minute period, have Poisson distributions, as would be expected. If a Poisson distribution of the noise peaks is assumed, the positions of the measured peaks on 7, 10, and 12 April, expressed in solar radii between 0 and 13, are bunched to such a degree that the probabilities are on the order of  $10^{-4}$  that they could be caused by solar noise alone. Since the position of the bunched peaks for each day corresponds to the expected range of the reflecting regions, the probabilities that solar echoes were obtained on each day are very nearly unity, being about 1 to  $10^{-5}$ . This result is for each day considered independently. Since similar results were obtained on all three days, the total probability of success is even much nearer unity.

From a preliminary analysis of the spectrum received it appears that the echo energy is spread over at least 2000 cycles per second. Solar rotation alone could account for much of this Doppler broadening, but gross motions in the solar corona would also be expected to produce a wide echo spectrum.

### **Conclusions**

There is a growing interest in the potentialities of probing the solar system with man-made radio waves. An obvious name for this field of investigation is "radar astronomy." With the added versatility inherent in the control of the transmitted waves, it is expected that much will be learned which will complement and extend knowledge gained from passive visual and radio astronomy, and from rocket probe measurements.

The scientific information about the sun gained from the first radar experiments, described above, is very limited. However, it is now possible to plan with confidence the systems and test procedures needed for more meaningful radar studies of our dynamic sun. From the time variability of the echo strength, delay, polarization, and spectrum, much will surely be learned about the constantly changing solar phenomena which affect so vitally the earth and its surroundings. More sensitive installations that will be suitable for solar and other studies in radar astronomy are now under construction at several locations, including Stanford University.

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# Cosmic-Ray-Produced Silicon-32 in Nature

Silicon-32, discovered in marine sponges, shows promise as a means for dating oceanographic phenomena.

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The nuclear transmutations resulting from the interaction of cosmic rays with nuclear species in the atmosphere have produced a variety of radioactive products detectable on the surface of the earth. Such isotopes as C14, H3, Be10, and P<sup>32</sup> have been found, and their individual distributions and concentrations in the various geological domains have led to many significant concepts and contributions in geochemistry, geophysics, and geochronology (see, for example, 1).

This article (2) concerns still another

isotope produced by cosmic rays-Si<sup>32</sup>, which we have detected in the marine environment. It is thought that this isotope is produced from the nuclear spallations of argon by cosmic rays. It has a half-life of roughly 710 years (3). Any Si<sup>32</sup> that reaches the earth from the atmosphere will be rapidly diluted with stable silicon, and the resulting specific activity of Si<sup>32</sup> will be very small. However, Si<sup>32</sup> decays by negatron ( $\beta^{-}$ ) emission to P<sup>32</sup>, which is a negatron emitter with a half-life of 14.3 days. This makes

it possible to detect Si<sup>32</sup> by milking and by counting the P<sup>32</sup> daughter from large amounts of silicon.

The principal exchange reservoir for Si<sup>32</sup> is probably the marine hydrosphere which most likely receives Si<sup>32</sup> via oceanic rains. The small amounts of silicon in surface marine waters should vield a relatively high specific activity of Si<sup>32</sup>, whereas the fallout on land will be so diluted by exchange and other chemical interactions with the exposed crustal materials that the detection of this nuclide will be extremely difficult. We estimate the average concentration of Si<sup>32</sup> to be 2.6  $\times$  10<sup>-5</sup> disintegrations per minute, per liter of sea water, or 8 disintegrations per minute, per kilogram of silicon, for a hypothetical thoroughly mixed ocean. The amount of sea water required to yield 1 disintegration per minute, an activity conveniently detectable, is  $3.8 \times 10^4$  liters. Since the handling of such an amount of water for the extraction of silicon presents many difficulties, Si<sup>32</sup> was sought initially in siliceous (opaline) sponges, which derive

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their silicon from sea water and are available in abundance, especially on the sea floors of the continental shelf. Three sponges trawled from the Gulf of California in 1956 were analyzed.

