Radioactivity in the River Water, Brackish-Water, Sea Water and the Laver at River Mouth, Derived from Radioactive Rain Water

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(Tables 1–3; Text-figs. 1–4)

INTRODUCTION

The contamination of the river water by fission products of atomic bomb tests has been surveyed by HIYAMA *et al.* in Japan.[1] But they could not find out any remarkable radioactivity in the river water. And yet nothing has hitherto been reported concerning the effects of radioactivity which derived from fission products in the river water caused through rainfall upon laver (*Porphyra tenera* KJELLM.) which was cultured at river mouth.

In 1954, ARASAKI & NOZAWA[2] surveyed the contamination of laver by radioactivity of fission products, cultured at the northern part of Tokyo Bay and near the river mouth of Shiogama Bay, and no radioactivity was found out.

In this investigation, from Apr. 1956 to Feb. 1957, the radioactivity of rain water has been surveyed. Then it was made evident that considerable fission products were contained in every rain water that they have recently had. On the other hand, a radiological investigation into the river water was carried out, while the investigation was continued about the radiological contamination of the sea water sampled at a few stations in the Seto Inland Sea and at one station in the Strait of Hōyo. And whenever the rain containing fission products was falling, radioactivity in the water of the river Ashida was found out. In order to investigate the state of radiological contamination of the river water in detail, the water of the river Ashida was sampled very carefully for three months from Dec. 1956 to Feb. 1957 and the amounts of its radioactivity were measured. And at the same time the brackish-water at the mouth of the river Ashida and the sea water off the mouth of that river were surveyed.

It was reported by SAITO & SAMESHIMA (1956)[3] and TANAKA (1956)[4] that in their laboratory the marine algae immersed in the sea water containing fission products were remarkably contaminated by radioactivity. A radiological investigation of the laver, cultured at the mouth of the river Ashida, was conducted because it seemed probable that the laver, cultured at the mouth of the river Ashida, might have been exposed to radioactive river water.

MATERIAL AND METHODS

In order to investigate radioactivity in the water of rain and the river, samples were taken from Apr. 1956 to Feb. 1957 as follows. Rain water was gathered in a glass basin using a vinylon sheet (1 m. × 1.5 m.) in the yard of the writers' laboratory in the vicinity of Fukuyama City. In this case, the water of a litre of each rain which fell was sampled at the

very beginning of each rainfall. The water of the river Ashida was also sampled from the surface of the stream at the following survey point at every few hours immediately after it began to rain. These samples of the water of rain and the river were treated at once as described below, and then they were counted. The survey was continued till the radioactivity could not be found out in the river water.

A survey point was selected at Kashima-bridge base on the river Ashida, which flows through the western part of Fukuyama City into the Seto Inland Sea, about 8 km. upstream from the mouth of the river. The river has the width of about 300 m., the depth of water about 50 cm., the volume of running water about 10 m^3 per second and the width of its watercourse about 50 m. as usual. And the river water sampled there was always fresh and free from any influence of sea water. Of course when the rain did not fall, or in case of the amout of rainfall was less than about 5 mm., the river water always remaind clear. It was scarcely muddy when the amount of rainfall was about 10 mm., while it was somewhat muddy when the amount of rainfall was about 40 mm., which was the maximum value while the survey was in progress. The survey point on the river was located 8 km. away from the writer's laboratory.

A litre of water was heated and condensed respectively in a large beaker. Then it was poured into a small beaker and was condensed up to 10 ml. or so. Being replaced drop by drop into a planchet, it was dried up. Moreover, a small quantity of $\frac{1}{2}$ N HCl which was used for washing the small beaker was added to the planchet and dried up. The planchet made of stainless steel, which was 6.24 cm^2 in area, was used. When the river water was somewhat muddy, 200 ml. of the sample as well as a litre of the river water was treated. Being dried up in the planchet under infra-red lamps, the sample was counted using the G.–M. counter. Self-absorption in the sample was very small at the densities in the planchet.

