

**Investigation of Environmental Impact of the
Accident of Fukushima Dai-ichi Nuclear Power Plant
by Using the Specific Activity of T and Several Ions in
Precipitation, Spring Water and Lake Water**

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Summary

The accident of Fukushima Dai-ichi nuclear power plant in Japan on 12-15th March, 2011 result in the release of large amounts of radioactive materials including tritium (T). It caused the serious contamination of local region and trace contamination throughout adjacent towns and prefectures. The subsequent transfer of radionuclides, mainly radioisotopes such as T and caesiums, caused radiation exposures after the accident. In order to investigate the effect of the accident of the nuclear power plant on environment, I employed the measuring method of tritium applying a distillation process and an electrolytic enrichment process. The specific activity of tritium (T concentration) in the water thus obtained was measured by liquid scintillation counter. This procedure was applied to monthly and short term precipitation in Niigata city, spring water in both Fukushima and the adjacent prefecture, and to lake water in both Fukushima prefecture and Niigata city. Furthermore, I also measured the concentration of ions (Na^+ , K^+ , Ca^{2+} , Mg^{2+} , NH_4^+ , SO_4^{2-} , NO_3^- , and Cl^-) in precipitation, spring water and lake water. From the above-mentioned, the following matters have been obtained:

- (1) The variation of T concentration in recent years can be clarified.
- (2) The influence of the accident of Fukushima Dai-ichi Nuclear Power Plant on environment in Niigata city almost disappeared after four months from the accident.
- (3) As to short precipitation on 15th March, 2011, the activities of radiocaesiums were not detected, but the activity of T was detected significantly.
- (4) The effect of the accident on spring water in mountains around Fukushima prefecture is so small.
- (5) The influence of the accident of Fukushima Dai-ichi nuclear Power Plant to the adjacent lakes (e.g., in Fukushima prefecture and Niigata city) was quantitatively clarified.
- (6) The effect of the accident of Fukushima Dai-ichi nuclear Power Plant on Fukushima prefecture is gradually decreases.

CHAPTER 1

GENERAL INTRODUCTION

1.1 Background for the study

In recent years, the radioactive materials released including tritium (T) by damaged reactors at Fukushima Dai-ichi nuclear power plant has received a huge amount of attention in Japan and the world, and also caused a great deal of environmental problems. Lots of thus radioactive materials including tritium (T) have been released to the environment, it can transfer to man by inhalation, passage through skin, and by ingestion in food and drinking water¹⁻⁶).

Precipitation shows an important removal path of particles and soluble gases from the atmosphere⁷), and the chemical composition in precipitation also shows a significant role in reflecting change in anthropogenic emissions of element, atmospheric chemical transformations with weather patterns⁸).

In this study, the followings were measured to investigate the effect of the accident of Fukushima Dai-ichi nuclear power plant in environment.

①the specific activity of T and the ions concentration in precipitation in Niigata city; ②spring water in both Fukushima and the adjacent prefecture; ③lake water in both Fukushima prefecture and Niigata city

1.2 Overview of tritium

Tritium, the heaviest and only radioactive isotope of hydrogen, is

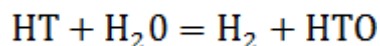
increasing in importance in energy and environmental considerations. This nuclide is widely distributed throughout man's environment because of its ubiquitous form as tritiated water and of its persistence in the environment. The nuclide is produced copiously by the military nuclear program, and tritium as in the form of water was distributed worldwide by the nuclear weapons tests that carried out in the open atmosphere⁹⁾, particularly during the period 1954 to 1962.

1.2.1 Physical and chemical properties of tritium

The radioactive of tritium was established in 1939 by Alvatez and Cornog¹⁰⁾. They found that the radioactive is of very short range and that half-life is long, and in subsequent work, they found establish its physical half-life at 12.3 years. Tritium decays by the emission of a beta particle with maximum energy of 18.6 keV, an average of 5.7 keV, and decays to form of ^3He . Table 1.1 lists the important physical properties of the isotope.

Tritium is closely the reactions of ordinary hydrogen, but its relatively large mass differences make isotopic effects. Because of the importance of water in the life processes, its isotopic exchange with tritium is of special concern. Libby^{11),12)} calculated theoretical equilibrium constants for a number of tractions involving the isotopic variants of hydrogen gases and their oxides. Black and Taylor¹²⁾ experimentally determined the

equilibrium constants for the reaction:



over the temperature range 16 to 303 °C, and they compared quite well with the theoretically derived values of Libby (Table 1.2). The experimental values were consistently higher, but they were within the predicted limits of accuracy estimated of Libby. The experimental values can be summarized by the equation:

$$\log K = 0.292 \log T + (336.5/T) - 1.055$$

Swain and Schaad¹⁴⁾ derived an equation to describe the relation between the isotope effects of tritium and an deuterium based on quantum mechanics considerations:

$$\frac{K_{\text{H}}}{K_{\text{T}}} = \alpha \left(\frac{K_{\text{H}}}{K_{\text{D}}} \right)^{1.442}$$

where K_{H} , K_{T} , and K_{D} are molecular constants pertaining to hydrogen, tritium, and deuterium, respectively. The value of α has a low-temperature limit of 1,000 °C and a high-temperature limit of 1,050 °C. Bigeleisen^{15),16)} developed the theory of relative tritium–hydrogen and deuterium–hydrogen isotope effects for both kinetic and equilibrium processes, and also found that kinetic isotope calculations yield exactly the same results as calculations of equilibrium isotope effects, and that they are the same as those obtained from collision theory.

Tritium is produced by a number of nuclear reactions, including fission. Some of the more important sources are ternary fission and neutron irradiation of deuterium, lithium, and boron.

1.2.2 Source of tritium production and its release

Tritium is produced both by ternary fission and by neutron reactions with light elements such as boron in control rods or dissolved as “burnable poisons” in the coolant of pressurized water reactors. Most of the fission product tritium produced is retained within the fuel and only a minor fraction passes to the coolant or is released with gases or liquids to the environs of the reactor.

In 1938 Sherr, Smith, and Bleakney¹⁷⁾ concluded that if tritium is present in ordinary water its concentration must be less than 1 tritium atom per 10^{12} hydrogen atoms. The relative concentration of tritium in atmospheric hydrogen is about 10^3 to 10^4 times in rain water^{18-20),21)}, but the quantity of hydrogen gas in the atmosphere is a factor of 104 less than the average quantity of water vapor²²⁾.

Tritium is produced by a number of different types of nuclear activity. Production at heavy-water reactors currently creates the most serious operational problems. As reactor-fuel reprocessing plants are established, the tritium produced by fission will eventually be released to the environment, the yield of tritium produced as a fission product in reactors

is 0.01%^{23),24),25)}. The status of tritium in the coolant of light-water reactor is still not clear. The possibility of diffusion of fission-product tritium into the coolant is significant, tritium contamination problems might arise at reactor sites. In the event, the amount of tritium released during fuel reprocessing would be lowered by whatever fraction had been lost by diffusion. Further studies of the tritium released to the coolant in light-water reactors and of the tritium retained in the fuel and released during reprocessing will be required before quantitative evaluations of tritium releases are possible. Furthermore, it is quite likely that both the production and release of tritium in light-water reactors will depend on the type of fuel and cladding used and the energy spectrum of the neutron flux.

Production of the residual tritium in thermonuclear weapons and reactors could present a very considerable source of release. However, the possibility of developing thermonuclear reactors in the near future seems rather remote. Also, the value of tritium as a fuel makes its recovery economically attractive, so little would be allowed to escape.

1.2.3 Procedure for tritium enrichment

The natural concentration of tritium in the environment is so low that it cannot be measured directly^{26), 27)}. Even with the increase in tritium levels due to thermonuclear testing, the concentration of environmental tritium

remains so low that its direct measurement is only rarely possible^{28), 29)}. Several methods have been developed, particularly since 1939, which permit the separation, or at least the concentration, of many isotopes³⁰⁾.

For greater enrichment electrolysis must be carried out in several stages, with distillation of the sample between stages to keep the concentration of the hydroxide in the optimum range³¹⁻³⁴⁾. With five stages of electrolysis, the tritium concentration can be increased³¹⁾ by factor of 2,000 to 20,000.

Electrolysis is a relatively simple procedure for modest enrichments of tritiated water where small volumes are sufficient. For large-scale continuous separations, thermal diffusion seems to have an average, especially if the tritium exists as the gas. Gas chromatograph appears to have considerable promise as a laboratory procedure for enrichment for tritium analysis. Although gas chromatograph is a complex method, the recovery and the degree of enrichment are quite high. Furthermore, tritiated isotopes of simple molecules other than the gas can be separated.

1.2.4 Movement of Tritium in the environment

Natural tritium is produced primarily in the atmosphere; production at the earth's surface is quite low. About two-thirds of the natural tritium U is produced in the stratosphere³⁵⁾. Thermonuclear explosions at high altitudes in tropical regions introduced tritium directly into the

stratosphere^{36), 37)}, where mixing processes are very low.

As already mentioned, the residence time for tritium in the troposphere is only a matter of a few weeks rather than a few months to a few years, as in the stratosphere. Consequently, the fallout of tritium released to the troposphere is limited to the general latitude of release. Some lateral mixing occurs, and some possibility of lateral tropospheric mixing exists, even across the equator³⁸⁾, but, in general, the concern with tropospheric releases is limited to local areas of discharge.

Tritium can reach surface waters by a variety of pathways: directly through precipitation, through molecular exchange with the atmosphere, by release of tritiated water from nuclear plants, and indirectly from runoff or by influx to streams from groundwater. Surface water, which contain over 99% of the total water, constitute the greatest reservoirs of water on earth^{39), 40)}.

Tritium released to the environment is usually converted to the oxide form quite rapidly and is dispersed like ordinary water. The relatively large mass difference between ordinary water and tritiated water has some influence on its properties. For example, the vapor pressure of T₂O is lower than that of ordinary water⁴¹⁾; hence, tritium would tend to become enriched in the condensed phase. In general, the circulation of tritium would be expected to closely parallel the circulation of water.

1.2.5 Projected tritium production in a nuclear reaction and its impact on local and worldwide populations

Although tritium is produced by a number of reactions, the amount of tritium produced by fission probably exceeds the amount produced from all other sources in light-water-cooled reactors. Fission-product tritium⁴²⁾,⁴³⁾, is produced with a yield of about 0.01%. Some tritium may penetrate fuel-element cladding and escape into the coolant⁴⁴⁾, but most of it is apparently retained until the fuel is reprocessed⁴⁵⁾,⁴⁶⁾. Other possible major sources of tritium include heavy-water reactor stations, thermonuclear detonations, and, perhaps, thermonuclear reactor.