The experimental techniques and the results are presented in the next section. In the third section, an estimate of the rate of production of Si<sup>30</sup> in the atmosphere from the available cosmic ray and accelerator data is presented. The applications of Si<sup>30</sup> as a tracer for studying geochemical and geophysical processes with special reference to oceanography are discussed in the last section.

## **Experimental Techniques and Results**

Chemical procedure. The sponges were first cleansed of organic matter and foreign materials, not incorporated in the opaline structure, by digestion with nitric acid, followed by repeated washings with water and acetone. Approximately 200 grams of this material was allowed to stand for 3 months to allow the  $P^{32}$  to build up.

The milking procedure was initiated by treating the sample (after grinding) with a 50 percent solution of sodium hydroxide containing a known amount of carrier phosphate, in a polypropylene beaker. The solution was heated to speed up the dissolution. Hydrogen peroxide was added to remove any interstitial organic matter which was released during the breakdown of the opal. At this point approximately 95 percent of the silicon was in solution: the remaining 5 percent was treated with hydrogen fluoride and sulfuric acid, and heated to near-dryness; finally the residue was dissolved in HNO<sub>3</sub>.

The dissolved silicon in the NaOH solution was dehydrated to the SiO<sub>2</sub> form by the addition of 400 ml of concentrated hydrochloric acid and filtration through a Büchner funnel. The silica was dried at 130°C and washed thoroughly with dilute HCl to remove any adsorbed phosphorus compounds. The filtrate, washings, and the solution resulting from the treatment with HF and H2SO4 were combined and evaporated until the sodium chloride had crystallized out. The salt was removed by filtration through a sintered glass funnel and washed with concentrated HCl. These evaporation steps were repeated twice to remove the bulk of the NaCl. The final filtrate was taken to dryness, digested with 6N HCl, and dehydrated. This step was repeated twice to

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render insoluble any remaining traces of silica. The residue was heated with perchloric acid to dryness, and dissolution was effected in 20 ml of concentrated HNO<sub>8</sub>.

The phosphorus was extracted from the above solution and purified by the following steps carried out in accordance with often published standard procedures (4): (i) phosphoammonium molybdate precipitation; (ii) magnesium ammonium phosphate precipitation; (iii) elimination of cations by passage through a Dowex-50 cation exchange resin in the H-form; (iv) magnesium ammonium phosphate precipitation; and (v) ignition to magnesium pyrophosphate. This solid was assayed for its radioactivity.

After an initial counting, the sample was subjected to additional chemical purification by a repetition of the last four steps. The silica recovered in the dehydration step was milked again after a period of 28 days.

Counting techniques. The samples were deposited over an area of approximately 6 cm<sup>2</sup> on two split cylindrical copper supports and counted in a cylindrical geometry with a thin-wall, flow-type Geiger tube with "Q gas" (98.7 percent He; 1.3 percent isobutane). The details of the counter have

been described by Martell (5). The counter was housed in a shield which provided a minimum of 8 inches of steel on all sides and was operated in anticoincidence with a ring of large cosmic ray counters to eliminate the recording of cosmic ray µ-mesons. The background counting rate with blank copper supports was 0.26 count per minute. The counting efficiency for hard beta rays was determined by assaying a weighed sample of potassium chloride for  $K^{40} \beta$ -radiation ( $E_{max.} = 1.34$  Mev) under conditions similar to those employed for our samples. The over-all counting efficiency, which includes the counting geometry, back-scattering from the support, and absorption in the counter wall, was found to be 37 percent.

Several blank runs were made to ascertain the amount of contamination arising from chemical reagents and dust in the laboratory. The net counting rates of the blanks varied between  $0 \pm 0.05$  and  $0.05 \pm 0.04$  count per minute. A small, but positive, blank of 0.025 count per minute resulting from man-made or natural activities is indicated.

*Results.* The chemical yields of phosphorus and the activities of  $Si^{32}$  are summarized in Tables 1 and 2, respectively.

Table 1. Chemical yields.						
Sample No.	Remarks	Dry sample weight (gm SiO <sup>2</sup> )	Amount of carrier added (mg Mg2P2O7 eq.)	Yield (%)		
1	First milking	200	120	92		
1R	First milking recycle			83		
2	Second milking	190*	135	66		

\*Five percent of sample 1 did not dissolve in NaOH treatment. It was treated with HF and  $\rm H_2SO_4$ , and therefore was not available for the second milking.

