

CONCENTRATIONS OF COSMOGENIC Si^{32} IN DEEP PACIFIC AND INDIAN WATERS BASED ON STUDY OF *GALATHEA* DEEP SEA SILICEOUS SPONGES

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INTRODUCTION

With a view to understand the nature of large scale circulation of water in the oceans, we have measured the concentrations of cosmogenic Si^{32} in the Pacific and Indian Ocean waters. Our method of obtaining this information is based on the fact that certain marine life-forms secrete opal from dissolved silicon for their skeletons. The results of radiochemical analyses of deep sea opaline materials and their implications to certain items of nuclear physics/geochemical interest are discussed.

Silicon-32 is produced in the atmosphere in spallations of argon nuclei by primary and secondary cosmic ray particles (LAL *et al.*, 1960; LAL & PETERS, 1967). The half-life of Si^{32} , unfortunately, has not yet been determined with any precision: however, it can be said with some certainty that it lies somewhere between 300-600 yrs. (GEITHOFF, 1962; HONDA & LAL, 1964; JANTSCH, 1967). The half-life of Si^{32} for which 500 yrs. has been considered to be a good working value (KHARKAR *et al.* 1966b) is ideal for studying diffusion/advection rates in the ocean, since radiocarbon studies indicate mean oceanic turnover times of the order of 700-1500 yrs. for the principal oceans. The only other radioactive tracer for which one can study the natural distribution of both the tracer and its stable isotope is C^{14} . Relatively, the half-life of C^{14} is, therefore, fairly long (5730 yrs.) which implicates a high precision in its measurement in oceans in order to obtain meaningful answers to the problem of oceanic circulation. Besides C^{14} and Si^{32} the only other naturally occurring radioisotope, Ra^{226} , has a fairly

suitable half-life (1600 yrs.); however, it has no stable isotope to allow a normalisation of biophysical fluxes within the oceans.

The application of Si^{32} to oceanic circulation is, however, confronted with one difficulty – its small cosmogenic production rate which necessitates analyses of fairly large volumes of water, of the order of 10-100 metric tons. The first detection of Si^{32} (LAL *et al.*, 1960) made use of the fact that *Hyalospongiae* (Hexactinellidae), the opal-rich marine sponges, formed a natural agency for the enrichment of dissolved marine silica. This fact was later utilised to study the global specific activity of Si^{32} in continental off-shore surface waters of several oceans and seas (KHARKAR *et al.*, 1966a). In order to obtain an insight into the rates of mixing of deep water with surface waters, it becomes necessary to study specific activity of Si^{32} in deep waters between 1000-5000 meters where a considerable variation would be expected because of short half-life of Si^{32} . The study of deep waters, basing on the sponge method, however, becomes problematic because of rarity of such samples. The present experiments rest on the fact that several precious samples of siliceous sponges trawled from the deep depths of Pacific and Indian oceans during the Galathea Expedition were made available to us by the courtesy of Dr. TORBEN WOLFF. For this reason, the authors consider it appropriate and a privilege to report the results of Si^{32} study of deep sea sponges in this journal.

EXPERIMENTAL

1. Description of samples studied

A total of 14 Hyalospongiae and two Demospongiae specimens from depths exceeding 500 meters were analysed in the present work. Out of these, 12 specimens were collected by the *Galathea*. In Table 1, we have listed relevant data on these samples along with the information on their classification and regions of occurrence (for completeness a brief description is given in each case of the appearance

of the sponges). The relevant data of the remaining 3 sponge samples are summarised in Table 2; in these cases the information on species is lacking.

It may be of some interest to note here that the Galathea Expedition counted 31 species of Hyalospongiae, out of which 14 were new (LÉVI, 1967). In the present work, 4 of these new species are represented by samples TF 67, 70, 76 and 77; two new species of Demospongia are represented by TF 66 (see Table 1).

Table 1. Relevant data on "Galathea Expedition deep-sea siliceous sponges" analysed¹.

