On the Distribution of Radioactivity in the Sea around Bikini Atoll in June, 1954.

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Abstract

By the radioactivity in the air and sea water caused by the US hydrogen bomb tests in the area of Bikini-Eniwetok Atolls, 1954. The radioactivity contamination to fishes and fishing boats were brought about. The observation of the actual state of the radioactivity around Bikini-Eniwetok Atolls was carried out aboard the survey boat "Shunkotsu-maru". The maximum radioactivity in sea water was 7025 cpm/l at 570 km west of Bikini and the influence of the radioactivity was recognized as far as about 2000 km from Bikini Atoll. Some oceanographic discussions on the horizontal as well as vertical distributions of the radioactivity in the sea are given.

1. Introduction

Leaving Tokyo Harbour on 15th May, 1954, the observation were carried out until 4th July aboard the survey boat the "Shunkotsu-maru" belonging to the Ministry of Agriculture and Forestry in the sea area near Bikini Atoll where H-bomb tests had been done by U. S. Atomic Energy Commission, to make clear the state of the radioactive contamination to living matter, sea water, the air etc. The purpose of this "Japanese Bikini Expedition" was to obtain a counter-plan against the serious damage fell on the Japanese pelagic fisheries due to the radioactivity carelessly scattered in this sea area since the unfortunate affair of the fishing boat the "Fuku-ryu-maru", the "Lucky Dragon" on 1st March, 1954.

In this expedition the present authors took charge of making a plan and observations on the radioactive contamination of sea water and the air. Two of the authors (SUGIURA and KAMEDA) participated in the expedition as observers. The fairly strong radioactivity was detected in wide area around Bikini Atoll, though U. S. Atomic Energy Commission had declared that the presence of the radioactivity in the area was negligible [1]. In this paper the authors intend to report the results of observations and some oceanographic discussions regarding the distributions of the radioactivity near Bikini Atoll.

2. The method of measurement of radioactivity in sea water

It is difficult to measure the radioactivity in sea water directly by simple evaporation method owing to its high salinity, therefore, the carrier method has been adopted. After adding 2 gr of solid ammonium chloride, 1 ml of the aqueous solution of iron alum (86.3 gr/l) and 1 ml of barium chloride solution (17.8 gr/l) to 1 litre of sea water, water is heated to 60~70°C stirring well. A few drops of an alcoholic solution of phenol-phthalein (1%) are added as an indicator, then ammonium

hydroxide solution (1:1) is carefully dropped in with a pipet until a faint pink colour appears. After two minutes boiling, the precipitate is settled on standing for a couple of hours at room temperature. The precipitate is filtered under suction on a round filter paper (dia. 3.5 cm) laying above the glass filter (Fig. 1). The precipitate remained on a filter paper is dried in an air bath or a desiccator holding in a small brass case (Fig. 2) to avoid the folding of the surface. Measurements of radioactivity are done by putting the precipitate kept in a brass case under a mica window of G-M counter. The G-M counter used aboard the "Shunkototsu-maru" is Radiation Counter, Model 32 manufactured by Science Research Institute Ltd., Tokyo. The thickness of mica window is 3.7 mg/cm^2 and the distance between the mica window and the surface of the precipitate is 1.5 cm. After the measurement has been done, the precipitate is covered by a polyethylene film to protect the surface. It was confirmed that at least 70~80% of the total activity in sea water could be transferred to the precipitate by preliminary tests using the fission materials fell aboard the fishing boat the "Fukuryu-maru". Therefore, the actual activities might be 25~40% higher than observed values. To obtain the counting efficiency the aqueous solution containing Ce 144 of which activity had been referred to the

Fig. 1. The apparatus for filtration.

Fig. 2. A small brass case to avoid the folding of the surface of a filter paper.

standard RaE was treated chemically as mentioned above, and its radioactivity was measured under the same geometrical conditions. Thus, the efficiency of the measurement was found to be 7.7%, that is, 1000 counts per minute is equivalent to about $5.9 \text{ m}\mu\text{C}$. The maximum activity in sea water obtained aboard by this method was 7025 cpm/l which was found at the depth of 75 m, at 450 km west of Bikini Atoll on 21 st June.

The second was 6050 cpm/l found at the depth of 20 m at 150 km west of the atoll on 12 th June. It was confirmed that most parts of the radioactive materials were dispersed in water as true solutions, ionic or eu colloidal, since more than

99% of the radioactivity passed through a filter paper (pore size 0.5μ , No. 4 Filter Paper, Tōyō Filter Paper Manufacturing Co., Tokyo).

It is rather surprising that the activity in sea water near Bikini and Eniwetok Atolls was often stronger than, or at least comparable to, that of White Oak Lake receiving waste effluent from nuclear reactors of Oak Ridge National Laboratory USA. The latter was 2710~5190 cpm per litre on an average [2].