Table 2. Counting data.						
Period of milking	Net sample counting rate (count/min)	Time elapsed between end of milking and counting (day)	Estimated No. of disintegrations of Si <sup>82</sup> per minute, per kilogram of Si*			
3 mo	$0.52 \pm 0.07$	2	$20.0 \pm 2.7$			
Sample No. 1R						
	$\begin{array}{c} 0.36 \pm 0.04 \\ 0.41 \pm 0.05 \\ 0.28 \pm 0.04 \\ 0.21 \pm 0.04 \\ 0.11 \pm 0.03 \end{array}$	$ \begin{array}{c} 4 \\ 7 \\ 13 \\ 20 \\ 28 \end{array} $	19.9 ± 1.7			
Sample No. 2						
28 days	$0.26 \pm 0.04$	2	$18.9 \pm 2.9$			
			Mean $19.6 \pm 1.3$			

\* Self-absorption in the sample was assumed to be 6 percent in all cases.

The observed activity of the samples can be unambiguously attributed to the presence of  $P^{32}$  on the basis of the following four independent checks on the nature of the radiation:

1) Half-life. The activity of sample 1R was followed for a period of two half-lives of  $P^{32}$ . The counting data are assembled in Table 2. The observed half-life of the activity,  $13 \pm 4$  days, is in good agreement with the literature value for  $P^{32}$ , 14.3 days.

2) Chemical behavior. Sample 1 was recycled through the purification steps 1 to 4 with the result that the specific activity of the recycled sample remained unchanged within the errors of measurement (Tables 1 and 2).

3) Beta energy. Absorption measurements of sample 1, with vinyl acetate absorbers of thicknesses 51 and 102 mg/cm<sup>2</sup>, yield a half-thickness value of

 $75 \pm 20 \text{ mg/cm}^{\circ}$ . The P<sup>se</sup> half-thickness for cylindrical counting geometry had previously been found to be 84.3 mg/cm<sup>2</sup> of aluminum (6). The vinyl acetate half-thickness, in milligrams per square centimeter, is expected to be about 13 percent higher than that for aluminum absorbers. The thinness of the samples, coupled with appreciable backscattered radiation, on the other hand, should yield a slightly smaller half-thickness. In view of this, the energy of the beta radiation is consistent with that of P<sup>se</sup> (7).

4) Growth of daughter activity. The estimated disintegration rates of  $Si^{20}$  from the first and second milking are concordant.

These results prove conclusively that the observed activity is due to  $P^{a_2}$  and that it arises from the decay of the parent nuclide Si<sup>a2</sup>.



Fig. 1. Vertical distribution of silicic acid in the Atlantic and Pacific oceans. Broken curve: North Atlantic Ocean, latitude  $47^{\circ}24'N$ , longitude  $07^{\circ}52'W$  [data from Armstrong (18)]. Solid curve: North Pacific Ocean, latitude  $26^{\circ}22.4'N$ , longitude  $168^{\circ}57.5'W$  [Goldberg (19)].

#### **Rate of Production of Silicon-32**

The production rate of Si<sup>32</sup> can be calculated from the available cosmic ray data, in different regions of the atmosphere, on (i) the star production rates in photographic emulsions and cloud chambers; (ii) the frequency distribution of stars as a function of the number of charged particles emitted; and (iii) the observed variation in the intensity of slow and fast neutrons. Lal et al. (8) have calculated the production rates of P<sup>32</sup>, P<sup>33</sup>, and S<sup>35</sup> in spallations of atmospheric argon for all regions of the atmosphere from such data. By their procedure, the global production rate of Si<sup>32</sup> is computed to be 1.7  $\times$  10<sup>-4</sup> atom/cm<sup>2</sup> column per second.

