Fallout Radioactivity due to the Underground Nuclear Test of U. S. S. R. (Jan. 15, 1965)

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ABSTRACT

Fallout radioactivity due to a leakage of the underground nuclear explosion test of U. S. S. R. (Jan. 15, 1965) was investigated. Abnormal increase of radioactive fallout was observed at first on the early morning of Jan. 20 at Nagaoka, Niigata Pref. In the fallout after Feb. 10, the influence by short lived nuclides could not be found.

Gamma-ray spectrum of rain-snow sample of Jan. 20 coincided well with that of $^{140}$Ba+$^{140}$La, and the counting rate of gamma-rays decayed to a half in every 13 days through the beginning of March. Most part of gamma-activity was considered to be attributed to $^{140}$Ba+$^{140}$La in the transient equilibrium state.

Radioactive product, $^{140}$Ba, was used as a checking source and $^{140}$Ba+$^{140}$La content in each fallout sample collected after Jan. 20 was estimated. After subtraction $^{140}$Ba+$^{140}$La activity from the total activity in each sample, the residual decayed almost exponentially, with a half-life of 51~53 days, extending for a fairly long period of time. Maximum beta-ray energy was estimated to be 1.5~1.6 MeV at the end of March when $^{140}$Ba+$^{140}$La was considered practically to have been extinct. Thus a chief component of the residual was thought to be $^{89}$Sr.

Cumulative deposition of $^{140}$Ba+$^{140}$La and other nuclides during the period from Jan. 19 to Feb. 9 was estimated to be about 110 mCi/km$^2$ and 270 mCi/km$^2$ respectively as values of Feb. 15.

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INTRODUCTION

It was reported that an underground nuclear explosion test was carried out by U.S.S.R. at Semiparatinsk district on Jan. 15, 1965, when its radioactive material leaked out above the ground. Late in January 1965, the radioactivity in rain water and air-borne dusts increased unusually at Nagaoka, Niigata Pref.

The decay rates and gamma-ray spectra of the radioactive fallout samples in those days have been studied in detail, some comments on which are given in this paper.

SAMPLING AND MEASUREMENT

Sampling of rain-snow water (fell during one day period starting at 9 h in the morning and ending at 9 h on the following morning) was made by means of a steel funnel 1 m x 1 m. Sampling of air-borne dusts was made by using an electric dust collector during an hour period from 9 h to 10 h every morning; on the other hand, dry fallout for 72 hours was collected on gummed-paper once every three days. Their radioactivities were measured with GM counter and a single channel gamma-spectrometer.

EXPERIMENTAL RESULTS

1. Day to day change in amounts of artificial, radioactive fallout The radioactive contamination by daily rain (mCi/km²) and air-borne dusts (μCi/m³) from January to March is shown in Fig. 1.

With respect to rain water, that of Jan. 19 (collected at 9 a.m., 20th) gave one order higher contamination than the preceding rains; in addition, by measuring the decay rate of the radioactivity, it was found to be of different character from the preceding ones.

With respect to air-borne dusts, abnormal increase of radioactivity was found for the first time in dust of Jan. 20.

In the precipitation amounting 25.3 mm on Jan. 19 about 2 mm fell on 19th at midnight and the remainder fell after 0 h, 20th; on the other hand, the radioactivity (cpm/l) of rain water on Jan. 20 (collected at 9 a.m., Jan. 21st) jumped up abruptly about 37 times that of the day before. Therefore, it seems that the abnormal increase of radioactive fallout at Nagaoka appeared in the early morning of Jan. 20.

The daily cumulative contamination by rain and dusts had both the peak on 20th and the minimum on 25th, Jan. and afterwards it increased for a while and reached the constant level after about Feb. 3.

The radioactivity of the dry fallout collected on gummed-paper exhibits the same tendency as the above.

In order to observe how long the abnormality in the radioactivity in rain water in the last decade of January continued, the beta-decay characteristics of the daily
radioactive fallout are shown in Fig. 2.

As shown in Fig. 2, the later are the dates of collection of fallout, the gentler become the slopes of the decay curves, and after Feb. 10 slopes are not much different from those of fallout before the middle of January, which seems to show that in the fallout after Feb. 10 the influence by short lived nuclides faded away.

Some of the beta-decay curves of the main samples are shown in Fig. 3. The radioactivity of the rain water of Jan. 19 decayed rapidly at the beginning and then diminished gradually, and that of Jan. 20 decayed slowly at first and diminished rapidly after two or three days and then decayed slowly again. The radioactivity of the rain of Jan. 21 increased at the beginning and reached the maximum on Jan. 25 and then diminished.
The radioactivity of the air-borne dusts of Jan. 20 and 21 decayed particularly fast in the early stage, but that of 22nd and 23rd Jan. decayed somewhat slowly at first and that of 24th remained on the constant level till January 28.

