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Radioactive Contamination of the Indo-Antarctic Ocean Water in Each Earlier Period in 1961 and 1962

NAGAYA, Yutaka* SHIOZAKI, Masaru* and Yoshio SETO*

長屋　裕　塩崎　愈　背戸義郎

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ABSTRACT

Significant increase of Ce-144 concentration was detected in the Indo-Antarctic surface sea water collected in early 1962 (mean 0.57 $\mu\mu c/l$), comparing with those of early 1961 (mean 0.12 $\mu\mu c/l$), and was supposed to be caused by the stratospheric fallout injected by the resumption of nuclear explosions by USSR on the northern polar region during September to November, 1961. On the other hand, no increase of Sr-90 concentration was detected in these samples (means 0.07 and 0.06 $\mu\mu c/l$) and that of Cs-137 concentration was slight (means 0.13 and 0.15 $\mu\mu c/l$).

The horizontal distributions of the radioactive nuclides in surface water indicate the decreasing Sr-90 and Cs-137 concentrations with increasing latitude, but no latitudinal variation of Ce-144 is detected.

By comparison of Ce-144 concentrations in early 1961 and 1962 samples, only 6 months of minimum time is estimated for the transport of stratospheric fallout debris from the northern polar region to the southern hemisphere, and this value is considerably shorter than those reported by other workers.

INTRODUCTION

A part of fission products debris released by the thermo-nuclear explosions in the northern hemisphere is known to deposit on the southern hemisphere. Radioactive fallout in Antarctic snow[1], Sr-90 in precipitations on Australia[2,3,4,5,6] and New Zealand[7], and fission products deposition on the southern hemisphere[8] have been reported. These data disclosed that the fallout level in the southern hemi-

* Hydrographic Division, Maritime Safety Agency, Tokyo, Japan

sphere was considerably low in comparison with that of the northern hemisphere and no fission products released by polar explosions in 1957 or 1958 was detected during 1959 through 1961 on the southern hemisphere.

A few data of the radioactive contamination of the southern hemisphere sea water during 1960 through 1961 have been reported. Folsom and Mohanrao[9] and Schroeder and Cherry[10] reported Cs-137 in the Indian Ocean, Higano *et al*[11] reported Sr-90 and Cs-137 in the Indian Ocean, and Rocco and Broecker[12] reported Sr-90 and Cs-137 in the South Atlantic, the Antarctic and the eastern South Pacific Oceans.

Authors have measured the concentrations of Sr-90, Cs-137 and Ce-144 in surface water of the Indo-Antarctic Ocean collected on early periods of 1961 and 1962, and the comparison was made with the data obtained by other workers.

ANALYTICAL PROCEDURES

40 liters of surface water were collected at each stations shown in Fig. 1 and 40 m*l* of concentrated hydrochloric acid were added as preserver.

