Observation of Tritium Behavior Both in Monthly Precipitation in Niigata City and in Spring Water at Top of Mt. Zao and Some Mountains in Fukushima Prefecture After the Fukushima Dai-ichi Nuclear Power Plant Accident

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In 2011, the released radioactive materials including tritium (T) from damaged reactors at Fukushima Dai-ichi nuclear power plant has caused much environmental problems. After the Fukushima Dai-ichi Nuclear Power Plant Accident, the specific activity of T in precipitation was increased in Niigata city. In this study, each monthly precipitation (imitative ground infiltrated precipitation) was collected at the rooftop of Niigata University. Such a sample was subsequently enriched by electrolytic enrichment method (SPE method), and the specific activity of T of each enriched sample was measured by low-background liquid scintillation counter. In addition, the relation between specific activity of T and nssCa\(^{2+}\) (i.e., non-sea-salt Ca\(^{2+}\)) concentration in monthly precipitation was also discussed. Moreover, the concentrations of cations, i.e., Na\(^+\), K\(^+\), Ca\(^{2+}\), and Mg\(^{2+}\), in monthly precipitation and the specific activity of T in spring water at top of Mt. Zao (and other mountains in Fukushima prefecture) were also measured. Based on the above-mentioned matters, it was found that the influence of the Fukushima Dai-ichi Nuclear Power Plant Accident on environment in Niigata city almost disappeared after four months from the accident.

Key Words: Fukushima Dai-ichi Nuclear Power Plants, monthly precipitation, tritium, non-sea-salt Ca\(^{2+}\), spring water, ion concentration in precipitation

1. Introduction

In recent years, the Fukushima Dai-ichi Nuclear Power Plant Accident was focused the attention in Japan and in the world because the accident released a lot of radioactive materials including tritium (T), and also caused much environmental problems. Thus T directly reached some surface water as precipitation, and indirectly reached some rivers from ground water.\(^1\)

Tritium is naturally produced by the interaction between \(^{14}\)N (or \(^{16}\)O) and \(^{1}\)n. When T was naturally produced by the above-mentioned manner, the specific activity of T in precipitation is almost 0.5–1.0 Bq\(\cdot\)kg\(^{-1}\) in Niigata city. On the other hand, T also produced by nuclear bomb test. When atmospheric nuclear bomb tests were carried out in 1960s, the maximum value of the specific activity of T exceeded 200 Bq\(\cdot\)kg\(^{-1}\) in Japan. From Partial Test Ban Treaty (1963) was put into operation, the specific activity of T continuously decreased and attained the environmental revel (0.5–1.0 Bq\(\cdot\)kg\(^{-1}\)),\(^2\) recently.

However, after the Fukushima Dai-ichi Nuclear Power Plant Accident, the specific activity of T in precipitation was suddenly increased in Niigata city. There are two processes of the T-production in nu-
clear reactor, one is from the ternary fission, and the other is the reaction between deuterium and neutron.

Besides, the specific activity of T in precipitation changes with the variation of weather conditions, and also changes with different places and different seasons in Japan. In order to measure the specific activity of T in an environment water, we aim to get the reference value of concentration of T in an environment water, and to analyze the origin of the air mass characteristics causing precipitation. In addition, to clarify the behaviors of some trace elements, some ground waters were collected.

In the former paper,3) we observed the effect of the Fukushima Dai-ichi Nuclear Power Plant Accident on some spring waters in Mt. Zao and in some mountains in Fukushima prefecture, and clarified that the effect was so small.

Considering the above-mentioned, in order to investigate the influence of T on environment in Niigata city after the Fukushima Dai-ichi Nuclear Power Plant Accident, we tried to research the influence of T on Niigata city in monthly precipitation and to clarify the variation of the specific activity of T in precipitation.

2. Experimental

2-1 Samples

The location of the sampling point in Niigata city is shown in Fig. 1. Each water sample of monthly precipitation was collected at the rooftop of faculty of Engineering, Niigata University (37.87°N, 138.94°E). The plastic sampler4) is used for monthly collection of precipitation. The collected samples are similar to infiltrated groundwater,5) because the collected samples will overflow up to 90% from the precipitation.2) In this study, the water samples were collected at the point (shown in Fig. 1) from 2011 to 2015.