3. The distribution of radioactivity in sea water around Bikini Atoll

a) Horizontal distribution

Fig. 3 shows the horizontal distribution of the activity in the surface water near Bikini Atoll. There must have been some changes in the activity due to the time variation of the sea condition, the natural decay and the loss of the radioactivity during the course of observation. However, since the causes of changes were much complicated, and the correction were almost impossible, the distribution was drawn by using directly the observational data without giving any correction.

As shown in Fig. 3 the radioactive water extended on the WNW direction leaving near Bikini Atoll while a branch spread out to WSW. The cause of this branching is not clear. But, as explained below, it seems that there was a weak discontinuous boundary along the line passing Bikini and Eniwetok Atolls. The water B_1 (Fig. 4) on the northern side of this line had on an average the

Fig. 3. The horizontal distribution of the radioactivity of the surface water. Number expresses the radioactivity in cpm/l.

Fig. 4. The course lines of observation, the water systems, and the current speed and its direction.

temperature, 27.1°C, the chlorinity, 19.23‰ and σ_t , 22.51 at 50 m depth. On the other hand, the water system B_2 on the southern side of the line had T, 27.0°C, Cl, 19.04‰ and σ_t , 22.29. Thus, it seems that there was a slight but definite jump in the properties between two water systems, which might be one of the causes of the branching.

The radioactive water flowing on the main direction of the North Equatorial Current extended sooner toward the west than in the southern part where the flow rate was smaller because the latter was near the boundary between the North Equatorial Current and the Equatorial Counter Current and there was also the oppression due to the north-eastward inclination of the Equatorial Counter Current. Thus, on the WNW direction the activity of 100 cpm/l reached as far as 2000 km from Bikini Atoll, while on the WSW direction the extension was only 1000 km.

It is to be noted that the radioactivity was also detected at the position 700~800 km from Bikini on the ENE direction. This is the end part of the radioactivity distribution turning to the east at about 350 km north of Bikini Atoll (Fig. 4). To clarify the cause of this turning on the backward direction of the North Equatorial Current may be one of the important problems.

Though it is not clear at present, whether it was the extension of the easterly current along the northern boundary of the North Equatorial Current which was found recently between 180°E~140°W [3] or it was only the local eddy, several discussions will be done below on this point.

When we investigate the horizontal distribution of water systems near Bikini Atoll by using the T-S diagrams of waters at the depth of 50 m below the surface, we can find out that there are at least four different types of waters (Fig. 4). The first one A is highly saline water (chlorinity, 19.3‰, water temperature, 27.5°C, σ_t , 22.77) covering the area northern than 15°N.

In the southern part of the atoll there is the second type of water C which belongs to the Equatorial Counter Current with smaller chlorinity (Cl, 18.9‰) and higher temperature (T, 28.0°C). The third one B belongs to the North Equatorial Current representing the intermediate property between A and C . In the southern part of the water C , there extends the water D of the South Equatorial Current with higher chlorinity (Cl, 19.6‰) and higher temperature (T, 28.0°C).

The water B may be classified into two sub-groups B_1 and B_2 as mentioned above. As shown in Fig. 4 the east part of B was comparatively broad and it became narrower on approaching to the west. It was probably due to that at the time the boundary between waters B and C somewhat inclined to the north. Therefore, the North Equatorial Current was pressed by the Equatorial Counter Current and the backward current flowing toward the north-east direction of Bikini Atoll would have been resulted.

The observation of the current direction obtained *in situ* also suggests the presence of counter or eddy current in the same area. It is, however, not sure whether this current would form the origin of the general easterly current as mentioned above.

On the other hand, a remarkable loop of the isochlor lines shown in the surface distribution of the chlorinity (Fig. 5) in the northern area of Bikini also indicates the presence of eddy flow.

As shown in Figs. 6 and 7, in the vertical distributions of phosphate and the dissolved oxygen along 15°N there was the subsidence in the area near 15°N,

165°E which was probably due to the anticyclonic eddy.

b) Vertical distributions of the radioactivity in sea water

Figs. 8, 9 and 10 show the distributions of radioactivity of sea water on the vertical section perpendicular to the North Equatorial Current along the lines respectively at 150 km, 570 km and 1300 km west of Bikini Atoll. The profile at 150 km west of Bikini shows already the branching of the activity in the direction of N-S. The dotted lines in the figures represent respectively the latitude of the sites of Bikini and Eniwetok Atolls. The strong activity is separated on both sides of these dotted lines and the distance of two maxima is about 100 km. The depth which shows the activity of 100 cpm/l is at about 80 m below the surface in the northern and southern parts and in the middle part it sinks down to about 120 m. It is noteworthy that the activity decreases rapidly down to only 28 cpm/l at 200 m depth. This corresponds to the depth of thermocline which was about 150 m in this area.