In these investigations, the treated samples of the water of rain and the river have been kept in the constant conditions to obtain the rate of physical decay in the radioactivity of the samples. To determine whether or not the radioactive substances found out in the water of rain and the river Ashida are the same, decay curves of both samples were prepared.

The brackish-water at the mouth of the river Ashida and the sea water some miles away from the river mouth were sampled when it was raining or within a few hours after the rain stopped. Three survey stations at the river mouth were selected along the centre line of the stream at a distance of 500 m. upstream from the mouth of the river. The sea water was sampled three times a month at the three stations located each at a distance of 1 mile from the mouth of the river Ashida. These stations were situated at the northern part of the Island of Hashiri in the Seto Inland Sea. The samples of the sea water were treated as follows:

Each solution of NH_4Cl(2g/l), $FeNH_4(SO_4)_2 \cdot 12H_2O$(86.3g/l) and $BaCl_2 \cdot 2H_2O$(17.8g/l) was added in order 1 ml. by 1 ml. to the sample of a litre of the sea water and was heated up to 60°~70°C, and then aqueous solution of ammonia was added to it till it showed a little pink colour, using phenolphthalein as indicator. After having been boiled for a few minutes, it was cooled down to the room temperature. Then it was filtered through the Tōyō filter paper No. 5A. The deposit on the filter paper was dried carefully under infra-red lamps and then the deposit was counted as it was on the filter paper.

To investigate the contamination of the living laver cultured at the mouth of the river Ashida, the writers measured the amouts of radioactivity of the living laver collected during

the rainfall or within a few hours after the rain stopped and that of the living laver collected many days after it had stopped raining. In this study the samples of the living laver were taken at random from the collectors for laver which were laid at the mouth of the river Ashida. And at the same time the brackish-water near the collector was taken as one of the preceding samples. The samples of the living laver were put in the ceramic crucibles and then they were dried under infra-red lamps, keeping the temperature of crucibles about 110°C. The samples in the crucibles were put in a muffle furnace, and then they were heated overnight at about 550°C. The ash obtained in crucibles was ground and spread evenly on the planchet with the aid of distilled water, and it was dried under infra-red lamps and counted.

In order to investigate the dried laver, "asakusanori", one of the favorite Japanese foods, exposed to radioactive river water during its living periods, the writers measured the amounts of radioactivity in the ash of the two samples of dried laver which were cultured in 1954 and 1957 at the mouth of the river Ashida. One of the samples was cultured and dried into "asakusanori" in Feb. 1954 before the atomic bomb tests at Bikini Atoll in the Marshall Islands, the other was collected and dried into "asakusanori" on 6th Feb. 1957 after the atomic bomb tests on the same grounds. Considering the natural radioactivity of K^{40} included in laver, the amouts of radioactivity of both samples were compared with each other.

In the case of the dried laver, "asakusanori", it was broken into flinders and put in the ceramic crucible. It was ashed in a muffle furnace in the same way as in the case of the living laver. The ash was weighed and transferred into a planchet and treated by the same method as in the living laver, and then it was counted.

In the ash of laver, self-absorption curve was prepared to determine the self-absorption of the ashed sample of laver.