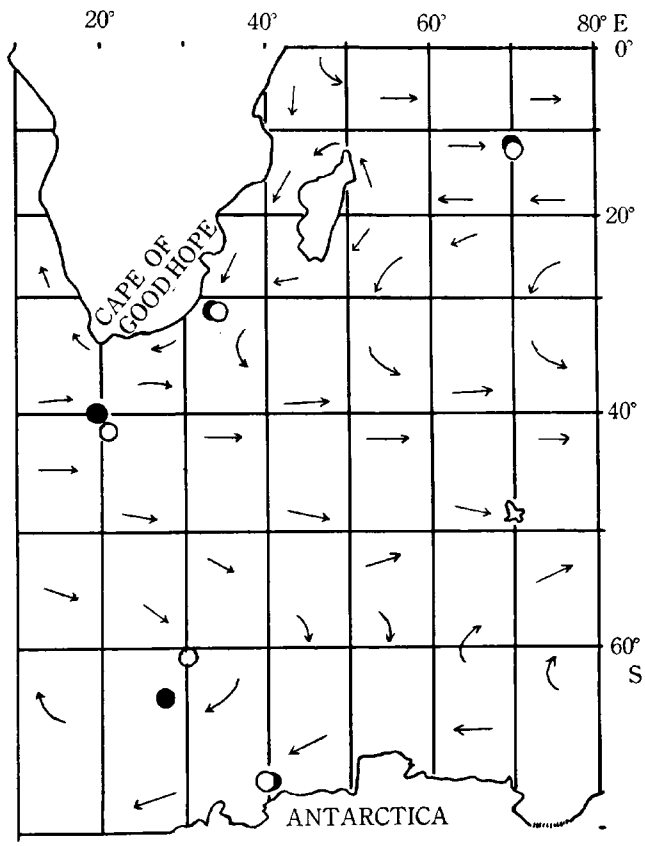

Fig. 1. Sampling stations
Dots : 1961 stations
Open circles : 1962 stations

These stations were located in the area of 10°S to 67°S and 20°E to 70°E, and made five pairs of samples collected on approximately same latitudes and longitudes. Beside, sampling was carried out during the same period (January to March) of 1961 and 1962 except a sample on April 8, 1961. Therefore, these samples are capable to be compared for the latitudinal and annual variations of radioactive contamination without the considerations for the seasonal variation of the fallout.

Sr-90 was separated from calcium by fuming nitric acid method, with natural strontium, after precipitated as carbonates, and measured by the milking of daughter nuclide Y-90. Cs-137 was separated from sea water by use of nickel ferrocyanide and ammonium molybdophosphate, and measured as platinochlorate. Ce-144 was precipitated as hydroxide, and purified by solvent extractions using tri-n-butyl phosphoric acid and methyl-iso-butyl keton. The chemical recovery rate of each nuclide was measured gravimetrically for each sample, and the rates were generally high and ranged from 60% to 89%. The details of analytical procedures were described in previous reports[11, 13, 14].

All the nuclide samples were measured by a 4-pi low-background gas flow counter, having the counting efficiency of 74% for Y-90 (hydroxide ppt.), of 41% to 43% for Cs-137 (platinochlorate ppt.) and of 28% to 34% for Ce-Pr-144 (oxalate ppt.).

The nuclides were identified with their chemical behaviors and decay rate measurements, and in cases of Ce-144 in 1962 samples, gamma ray spectrometry was used to confirm the purity of samples.

The contamintions by reagents and in analytical processes were negligible.

RESULTS

Results of analysis and the latitudinal variations of nuclides are shown in Table 1 and Fig. 2.

Except the 67°S sample in 1962, Sr-90 concentration decreases with increasing latitude, from 0.1 $\mu\mu$c/l to 0.01 $\mu\mu$c/l. Mean Sr-90 concentrations of 1961 and 1962 samples except 67°S are 0.07 $\mu\mu$c/l and 0.06 $\mu\mu$c/l respectively, and as shown in Table 2, no significant increase during 1961 is indicated on each latitudinal pair of samples except 67°S.

Cs-137 concentration, except the 67°S sample in 1962, shows the similar tendency of latitudinal decrease, from 0.21 $\mu\mu$c/l to 0.03 $\mu\mu$c/l, to that of Sr-90. But significant annual increases are indicated on the latitudinal groups of samples and 20% of mean increase during 1961 is estimated for samples except 67°S. The mean concentrations of Cs-137 of 1961 and 1962 samples except 67°S are 0.13 $\mu\mu$c/l and 0.15 $\mu\mu$c/l respectively.

On the contrary to Sr-90 and Cs-137, Ce-144 concentration shows no latitudinal variation except 67°S sample in 1962, and marked increase are indicated on the latitudinal groups. The mean concentrations of 1961 and 1962 samples except 67°S

Table 1. Results of analysis

Latitudinal group	Lat. S	Long. E	Date	Sr-90	Cs-137 $\mu\mu c/l$**	Ce-144*
			1961			
12° S	11°36′	69°50′	Apr. 8	0.11 ± 0.007	0.14 ± 0.02	0.11 ± 0.03
31° S	31°17′	33°39′	Mar. 24	0.08 ± 0.007	0.17 ± 0.02	0.14 ± 0.03
41° S	40°03′	19°29′	Mar. 12	0.06 ± 0.005	0.12 ± 0.01	0.08 ± 0.03
61° S	62°49′	27°25′	Mar. 5	0.02 ± 0.008	0.07 ± 0.02	0.15 ± 0.03
Mean***				0.07 ± 0.003	0.13 ± 0.009	0.12 ± 0.02
67° S	67°05′	40°29′	Jan. 15	0.01 ± 0.005	0.03 ± 0.02	0.14 ± 0.03
			1962			
12° S	12°10′	69°55′	Mar. 20	0.11 ± 0.01	0.21 ± 0.01	0.57 ± 0.04
31° S	31°15′	34°06′	Mar. 10	0.07 ± 0.01	0.16 ± 0.01	0.58 ± 0.04
41° S	41°27′	21°07′	Feb. 24	0.04 ± 0.01	0.11 ± 0.01	0.55 ± 0.04
61° S	60°27′	30°23′	Feb. 18	0.03 ± 0.004	0.11 ± 0.01	0.59 ± 0.05
Mean***				0.06 ± 0.004	0.15 ± 0.005	0.57 ± 0.02
67° S	67°09′	39°47′	Jan. 6	0.09 ± 0.01	0.17 ± 0.01	0.71 ± 0.05