The estimated amount of flow of radioactivity was about 1×10^5 curie/hour passing through this section. On the vertical section at 570 km west of Bikini the distance of branching became broader and also the depth of 100 cpm/l activity was 200 m below the sea surface on an average. The flow of radioactivity was about 0.6×10^5 curie/hour. The distance between the former and the latter section was about 400 km and assuming that the speed of current was about 0.7 knots

Fig. 7. The vertical distribution of phosphate P in $\mu\text{g atoms/L}$.

Fig. 8. The vertical distribution of the radioactivity in sea water in the section perpendicular to the North Equatorial Current along the line at 150 km west of Bikini Atoll.

Fig. 9. The vertical distribution of the radioactivity in sea water in the section perpendicular to the North Equatorial Current along the line at 570 km west of Bikini Atoll.

Fig. 10. The vertical distribution of the radioactivity of sea water in the section perpendicular to the North Equatorial Current along the line at 1300 km west of Bikini Atoll.

on the direction perpendicular to the sections it would have taken about 2 weeks to flow between them. Thus, the activity passing through both sections would have been about the same when the rate of decay during two weeks was taken into consideration.

4. The relation between the radioactivity in sea water and the radioactive fallout

The fishing boat the "Fuku-ryu-maru" was situated at the position 150 km east of Bikini Atoll in the early morning on 1st March when her crews recognized small, white dust particles falling on the deck. The density of dusts covered the surface of the deck was not clearly known, however, it may be supposed that it was about 0.5 gr/m² or so. On the other hand, the radioactivity of the dust particles was regarded as about 1.4 curie/gr at the time of falling (3 hours after the detonation) [4]. Assuming that the mean diameter of dust particles was 0.1 mm, the rate of sinking of such particles in sea water is about 30 m/hour. Then, it takes about 1~1.5 sec to pass the water layer of 1 cm. Here, if we assume that at most 0.01% of the radioactive substances in the dusts could dissolve into sea water in one second, most part of radioactive materials would have removed to water from dusts before they reached 100 m depth below the sea surface. Thus, the mean activity in water near the sea surface around there would have been 3.1×10^{-6} curie/l at the time five or six hours after detonation. Needless to say, this was the activity of sea water in the area about 150 km apart from Bikini Atoll, therefore, the activity near and in the atoll would have been much more stronger. On the other hand, it was confirmed by the records of micro-barographs and tide gauges obtained in Japan [5] that the detonations had been carried out four or five times since 1st March (1st March, 27th March, 6th Apr., 26th Apr. and 5th May), it would be no wonder the wide sea area far from Bikini Atoll had been contaminated by fission materials as strongly as it was actually observed aboard the "Shunkotsu-maru" in June, 1954.

5. The property on the radioactive substances in sea water

The relation between the gross radioactivity $A_{(t)}$ of the fission materials and the time after detonation may be simply expressed by the next equation,

$$A_{(t)} = ct^{-\alpha}$$

where c and α are constants. From the decay curve of the radioactivity of the precipitates prepared aboard the "Shunkotsu-maru", it has been found that α is confined within the comparatively narrow limits of 1.3~1.6 (mean, 1.5) assuming the date of detonation was 5th May. In normal cases, α is about 1.2 [6], whereas it was 1.37 for the radioactive dusts fell on the deck of the "Fuku-ryu-maru".

In sea water some of the radioactive elements contained in the original fission materials might be preferentially precipitated owing to pH of sea water, (about 8.2~8.3). Therefore, the chemical composition should be somewhat different from the original fission materials, which might be the cause of the change in the value of α .

The radiochemical analysis of the fission materials in sea water have been furnished also by the present authors which will be reported in near future. In Table 1 the full data of the gross activity of sea water observed aboard the observation vessel are given.

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References

- [1] US Atomic Energy Commission, 1950: Radiobiological Survey of Bikini, Eniwetok and Likiep Atolls, July-August, 1949. AECD-3446.
- [2] Radioisotope Uptake by Aquatic Plants and Animals Living in Waste Effluents of Oak Ridge National Laboratory; A paper presented to U.S.-Japan Radiobiological Conference held in Tokyo, 1954.
- [3] CROMWELL, T., 1953: Circulation in a Meridional Plane in the Central Equatorial Pacific. Jour. Mar. Res., 12, p. 196.
BARNES, C.A., BUMPUS, D.F. and LYMAN, J., 1948: Ocean Circulation in Marshall Island Area. Trans. Amer. Geophys. Union, 29 (6).
- [4] KIMURA, K. *et al.*, 1954: On the Radioactive Substances Having Precipitated on the Fishing Boat the "Fuku-ryu-maru". Japan Analyst, 3, p. 335. (in Japanese).
- [5] YAMAMOTO, R., 1954: The Microbarographic Oscillations Produced by the Explosions of Hydrogen-Bombs. Bull. Inst. Chem. Res. Kyoto Univ. The Radioactive Dust from the Nuclear Detonation.
Central Meteorological Observatory, Tokyo, 1954: A Preliminary Report of Observations on the Atmospheric and Oceanic Oscillations caused by H-Bomb Tests. (in Japanese)
- [6] HUNTER, H.F. and BALLOU, N.E., 1951: Fission-Product Decay Rates. Nucleonics, 9 p. C-2.