A second estimate can be made by using the observed fallout of cosmicray-produced P<sup>32</sup> and the expected ratio of cross sections for the production of Si<sup>82</sup> and P<sup>32</sup> from argon at energies of interest in cosmic rays. The fallout of P<sup>32</sup> was measured at tropical latitudes during the years 1956-58 (9) to be  $3.4 \times 10^3$  atoms of P<sup>32</sup>/cm<sup>2</sup> per year. This value is shown to be predominantly due to the removal by wet precipitations of activity produced in the troposphere only, with a mean removal period of 40 days. By correcting this figure for the decay of P<sup>32</sup> in the troposphere and taking a value of 29 percent for the fraction of P<sup>32</sup> produced below the tropopause (8), a production rate for  $P^{32}$  of  $1.1 \times 10^{-3}$  atom/cm<sup>2</sup> per second is obtained. The ratio of the cross sections of  $Si^{32}$  and  $P^{32}$  is computed to be 0.2 from Rudstam's empirical relation (10) describing the cross sections for the formation of nuclides in nuclear spallations.

The ratio of formation cross sections for two isobars, (A,Z) and (A,Z'), depends on two parameters only:

$$\frac{(A,Z)}{\sigma} \frac{(A,Z')}{(A,Z')} = \exp \left\{ -R[(Z - SA)^2 - (Z' - SA)^2] \right. \\ \left. = \exp \left[ R(Z' - Z) \left( Z' + Z - 2SA \right) \right]$$

The values of the parameters R and S are found to be nearly insensitive to either the bombarding energy or the target nucleus for masses between 51 and 75. In order to approximate our situation, we have used the values observed in the bombardments of vanadium by 60-, 100-, 170-, 175-, and 240-Mev protons:  $R = 1.6 \pm 0.1$ ; S =0.468  $\pm$  0.001. Therefore the ratio of cross sections is

$$\sigma$$
 (Si<sup>32</sup>)  $/\sigma$  (P<sup>32</sup>) = exp [R(Z + Z' - 2SA)]  
= 0.2 ± 0.02

The production rate of Si<sup>32</sup> therefore

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amounts to  $0.2 \times 1.1 \times 10^{-3} = 2.2 \times 10^{-4}$  atom/cm<sup>2</sup> per second, which is in good agreement with the previous estimate  $(1.7 \times 10^{-4})$ .

We take  $2.0 \times 10^{-4}$  atoms/cm<sup>2</sup> per second as the average global production rate of Si<sup>32</sup> in the atmosphere. The corresponding inventory of Si<sup>32</sup> on the earth is 28 kilocuries, or 1.75 kg of Si<sup>32</sup>.

The production of Si<sup>32</sup> in any significant quantities by nuclear weapons seems quite improbable but cannot be entirely ruled out at present. It cannot result from surface shots by principal neutron capture reactions,  $(n, \gamma)$ , (n, p)(n, a), since the target nuclides involved are short lived and do not exist in nature. However, it could be produced in the bomb hardware by successive neutron capture reactions. Unfortunately we cannot estimate this contribution. The total amount of Sr<sup>00</sup> produced till the end of 1958 has been estimated to be 63.5 kg (11). If Si<sup>32</sup> has been produced in bombs, its natural inventory could be raised by 10 percent if it amounts to as much as  $\sim 1$  percent of Sr<sup>90</sup> in nuclear weapons.

#### **Geochemical Considerations**

The geochemical behavior of silicon has been extensively studied not only because it is one of the principal rockforming elements but also because it is involved both in the biological and inorganic reactions in the major sedimentary cycle. The following brief summary of present knowledge of silicon geochemistry provides an entry into the problem of the distribution of Si<sup>32</sup> among the earth's geological domains following its formation.

Crustal rocks are decomposed by various weathering agents, resulting in the solution of silicon, the formation of new minerals containing silicon, and the accumulation of resistant minerals such as quartz, feldspar, and so on (see, for example, Pettijohn, 12). The two groups of solid phases are transported to the marine environment through the atmosphere and the hydrosphere. The river load of solids is deposited primarily in the near-shore areas, while the eolian minerals, originating mainly from the arid desert areas, fall to the sea floor in mid-latitudinal belts extending across the oceans (13).