* Values were corrected for date of collections.
** Precision criterion was derived from the square root of the total count.
*** 67° S sample was omitted.

are 0.12 $\mu\mu c/l$ and 0.57 $\mu\mu c/l$, and 500% of annual increase during 1961 is estimated.

Relatively high radioactive contaminations of 67°S sample in 1962 are likely due to the contributions of melting snow and ice containing the fallout precipitated up to 1961, because this sampling station was located in pack ice region and near shore of the Antarctic continent. The lower chlorinity of surface sea water around 67°S station[15], shown in Fig. 3, supports this surmise.

No Ce-141 was detected in Ce-144 samples, and this could be accounted for the fact that the half-life of Ce-141 (32.5 days) is shorter than that of Ce-144 (285 days) and the chemical analysis was carried out 20 months after the collections.

DISCUSSION

By comparison of the sampling data with the chronological data of nuclear explosions, it is made clear that the 1961 samples were collected in the last period of nuclear moratorium, and on the other hand, the 1962 samples were collected after the resumption of USSR nuclear explosions in September, 1961 on Novaya Zemlya, and before the beginning of US nuclear tests on the equatorial Pacific in April, 1962.

Therefore, as the fallout produced by the nuclear explosions prior to the moratorium was reported to be very low on the southern hemisphere during 1960 through 1961[2,3,4,7,8] and the nuclear explosions in Sahara in 1960 and 1961 were of

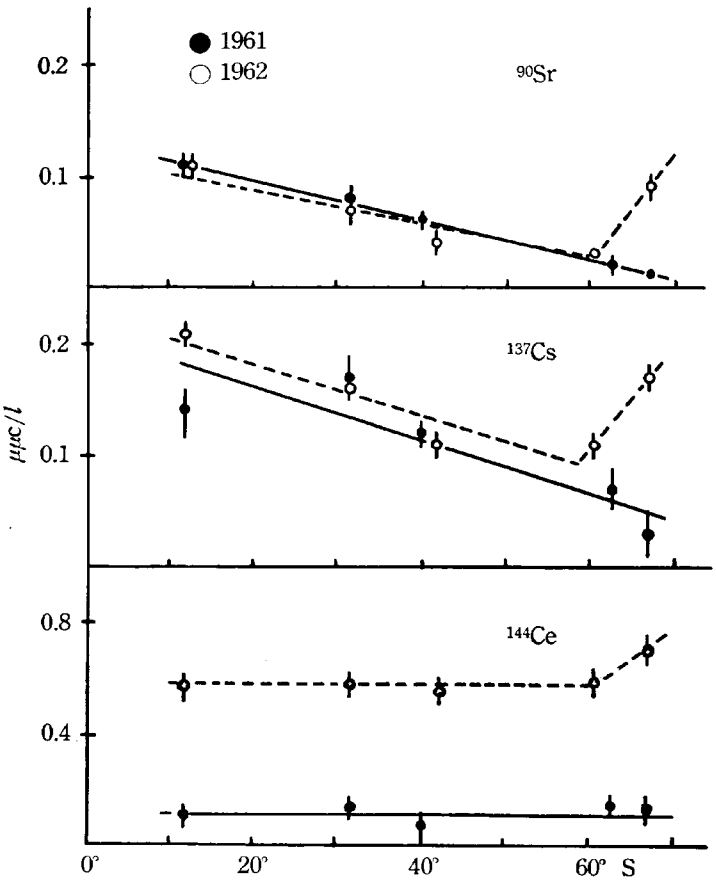

Fig. 2. Latitudinal distributions of nuclides in the Indo-Antarctic Ocean

Table 2. Rates of increase in the nuclide concentrations

Latitudinal group	Sr-90	Cs-137 1962 ~ 1961	Ce-144
12° S	1.0 ± 0.1	1.5 ± 0.2	5.2 ± 1.5
31° S	0.9 ± 0.3	0.9 ± 0.1	4.1 ± 0.9
41° S	0.7 ± 0.2	0.9 ± 0.1	6.9 ± 2.4
61° S	1.5 ± 0.5	1.6 ± 0.5	3.9 ± 0.9
Mean*	1.0 ± 0.2	1.2 ± 0.1	5.0 ± 0.8
10-40°N**	1.0 ± 0.04	1.6 ± 0.05	3.4 ± 0.1

* 67° S samples were omitted
** North Pacific sea water[16]

tropospheric scale resulting no significant fallout on the Indo-Antarctic Ocean, highly significant increase of radioactive contamination in the samples of each latitudinal group is supposed to be due to the fallout produced by USSR nuclear tests on the northern polar region in late 1961.