Classification	Description of sponges	Remarks	Location, water depth and collecting date	Distribution	Our code no.
<i>Euplectella timorensis</i> Ijima	Tubular, 40-70 mm in diameter. Sieve-like appearance. Supported by a very regular lattice-work of spikes. Surface full of orifices of 1 to 1.5 mm diameter	—	East of Cebu, Philippines (10°27'N, 124°18'E) 810 m 25 July 1951	—	T. F. 74
<i>Mixture of two species</i>					
(1) <i>Pheronema pilosum</i> Lévi	A greyish green sponge, 23 mm long, cylindrical, with a heavy convex base of 20 mm diameter and a large central opening covered by many irregular spicules all over. Very spiky	Resembles <i>Pheronema carpenteri</i> of the Atlantic Ocean (A new species)	Strait of Malacca (6°38'N, 96°00'E) 1140 m 9 May 1951	Strait of Malacca	T. F. 76
(2) <i>Pararete farreopsis</i> (Carter)	Found in many fragments. A tubular sponge, with the inner surface very smooth	—	East of Cebu (10°27'N, 124°18'E) 810 m 25 July 1951	Philippines and Indonesia	T. F. 76
<i>Oonema bianchoratum</i> (Wilson)	Numerous specimens, but many in fragments. The smallest one was thick, yellowish and ovoid, measuring 13/6/3 mm. The elongated and ovoid ones measured 40/12/15 mm. Better developed and still fixed by the peduncle were 50/5/23 mm	—	Off Costa Rica (9°23'N, 89°32'W) 3570 m 6 May 1952	East Pacific	T. F. 71
<i>Cyliconema polycaulum</i> (Lendenfeld)	Globular or ovoid. Most samples are 30 mm long and 25 mm in diameter. The opening is elliptic, 5-7 mm long, without periphe-	The re-opening of small tails and the small dimensions of the thorns are peculiarities of the genus <i>Cyliconema</i>	Mindanao Sea, Philippines (8°48'N, 124°09'E) 1500 m 16 Aug. 1951	East Pacific and Philippines	T. F. 90

Classification	Description of sponges	Remarks	Location, water depth and collecting date	Distribution	Our code no.
	<p>ral cushions. Peduncle is of divergent spike bundles.</p> <p>Most specimens had their skin cover destroyed</p>				
<i>Prionema spinosum</i> Lendenfeld	<p>Found in many fragments, carried by a thick peduncle; greyish or brown. A typical sponge measured 90/60 mm at top and was 50 mm high</p>	<p>The spike structure is typical of this species. <i>Prionema crassum</i> is a closely resembling species</p>	<p>Gulf of Panama (5°49'N, 78°52'W) 3270-3670 m 13 May, 1952</p>	<p>East and Central Pacific</p>	T. F. 75
<i>Leptonema choaniferum</i> Lévi	<p>Soft, weak sponge with a velvety surface, ovoid, yellowish rosy colour, measures 35 mm high, 12 mm wide, narrowing at both extremities like a conch-shell</p>	(A new species)	<p>North of Madagascar (5°25'S, 47°09'E) 4820 m 10 Mar. 1951</p>	<p>West Indian Ocean</p>	T. F. 67
<i>Corynonema natalense</i> Lévi	<p>Irregular shape, form is conical and flat with two peduncles of 200 mm length. Thickness 510 mm. One intact specimen measured 60/40/20 mm</p>	<p>The skeleton is made of heterogeneous elements. Its thick pinules are of <i>Corynonema</i> characteristic (A new species)</p>	<p>Off Natal (25°36'S, 35°21'E) 730 m 21 Feb. 1951</p>	<p>West Indian Ocean</p>	T. F. 77
<i>Oonema bipinnulum</i> Lévi	<p>Cylindrical, measuring generally 90/15/20-25 mm. The upper region is cap-shaped, and the axis is elongated into a short, cylindrical bag of 10 mm length and 5 mm diameter. One sample with an unusually long (600 mm) peduncle</p>	<p>Beautiful specimen with spicules very similar to <i>Oonema bianchoratum</i>. Obtained in many varieties (A new species)</p>	<p>Kermadec Trench (36°38'S, 178°21'E) 2470 m 25 Feb. 1952</p>	<p>Kermadec Trench</p>	T. F. 70
<i>Caulophacus schulzei</i> Wilson	<p>Beautiful sponges, obtained in various sizes. The smallest one is clean grey and thick and measures only 10 mm high and 2-10 mm in diameter. Peduncle is very rigid. As heads enlarge in diameter, heights decrease, and the heads get flat. Some look like mushrooms, some are with lots of orifices like a sieve and some are like corals</p>	—	<p>Tasman Sea (45°47' to 43°58'S, 164°39' to 165°24'E) 4390-4510 m 14-15 Jan. 1952</p>	<p>Tasman Sea and East Pacific</p>	T. F. 72 and T. F. 73

