4	0.110
10°–20°N	4.2	0.069
20°–30°N	4.1	0.064
30°–40°N	5.0	0.070
40°–50°N	8.3	0.101
50°–60°N	12.2	0.120
60°–70°N	13.8	0.100
70°–80°N	11.3	0.050
80°–90°N	10.7	0.016
0°–90°N		0.70
	Southern Hemisphere	
0°–10°S	4.9	0.083
10°–20°S	1.8	0.030
20°–30°S	2.0	0.031
30°–40°S	2.7	0.038
40°–50°S	3.6	0.044
50°–60°S	3.1	0.031
60°–70°S	3.2	0.023
70°–80°S	2.9	0.013
80°–90°S	2.7	0.004
0°–90°S		0.30
	Both hemispheres	
Total		1.0

There is some confirmation of the theory that the tropopause gap, the region in middle latitudes where the high tropical tropopause overlaps the lower polar tropopause, is the chief route of transport of radioactive debris from the stratosphere into the troposphere. Samples taken in this region show a gradual decrease in activity in passing from the polar stratosphere laterally through the gap and into the tropical troposphere. The movement of air through the gap is promoted by the generally turbulent character of the region, as exemplified by its frequent association with the jet stream. The marked increase in turbulence in the gap during winter months may be a major cause of the observed spring maximum in the rate at which fallout reaches the ground (*8*).

Residence Half-Time of Debris

The residence half-time of nuclear debris in the stratosphere may by calculated by comparing the measured stratospheric inventory of strontium-90 with estimates of the total quantity injected into the stratosphere since the testing of nuclear weapons began. It is assumed, for purposes of this calculation, that the "drip" of debris out of the stratosphere can be approximated by first-order kinetics. Although this is an oversimplification, it does yield a first approximation to the truth. According to data released by the Atomic Energy Commission on nuclear bomb yields (*9*), the total fission yield for air bursts from 1945 through 1958 was 37.8 megatons. Presumably about 15 megatons of this amount were produced by the Soviet test series during the fall of 1958 (*2*). The total fission yields for ground-surface and water-surface bursts during the same period were 21.5 and 32.6 megatons, respectively. If it is assumed that 100, 20, and 30 percent of the debris from air, land-surface, and water-surface tests, respectively, stabilizes in the stratosphere, it appears that about 37 megatons of fission products had been introduced into the stratosphere by October 1958, when the Russian fall test series began. To explain the presence of only 1.0 megacurie of strontium-90, taken to be equivalent to 10 megatons of fission products, by mid-1958, one must assume a residence half-time in the stratosphere of less than one year, equivalent to a mean residence time of

less than 18 months. If it is assumed that debris introduced into the polar stratosphere by Russian tests has a different residence time than debris introduced in the tropical stratosphere by United States and British tests, residence half-times of 4 to 9 months for Russian debris and 9 to 15 months for United States and British debris may be derived. These values appear to be applicable to the debris which has been introduced into the stratosphere during the past four years; debris introduced at higher altitudes by the 1954 Castle series of high-yield tests may have had a longer residence time.

Alexander (*10*) has estimated that the total ground inventory of strontium-90 by 1 October 1958 was 3.2 megacuries. Thus, the ground inventory in July 1958 was probably about 3.0 megacuries. The estimates of yields give a stratospheric injection of 37 megatons of fission products, or about 3.6 megacuries of strontium-90, by July 1958. The values 1.0 megacurie for the stratospheric burden and 3.0 megacuries for the ground burden agree reasonably well with this value for total injection, in view of the uncertainties in these estimates.

Because the residence time of most nuclear debris in the stratosphere is relatively short, the radioactive decay of the debris during its storage there is negligible for long-lived nuclides such as strontium-90 and cesium-137 and is much less than was previously expected for nuclides such as strontium-89 and cerium-144. As a result, stratospheric storage cannot appreciably lessen the ultimate dietary and skeletal contamination of the human population. On the other hand, predictions of the surface concentrations of strontium-90 to be expected during the next few years as a result of further stratospheric fallout have generally been too high because of the overly high estimates of the current stratospheric inventory.