Rocco and Broecker[12] reported Sr-90 concentration in the southern hemisphere surface sea waters, which were collected in early 1961, as 0.03 to 0.04 $\mu\mu c/l$, and these values are rather lower than those obtained by authors for the Indo-Antarctic surface water on the same period. Miyake[17] estimated the average Sr-90 concentration of 0.05 $\mu\mu c/l$ in the southern hemisphere

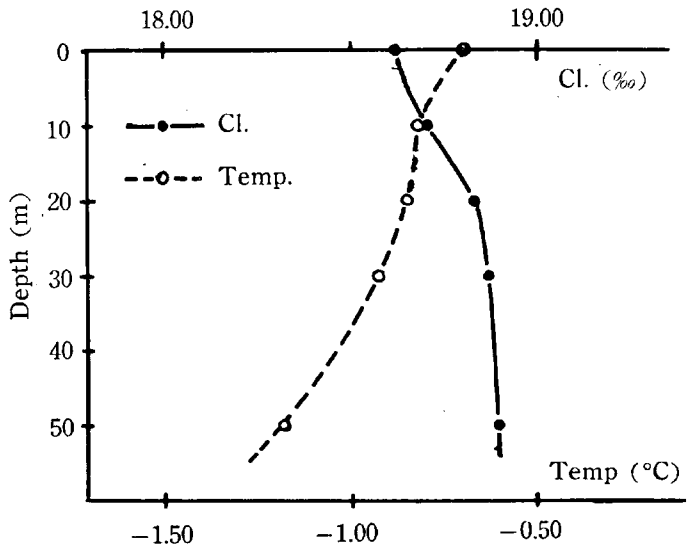

Fig. 3. Vertical distributions of mean chlorinity and mean temperature around the 67°S station during January to March

sea water, which was originated from the stratospheric fallout prior to 1961, assuming the thermocline thickness of 100 meters and that the most of radioactive fallout remains within the thermocline for a few years. This estimation agrees well with the average Sr-90 concentration of 0.07 $\mu\mu c/l$, in the Indo-Antarctic sea water collected in early 1961, and slight difference of the concentrations could be accounted for the gradient of Sr-90 distribution within the thermocline.

As no significant annual increase of Sr-90 is indicated on each latitudinal group of samples, it is clear that Sr-90 in the fallout during 1961 was of such low level that only able to compensate the losses by the vertical diffusion and mixing of water masses. This results agrees with Sr-90 deposition data on the southern hemisphere[2,3,4,7,8] during 1961, which are reported as approximately 0.5 to 1 mc/km^2, as shown in Table 3, and could be converted to 0.005 to 0.01 $\mu\mu c/l$ of increase in sea water within the the thermocline of 100 meters deep.

The mean Cs-137 concentrations of 0.13 and 0.15 $\mu\mu c/l$ in the Indo-Antarctic surface water in early 1961 and 1962 are higher than those reported by Folsom

Table 3. Sr-90 deposition on the earth's surface during 1961* (mc/km^2)

Northern	80–50°N	50–40°N	40–30°N	30–20°N	20–10°N	10–0°N
hemisphere	1.37	2.05	1.64	1.37	0.64	0.75
Southern		50–40° S	40–30° S	30–20° S	20–10° S	10–0° S
hemisphere		0.72	0.96	0.87	0.52	0.58

* Data from HASL-131[8]

and Mohanrao[9] (0.02 $\mu\mu c/l$ in the Indian ocean during 1959 to 1962), and by Rocco and Broecker[12] (0.03 $\mu\mu c/l$ in the Antarctic and 0.04 $\mu\mu c/l$ in the South Atlantic on early 1961), but agrees fairly with that reported by Schroeder and Cherry[10] (0.09 $\pm$ 0.05 $\mu\mu c/l$ in the seas off the Cape of Good Hope in mid 1961).