Mixture of two species

(1) <i>Chondrocladia multichela</i> Lévi	Peduncle 100 mm long and 5 mm in diameter, formed by longitudinal spicules. It ends with a globular head, 25 mm in diameter and carrying tentacle-like expansions, 30-50 mm in length. The whole sponge, especially the head and lateral expansions, surrounded by a detachable envelope	This species is similar to <i>C. gigantea</i> Hansen and <i>C. yatsui</i> Topsent and is distinguished by the spicule size. (A new species)	Off Kenya (3°23'S, 44°04'E) 3960 m 13 Mar. 1951	Off Kenya	T. F. 66
(2) <i>Cladorhiza nematophora</i> Lévi	Tiny sponge, 12 mm in diameter. Head yellowish orange, carried by a tiny peduncle of 1.5 mm diameter. Head with several extensions (length 12 mm, diameter 0.5 mm) on sides.	(A new species)			T. F. 66

1. These samples were very kindly made available to us by Dr. TORBEN WOLFF of the Zoological Museum, Copenhagen.

2. Chemical procedures for milking and counting of P³² activities from Si³² in sponges

In view of the wide geochemical applications of Si³² in glaciology (DANSGAARD *et al.*, 1966; CLAUSEN *et al.*, 1967), in hydrology (NIJAMPURKAR *et al.*, 1966; LAL *et al.*, 1970), in meteorology and in oceanography (KHARKAR *et al.*, 1963; KHARKAR *et al.*, 1966a and b; LAL, 1969; SOMAYAJULU 1969a), the chemical and counting procedures for the measurement of Si³² activity by milking its 14.3 days

daughter activity, P³², are fairly standardized. We therefore will not discuss these at any length but only indicate the magnitude of P³² activities found in the present work in order to give some idea of the reliability of the present results.

The P³² activities milked from the siliceous sponge samples, were counted on low-level counters (LAL & SCHINK, 1960) over periods of the order of 3-5 half-lives of P³². As an illustration, the gross counting rates (including the counter back-ground), are shown in Fig. 1 for four of the samples.

Table 2. Relevant data on three deep-sea siliceous sponges analysed for Silicon-32.

Code No.	Location	Date	Appearance	Source of sample
LC II-25	Off S. California (32°40'N, 117°20'W) 1080 m	29 May 1960	Pin-cushion	SAM HINTON, Scripps Institution of Oceanography, La Jolla, California
C	N.E. of Guadalupe I. (31°17'N, 117°36'W) 2110 m	13 Feb. 1960	Needle like	E. D. GOLDBERG, Scripps Institution of Oceanography, La Jolla, California
B	N.E. of Guadalupe I. (29°31'N, 117°17'W) 540-820 m	13 Feb. 1960	Needle like	E. D. GOLDBERG, Scripps Institution of Oceanography, La Jolla, California