Table 1. Radioactivity in sea water at various depths near Bikini-Eniwetok Atolls, 1954.
1000 cpm is equivalent to about 5.9 μC .

Date	St. No.	Latitude Longitude	Depth <i>m</i>	Activity cpm/l	Date	St. No.	Latitude Longitude	Depth <i>m</i>	Activity cpm/l	
May, 31	6	13°51'N 172°56'E	0 100 210 430	450 34 5 0	June, 12	17-7	11°59'N 163°53'E	0 20 40 50 60 86 105 200	5540 6050 923 356 375 55 15 64	
June, 1	7	10°44 173°03	0 5 10 20 30 49 92	25 27 27 27 27 36 0		17-8	12°09 163°57.3	0 20 40 50 60 82 102 196	260 1091 452 580 346 39 32 49	
12	17-1	10°55 163°51	0 25 54 75 98 210 300	84 262 5780 67 25 28 23		13	18	13°23 164°25	0 20 40 50 60 83 101 193	70 57 61 84 58 38 10 59
	17-2	11°17.3 163°49.3	0 20 40 50 60 94 105 192	1610 982 169 50 23 12 8 27		14	19	15°57 165°22	0 25 50 100 213 394 576	654 603 420 24 64 16 0
	17-3	11°18 163°50	0 20 40 50 60 83 101 194	566 810 1160 503 273 1283 368 11		19	20	15°03 163°15	0 25 50 75 106 199 480	478 426 408 22 8 0 0
	17-4	11°29 163°51	0 20 40 50 85 100 195	859 641 1563 1683 840 939 59		20	21-1	12°42 161°26	0 25 50 75 106 193 489	1260 1233 331 187 24 34 48
	17-5	11°39 163°52	0 20 40 50 60 86 105 202	1539 1841 1792 1221 517 182 25 65		21-2	12°18 161°03.5	0 25 50 74 93 175	1829 1684 1237 1840 343 20	
	17-6	11°49 163°52.5	0 20 40 50 62 80 95 200 290 475	1792 1875 516 470 216 48 20 50 110 116		21-3	11°54 160°46	0 25 50 79 102 195	457 680 1242 19 14 83	

Date	St. No.	Latitude Longitude	Depth <i>m</i>	Activity cpm/l	Date	St. No.	Latitude Longitude	Depth <i>m</i>	Activity cpm/l
June, 20	21-4	11°30'N 160°29'E	0	421	June, 23	23	05°03'N 155°53'E	0	16
			25	487				26	7
			48	1056				50	9
			70	879				72	6
			98	168				99	5
			180	105				189	13
	21-5	11°05 160°12.5	0	360		24	03°24.5 155°31	500	13
			24	480				0	0
			48	1400				25	0
			82	7025				49	5
			103	1620				72	9
			194	231				106	7
	21-6	10°42 159°55	0	93		26	08°59 152°51	170	0
			25	46				515	0
			49	390				0	10
			81	464				25	10
			104	55				49	0
			206	130				73	0
21	22-1	10°21 159°35	0	208		27	12°00 151°10	95	tr.
			10	208				180	tr.
			25	235				565	0
			48	775				0	10
			70	330				25	7
			80	60				50	9
			115	44				75	69
			200	95				102	7
			280	38				193	tr.
			360	6				485	tr.
	22-2	09°58.5 159°15	405	13		28	14°46 149°56	0	350
			485	22				25	185
			715	43				49	184
			0	425				72	180
			25	905				106	0
			50	1225				185	0
			75	295				450	0
			96	240		29	18°00 148°38.5	0	7
	22-3	09°35 158°57	184	97				27	3
			0	1890				51	3
			25	2016				74	3
			48	1349				110	3
			76	349				186	2
			99	120				495	0
	22-4	09°13 158°41	198	25		30	21°00 147°21	0	0
			0	78				25	7
			25	850				50	10
			49	394				74	5
			73	157				98	0
			95	157				198	0
			175	0				525	0
			455	2					