A cursory look at the potential environmental hazards that are likely to be caused by increasing releases of tritium resulting from the growth of a nuclear power economy based on light-water reactors suggests that the general increase in the worldwide concentration of tritium is not like to result in a significant increase in hazards. Local environmental contamination problems may be significant, but they appear to be of lower order of magnitude⁴⁷⁾,⁴⁸⁾, than those arising from the release of ⁸⁵Kr. The magnitude of the local contamination problem will depend on the nature, mode, and quantity of the releases, as well as on local environmental conditions.

1.3 Purpose of this work

Based on the mentioned in the preceding sections, the following five purposes were considered.

- (1) Observing the reference value of T concentration in precipitation
- (2) Observing the seasonal variation of T concentration
- (3) Obtaining the relation between the concentration of T and several ions
- (4) Obtaining the effect of the accident of Fukushima Dai-ichi Nuclear Power Plant on environment.
- (5) Investigating the environmental influences of Niigata city after Fukushima Dai-ichi Nuclear Power Plant

1.4 Structure of this report

This thesis has following 5 chapters.

In Chapter 1, the general introduction was stated.

In chapter 2, Imitative ground infiltrated precipitation (long-term precipitation) of Niigata city, that monthly collected precipitation from 2011 to 2015, were measured. In addition, short term precipitation (hourly collected precipitation) was also collected after the accident of Fukushima Dai-ichi Nuclear Power Plant on March 15th, 2011, and major ion (Na^+ , K^+ , Ca^{2+} , Mg^{2+} , NH_4^+ , SO_4^{2-} , NO_3^- , and Cl^-) concentrations in precipitation were also measured. Based on the above –measured, the

effect of the accident of the Fukushima Dai-ichi nuclear power plant on the environment was quantitatively discussed. Furthermore, the relation between the specific activity of T and concentration of non-sea-salt (nssCa^{2+}) in monthly precipitation was also mentioned.

In chapter 3, spring water were collected from Fukushima and Yamagata prefecture, these samples belonged to four different mountains. Each collected sample contained 9 quantities such as T concentration and major ions (K^+ , Ca^{2+} , Mg^{2+} , Na^+ , NH_4^+ , Cl^- , SO_4^{2-} , and NO_3^-), and were measured and discussed.

In chapter 4, lake waters were collected from Fukushima Prefecture and Niigata city, totally 8 sampling points. These samples obtained in spring and in autumn, respectively. Besides, dissolved Oxygen (DO), pH, and ORP in the water were immediately measured at each sampling point.

In chapter 5 (the final chapter), the mentioned in chapters 2 ~ 4 were summarized.

Table and Figure

Table 1.1 Physical properties of tritium

Tritium	
Half-life	12.3 years
Specific activity	29.2 Ci/milliatom
Maximum energy of radiation (β^-)	18.6 keV
Mean energy of radiation	5.7 keV
Decay product	$^3\text{He}^+$ (stable)
Maximum penetration of radiation	
Air	ca 6 mm
Water	ca 6 μm
Glass/concrete	ca 2 μm

Table 1.2 Equilibrium constants for $\text{HT} + \text{H}_2\text{O} = \text{H}_2 + \text{HTO}$

Temperature	Experimental equilibrium constant	Theoretical equilibrium constant
16	6.75 ± 0.04	6.47
20.2	6.47 ± 0.02	6.24
25	6.25 ± 0.05	6.01
56.2	5.05 ± 0.05	4.84
79.6	4.37 ± 0.05	4.23
111.3	3.76 ± 0.04	3.64
158.5	3.10 ± 0.06	3.03
217.1	2.64 ± 0.04	2.54
302.9	2.17 ± 0.02	2.08

CHAPTER 2

Precipitation From 2011 to 2015 in Niigata city

2.1 Introduction

On 12th March, 2011, the nuclear accident occurred at Fukushima, Japan after large earth-quake (Higashinihon disaster). In the accident, increasing of T concentration has lots of worried and the influence on the environment should be considered. After the accident of Fukushima Dai-ichi nuclear power plants, the specific activity of T in precipitation was increased in Niigata city. Considering the above-mentioned matters, in this study, monthly and short-term precipitations which collected from Niigata city are discussed in detail. In addition, the concentration of ions (Na^+ , K^+ , Ca^{2+} , Mg^{2+} , NH_4^+ , SO_4^{2-} , NO_3^- , and Cl^-) in precipitation were measured also. In the previous report⁴⁹⁾, the behavior of T is similar to one of non-sea-salt Ca^{2+} (nssCa^{2+}), therefore, the relation between specific activity of T and nssCa^{2+} concentration in monthly precipitation was also discussed. Based on the above-mentioned matters, in order to investigate the influence of T on environment in Niigata city after the accident of Fukushima Dai-ichi nuclear Power Plant, 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.

2.2 Experimental

2.2.1 Samples

Each water samples of monthly precipitation (imitative ground infiltrated precipitation)^{50), 51)} and short-term precipitation were collected from the roof top of Niigata University (37.87 °N, 138.94 °E) (see Fig.2.1). The white plastic sampler⁵²⁾ is used for collected monthly precipitation, as shown in Fig.2.2, the collected samples is similar to infiltrated groundwater by using sampler like this, because the sample will overflow up to 90% when collect precipitation. Fig.2.3 shows collected short-term precipitation in the blue large plastic sampler on March 15th, 2011, and the sample waters was collected hourly.

Each short precipitation was collected in large plastic sampler every onr hour on 15th May, 2011 at Niigata University (37.55 °N, 139.03 °E). And each point (measured air dose rate) is shown in Fig.2.4.

2.2.2 Sample pretreatment and measurement

For distillating sample water, GR grade Na₂O₂ from Wako Pure Chemical and KMnO₄ were supplied in the distillation bottle. After being distilled, each sample was subsequently enriched by electrolytic enrichment method (SPE method)⁵³⁾⁻⁵⁶⁾. After electrolytic enrichment, 30 mL of the enriched sample was introduced into 145 mL polyethylene

scintillation vial, and 70 mL of the liquid scintillation cocktail (ULTIMA GOLDTM LLT, provided from Perkin Elmer, USA) was added. After that, the vial was put in the darkroom at low temperature for a week, then the specific activity of tritium was measured by low-background liquid scintillation counter (LCS, Aloka Co., Ltd., Japan).

The radioactivity of short-term precipitation was measured using a p-type high-purity germanium detector (GC4020-7500SL, Canberra, USA) with a multi-channel analyzer (Lynx, Genie 2000, Canberra, USA).

In addition, the ions concentration are detected by Ion chromatograph, but among them, Na⁺ and K⁺ are determined by flame spectrophotometry; and Ca²⁺ and Mg²⁺ are by atomic absorption spectrophotometry. The analyzed radionuclides by gamma-ray spectrometry were ¹³⁷Cs (661.7 keV) and ¹³⁴Cs (604.7 or 795.6 keV).

2.3 Results and discussion

2.3.1 Monthly precipitation

The specific activity of T from 2011 to 2015 in monthly precipitation, i.e., imitative ground infiltrate precipitation is shown in Fig.2.5.

In this figure, the specific activity of T obviously increased in March, 2011, the value is about 2.5 Bq · kg⁻¹, it is about 3 - 5 times higher than previous level at Niigata city (0.51 ± 0.21 Bq · kg⁻¹) (Table 2.1). In general,

the specific activity of T will increase in Niigata city in the spring due to spring peak⁵⁷⁾.

In addition, in previous report⁵⁸⁾, the high specific activity of T disappeared in a few weeks after the accident. Nevertheless, this research showed that the specific activity of T almost disappeared after four months from the accident.

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

2.3.2 Calculations of $ssCa^{2+}$ and $nssCa^{2+}$ in precipitation

As to the sea salt contribution, other sources that contribution to the concentration of Ca^{2+} can be estimated using the concentration of non-sea-salt Ca^{2+} ($nssCa^{2+}$). 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^{49), 59)}:

$$nss[Ca^{2+}] = [Ca^{2+}]_{total} - 0.038 \times [Na^+]_{marine} \quad \textcircled{1}$$

$$ss[Ca^{2+}] = [Ca^{2+}]_{total} - nss[Ca^{2+}] \quad \textcircled{2}$$

2.3.3 The relation between T and $nssCa^{2+}$

Figure.2.6 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+} concentration⁴⁹⁾, 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 is concluded that T thus observed is from the accident.

In order to the further the relation between T and nssCa^{2+} , we compared T specific activity with ssCa^{2+} and nssCa^{2+} in monthly precipitation in monthly precipitation from 2011 to 2015 in each of the four seasons (shown as Fig.2.7-2.10). The abscissa stands for specific activity of T, and the ordinate stands for ssCa^{2+} and nssCa^{2+} concentrations.

Fig 2.7 shows the dates in spring. Comparing the specific activity of T in Niigata city from 2011 to 2015 ($0.51 \pm 0.21 \text{ Bq} \cdot \text{kg}^{-1}$), there is a great change in spring due to spring peak. Besides that, as specific activity of T move the nssCa^{2+} concentration is scattered and the minimum and maximum vary widely. In spring, a mount of transported sand from china by continents air mass, the main component is CaCO_3 that are present in monthly precipitation in Niigata city, it gets high concentrations of nssCa^{2+} .

Fig 2.8 shows the dates in summer. In summer, as specific activity of T move the ssCa^{2+} concentration seems to have little effect, but nssCa^{2+} concentration increases. Summer is rainy season in Niigata city, through

our previous reports, at this time, the precipitation contains high concentration T, nssCa^{2+} and so on. In addition, after the rainy season, the specific activity and nssCa^{2+} concentration will be decreases.

Fig 2.9 and Fig 2.10 shows the dates in autumn and winter. The monthly pooled samples of precipitation in Niigata city is a mixture of continents and marine air mass, or at least contains an admixture of a certain percentage of marine vapor in autumn and early winter. The marine air mass crossing Niigata city contains lots of seawater vapor, and decreases T specific activity in precipitation. As specific activity of T move, the ssCa^{2+} and nssCa^{2+} concentration were little seasonal variation in autumn, and becomes scattered in winter.

In other words, it indicates that the accident of Fukushima Dai-ichi Nuclear Power Plant had not affected the concentration of nssCa^{2+} , but led to the increasing of the specific activity of T. Moreover, it may be that the concentration of nssCa^{2+} is to be one of indicators of the specific activity of T in Niigata city and around site.

2.3.4 Ion concentrations

Concentrations of ions in precipitation were measured from 2011 to 2015. The variations of them depending on time are shown in Fig.2.11 (a-h), respectively.

From our previous research⁶⁰⁾, the concentration of Ca^{2+} is similar

with specific activity of T. However, it is found some interesting features by the figure, i.e., the trends of Ca^{2+} concentration is very similar to Ca^{2+} concentration, i.e., increases in later winter and spring, and decreases during autumn and early winter. It seems that using the date of Ca^{2+} , the behavior of T can be deduced and the movement of precipitation can be also deduced in Niigata region.