Each water sample of spring water was collected at Mt. Zao in Yamagata prefecture (next to Fukushima prefecture) or each mountain in Fukushima prefecture (Mt. Azuma, Mt. Adatara, or Mt. Bandai). Each location was shown in the reference,3) and the sampling date in spring water is shown in Table 1.

2-2 Sample pretreatment and measurement

For distilling sample water, GR grade Na₂O₂ and KMnO₄ (both from Wako Pure Chemical) were supplied in the distillation bottle. After the distillation, each sample was subsequently enriched by applying
the electrolytic enrichment method (SPE method) \(^6\)–\(^9\). After that, 30 mL of the enriched sample was introduced into 145 mL polyethylene scintillation vial, and 70 mL of the liquid scintillation cocktail (ULTIMA GOLD \(^\text{\textregistered}\) LLT, provided from Perkin Elmer, USA) was added. Then, the vial was put in the darkroom at low temperature for a week, the specific activity of T was subsequently measured by low-background liquid scintillation counter (LCS, Aloka Co., Ltd., Japan).

Moreover, Na\(^+\), K\(^+\), Ca\(^{2+}\), and Mg\(^{2+}\) were also measured. Na\(^+\) and K\(^+\) are determined by flame spectrophotometry; Ca\(^{2+}\) and Mg\(^{2+}\) are by atomic absorption spectrophotometry.

### 3. Results and discussion

#### 3.1 Monthly precipitation

The specific activity of T from 2011 to 2015 in monthly precipitation, i.e., imitative ground infiltrate precipitation, \(^2\)–\(^5\) is shown in Fig. 2.

In this figure, the specific activity of T obviously increased (i.e., \(2.60 \pm 0.13 \text{ Bq kg}^{-1}\)) in March, 2011, and the value is about 3–5 times higher than previous level at Niigata city (\(0.6–1.0 \text{ Bq kg}^{-1}\)) \(^2\) (Table 2). After this month, the specific activity of T was gradually decreased to July. In general, the specific activity of T will increase in Niigata city in the spring due to spring peak. \(^10\)

In addition, in previous report, \(^11\) the high specific activity of T disappeared in a few weeks after the accident. Nevertheless, this research shows that the specific activity of T almost disappeared after four months from the accident.

Moreover, comparing with several other years, it is found that the value of the specific activity of T from 2012 to 2015 shows the value of average year.

#### 3.2 Calculations of ssCa\(^{2+}\) and nssCa\(^{2+}\) in precipitation

As to the sea salt contribution, other sources that contribute to the concentration of Ca\(^{2+}\) can be estimated using the concentration of non-sea-salt Ca\(^{2+}\) \((\text{NSSCa}^{2+})\). \(^6\) Using Na\(^+\) in the sea-salt as tracer and assuming that the soluble Na\(^+\) has no source other than marine origin, the concentration of nssCa\(^{2+}\) are calculated by the following equations \(^2\), \(^12\):
The relation between T and nssCa$^{2+}$

Figure 3 shows the concentration of nssCa$^{2+}$ from 2011 to 2015 in monthly precipitation. In Niigata city, the increasing T concentration generally accompanies the increasing nssCa$^{2+}$ concentration, but in this figure, the concentration of nssCa$^{2+}$ was no significant change comparing with other years. Moreover, the T observed is no accompanying with increasing Ca$^{2+}$ concentration. From the above-mentioned, it concludes that T thus observed was caused by the accident.

In other words, it indicates that the Fukushima Dai-ichi Nuclear Power Plant Accident had not affected the concentration of nssCa$^{2+}$, but led to the increasing of the specific activity of T.