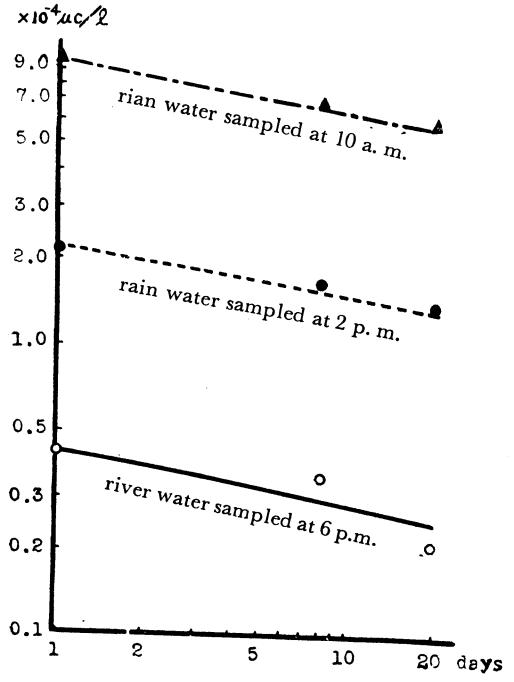
The radiation counter (G.-M. tube system) used in this measurement is a product of Tokyo Shibaura Electric Co., Ltd. Its form is RGD-100-B. The weight of mica of this G.-M. counter tube is about 1.9 mg/cm^2. The distance from the window of mica to the sample was always kept 10 mm.

The values for the amounts of radioactivity of the sample were determined by making clear the relation between curies and counts/minute obtained by measuring the amounts of radioactivity of a standard source produced by Scientific Research Institute. The standard source was put into the planchet and was counted at 10 mm. off the mica of the window of the G.-M. counter tube in the same way as the samples were counted and the counts/minute were compared with known numbers of disintegration per minute of this standard source and counts/minute could be readily converted into microcuries. The correction was made for the geometrical difference between the standard and the sample. It was also made for back-scatter, but no correction was made for self-scatter. The relation between microcuries and counts/minute was determined by the next formula:

$$M\mu c = 4.02 \times 10^{-6} \times Mc.p.m. \text{ (net counts)}$$

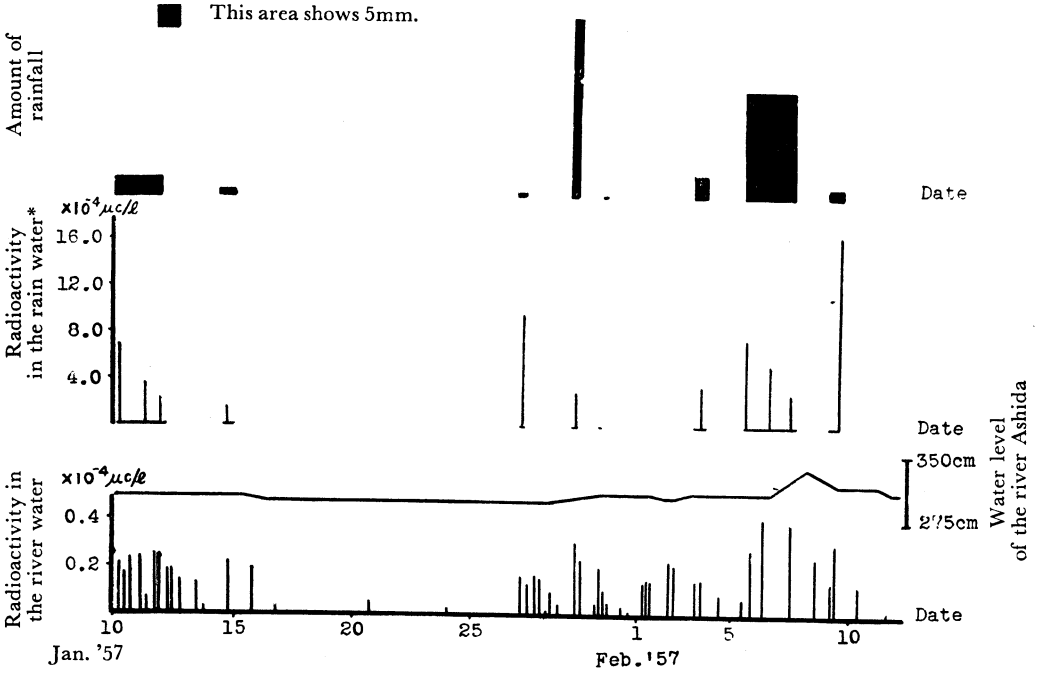
RESULTS AND DISCUSSION

A) Radiological contamination of the river water by rain water including fission products