The dissolved silicon exists nearly entirely as undissociated monomeric silicic acid, Si(OH)<sub>4</sub>, in both river and 5 FEBRUARY 1960 marine waters and is in an undersaturated state (14). Typical depth distributions in sea water are shown in Fig. 1. The acid is depleted in the photosynthetic zone (upper 100 meters or so) by microscopic organisms such as diatoms, radiolaria and silicoflagellates, which incorporate it into their opaline skeletons. This effect is extended to greater depths by the wind mixing in the upper layers of the oceans, such that waters down to 300 or 400 meters or more can have minimal values of silicon. The combustion of the plant materials which settle from the euphotic zone results in the dissolution of the silicon dioxide at intermediate depths. A maximum in the depth profile arises from the greater amounts of regeneration in warmer waters-that is, depths less than 1000. meters or so.

On the basis of the chemical properties of silicon, it is probable that Si<sup>32</sup> is involved primarily in the solution geochemistry of silicon. From the chemistry of silicon, we expect that whatever molecular species are formed initially after the production of single atoms of Si<sup>32</sup>, interactions with water droplets during condensations will finally lead to the formation of Si(OH)<sub>4</sub>. Single molecular species of silicon such as SiO<sub>2</sub> will be unstable with respect to Si(OH). since they cannot form aggregates. Such a situation is probably not too dissimilar to that of cosmic-ray produced Be<sup>7</sup>, which is found to exist in rain water as soluble species.

With the foregoing discussions in mind, we can consider the radiosilicon balance in the oceans. Simplified models for oceanic circulation for the case of



Fig. 2. Simplified model of silicon reservoirs in the exchangeable system.

 $C^{14}$  have recently been discussed (15, 16). We adopt the model of mixing and transfer processes in the oceans proposed by Craig (16). The oceans are divided into two principal reservoirs called the mixed (m) and deep (d) layers which are assumed to be well mixed internally. A steady state is reached by the exchange or transfer of silicon between the reservoirs (Fig. 2). The mixing cycle of soluble silicon is considered. As usually is the case in such models, the exchange of isotopes of very low abundance is governed by first-order kinetics, and the exchange rates of dominant species can thus be calculated. The following definitions are used:  $N_m$  and  $N_d$  are the amounts of silicon in the mixed and deep layers, respectively;  $N^*_m$  and  $N^*_a$  are the

amounts of Si<sup>32</sup> in the mixed and deep layers, respectively; R and  $R^*$  are the rates of introduction of silicon and Si<sup>32</sup> into the oceans, respectively;  $k_{m-d}$  and  $k_{a-m}$  are the rate constants for the mixing of soluble silicon between the mixed and deep layers, and the deep and mixed layers, respectively; K is the rate constant for removal of silicon from ocean to the sediment; B is the rate constant for removal of silicon from the mixed layer by biological precipitation of solid siliceous phases (part of the precipitated silicon which does not dissolve at greater depths in the oceans reaches the sediments directly; see Fig. 2); and  $\lambda$ is the disintegration constant of Si<sup>32</sup>. Therefore,  $\lambda = 0.693/710 = 1 \times 10^{-3}$ yrs<sup>-1</sup> (3).

By our definitions, we obtain the following relations for a steady state:

$$R \equiv (N_m + N_d) K \tag{1}$$

 $\frac{\mathrm{d}N_m}{\mathrm{d}t} = O = R - k_{m-d} N_m + k_{d-m} N_d - B N_m (2)$ 

$$R^* = (K + \lambda) (N^*_m + N^*_d)$$
 (3)

$$\frac{dN^{*}_{m}}{dt} = O = R^{*} - k_{m-d} N^{*}_{m} + k_{d-m}N^{*}_{d} - BN^{*}_{m} - \lambda N^{*}_{m} (4)$$

Solving these equations, we find:

$$\frac{N_m}{N_a} = \frac{K + k_{d-m}}{B + k_{m-a} - K}$$
(5)

$$\frac{N^*_m}{N^*_d} = \frac{K + \lambda + k_{d-m}}{B + k_{m-d} - K}$$
(6)

The ratio of the specific activities in



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the mixed and deep layers is readily obtainable as:

$$\frac{N^*_m/N_m}{N^*_d/N_d} = \frac{K+\lambda+k_{d-m}}{K+k_{d-m}}$$
(7)

Goldberg and Arrhenius (17) have computed a value of  $10^{-4}$  yr<sup>-1</sup> for K. Since this refers to both the soluble and particulate forms of silicon, a reduction by a factor of 2 of the rate of introduction of silicon based on an induction of Rex and Goldberg (13) that up to 50 percent of the silicon enters the oceans in solid phases, gives a more realistic figure of  $5 \times 10^{-5}$  for K. Since K is many orders of magnitudes smaller than other exchange coefficients, uncertainty in K does not affect our model calculations.