Moreover, as to monthly precipitation, the variations of some ion concentrations change with specific activity of T, through the figures, Mg^{2+} and Na^+ in all seasons shares very similar variations. From the above-mentioned, that these ion concentrations may be one of the indicators of seasonal variations.

2.3.5 Short-term precipitation

The short-term precipitation was collected on 15th March, 2011, and specific activity of T were measured (Fig.2.12). In addition, the concentration of radiocaesiums were also measured (Table 2.2), and air dose rates ($\mu\text{Sv/h}$) in Niigata city at the same time are shown in Fig.2.13, The detail obtained by using monitoring post⁽⁶¹⁾. As to these samples, radiocaesiums were not detected, but T concentration was increasing during 17:30-18:30. The increasing of the air dose rate in Niigata city was not observed clearly, but in Agano town and Minamiuonuma city, the rates increased. From these results, a few radioactive plume reached in

Niigata city via Minamiuonuma city. It seems that radiocaesiums were almost washed out in Agano town and Minamiuonuma city. However, only T was reached at Niigata city, because T is easy to form HTO, and diffused easily than other radionuclides.

2.4 Conclusions

The above discussion can be concluded as follows:

- (1) The specific activity of T in monthly precipitation became 3 - 5 times higher than previous level at Niigata city after the accident of Fukushima Dai-ichi Nuclear Power Plant, but after five months, it became to average level ($0.51 \pm 0.21 \text{ Bq} \cdot \text{kg}^{-1}$).
- (2) The nssCa^{2+} concentration was not a significant change after the accident of the Fukushima Dai-ichi Nuclear Power Plant. It is considered that the concentration of nssCa^{2+} is to be one of indicators of the specific activity of T in Niigata city and around site.
- (3) As to each short precipitation sample collected on March 15th, 2011, the T concentration of each sample gathered during 13:30~17:30 was about 2Bq/kg, but the sample during 17:30~18:30 was about 12Bq/kg. So, Niigata city was temporarily affected by the accident .

(4) After the accident, released radiocaesiums seemed to be washed out in Minamiuonuma city and Agano town. However, only T was reached Niigata city because T is diffused easily than other radionuclides.

Table and Figure

Table 2.1 Specific activities of T [$\text{Bq} \cdot \text{kg}^{-1}$] in monthly precipitation from 2011 to 2015 in Niigata city.

Month/Year	2011	2012	2013	2014	2015
Jan	--	0.71 ± 0.07	0.35 ± 0.05	0.40 ± 0.05	--
Feb	0.51 ± 0.06	0.66 ± 0.07	0.53 ± 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	0.68 ± 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	0.45 ± 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	--	0.24 ± 0.04	0.40 ± 0.05
Oct	0.38 ± 0.05	0.35 ± 0.05	0.85 ± 0.08	0.48 ± 0.06	0.27 ± 0.04
Nov	--	0.36 ± 0.05	0.32 ± 0.05	0.52 ± 0.46	0.20 ± 0.04
Dec	0.41 ± 0.05	0.11 ± 0.03	0.23 ± 0.04	0.32 ± 0.05	--

Table 2.2 Each specific activity of T, radiocaesiums, and Air dose rate in short term precipitation in Niigata city

	T/Bq kg ⁻¹	¹³⁴ Cs/Bq kg ⁻¹	¹³⁷ Cs/Bq kg ⁻¹	Air dose rate/μSv·h ⁻¹
2011 3.15 13:30~14:30	1.3±0.2	< 0.1	< 0.1	0.049
2011 3.15 14:30~15:30	2.0±0.1	< 0.1	< 0.1	0.050
2011 3.15 15:30~16:30	2.6±0.1	< 0.1	< 0.1	0.048
2011 3.15 16:30~17:30	2.1±0.2	< 0.1	< 0.1	0.049
2011 3.15 17:30~18:30	12±0.5	< 0.1	< 0.1	0.051



Fig.2.1 The location of Niigata City, Japan (37.87° N, 138.94° E).



Fig.2.2 The plastic sampler used for collection of monthly precipitation.



Fig.2.3 The large plastic sampler used for collection of short term precipitation.

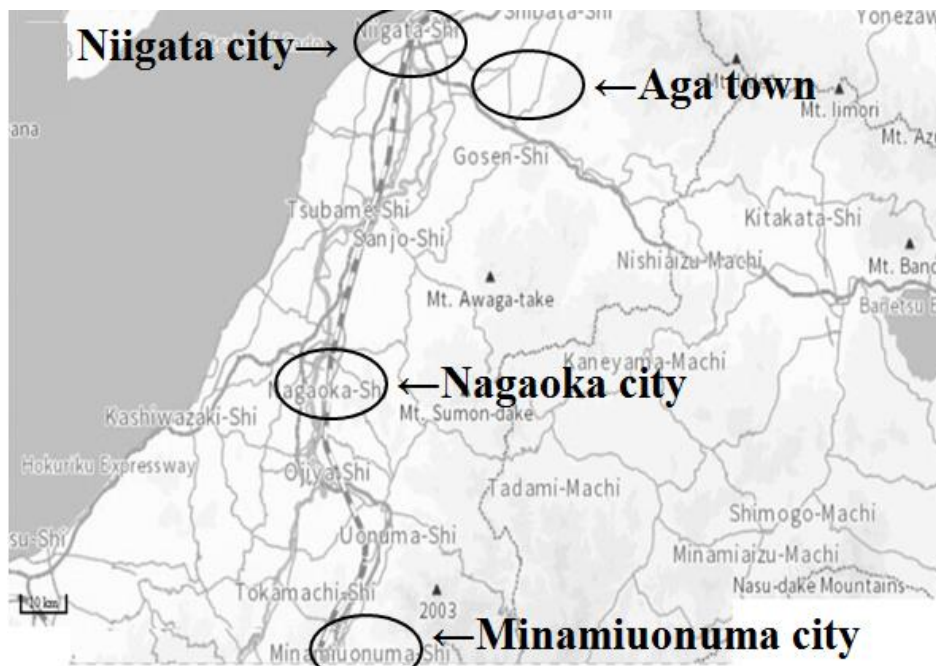


Fig.2.4 The measuring sites of air dose rate.

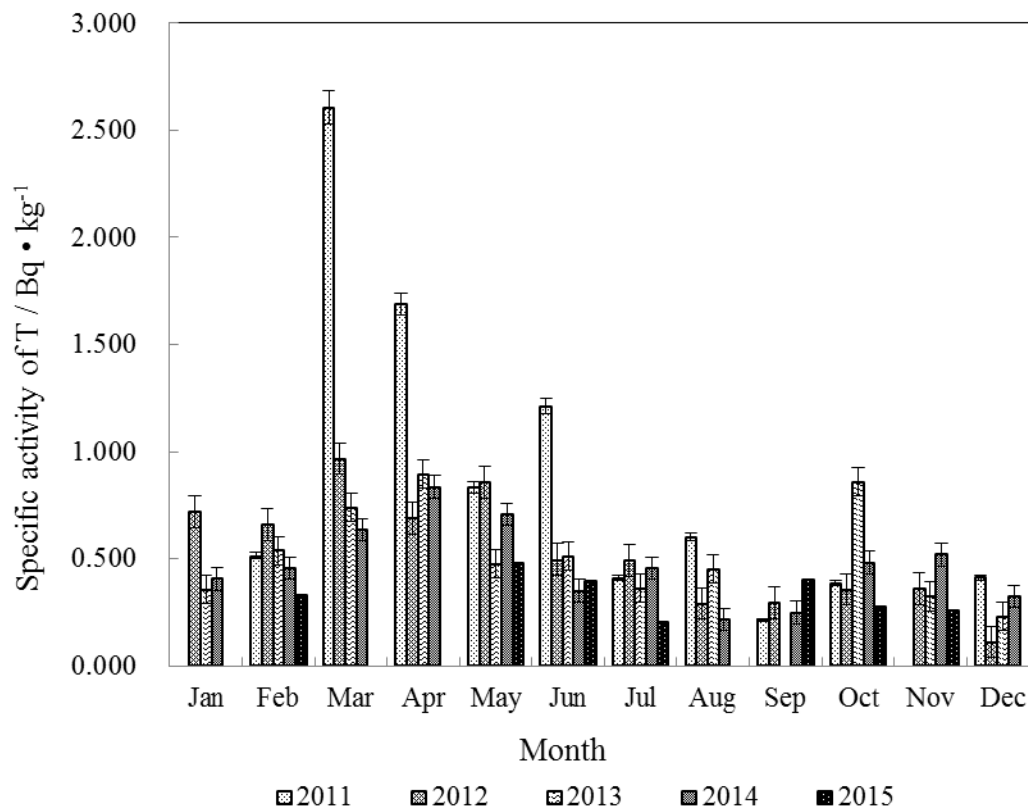


Fig.2.5 Specific activity of T in monthly precipitation from 2011 to 2015 in Niigata city (at the rooftop of faculty of Niigata University), Japan.

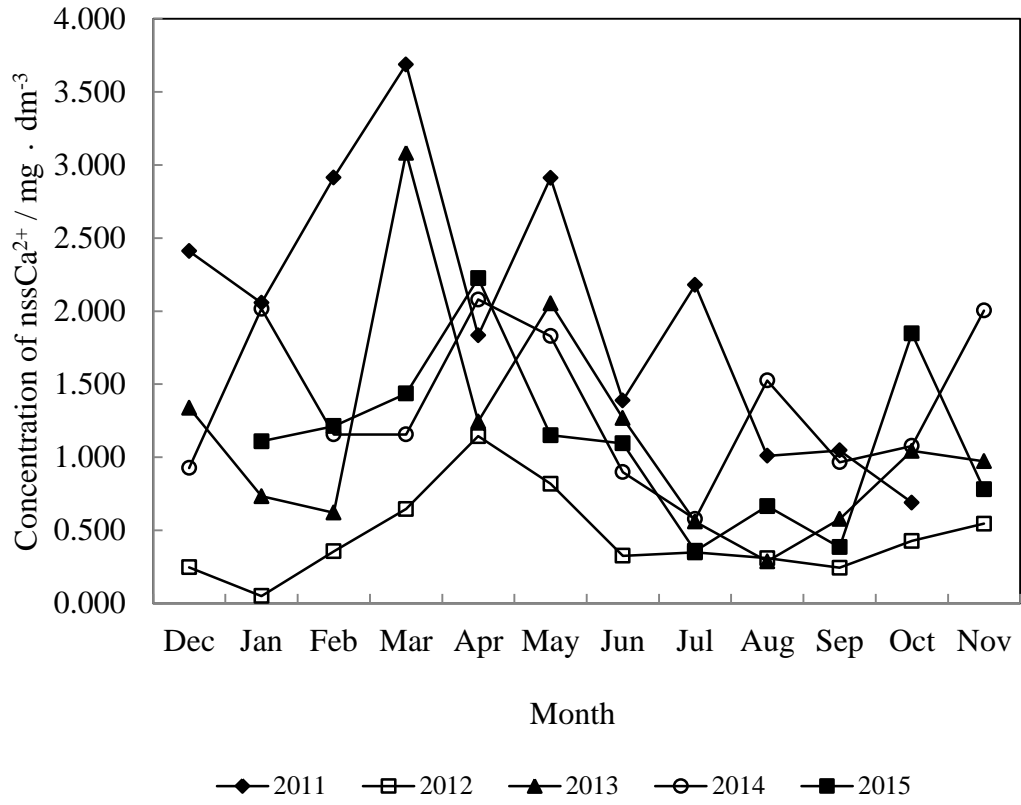


Fig.2.6 The concentration of nssCa²⁺ from 2011 to 2015 in monthly precipitation.