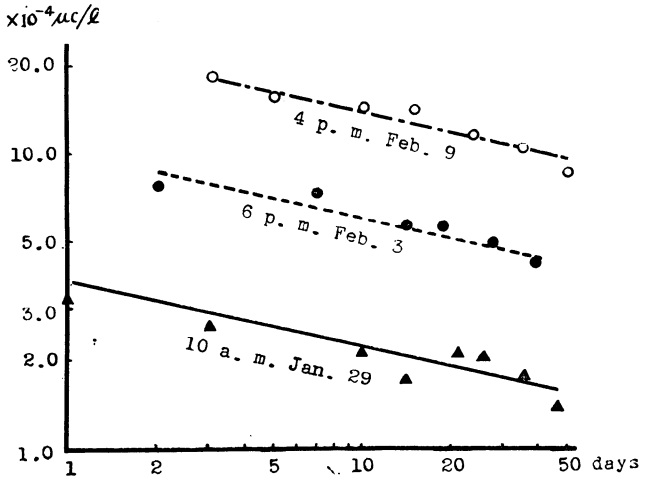
Text-fig. 1 shows the physical decay of radioactivity in the sample of the rain water and hat of the river water on 4th Dec. 1956. In Text-fig. 1, the slopes of the three decaying curves obtained by treating the water of the rain and the river sampled at 10 a. m., 2 p. m. and 6 p. m. respectively on the same day of Dec. 1956 were so similar that the radioactivity, found out in the river water, would be derived from fission products of atomic bomb tests which were contained in the rain water.

Text-fig. 1.
Decay curves of radioactivity of fission products in the water of the rain and the river Ashida sampled on 4th Dec. 1956.



Text-fig. 2. Radioactivity in the rain water and the river warer, water level of the river Ashida and the amount of rainfall in Jan. and Feb. 1957.
*The length of the base-line shows the time of rainfall continued.

Text-fig. 2 shows a part of the results of the radiological survey of the rain water gathered in the yard of the laboratory and that of the water of the river Ashida from Jan. to Feb. 1957. The full line shows the water level of the river Ashida at Kashima-bridge base and the filled column shows the amout of rainfall at Fukuyama. On 1st Feb. the amounts of radioactivity in the water of the river Ashida was $0.15 \pm 0.11 \mu c/l$ and the change of the water level was observed on the same day and on 3rd Feb., but it had not rained in the Fukuyama districts from 31st. Jan. to 3rd Feb. It is therefore supposed that the radioactivity in the river water on 1st Feb. was caused by the rainfall over some part of the basin of the river Ashida.



Text-fig. 3. Decay curves of radioactivity of fission products in the rain water in Jan. and Feb. 1957.

As shown in Text-fig. 2, the amouts of radioactivity in the river water were almost constant during a rainfall and it began to decrease soon after it had ceased to rain. The radioactivity could not be found out after about two days, and usually the state of no remarkable radioactivity in the river water continued till the next rainfall. Text-fig. 3 shows the decay curves of radioactivity, which indicate that the amounts of radioactivity in the rain water decrease to half after about 30 days. From these results it may be seen that the decrease of radioactivity in the river water described above is not caused by the physical decay of radioactive elements in fission products in the river water but caused by the effect that the amounts of fission products in the river water were decreased. The reason for the decrease is supposed to be that the radioactive substances were washed away downstream or precipitated on the river bed or absorbed by soil or aquatic organisms in the stream.

B) Radioactivity in the water at the mouth of the river Ashida and Seto Inland Sea

The results of measuring the amounts of radioactivity in the brackish-water and the sea water off the mouth of the river Ashida is shown in Table 1.