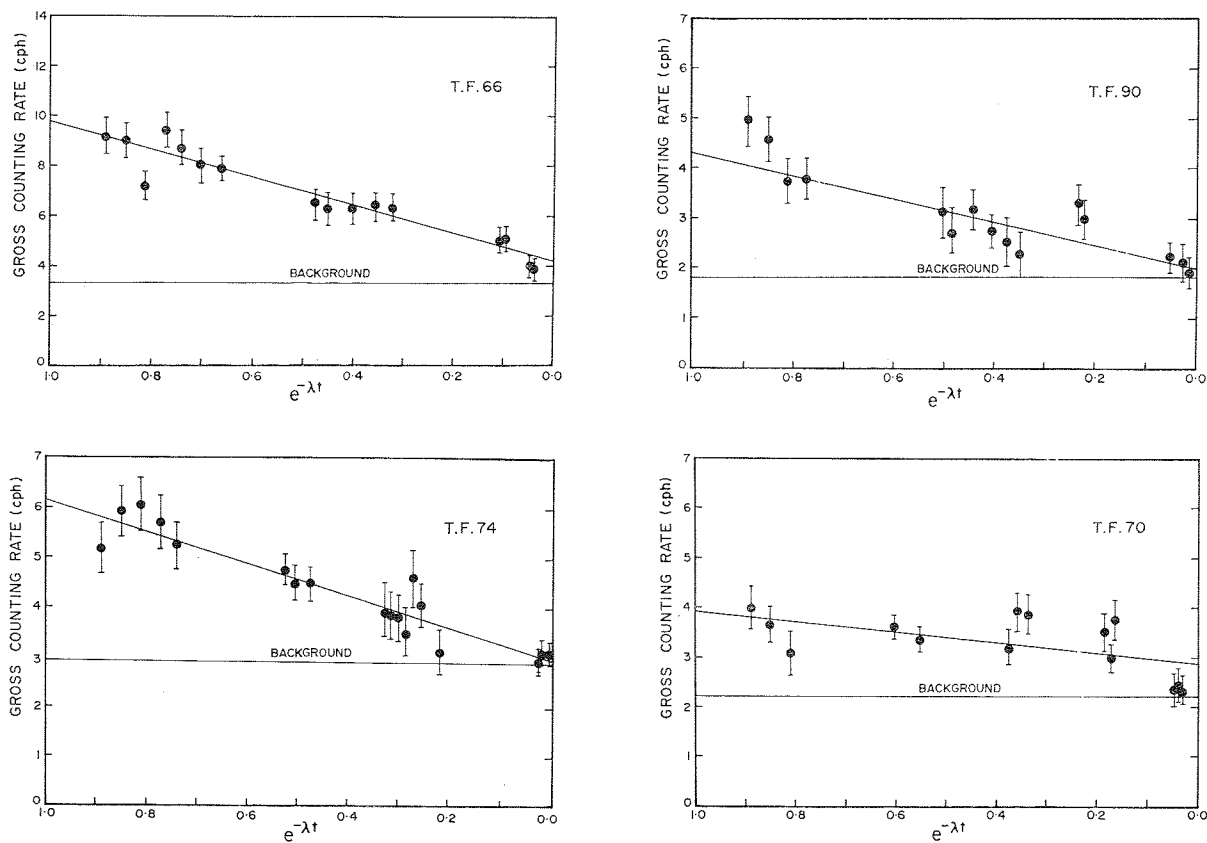


Fig. 1. Observed decay of P^{32} activities, milked from deep siliceous sponges T. F. 66, T. F. 74, T. F. 90 and T. F. 70 (see Tables 1 and 3 for other details). The observed gross counting rates (in unit of counts per hour) are plotted. The mean background rate is shown by the solid line.

From our experience, we have learnt that P^{32} counting rates exceeding 0.5 counts/hr. (called cph henceforth) can be detected and for signals exceeding 5 cph, the measurement can be carried out with an accuracy of better than 15 per cent. In the present case the level of activities are in the range of 2-5 cph in half the cases while for the others they are small, being 0.5-1.0 cph. In the latter cases, the statistical errors assigned are of the order of 30-50 per cent (see Table 3).

In order to ascertain the validity and reproducibility of our measurements, two of the samples were re-milked and in the case of four of the samples they were combined in pairs and re-milked: the results for the composite samples (TF 66, TF 67) and (TF 72, TF 73) are also given in Table 3. The composite sample, TF 66, 67 was re-milked (see Table 3). In all these cases, except for the composite sample TF 72, 73 where the counting errors remained large and no specific conclusion could be drawn, a good agreement is seen in other cases indicating the validity of the various results presented in Table 3.

3. Discussion of results

The experimental results on the specific activities of Si^{32} in 14 deep sea hyalosponges, summarised in Table 3, are presented in Fig. 2 on a Goode's Homoloxine equal-area projection map. The numbers in bold letters indicate the mean values for the specific activity of Si^{32} . An analysis of these results reveal the following features:

- (i) The specific activities in the Pacific Ocean samples show a general decrease with increasing depth, ranging between 25 and 5 dpm/kg SiO_2 for depths 700-4500 m, respectively.
- (ii) The specific activities are independent of the type of siliceous sponge, for example, TF 90, 76 and 74 which are all different species and represent samples from 700-1500 m depths, have specific activities of 23.6, 20, and 25.5 respectively. This would be expected also since the opal in the skeletal material of the siliceous sponges are known to be ingested from the dissolved silicon present in sea water.
- (iii) The results for the deep Indian Ocean samp-

Table 3. Specific activity of Si³² in marine siliceous sponges (water depth > 500 m).