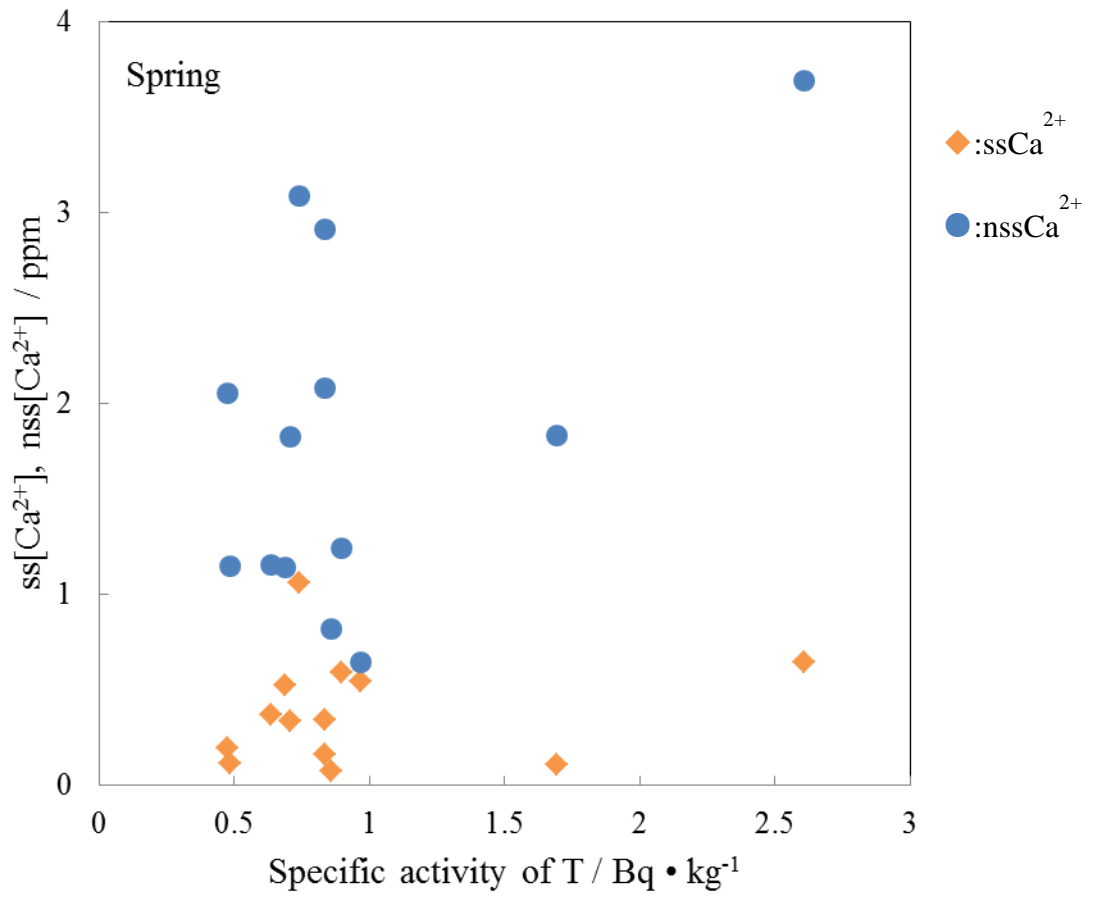


Fig.2.7 Specific activity of T vs. the concentrations of ssCa²⁺ and nssCa²⁺ in spring (Mar. to May) from 2011 to 2015.

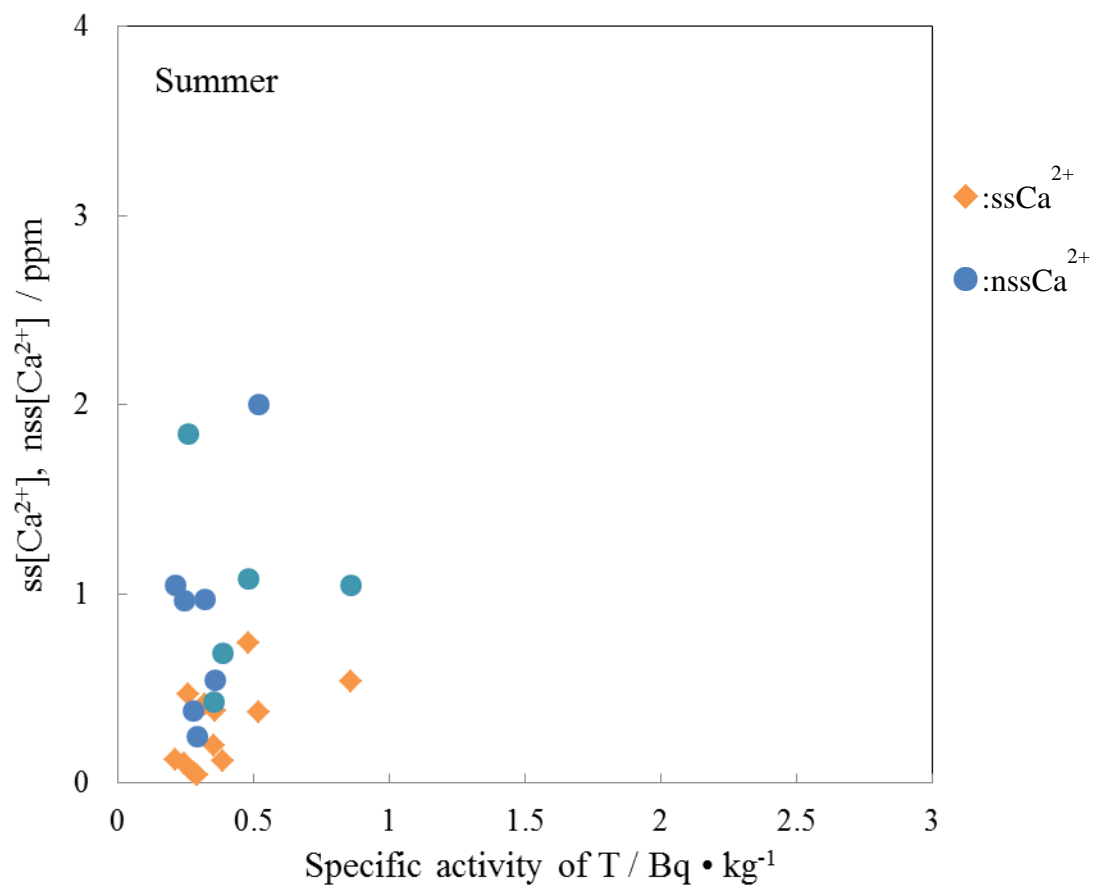


Fig.2.8 Specific activity of T vs. the concentrations of ssCa²⁺ and nssCa²⁺ in summer (Jun. to Aug.) from 2011 to 2015.

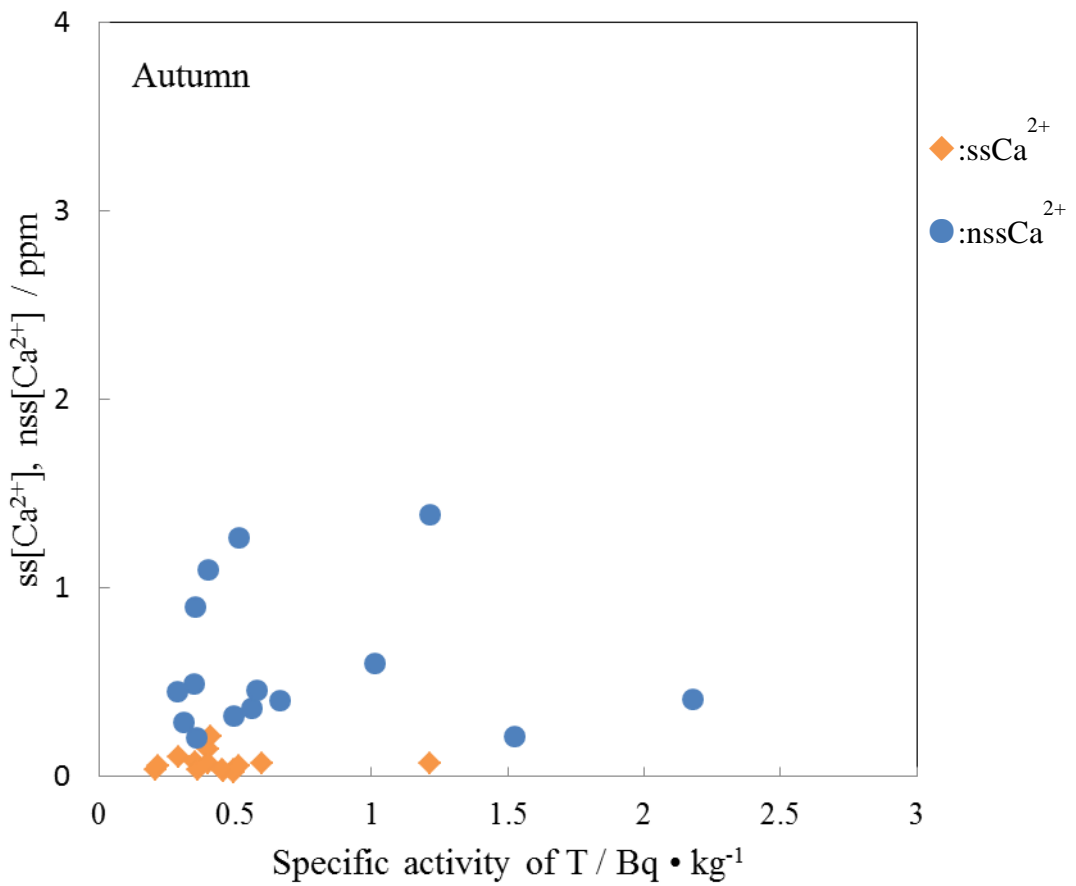


Fig.2.9 Specific activity of T vs. the concentrations of ssCa²⁺ and nssCa²⁺ in autumn (Sep. to Nov.) from 2011 to 2015.