2. Main components The gamma-ray spectrogram of a sample prepared by drying up 23.8 litres of rain-snow water which fell on Jan. 20 was taken on Feb. 1 using an NaI scintillator and is reproduced in Fig. 4. It coincides well with the gamma-ray spectrum of $^{140}$Ba+$^{140}$La. The gamma-ray spectrum of another sample made from 50.3 litres rain during the days from Jan. 21 to 24 was also similar in shape to that of $^{140}$Ba+$^{140}$La.

There appeared no energy peak higher than 1.6 MeV in the spectra mentioned.
As shown in Fig. 5, the counting rate of the gamma-rays at 1.6 MeV decays to a half in about every 13 days, and the counting rate of the total intensity of gamma-rays also decreases to a half in every 13 to 14 days till the beginning of March and thereafter it decays a little slowly.

From the facts mentioned above most part of gamma-rays is considered to be attributed to $^{140}$Ba+$^{140}$La in the transient equilibrium state, and contribution from other nuclides is regarded to be negligible till the beginning of March.

Accordingly, radioactive product, $^{140}$Ba, which is in the transient equilibrium state with $^{140}$La was taken, and beta-counting rate and 1.6 MeV gamma-ray peak were determined under the same condition as in the fallout samples. Thus, by using the rule of three contribution of $^{140}$Ba+$^{140}$La to total beta-activity of fallout was estimated as follows: on 31st January, the ratios of the activity of $^{140}$Ba+$^{140}$La to the total activity of rain water which fell on 20th and 21st January were respectively 39% and 44%. In contrast to this value, Koyama* obtained the following result from radiochemical analysis: about the rain of Jan. 21 fell at Niigata that

* Koyama, S. The 7th annual meeting of Japan Radiation Research Society held in Tokyo on May 14～15, 1965.
$^{140}$Ba and $^{140}$La contribution was 16\% and 18\% respectively on Jan. 30.

Furthermore, after subtraction of decay curve of $^{140}$Ba + $^{140}$La from that of fallout samples stated above, the residuals decayed almost exponentially extending for a fairly long period of time as can be seen in Fig. 6. Its apparent half-life was about 51~53 days, and the most part of the residual seemed to consist of a single nuclide.

Accordingly, a measurement of its beta-ray absorption was made using aluminium absorbers at the end of March, when $^{140}$Ba + $^{140}$La was thought practically to have been extinct, and the maximum beta-ray energy was estimated to be 1.5~1.6 MeV by means of the Feather method and the Harley method.

Isotopes which have a half-life of 51~53 days and beta-energy of 1.5~1.6 MeV are expected to be $^{89}$Sr and $^{91}$Y, in which the former will probably be a chief component* judging from its half-life although the coexistence of $^{91}$Y is conceivable so far as the fission yields of $^{235}$U are concerned.

* As a result of radiochemical analysis of rain water which fell on 21 Jan. at Niigata, Koyama gave a value of 60\% of $^{89}$Sr to the total activity measured on 30 Jan. (Presented at the 7th annual meeting of Japan Radiation Research Society, Tokyo, 1965)
Furthermore, several samples were processed by drying up several or some ten litres of rain-snow water which fell during the days from Jan. 19 to Feb. 5. A photopeak of their gamma-ray spectra at around 1.6 MeV was obtained, and $^{140}$Ba + $^{140}$La content was estimated. The values of which, reduced on Feb. 15, are given in Table 1.

There is a close correlation between the contents of $^{140}$Ba + $^{140}$La in Table 1 and the slopes of curves in the early stage as shown in Fig. 2.

From Table 1 and the slopes of the decay curves in Fig. 2, contributions of $^{140}$Ba + $^{140}$La to total beta-activity of the rain water on Jan. 26~27, Jan. 30~Feb. 2 and Feb. 6~9, when calculated as values of Feb. 15, are roughly 30%, 30% and 5% respectively.

Thus cumulative deposition of $^{140}$Ba + $^{140}$La during the period from Jan. 19 to Feb. 9 is estimated to be about 110 mCi/km², as the value of Feb. 15

DISCUSSION

Slopes of the decay curves of the radioactive samples mentioned above seem to be affected by $^{140}$Ba + $^{140}$La content in the rain water till the beginning of April. But even after the middle of April when $^{140}$Ba + $^{140}$La practically becomes extinct, there is a variation in slopes of decay curves according to the date of precipitation.
as can be seen in Fig. 2. Major reason for it is likely that there is a variation in
the ratio between the new (due to the nuclear explosion test carried on Jan. 15)
and the old radioactive fallout (due to the past nuclear explosion tests) in individual
sample, assuming that in the new fallout fractionation of nuclides, whose half-lives
are very long, does not occur or can be regarded as negligible. A plausible reason
for this is seen in the decay curves of radioactive rain water which was collected
on 20th and 21st January.
Table 1. $^{140}\text{Ba}+^{140}\text{La}$ contained in daily precipitation (values are reduced
to those on Feb. 15)