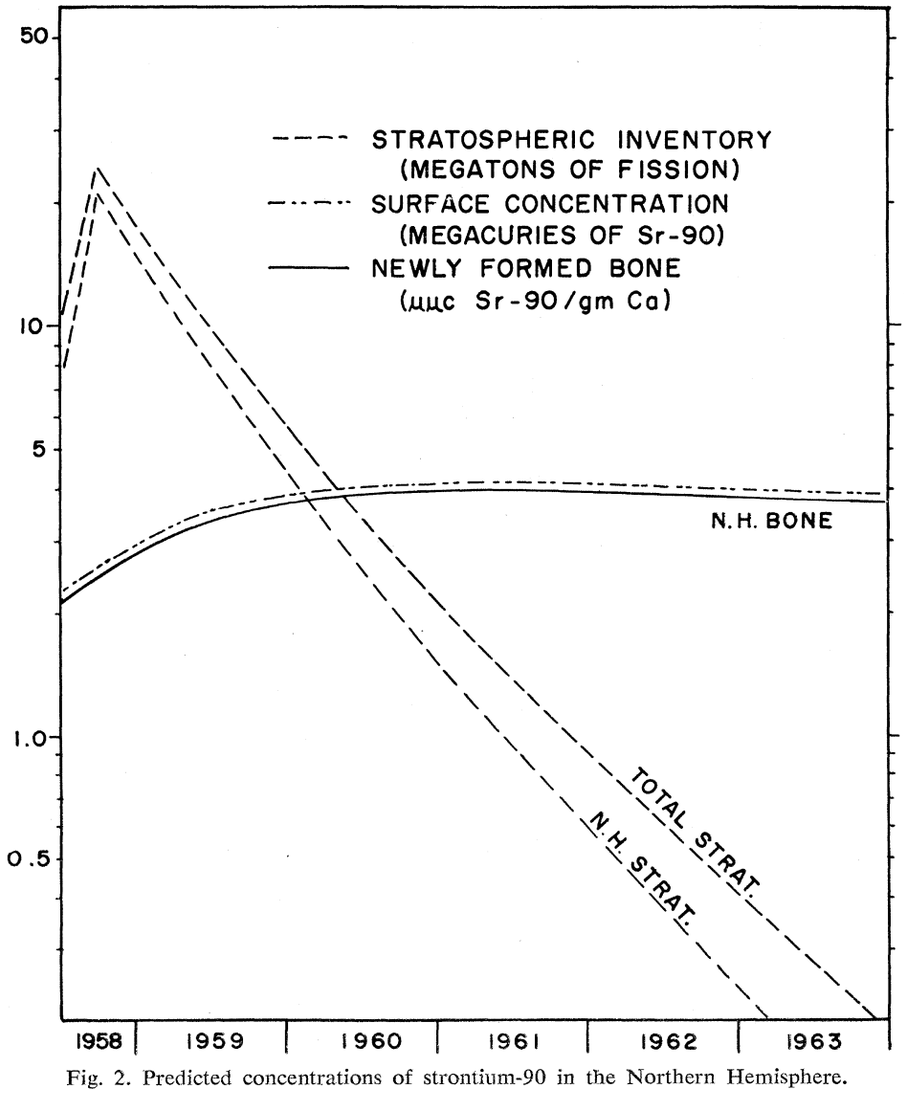

Fig. 2. Predicted concentrations of strontium-90 in the Northern Hemisphere.

Predicted Levels in Human Beings

A calculation may be made of the strontium-90 concentrations in human bones to be expected in the future if the testing of nuclear weapons in the atmosphere is not resumed. Kulp *et al.* (*11*) have estimated that a concentration of about 2.2 micromicrocuries per gram of calcium existed in newly formed bone in the North Temperate Zone in July 1958. It is assumed that the dietary levels of strontium-90 are directly proportional to the total quantity of strontium-90 in soil. As a result of the Soviet tests in late 1958, the total stratospheric burden reached more than 2 megacuries by November 1958. It is assumed that all debris from the Soviet tests will be deposited in the Northern Hemisphere and that the residence half-time is 6 months for Soviet debris and 12 months for U.S.–British debris. As shown in Fig. 2, the concentration of strontium-90 in newly formed bone will continue to rise for one or two years, passing a maximum of almost 4 micromicrocuries per gram of calcium during 1961. After the maximum is reached, the concentrations should decrease at a rate close to the 28-year half-life of strontium-90.

If, as is quite possible, dietary levels of strontium-90 are more dependent upon the rate of fallout than upon the cumulative surface burden, the maximum concentrations in newly formed bone should have been reached in 1959, and concentrations should decrease fairly rapidly during the next few years. Thus, no great increase in skeletal burden is to be expected as a result of future stratospheric fallout from the bomb tests conducted through the end of 1958 (*12*).

Note added in proof. Data from the analyses of 1029 samples collected since 1 January 1959 have been used to calculate a mean stratospheric strontium-90 burden of 0.8 Mc for January to August 1959. This is lower than the burden predicted in Fig. 2, suggesting that (i) the fall 1958 Soviet injection was less than 15 Mc of fission products, (ii) more than half of the debris from this Soviet injection fell out within six months, and (iii) at least half of the debris injected by the U.S. and Great Britain during 1958 fell out within 12 months.