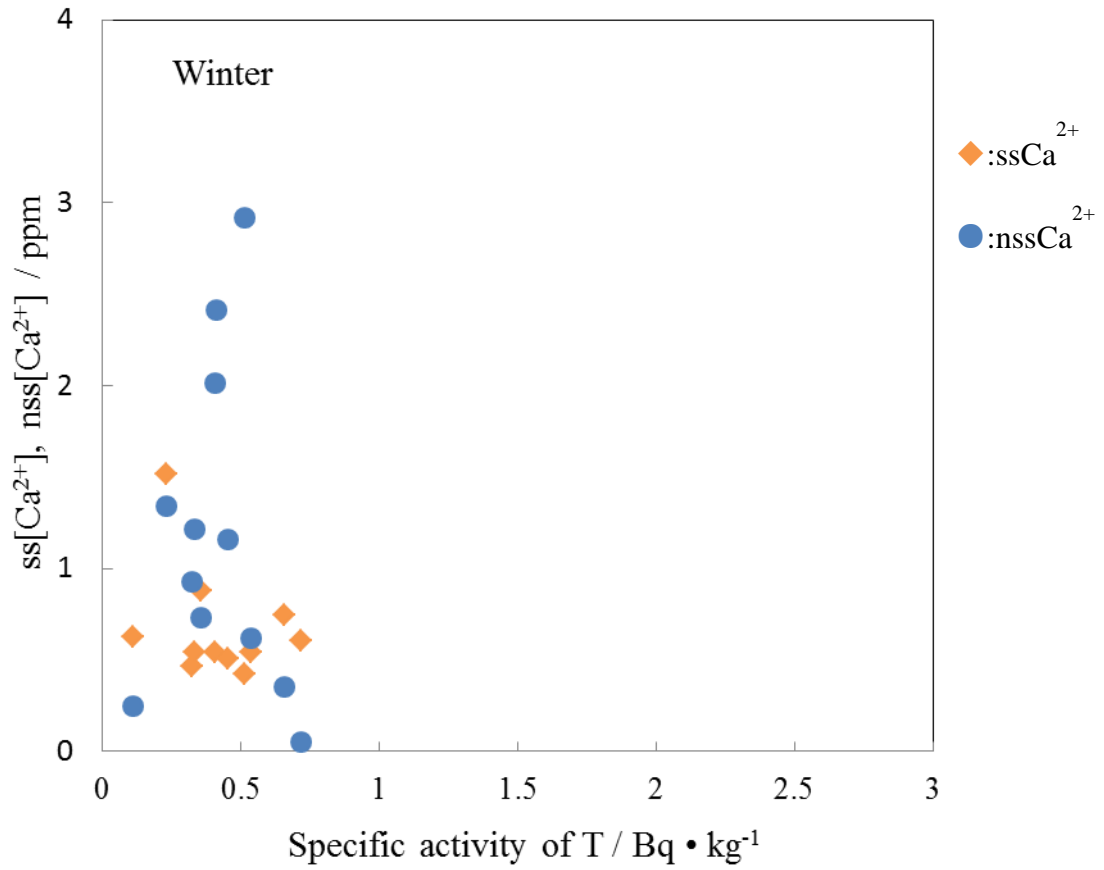
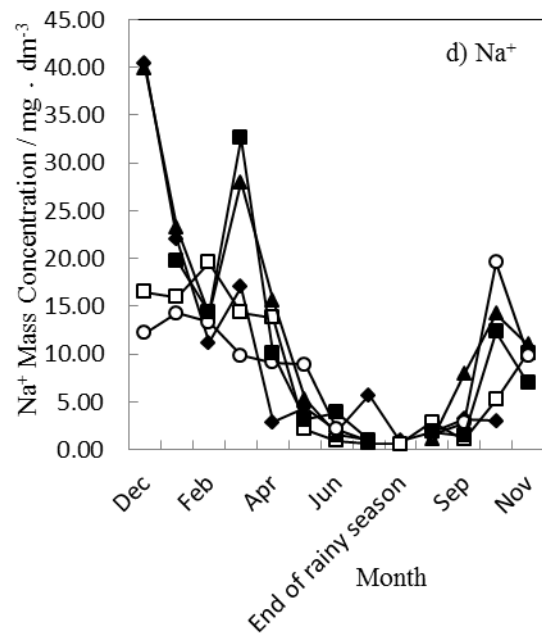
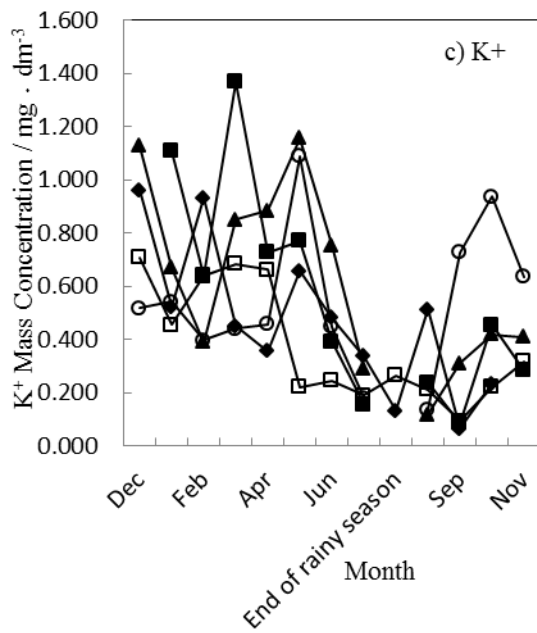
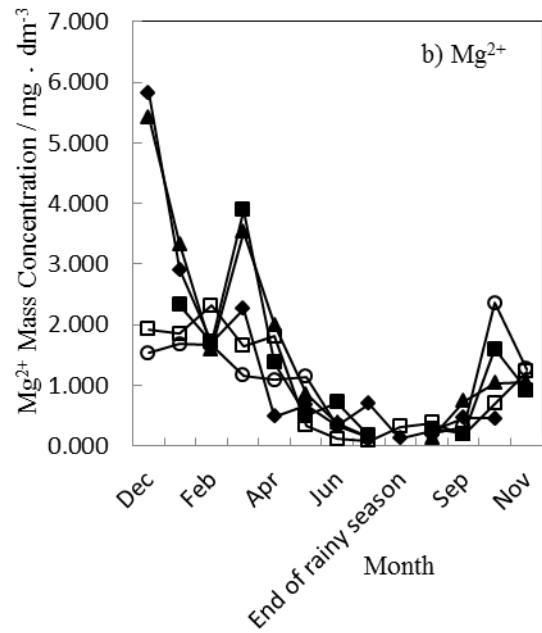
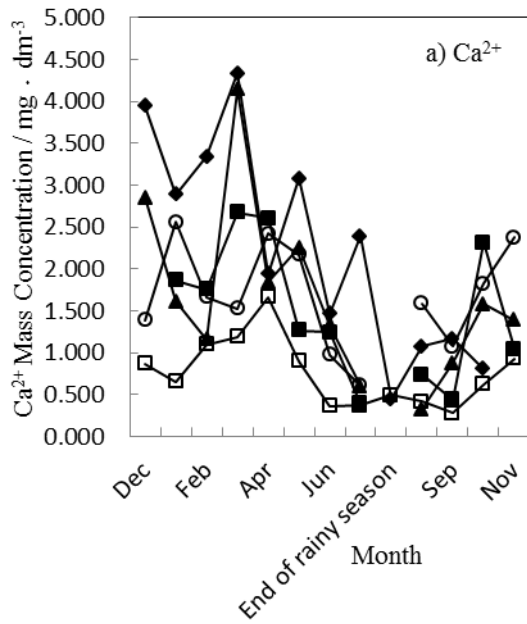


Fig.2.10 Specific activity of T vs. the concentrations of ssCa²⁺ and nssCa²⁺ in winter (Dec. to Feb.) from 2011 to 2015.



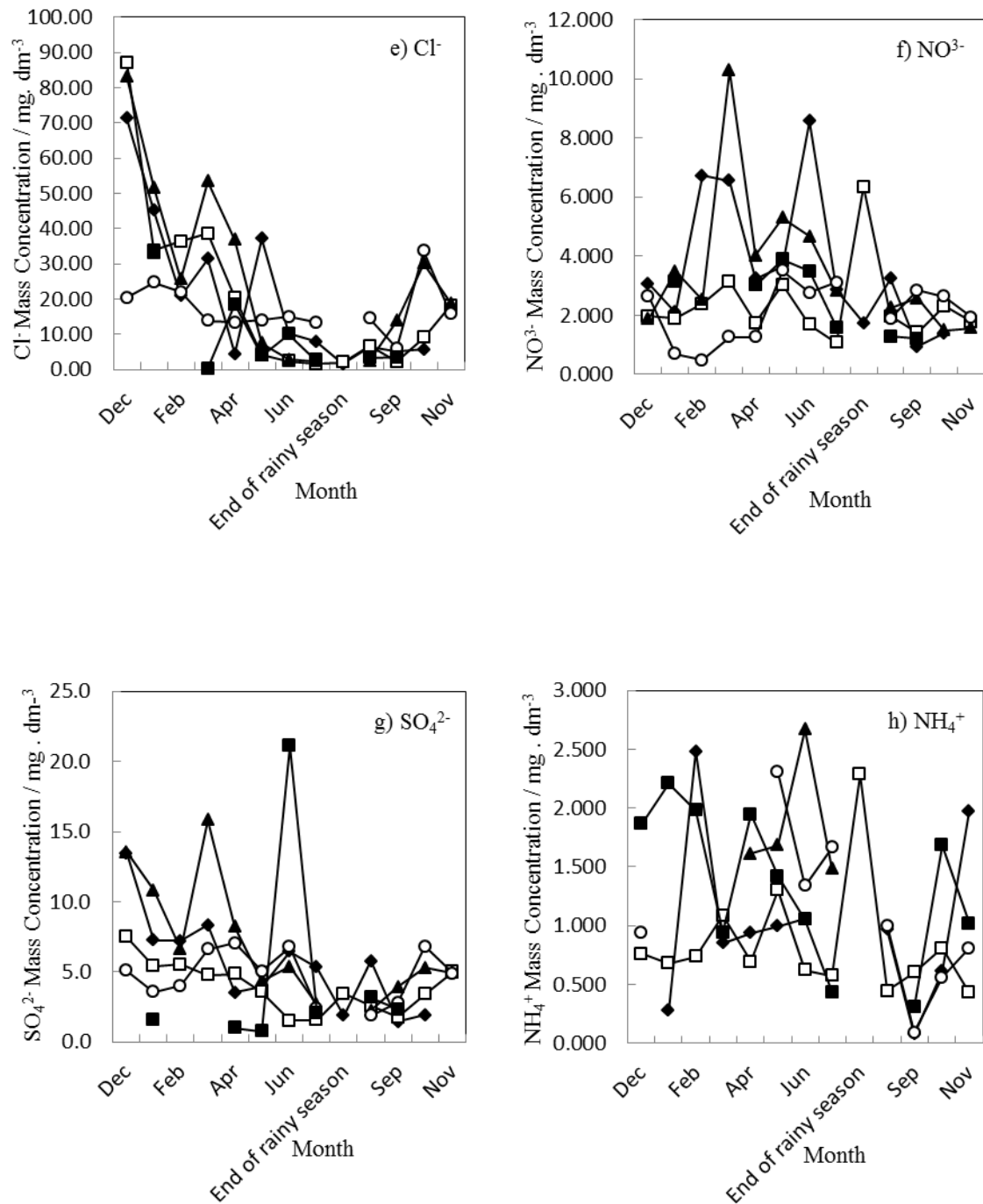


Fig.2.11 The concentrations of several ions from 2011 to 2015 in monthly precipitation.