<table>
<thead>
<tr>
<th>Date</th>
<th>Amount of precipitation (mm)</th>
<th>Total activity ($\mu$Ci/l)</th>
<th>$^{140}\text{Ba}+^{140}\text{La}$ activity ($\mu$Ci/l)</th>
<th>Ratio (%)</th>
<th>Daily contamination by $^{140}\text{Ba}+^{140}\text{La}$ (mCi/km²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jan. 19</td>
<td>25.3</td>
<td>308.3</td>
<td>63.2</td>
<td>20.5</td>
<td>1.60</td>
</tr>
<tr>
<td>20</td>
<td>20.4</td>
<td>12684.1</td>
<td>3444.7</td>
<td>27.2</td>
<td>70.27</td>
</tr>
<tr>
<td>21</td>
<td>2.2</td>
<td>11336.1</td>
<td>3493.7</td>
<td>30.8</td>
<td>7.69</td>
</tr>
<tr>
<td>22</td>
<td>38.6</td>
<td>1618.7</td>
<td>363.6</td>
<td>22.5</td>
<td>14.03</td>
</tr>
<tr>
<td>23</td>
<td>8.7</td>
<td>2245.3</td>
<td>632.4</td>
<td>28.2</td>
<td>5.50</td>
</tr>
<tr>
<td>24</td>
<td>8.7</td>
<td>385.6</td>
<td>126.5</td>
<td>32.8</td>
<td>1.10</td>
</tr>
<tr>
<td>25</td>
<td>3.1</td>
<td>566.5</td>
<td>200.9</td>
<td>35.5</td>
<td>0.62</td>
</tr>
<tr>
<td>28</td>
<td>3.6</td>
<td>1761.5</td>
<td>466.6</td>
<td>26.5</td>
<td>1.68</td>
</tr>
<tr>
<td>29</td>
<td>4.3</td>
<td>1012.5</td>
<td>285.1</td>
<td>28.2</td>
<td>1.23</td>
</tr>
<tr>
<td>Feb. 3</td>
<td>9.5</td>
<td>161</td>
<td>57.5</td>
<td>35.1</td>
<td>0.55</td>
</tr>
<tr>
<td>4</td>
<td>16.6</td>
<td>202</td>
<td>24.2</td>
<td>12.0</td>
<td>0.40</td>
</tr>
<tr>
<td>5</td>
<td>28.9</td>
<td>110</td>
<td>14.7</td>
<td>13.4</td>
<td>0.42</td>
</tr>
</tbody>
</table>

Thus, by assuming (1) variations in slopes of decay curves after the middle
of April are simply due to the variation in the ratio of the new radioactive fallout
to the old one, (2) mean half-lives of the new radioactive fallout and the old one are
53 days and 440 days respectively, and by using the slopes of the decay curves on
May 1, the ratio (new to old) was estimated concerning each radioactive fallout
from Jan. 19 to Feb. 9.

By using the value of ratio, concentration of radioactive substances of rainfall
and the amount of precipitation every day, daily gross contamination by the new
radioactive fallout was calculated, and cumulative deposition of nuclides other than
$^{140}\text{Ba}+^{140}\text{La}$ during the period from Jan. 19 to Feb. 9 was estimated to be about
100 mCi/km² as the value of May 1 and about 270 mCi/km² as the value of Feb. 15,
the main component of which was probably $^{89}\text{Sr}$.

As stated above, main components of these radioactive materials are thought
to be $^{140}\text{Ba}+^{140}\text{La}$ and $^{89}\text{Sr}$ (or mixture of $^{89}\text{Sr}$ and $^{91}\text{Y}$). Therefore, the reason why
the radioactivity of the rain which fell after Jan. 21 increased in the early stage,
may be explained by the assumption that $^{140}\text{Ba}$ and $^{140}\text{La}$ have not reached yet the
transient equilibrium state.

In the process of generation of $^{140}\text{La}$ from $^{140}\text{Ba}$, the total radioactivity is
expected to attain the maximum value after four days. Therefore, from the fact that every rain water had its peak of radiation three to four days after precipitation it seems that there was almost no $^{140}\text{La}$ in case of each precipitation. That is, it can be concluded that $^{140}\text{La}$ had been formed from $^{140}\text{Ba}$ during several days after 21 Jan. and that the residual component rich in $^{140}\text{Ba}$ fell mixed in rain water.

On the other hand, the decay curve of the air-borne dusts of Jan. 20 was analyzed graphically and components of the dusts were divided into three as shown in Fig. 7: (a') $^{89}\text{Sr}$ or mixture of $^{89}\text{Sr}$ and $^{81}\text{Y}$, (b') $^{140}\text{Ba}$ and $^{140}\text{La}$ in the transient equilibrium state, (c) a component rich in $^{140}\text{La}$. Thus the dusts of Jan. 20 seemed to contain excessive amount of $^{140}\text{La}$. The same can be said of the dusts of Jan. 21. But the dusts of Jan. 24 seemed to contain very little amount of $^{140}\text{La}$. Radioactivity of the rain water of Jan. 19 also decayed rapidly in the early stage.

From these it can be concluded that the daughter, $^{140}\text{La}$ has a tendency to fall earlier than the parent, $^{140}\text{Ba}$.

![Fig. 7. Analysis of decay curve (air-borne dust, Jan. 20, 1965)](image-url)