T. L. F. R. code no.	Ocean, location	Depth of sample (metres)	Wt. of SiO ₂ taken for milking (gms)	Net P ³² activity observed (c. p. h.)	dpm Si ³² per kg SiO ₂
LC II-25	Pacific, off S. California	1080	43.0	2.9	6.5 ± 2.0
C	Pacific, N. E. of Guadalupe I.	2110	12.0	2.5	12.8 ± 2.0
B	Pacific, N. E. of Guadalupe I.	540-820	7.3	0.7	7.2 ± 2.0
T. F. 74	Pacific, Philippines	810	7.2	3.2	23.6 ± 2.3
T. F. 76	Pacific, Strait of Malacca	810-1140	6.1	1.9	20.0 ± 2.0
T. F. 71	Pacific, off Costa Rica	3570	a) 6.35 ¹ b) 6.20 ¹	0.93 0.7	9.2 ± 3.0 10.0 ± 3.0 <i>Mean: 9.6 ± 2.1</i>
T. F. 90	Pacific, Mindanao Sea	1500	a) 7.4 b) 4.4	2.3 1.9	27.6 ± 3.0 23.4 ± 4.6 <i>Mean: 25.5 ± 3.0</i>
T. F. 75	Pacific, Gulf of Panama	3270-3670	9.0	0.8	7.5 ± 4.0
T. F. 70	Pacific, Kermadec Trench	2470	6.0	1.1	11.1 ± 5.0
T. F. 72	Pacific, Tasman Sea	4510	9.2	0.52	4.25 ± 2.5
T. F. 73	Pacific, Tasman Sea	4390	2.1	0.1	2.1 ± 6.0
Composite Sample T. F. 72 (6.5 g) and T. F. 73 (1.0 g)	Pacific, Tasman Sea	4390-4510	7.5	0.75	6.8 ± 2.0
T. F. 66	Indian Ocean, off Kenya	3960	10.0	3.0	13.2 ± 1.8
T. F. 67	Indian Ocean, N. of Madagascar	4820	18.0	5.7	20.4 ± 2.8
Composite Sample T. F. 67 (10.0 g) and T. F. 66 (17.4 g)	Indian Ocean, N. of Madagascar	3960-4820	a) 27.4 b) 25.0	4.2 4.1	10.5 ± 1.0 9.7 ± 1.6 <i>Mean: 10.1 ± 1.0</i>
T. F. 77	Indian Ocean, off Natal	730	33.0	2.7	4.85 ± 1.0

1. Entries marked a) and b) refer to 1st and 2nd milkings respectively.

les seem to be systematically higher than for the Pacific samples (see Fig. 3). This is quite in line with expectations, since Si³² specific activities in the Atlantic Ocean would be high because of low silicon stable concentrations of the Atlantic waters and since the Indian Ocean waters which contain admixture of the Atlantic and the Pacific, lie in between.

(iv) The present results based on siliceous sponges can be compared with the recent direct determinations (SOMAYAJULU, 1969b; SOMAYAJULU *et al.*, 1970) of Si³² specific activity in Pacific waters at latitude 31°41'S and longitude 177°16.2'W: the

mean profile curve based on these results is shown in Fig. 3. The direct measurements for the Pacific waters became possible due to development of an *in-situ* silicon extraction technique (LAL *et al.*, 1964) and a butterfly sampler to flush water through the ferric-hydroxide grains suspended on marine spongin fibres. Till the development of this method, the analysis on deep waters, though attempted by bringing on board large amounts of sea water from great depths and processing them (SCHINK, 1962), was not practical on a routine basis. It should, however, be noted that the results for the deep sea became available first by the analyses of the Galathea