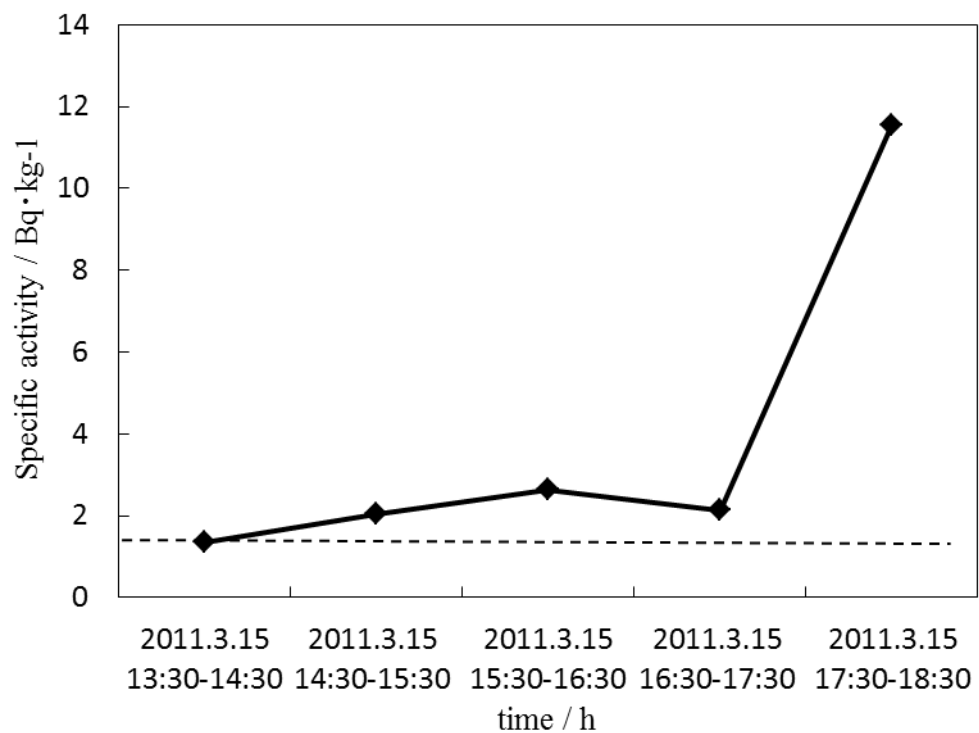


Fig.2.12 Specific activity of tritium in precipitation per hour on March 15th, 2011, rooftop of faculty of Niigata University, Niigata, Japan.

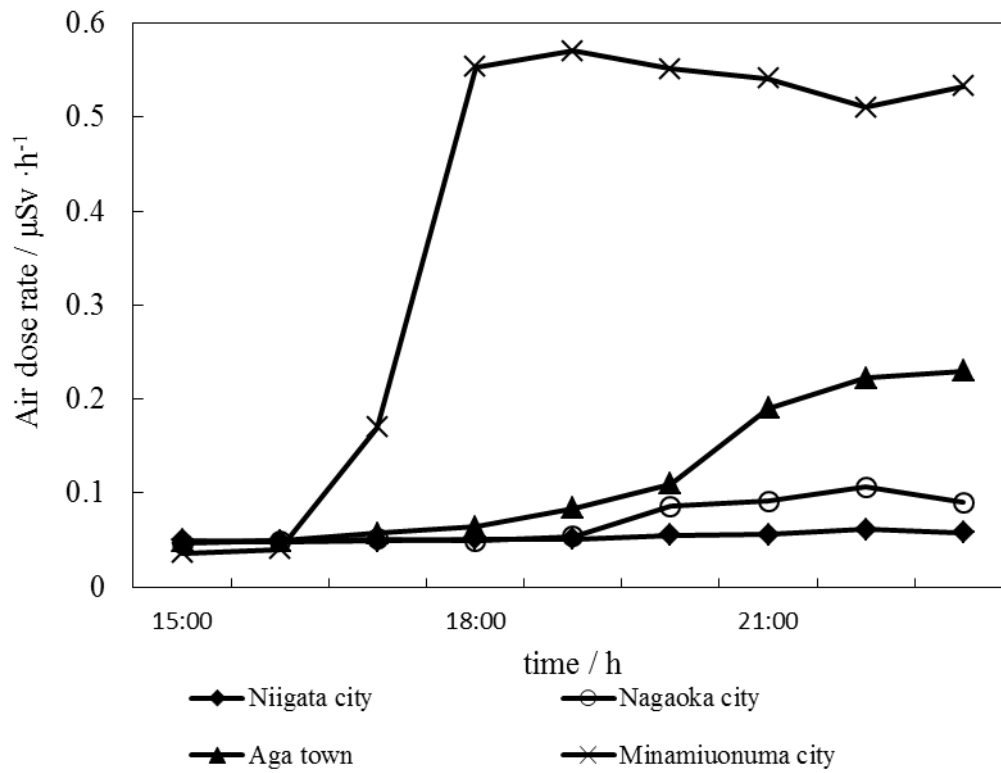


Fig.2.13 Air dose rate in Niigata city on 15th March 2011.

CHAPTER 3

Spring Water in Fukushima and Yamagata Prefectures

3.1 Introduction

In previous chapter, the influence of T in Niigata city already discussed, but it is necessary for investigation and analysis the specific activity of T in Fukushima. Because the detention time of the specific activity of T in spring water in mountains very long (about 2 to 3 years in Tohoku), so the influence of T will be not immediately apparent. Therefore, the sampling need to measured continuously. So in this chapter, the specific activity of T and major ions in spring water in Fukushima and Yamagata prefectures are discussed.

3.2 Experimental

3.2.1 Samples

Each water samples of spring water were collected at Mt. Zao in Yamagata prefecture (next to Fukushima prefecture) and some mountains in Fukushima prefecture (as shown in Table 3.1), and the location of each sampling point for collecting spring water is shown in Fig.3.1. In this study, the water samples were collected in August, 2012, September, 2013, and July, 2014, respectively.

3.2.2 Sample pretreatment and measurement

Each water sample thus obtained was analyzed with the same methods as shown in 2.2.2.

3.3 Results and discussion

3.3.1 The specific activity of T in sample water in each mountain

Each result of T measurements is shown in Fig.3.2. The broken line shows the average specific activity of T of spring water in August 28~31, 2012 (shown as broken line in Fig.3.2). In chapter 2, it is already known that the average specific activity of T in Niigata city is about 0.51 ± 0.21 Bq · kg⁻¹.

In previous report⁶²⁾, in general, the residence time of spring water in lower mountain and around the top of mountain is about 2 to 3 years in Tohoku. Comparing of specific activity of T in 2012, 2013 and 2014 with each other, it is found each specific activity of T of spring water is similar to each other.

In addition, it is also shown that the specific activity of T pf,in 2014 trends to be higher than that in 2013. Although the specific activity of T in 2014 is slightly higher, it may depend on variation of specific activity of T in the environment. Overall, the specific activity of T remained almost stable, so the difference may be affected by Fukushima Dai-ichi

nuclear accident, but it can be thought that the accident of influence of the Fukushima Dai-ichi nuclear power plant are so small.

3.3.2 Site differences

In this work, the samples collected from six differences site belong to Mt. Zao and Mt. Bandai Mt. Adatarara and Mt. Azuma those four mountains, respectively.

It is found that among the four mountains, the geographic location of “around the top of Zao” is higher than other five locations, it seems that the spring water in this place is similar to precipitation (as shown in Fig.3.2). So compared with other sampling sites, the spring water in this place had been less affected in the accident of Fukushima Dai-ichi nuclear power plants.

3.3.3 Comparing the specific activity of T in monthly precipitation with of T in spring water

As shown in Table 3.2, 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. The 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. Adatarara and Mt. Bandai, were higher comparing

with Niigata city and top of Mt. Zao. They indicate that Niigata city does not much affect by the accident of Fukushima Dai-ichi Nuclear Power Plant.

3.3.4 The concentrations of ions in each location

The major ions in spring water such as Na^+ , K^+ , Ca^{2+} , Mg^{2+} , NH_4^+ , SO_4^{2-} , NO_3^- , and Cl^- were measured in August, 2012, September, 2013 and July, 2014.

Measurement results of four different ions in spring water (at six different locations in Fukushima and Yamagata prefectures) are shown in Fig.3.3a-h, and the concentrations of ions are summarized in Table 3.3. From the figures, it is found that Cl^- concentration is high compared with other ions. In general, Cl^- concentration in sea water is higher than that in the precipitation, so the sample in spring water may be influenced by sea water.

The above-mentioned matter shows that Japanese atmosphere is strongly affected by sea water, especially in spring and winter seasons⁵⁷⁾. Moreover, the concentration of Na^+ and Ca^{2+} shows similar seasonal variation in six different locations, so it is infer they are subjected to similar influences from sea water.

In addition, It is also found that in Kinmeisui had much higher concentration of SO_4^{2-} than other five locations, SO_4^{2-} may came from Mt.

Bandai. Mt. Bandai is a famous volcano. The concentrations of Mg^{2+} and K^+ were lower but remain relatively stable.

The change of specific activity of T along with the ion concentrations change at the same time in spring water (especially Ca^{2+} concentration)⁴⁹⁾. It was through that the variations of ion concentrations in spring water for these three years show the similar yearly variation, and also show the effect of the accident is so small.

3.4 Conclusions

The above discussions can be concluded as follows:

- (1) The variation of specific activity of T in 2012, 2013 and 2014 was clarified.
- (2) The specific activity of T in spring water in each year nearly equals the annual average of normal T concentration in Niigata city ($0.51 \pm 0.21 \text{ Bq} \cdot \text{kg}^{-1}$) obtained in the same year.
- (3) Chloride ion concentration was higher in spring water, therefore it is considered that the sample in spring was influenced by sea water, and also considered that Japanese atmosphere seems to be strongly affected by sea water.