A plot of the ratio of the specific activities of Si<sup>32</sup> in the mixed and deep layers, calculated from Eq. 7, versus  $1/k_{a-m}$  is shown in Fig. 3. It is quite clear from the graph that this potentially measurable ratio is a quite sensitive indicator of the rate of mixing of deep waters into the mixed layer. It is of interest to note that this ratio is independent of the production rate of Si<sup>32</sup>. In the same figure, the expected specific activity of Si<sup>32</sup> in the mixed layer  $(\lambda N^*_m/N_m)$ , obtained by solving Eqs. 3 and 7, is plotted, taking for  $R^*$  a value of  $1.7 \times 10^{-4}$  atom/cm<sup>2</sup>/per second. The average silicic acid contents of deep and mixed layers are taken as 120 and 15 micromoles per liter, respectively.

The observed specific activity of 19.6 disintegrations per minute, per kilogram of Si, represents principally a value for the mixed layer, inasmuch as the sponges were retrieved from coastal waters of depths under 100 meters. This corresponds to an average mixing time (that is,  $1/k_{d-m}$ ) of more than 1500 years on the basis of the discussed model.

The quantitative significance of a photosynthesizing siliceous organism in the soluble silicon cycle in the sea can be readily seen by solving Eq. 5 for the biological removal constant B:

$$B = \frac{N_d}{N_m} (K + k_{d-m}) + (K - k_{m-d})$$
(8)

By taking a ratio of 49.6 for the volumes of deep to mixed layers (16), a value of 397 for Na/Nm is found. It is apparent that the second term is trivial compared with the first and can be neglected; furthermore, since K is small compared with  $k_{d-m}$ ,

$$B \approx 397 \ k_{d-m} \tag{9}$$

For likely values of  $k_{a-m}$ , say between  $2 \times 10^{-3}$  and  $5 \times 10^{-4}$  yr<sup>-1</sup>, values of B range between 0.8 and 0.2-that is, the silicon is removed from the mixed layer to deep layers or sediments due to biological activity in mean periods of 1 to 5 years.

In addition to studies of mixing times in the oceans, there are other obvious applications of this isotope as a tracer for studying problems in earth sciences: (i) the rates of accumulation of rapidly growing sediments containing large amounts of biogenous or hydrogenous siliceous phases; (ii) the individual characteristics of water masses within oceans with respect to mixing; (iii) the silicon cycle in the continental hydrosphere; (iv) changes in cosmic ray intensity with time during the last few thousand years; (v) ages of the polar ice caps.

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# Ross Granville Harrison, Experimental Embryologist

Ross Granville Harrison, born in Germantown, Pa., on 13 January 1870, died 30 September 1959. In his 89 years of life he encompassed an era of science in which he found absorbing interest. His early schooling was in Germantown and then in Baltimore. When he entered Johns Hopkins Uni-

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versity it was as an undecided undergraduate who was interested in gaining as much general knowledge as possible. He spent many hours browsing in the library. Later he had his own library facilities, and to the end of his intellectual life he enjoyed browsing in literature at a specialist's level.

Harrison's father was an engineer and was called upon to carry out many major engineering projects abroad. One of these was the construction of Russian railways, in the course of which he had to map part of the trans-Siberian railway in order to determine the characteristics of the railbed over this vast and then uncharted land. Ross' son, Richard Edes Harrison, is the recognized leader in modern cartography.

During Harrison's early childhood his mother died and his care devolved upon an aunt. This probably developed in him an early realization of his individuality. He felt early that he was the master of his own destiny. He realized that his father was abroad because of his business and professional problems, so he had no feeling of neglect.