Besides, mean Cs-137/Sr-90 ratio of 1.8 $\pm$ 0.2 in the Indo-Antarctic surface water on early 1961 agrees with those reported by Rocco and Broecker[12] for the southern hemisphere surface water in the same period, namely, 1.8 $\pm$ 0.2 in the South Atlantic, 1.2 $\pm$ 0.2 in the Antarctic and 1.6 $\pm$ 0.2 in the eastern South Pacific.

The increase of 0.02 $\mu\mu c/l$ of average Cs-137 concentration between 1961 and 1962 samples corresponds to 2 mc/km^2 of fallout Cs-137 during 1961, and this value agrees well with that estimated from Sr-90 deposition data, using Cs-137/Sr-90 ratio of 1.7 to 2.5 for the stratospheric fallout[18].

Assuming the ratio of Ce-144 depositions between the southern hemisphere and the northern hemisphere is similar to that of Sr-90 deposition, the average Ce-144 deposition rate in 10°S-50°S area during 1961 to early 1962 is estimated as 46.9 mc/km^2, as shown in Table 4. Although Ce-144 tends to become particulate or colloidal form in the natural sea water and to sediment rapidly with the debris on which radioactive cerium is attached[19], but assuming the newly deposited Ce-144 on the sea, namely, those deposited after September, 1961, remain within the thermocline at least for a few months, it is estimated that the mean Ce-144 concentration in the surface water of the southern hemisphere on March, 1962 was 0.44 $\mu\mu c/l$. This value agrees fairly with the mean Ce-144 concentration of the Indo-Antarctic sea water.

The latitudinal variation of southern hemisphere fallout during the moratorium is reported to be rather similar, but with lesser significance, to that of the northern hemisphere fallout; namely, more fallout on mid latitude zones than on low latitude [8, 20, 21]. But according to the Sr-90 and Cs-137 data of this report, the lower latitude zones were likely to be accumulated with more artificial radioactivity during the moratorium than the higher latitude zones. This is attributed to the transports of meridional direction by the currents and the changes by the divergence and

Table 4. Estimated Ce-144 deposition on the southern hemisphere during 1961 to early 1962

Mean Sr-90 deposition* (MC)			Mean Ce-144 deposition in 30-40°N**			Estimated Ce-144 deposition in 10-50°S	
30-40°N	10-50°N	ratio		mc/km^2	MC	MC	mc/km^2
0.06	0.12	0.5	Jan.-Aug.	5.76	0.21	0.42	2.77
			Sept.-Feb.	91.75	3.35	6.70	44.17
			Total	97.51	3.56	7.12	46.94

* HASL-131[5]

** HASL-131, Westwood, New Jersey[8]

convergence in the Indo-Antarctic Ocean, as shown in Table 1, resulting the modification of the latitudinal distribution of fallout nuclides in surface water during this period.

As the fallout produced on the polar region in 1957 and 1958 were not reported in the southern hemisphere environment[1,2,3,4,7], above mentioned Sr-90 and Cs-137 in sea water are supposed to be mainly due to the nuclear explosions on the equatorial Pacific.

Uniform latitudinal distribution of Ce-144 in the Indo-Antarctic sea water of 1961 is likely to be caused chiefly by its tendency to sediment rapidly as mentioned previously[19].

Although the estimated patterns of Sr-90 and Cs-137 depositions during January, 1961 to March, 1962 agrees with those of the fallout reported by other workers [2,3,4,5,6,7,8], intense increase of Ce-144 concentration of the Indo-Antarctic surface water indicates the highly significant deposition of relatively "young" fallout.

As mentioned previously, during the moratorium to March, 1962, the nuclear explosions with high fission yield, which are able to inject large amounts of fission products into the stratosphere were only of USSR in September to November, 1961 on the northern polar region, and the fallout released by these explosions are reported to be remarkably rich in radioactive cerium[22,23]. Therefore, the increase of Ce-144 in the surface water is likely due to the fallout originated from the resumption of USSR nuclear explosions.

This estimation suggests the minimum length of time of 6 months or less for meridional transport of the stratospheric fallout debris from the northern polar region to the southern hemisphere, and this value is about half of those estimated from Sr-90 deposition data by other workers[3,6,7].

The stratospheric fallout by 1957 and 1958 nuclear explosions in the equatorial Pacific or in the northern polar region are supposed to be too "aged" to cause the Ce-144 increase.

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