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6. This program depends for its success upon the work of many individuals. Personnel of the Defense Atomic Support Agency (formerly the Armed Forces Special Weapons Project) were responsible for the initiation and coordination of the work. Dr. F. H. Shelton, Lt. Col. R. W. Swanson, Lt. Col. H. C. Rose, Col. R. D. Maxwell, Maj. A. K. Stebbins, III, and Maj. L. E. Trapp contributed to that phase of the program. Personnel of the 4080th Strategic Reconnaissance Wing, Light, and of the Air Rescue Service of the U.S. Air Force performed the extremely difficult sample-collecting operation. At Isotopes, Inc., planning of the sampling program and interpretation of the results were carried out largely by Dr. D. R. Carr, Dr. J. L. Kulp, Dr. J. Spar, Dr. H. L. Volchok, and myself. The radiochemical analysis program was directed by Mr. P. W. Krey, and the evaluation of data on sampling efficiency was carried out by Dr. J. P. Friend. Many other scientists and technicians made substantial contributions. Advice and information were supplied by many other persons, notably Dr. J. Van den Akker of the Institute of Paper Chemistry and Prof. E. G. Reid of the Guggenheim Laboratory, Stanford University.
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John A. Anderson, Astronomer and Physicist

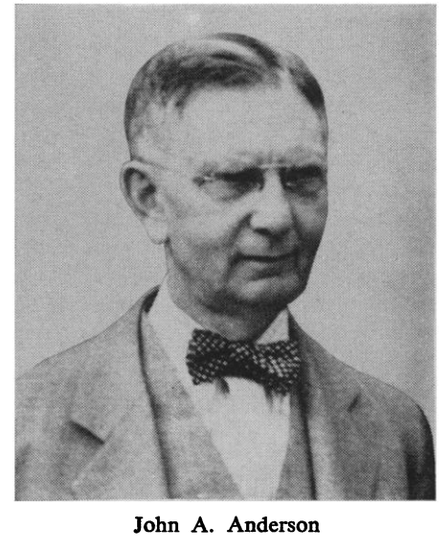
The 200-inch telescope on Palomar Mountain, California, was the work of many men. George E. Hale, of course, was responsible for the concept of such an instrument and for the enthusiasm that brought adequate financial support. The man chiefly responsible for its actual construction and for its present very successful operation was John A. Anderson.

Anderson was born at Rollag, Minnesota, 7 August 1876 and died suddenly in Altadena, California, 2 December 1959. He received his B.S. degree from Valparaiso University (Indiana), in 1900 and his Ph.D. from Johns Hopkins University in 1907. He remained at Johns Hopkins until 1916, advancing through the ranks of instructor, assistant professor, and associate professor. While in Baltimore he carried out various investigations on absorption and emission spectra and on the rotation of tourmaline by polarized light. He also took charge of the ruling machine constructed by Rowland for making spectroscopic gratings. Besides ruling a number of the finest gratings that had been produced up to that time, he developed methods for making replicas. With C. M. Sparrow he published a theory of the effect of groove form on the distribution of light by a grating. This was one of the important early steps toward the production of "blazed" gratings of high efficiency which play so important a role in current astronomical spectroscopy.

During the second decade of the present century, the Mount Wilson Observatory of the Carnegie Institution of Washington, under Hale's direction, was building up a group for the investigation of various fields of physics in order to obtain the fundamental data necessary for the interpretation of astronomical phenomena. In 1916 Anderson was invited to join this group of physicists, which included Harold Babcock and Arthur King. He immediately took charge of the large ruling engine which was then under construction.

Shortly after World War I Anderson developed the exploding-wire techniques for producing very high temperatures approximating those of the hotter stars. These high temperatures were obtained by discharging a large 50,000-volt condenser through a fine metallic wire; this procedure instantly evaporated the wire and raised the vapors to temperatures of 20,000°C or more. Since these explosions lasted only microseconds, Anderson and Sinclair Smith developed new techniques and instruments, including the rotating-mirror camera, the rotating mirror spectrograph, and the Kerr cell shutter, for studying them. These techniques and instruments came into common use during World War II in connection with atomic-bomb problems. Indeed, one of the original Anderson and Smith rotating-mirror cameras was used on that project and became the prototype of later models. During this same period Anderson contributed significantly to the development of Michelson's stellar interferometer and, in particular, to its application to the measurement of double stars. In collaboration with Harry O. Wood he developed the theory of the torsion seismometer. He designed equipment for, and participated in, eclipse expeditions in Spain in 1905, in Wyoming in 1918, in California in 1923, and in Sumatra in 1926.

In 1928 the International Education Board granted $6 million to California Institute of Technology for the construc-



John A. Anderson