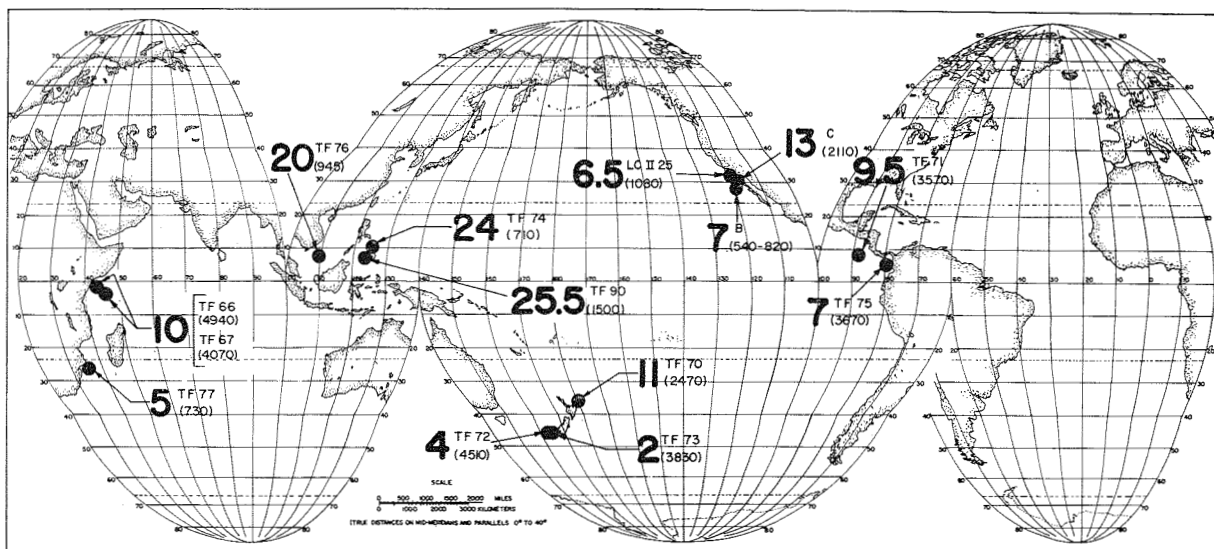


Fig. 2. Measured Si^{32} specific activities (dpm/kg SiO_2) in deep ocean waters (depths > 500 m) based on analyses of siliceous sponges. The two numbers given alongside each silicon-32 specific activity value refer to our sample code number (Tables 1 and 2) and depth in metres, respectively.

Expedition few years before the *in-situ* extraction technique was applied. It is therefore very gratifying to see that the very useful information on Si^{32} concentrations in deep waters, which can now be cross-checked with the direct results obtained, was based on the *Galathea* sponges.

The overall agreement between deep sea sponge results and direct measurements in South Pacific waters (at one station) is fairly good considering the fact that the sponges are from various locations in the Pacific. A direct comparison can be made in one case only; between the sponge value of $11.1 \pm$

5.0 dpm/kg SiO_2 for TF 70, which derives from the Kermadec Trench (37°S , 178°E), depth 2470 m, and the direct value of 17 ± 3 dpm/kg SiO_2 for waters at 2200-2300 m depth from 31°S , 177°W .

(v) The integrated Si^{32} activity in one sq. cm water column (dpm/cm²) based on the present results is found to be 3×10^{-2} .

In this computation we have considered the average depth distribution of stable silicon in the Pacific, since the sponge samples are fairly well distributed (see Fig. 2). It should also be noted that the calculation of the integrated activity is not sen-

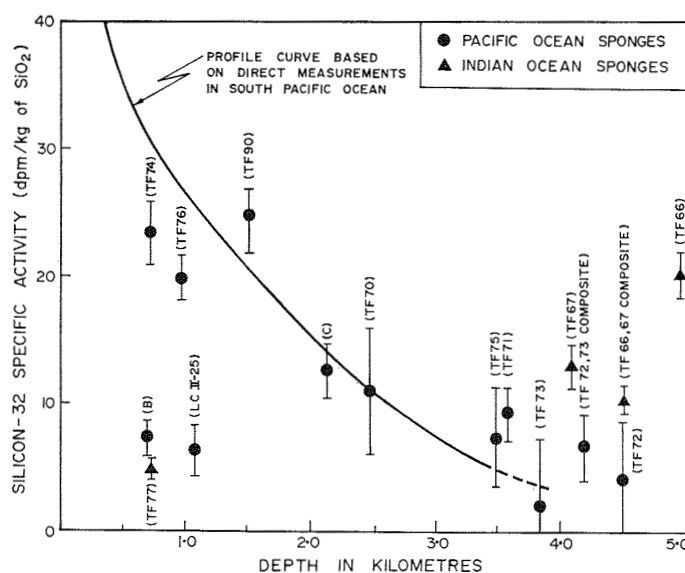


Fig. 3. Plot of silicon-32 specific activity as a function of the recovered depth of the siliceous sponge. Sample code number is given in parenthesis. The solid line is based on direct measurements in South Pacific Ocean waters (SOMAYAJULU *et al.*, 1970).