### 3.4 Comparing the specific activity of T in monthly precipitation and of T in spring water

As shown in Table 1, the specific activity of T in Mt. Zao was a little higher than that in Niigata city in 2012, and both of them are very close in 2014. Top of Mt. Zao is a higher location in Yamagata prefecture, so it seems that the spring water in this place is similar to precipitation. From Table 1, it is also found that the specific activity of T in Fukushima, i.e., Mt. Azuma, Mt. Adatara and Mt. Bandai, were higher comparing with Niigata city and top of Mt. Zao. They indicate that Niigata city does not much

### Table 2 Specific activities of T [Bq kg$^{-1}$] in monthly precipitation from 2011 to 2015 in Niigata city

| Month/Year | 2011  | 2012  | 2013  | 2014  | 2015  |
|------------|-------|-------|-------|-------|-------|
| Jan        | --    | 0.72±0.07 | 0.35±0.05 | 0.40±0.05 | --    |
| Feb        | 0.51±0.06 | 0.66±0.07 | 0.54±0.06 | 0.45±0.06 | 0.33±0.05 |
| Mar        | 2.60±0.13 | 0.96±0.08 | 0.74±0.07 | 0.63±0.07 | --    |
| Apr        | 1.69±0.11 | 0.69±0.07 | 0.89±0.08 | 0.83±0.08 | --    |
| May        | 0.83±0.08 | 0.85±0.08 | 0.47±0.06 | 0.70±0.07 | 0.48±0.06 |
| Jun        | 1.21±0.09 | 0.49±0.06 | 0.51±0.06 | 0.35±0.05 | 0.40±0.05 |
| Jul        | 0.40±0.05 | 0.49±0.06 | 0.36±0.05 | 0.46±0.06 | 0.20±0.04 |
| Aug        | 0.60±0.06 | 0.29±0.04 | 0.45±0.06 | 0.21±0.04 | --    |
| Sep        | 0.21±0.04 | 0.29±0.04 | --      | 0.25±0.04 | 0.40±0.05 |
| Oct        | 0.38±0.05 | 0.35±0.05 | 0.86±0.08 | 0.48±0.06 | 0.27±0.04 |
| Nov        | --      | 0.36±0.05 | 0.32±0.05 | 0.52±0.06 | 0.20±0.04 |
| Dec        | 0.41±0.05 | 0.11±0.03 | 0.23±0.04 | 0.32±0.05 | --    |
affect by the Fukushima Dai-ichi Nuclear Power Plant Accident.

3.5 Major ion concentrations in monthly precipitation

Figure 4 shows the ion concentrations in precipitation in Niigata city from 2011 to 2015. From our previous research, the behavior of the concentration of $\text{Ca}^{2+}$ is similar to that of the specific activity of $T$. However, it is found that the behavior of $\text{Ca}^{2+}$ concentration is very similar to nss$\text{Ca}^{2+}$ concentration (i.e., increases in later winter and spring, and decreases during autumn and early winter after the Fukushima Dai-ichi Nuclear Power Plant Accident).

Moreover, as to monthly precipitation, the variations of some ion concentrations change with specific activity of $T$, and $\text{Mg}^{2+}$ and $\text{Na}^+$ show very similar variations in all seasons. From the above-mentioned, it may find that the concentration of nss$\text{Ca}^{2+}$ is useful as one of indicators of the specific activity of $T$ in Niigata city and around site, and that these ion concentrations may be also useful as one of the indicators of seasonal variations.

4. Conclusions

The specific activity of $T$ and major ions were measured in monthly precipitation in Niigata city from 2011 to 2015, and the following quantitative conclusions could be obtained about the effects of the Fukushima Dai-ichi Nuclear Power Plant Accident.

1) In Niigata city, the specific activity of $T$ in monthly precipitation became 3–5 times higher than previous level after the Fukushima Dai-ichi Nuclear Power Plant Accident (i.e., $2.60\pm0.13 \text{ Bq kg}^{-1}$ in March, 2011), but, after four months, it became to
average level \(0.51 \pm 0.21 \text{ Bq} \cdot \text{kg}^{-1}\).

2) From the previous paper, it is obtained that the concentration of \(\text{nssCa}^{2+}\) mainly changes with specific activity of \(T\) in monthly precipitation in Niigata city. In this work, the increase of the concentration of \(\text{nssCa}^{2+}\) was not found. Consequently, it shows that the increase of the specific activity of \(T\) was affected by the Fukushima Dai-ichi Nuclear Power Plant Accident.

3) After four months, Niigata city is no more affected by the Fukushima Dai-ichi Nuclear Power Plant Accident.

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