This table shows that no remarkable radioactivity could be found out in the brackish-water and the sea water. It is supposed that the river water containing radioactive substances was influenced by the sea water at the river mouth. According to the results of periodical measuring of the amounts of radioactivity in the sea water from Jan. to Aug. 1956 at the station in the Strait of Hōyo, about 3 miles southwest off the Cape of Sada through which the Black Current comes into Seto Inland Sea and also at another several stations in the Seto Inland Sea, the writers could not recognize any remarkable radioactivity.

Table 1. Radioactivity of brackish-water at the mouth of the river Ashida and sea water off that river mouth.

Date of Sampling	Radioactivity ($\times 10^{-4}$ $\mu c/l$)				Time of low tide
	1000m upstream from the river mouth	500m upstream from the river mouth	the river mouth	1 mile off the river mouth	
12h. 11 Jan. '57	−0.08±0.11	−0.00±0.12	0.06±0.12	0.06±0.10	12h. 20m.
15h. 27 Jan. '57	0.02±0.11	—	—	—	15h. 35m.
12h. 28 Jan. '57	0.02±0.11	−0.04±0.11	0.12±0.12	—	16h. 25m.
14h. 29 Jan. '57	—	−0.01±0.10	—	—	17h. 05m.
15h. 30 Jan. '57	0.01±0.11	0.08±0.12	—	—	17h. 40m.
11h. 6 Feb. '57	0.05±0.12	0.08±0.11	—	−0.14±0.11	08h. 30m.
16h. 9 Feb. '57	—	—	—	0.09±0.11	11h. 40m.
15h. 12 Feb. '57	0.11±0.11	0.06±0.11	—	0.08±0.11	15h. 40m.
09h. 16 Feb. '57	—	—	—	0.07±0.11	06h. 00m.
15h. 22 Feb. '57	0.11±0.11	—	—	−0.03±0.11	10h. 55m.

C) Radiological contamination of laver at the mouth of the river Ashida by fission products in the river water

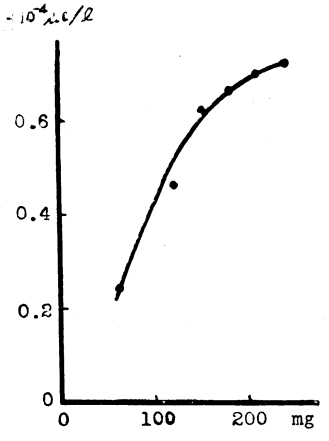


Fig. 4. Self-absorption curve of the ashed laver

In order to know the self-absorption in the ash of laver in this treatment, some quantity of the ashed sample of laver was counted. The results are shown in Text-fig. 4. Here the straight line, a part of the curve, shows that the values for the amounts of radioactivity of some quantity of the ashed sample can be generally converted into the values for the amounts of radioactivity per gram. Weights of ash of the linear range are from 0 to 150mg. as shown in Text-fig. 4. So the ash of laver was taken about 120mg. of each sample and was counted. So as to study whether or not the living laver is contaminated by radioactive substances in fission products in the river water, the ash of the living laver collected just when the radioactivity was found out in the river water was counted, and also the ash of the living laver collected just when no radioactivity was found out in the river water was counted. The radioactivity measured about the ash of the living laver mentioned above is shown in Table 2. Here, significant difference can not be recognized between the values for the amounts of radioactivity of the laver collected in these two cases described above.

The dried laver, "asakusanori", which was cultured in the different year at the mouth of the river Ashida was ashed and counted. One of the dried laver was collected and dried in Feb. 1954, which was not influenced by atomic bomb tests at Bikini Atoll, and the other laver cultured on the same ground on 6th Feb. 1957 was collected and dried after the

atomic bomb tests. The ash of the dried laver of these two cases was counted. These results are shown in Table 3.

Table 2.

a). Radioactivity in the river water, brackish-water and the laver, sampled at the river mouth when radioactivity was found out in the river Ashida.