sitive to lack of knowledge of specific activities of Si^{32} (dpm/kg SiO_2) in the surface waters which contain so little stable silicon. The value of 3×10^{-2} dpm $\text{Si}^{32}/\text{cm}^2$ compares well, within errors, with the value of 1.7×10^{-2} obtained by SOMAYAJULU (1969b). Additionally, it is also in good agreement with its estimated global fallout rate of 3×10^{-2} dpm/cm² based on measurements of its concentrations in wet precipitations (KHARKAR *et al.*, 1966b).

It should be pointed out here that since most of the Si^{32} inventory is in the deep sea and since all the samples reported herein were collected prior to 1952 (see Table 1), except for three samples in Table 2 which were collected in 1960, the results are free of any possible activity globally dispersed by nuclear weapons. This point is discussed, since DANSGAARD & CLAUSEN (1966) have evidenced the production of some Si^{32} in 1962 nuclear tests. Because of the close agreement between results based on Si^{32} concentrations in wet precipitations and its inventory in oceans we therefore conclude that no appreciable Si^{32} was present in the atmosphere when its fallout was measured (1961-1964).

4. General remarks on the implications of the present results

With respect to the nature of large scale circulation of water, we would like to bring out one point here and that concerns the average rate of overturn of the ocean as deduced from studies of C^{14} and Ra^{226} : the implications of the depth variation of Si^{32} specific activities to vertical diffusion/advection rates will be discussed elsewhere (SOMAYAJULU *et al.*, 1970). Basing on simple box-model type calculations, it can be shown that the ratio, R, of the mean Si^{32} specific activities (dpm/kg SiO_2) in the mixed layer (taken to be 0-100 m usually) and the deep-sea is given by LAL (1969):

$$R = 1 + (\tau_0/\tau).$$

In the above relation, τ_0 and τ , are the turnover times of the ocean and the mean life of Si^{32} respec-

tively. From direct measurements in Pacific waters (SOMAYAJULU, 1969b), Si^{32} specific activity of mixed layer is known to be 70 ± 15 (dpm/kg SiO_2) in contrast to the average value of about 15 dmp/kg SiO_2 as indicated from present measurements (see Fig. 3). Thus, considering values of the order of 1000-1500 yrs. for τ_0 , based on C^{14} studies, a good agreement between C^{14} and Si^{32} results exists only if the mean life of Si^{32} , τ , is taken to be of the order of 300-400 yrs.

It should also be pointed out here that based on the observed activity of Si^{32} in wet precipitations (KHARKAR *et al.*, 1966b), the production rate of Si^{32} in the atmosphere (LAL & PETERS, 1962) was estimated to be too small by about a factor of 3. In this comparison, the half-life of Si^{32} was assumed to be 500 yrs. If, therefore, its half-life is only 300 yrs., as also indicated by some spallation measurements (JANTSCH, 1967), the discrepancy between the theoretical estimates by LAL & PETERS (1962) and its fallout in wet precipitations, or its inventory in oceans based on present work, is considerably reduced.

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SUMMARY

Siliceous sponges collected in the Galathea Expedition have provided the first indications of the specific activity of cosmogenic Si^{32} in bathyal and abyssal depths of the Pacific and Indian Oceans. A few available recent direct measurements for the open South Pacific waters agree well with the values reported herein, based on analyses of siliceous sponges.

The present results yield a value for the integrated amount of Si^{32} (dpm values alone are considered

because its half-life is not accurately known as yet) in 1 cm^2 column which is compared with its fallout in wet precipitations during 1961-1964 and its theoretically estimated global production rate. The implications of the data on Si^{32} specific activities in hyalosponges to oceanic overturn rates are considered. It is shown that a close agreement with C^{14} data will result only if the half-life of Si^{32} is of the order of 300 yrs.

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