Table and Figure

Table 3.1 Sampling points of several mountains in 2012, 2013 and 2014

	Date			Location
1	2012.8.30	2013.9.13	2014.7.15	Hijikatadairaosyouzu (Mt. Azuma)
2	2012.8.30	2013.9.13	2014.7.15	Odennokami (Mt. Zao)
3	2012.8.30	2013.9.13	2014.7.15	Around the top of Zao (Mt. Zao)
4	2012.8.30	2013.9.13	2014.7.15	Kuraishidakeidosawafukin (Mt. Zao)
5	2012.8.28	2013.9.12	2014.7.15	Kinmeisui (Mt. Adatara)
6	2012.8.31	2013.9.12	2014.7.16	Ougonshimizu (Mt. Bandai)

Table 3.2 Specific activities of T [$\text{Bq} \cdot \text{kg}^{-1}$] from the monthly precipitation in Niigata city, and of the spring water at top of Mt. Zao and some mountains in Fukushima Prefecture.

Year \ Month	Monthly precipitation (rooftop of Niigata Univ.)	Spring water			
		top of Mt. Zao	Mt. Azuma	Mt. Adatara	Mt. Bandai
2012 Aug 28-31th	0.29 ± 0.04	0.48 ± 0.05	0.57 ± 0.06	0.58 ± 0.06	0.55 ± 0.06
2013 Sep 12-13th	--	0.36 ± 0.04	0.46 ± 0.05	0.34 ± 0.04	0.46 ± 0.05
2014 Jul 15-16th	0.46 ± 0.06	0.41 ± 0.05	0.60 ± 0.04	0.49 ± 0.04	0.70 ± 0.04

Table 2 The specific activity [Bq kg^{-1}] and several ion concentrations [mg dm^{-3}] in spring water from different mountains in Fukushima pref. and Yamagata one.

Location	Date	T	Na ⁺	Ca ²⁺	Cl ⁻	SO ₄ ²⁻
		Bq kg ⁻¹	mg dm ⁻³			
Hijikatadairaosyouzu (Mt. Agathuma)	2012.8.30	0.57±0.06	2.6	4.3	2.7	5.2
	2013.9.13	0.46±0.05	3.9	6.8	13	5.6
	2014.7.15	0.60±0.04	4.0	6.1	14	7.4
Ondenokami (Mt. Zao)	2012.8.30	0.45±0.05	9.8	15	2.9	14
	2013.9.13	0.47±0.05	4.5	7.9	13	8.4
	2014.7.15	0.50±0.05	2.9	4.2	13	5.0
around the top of Zao (Mt. Zao)	2012.8.30	0.48±0.05	3.5	2.5	3.0	14
	2013.9.13	0.36±0.04	3.9	4.3	14	13
	2014.7.15	0.41±0.05	3.2	3.7	12	7.2
Kuraishidakeidosawa (Mt. Zao)	2012.8.30	0.51±0.06	2.7	4.1	2.8	18
	2013.9.13	0.54±0.06	2.9	4.2	13	14
	2014.7.15	0.53±0.04	2.8	2.9	13	11
Kinmeisui (Mt. Adatara)	2012.8.28	0.58±0.06	2.2	3.8	2.1	25
	2013.9.12	0.34±0.04	3.7	4.2	13	22
	2014.7.15	0.49±0.04	2.5	4.0	12	18
Ougonshimizu (Mt. Bandai)	2012.8.31	0.55±0.06	0.3	2.7	2.2	8.3
	2013.9.12	0.46±0.05	2.8	3.8	14	5.6
	2014.7.16	0.70±0.04	2.6	3.5	12	6.2

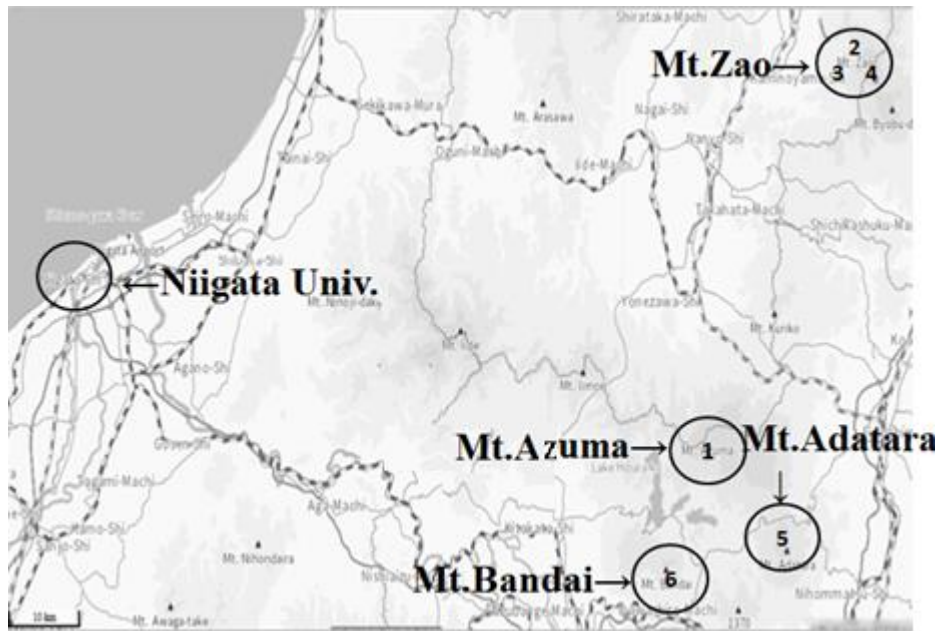
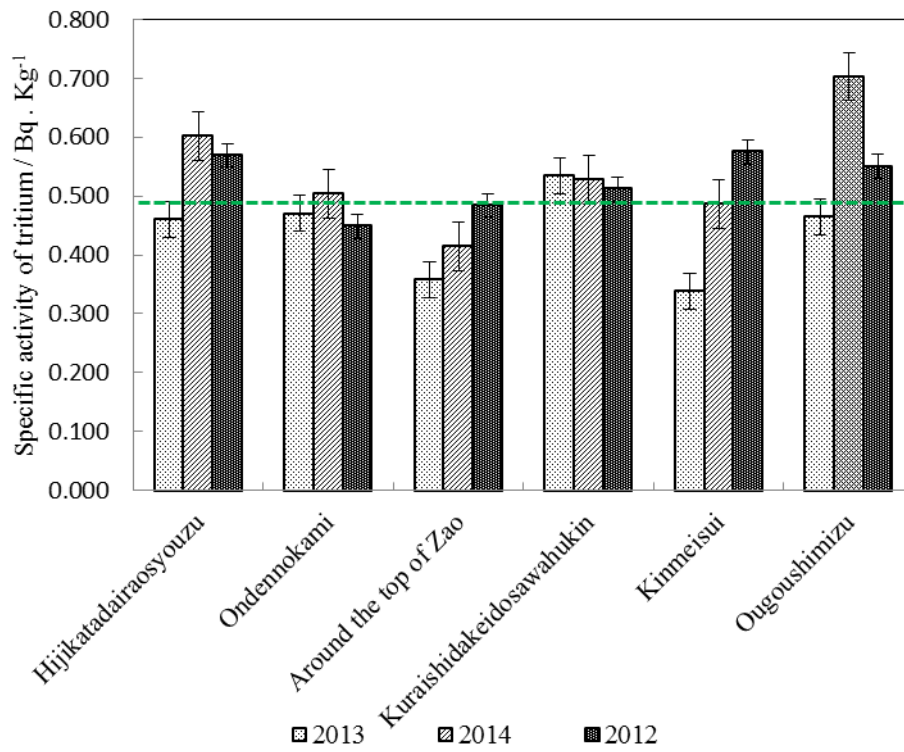


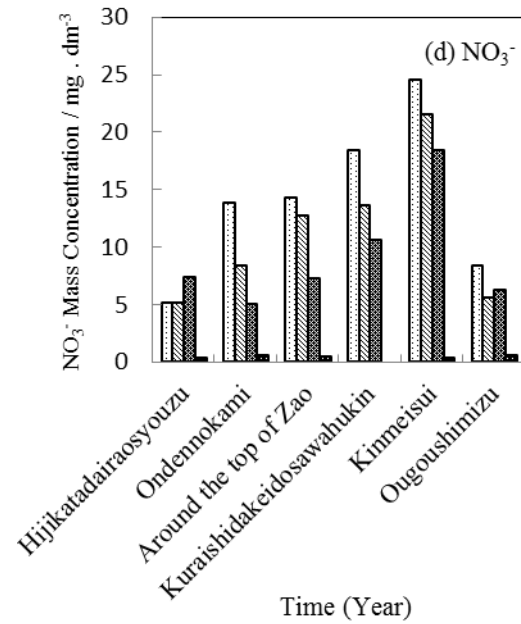
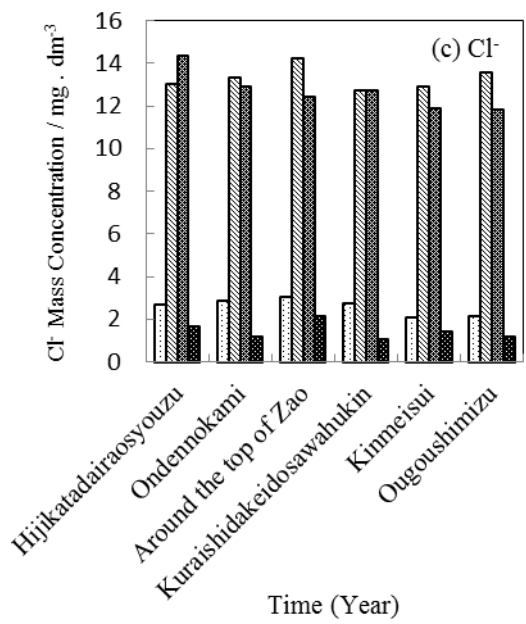
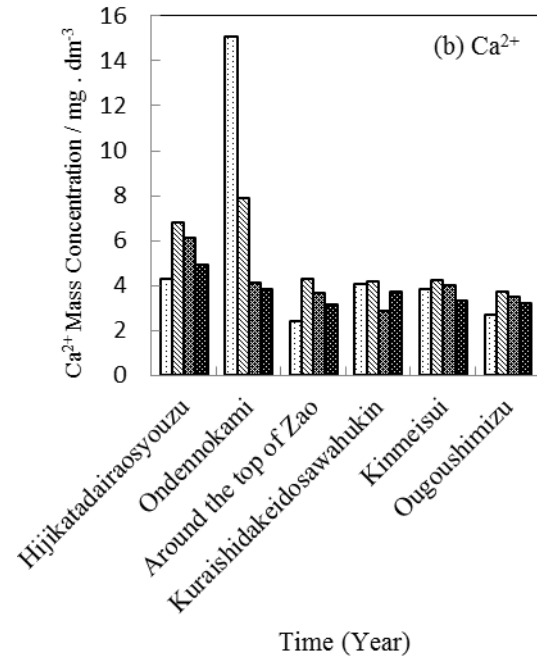
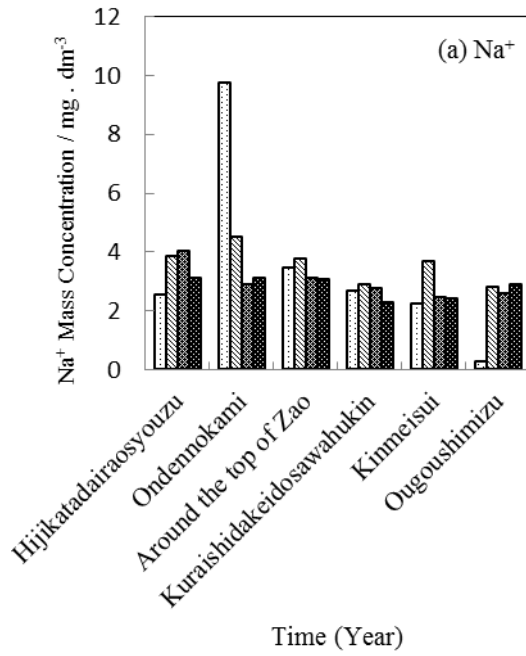
Fig.3.1 Spring water sampling sites in Fukushima and Yamagata prefectures.