Date of sampling	Radioactivity ($\times 10^{-4}\mu c$.)		
	Laver (per 120 mg.)	Brackish-water (per litre)	River water (per litre)
4 p.m. 27 Jan.'57	0.18±0.11	0.02±0.11	—
5 p.m. 27 Jan.'57	—	—	0.17±0.10
10 a.m. 29 Jan.'57	—	—	0.31±0.11
2 p.m. 29 Jan.'57	0.21±0.11	−0.01±0.10	—
10 a.m. 30 Jan.'57	—	—	0.20±0.10
3 p.m. 30 Jan.'57	0.49±0.12	0.01±0.11	—
10 a.m. 6 Feb.'57	0.05±0.12	0.05±0.12	0.41±0.11
11 a.m. 6 Feb.'57	0.17±0.12	0.08±0.12	—

b). Radioactivity in the brackish-water and the laver when radioactivity was not found out in the river Ashida.

Date of sampling	Radioactivity ($\times 10^{-4}\mu c$.)	
	Laver (per 120 mg.)	Brackish-water (per litre)
1 p.m. 12 Feb.'57	0.18±0.11	0.06±0.11
3 p.m. 22 Feb.'57	0.16±0.12	0.11±0.11
4 p.m. 22 Feb.'57	0.25±0.11	—
10 a.m. 11Mar.'57	0.34±0.11	0.03±0.11
2 p.m. 11Mar.'57	0.18±0.11	—

Table 3. Radioactivity in the "asakusanori".

(collected and dried up in Feb. 1954.)

No. of sample	Date of measurement	Radioactivity ($\times 10^{-4}\mu c$/120mg.)
1	5 Mar. '57	0.47±0.12
2	6 Mar. '57	0.45±0.12
3	6 Mar. '57	0.34±0.12
4	6 Mar. '57	0.29±0.11
5	8 Mar. '57	0.50±0.12
6	11 Mar. '57	0.49±0.12
7	13 Mar. '57	0.49±0.12
8	13 Mar. '57	0.51±0.12
9	13 Mar. '57	0.56±0.12
10	13 Mar. '57	0.52±0.12

(collected and dried up on 6th Feb. 1957.)

No. of sample	Date of measurement	Radioactivity ($\times 10^{-4}\mu c$/120mg.)
1	5 Mar. '57	0.48±0.12
2	5 Mar. '57	0.52±0.12
3	5 Mar. '57	0.43±0.12
4	5 Mar. '57	0.50±0.12
5	5 Mar. '57	0.45±0.12
6	5 Mar. '57	0.52±0.12
7	5 Mar. '57	0.62±0.12
8	11 Mar. '57	0.43±0.12
9	11 Mar. '75	0.49±0.12
10	11 Mar. '57	0.33±0.12
11	11 Mar. '57	0.36±0.12

From these results significant difference between the amounts of radioactivity of the laver before and after the atomic bomb tests cannot be seen. Referring to these two tables, it may be said that the increase of radioactivity either in the living or in the dried laver cultured at the river mouth can not be recognized.

SUMMARY

1) The radioactivity derived from fission products in the water of rain and the river Ashida has been surveyed, and the radioactivity was newly found out in the water of that

river at every rainfall from Dec. 1956 to Feb. 1957. During the rainfall, radioactivity was always detected in the water of the river Ashida.

2) When the rain stopped, the radioactivity in the river water began to decrease gradually and could not be found out after two days, and so on hereafter.

3) The radiological contamination of the brackish-water at the mouth of the river Ashida and also that of the sea water at the stations off the mouth of that river, at a few stations in the Seto Inland Sea and at the station in the Strait of Hōyo were surveyed respectively, and any remarkable radioactivity could not be found out in either case.

4) The radiological contamination of laver, cultured at the mouth of the river Ashida, has not been recognized from Jan. to Feb. 1957 even when the radioactivity was detected in the river water at every rainfall. During that term the values for the amounts of radioactivity in the river water was $(0.41 \pm 0.11) \times 10^{-4}\ \mu c/l$ at the maximum value.

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