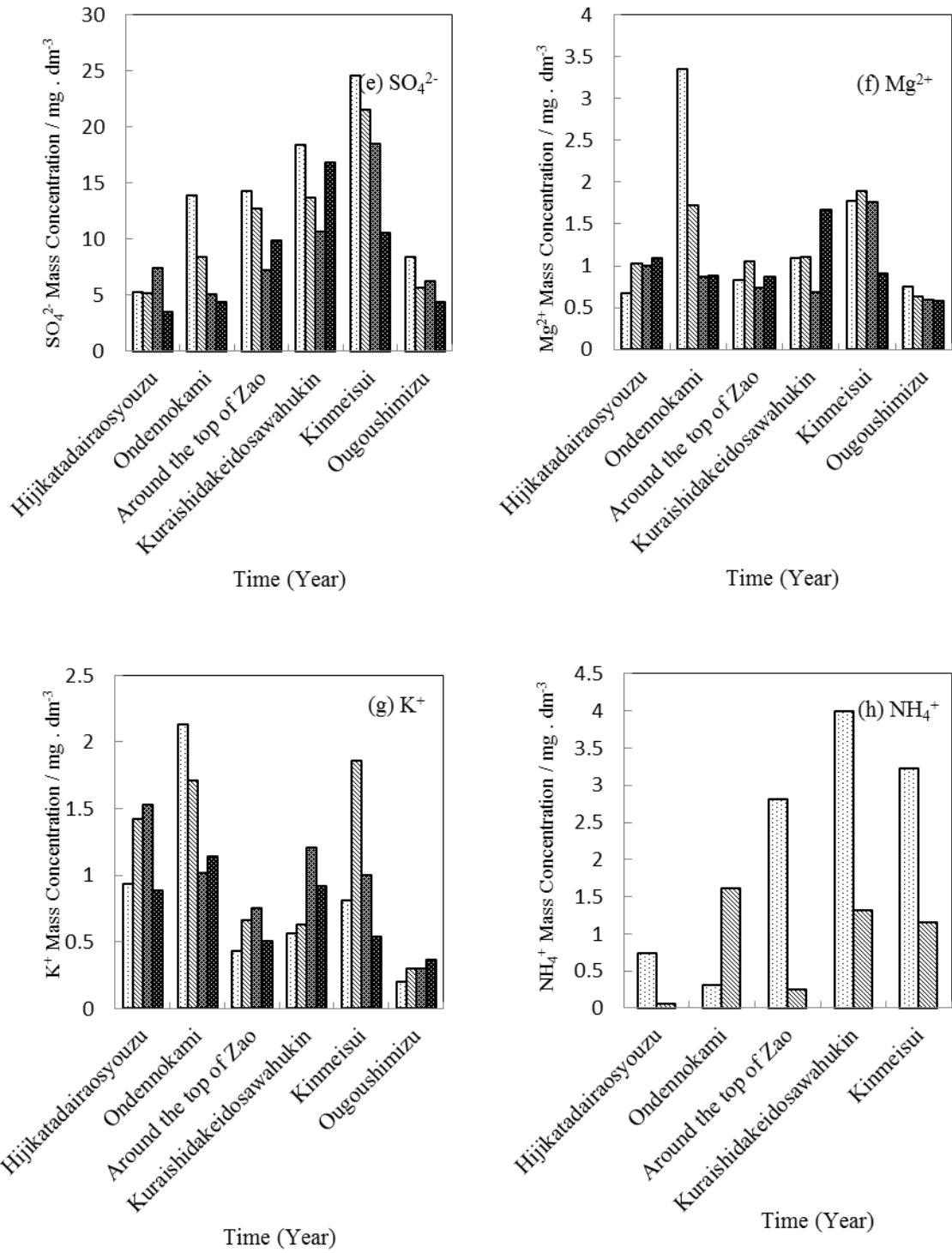
1. Hijikatadairaosyouzu (Mt. Azuma)
2. Ondennokami (Mt. Zao)
3. Around the top of Zao (Mt. Zao)
4. Kuraishidakeidosawafukin (Mt. Zao)
5. Kinmeisui (Mt. Adatarara)
6. Ougonshimizu (Mt. Bandai)



----- The average specific activity of T of spring water in August 28~31, 2012

Fig.3.2 Specific activity of tritium in each mountain.





□ 2012 ▨ 2013 ▩ 2014 ■ 2015

Fig.3.3 The concentrations of several ions in each location.

CHAPTER 4

Lake Water in Fukushima Prefecture and Niigata City

4.1 Introduction

In our previous research⁵⁸⁾, the specific activity of T in Niigata city was temporary increased by the accident, and the highest T content was also found in short term precipitation (the first rainfall) after the accident⁵⁹⁾. In this chapter, some lake waters and the sediments were collected in Fukushima prefecture and Niigata city, the specific activity of T and the concentration of ions were measured to investigate the effect of the accident on the environment after Fukushima Dai-ichi Nuclear Power Plant. Besides, dissolved Oxygen (DO), pH and ORP in the water were immediately measured at each sampling point also. Considering the above-mentioned, the following two aimed in this chapter: (1) to research the influence of the accident to several lakes in Fukushima prefecture and Niigata city; (2) to clarify the effect of the accident on Niigata city.

4.2 Experimental

4.2.1 Samples

In order to research the influence of the accident of Fukushima Dai-ichi nuclear power plant, we chosen four different lakes (Lake Inawashiro, Lake Akimoto, Goshikinuma and Lake Hayama) in Fukushima prefecture and two different lakes (Lake Toyonogata and

sakata lagoon) in Niigata city (as shown in Fig.4.1). Among them, Lake Inawashiro which contains Recreation park, Sakka beach and Tenjin beach these three spots. Besides, in all those sampling points, Lake Hayama is the nearest to the Fukushima Dai-ichi power plant.

These samples were collected in spring and autumn. The sample water was picked up about 5 meter from beach and depth is roughly 30 centimeter under surface. The sediment was also picked up using increment scoop (approximately 0 - 10 centimeter from the bottom of a lake). Sampling conditions and geographic are shows in Table 4.1 and 4.2. Moreover, DO, pH and ORP were immediately measurement at each sampling point.

4.2.2 Sample pretreatment and measurement

Each water sample thus obtained was analyzed with the same methods as shown in 2.2.2.

4.3 Results and discussion

4.3.1 Specific activity of T

Fig.4.2 shows the specific activity of T in each lake from 2013 to 2015. The figure shows that the specific activity of T at Lake Akimoto in May,

2013 showed higher than Sakata lagoon, and also higher than other samples in Fukushima prefecture. Lake Akimoto in Fukushima prefecture is surrounded by mountains, it seems a closed system same like Sakata lagoon in Niigata city, but one other thing to note is Sakata lagoon is not connected to Agano river but connected to Fukushima. So the figure indicates that the deposition of T is easily affected by the topography and may accumulate at the place where convection does not occur.

In addition, the figure also shows the accident of Fukushima Dai-ichi nuclear power plant gradually decreased in Fukushima prefecture.

Moreover, this research showed that the specific activity of T in Sakata lagoon and Lake Toyanogata were not large. In previous chapter (see chapter 2), the specific activity of T in precipitation in Niigata city is about was increased after the Fukushima Dai-ichi nuclear power plant, and the annual average of normal T concentration is about $0.5\text{Bq} \cdot \text{kg}^{-1}$. Therefore, based on the specific activity of T in Sakata lagoon and Lake Toyanogata, effect of the accident of Fukushima Dai-ichi nuclear power plant on Niigata city could not be observed.

4.3.2 Comparing the specific activity of T in monthly precipitation with of T in lake water

In order to further the effect of the accident of Fukushima Dai-ichi Nuclear Power Plant, we compared the specific activity of T in monthly precipitation and of in lake water, as shown in Table 4.3.

Overall, the specific activity of T had no significant difference in monthly precipitation between Niigata city and lake water in Fukushima prefecture and Niigata city. In previous chapter (chapter 1), we revealed that in the four month after Fukushima Dai-ichi Nuclear Power Plant, specific activity of T became to average level. So it is further indicates the effect of the nuclear accident has been go back to normal.

4.3.3 DO, pH, ORP and various ions

Dissolved Oxygen (DO), pH, Oxidation-Reduction Potential (ORP) and various ions in the sample were measured in lake water. DO, pH and ORP in the sample were directly measured at each sampling point (as shown in Table 4.4 and figure 4.2).

ORP is a common measurement for water quality. High ORP shows the pollution in water can filter completely. DO is an important parameter to measure when assessing water quality because of its influence on the organism living within a body of water. A DO level that is too high or too low can harm aquatic life and affect water quality.

At the sampling day, there were lots of plants in the lagoon and the water sampled was so muddy, therefore, combining each sampling point

the value of DO in Sakata lagoon was higher than others, that because at sampling day there were lots of plants in the lagoon, and the water sampled was so muddy. Moreover, the value of pH remained almost stable, as for ORP, based on current date, no available rules can explain changes about ORP, it affected by many factors, such as rain water, atmosphere and so on. In the figure, the value of ORP in May, 2013 and Jun. 2015 were higher than others, it may because there are much more rain water in this two months.

In addition, there are many hot springs near Goshikinuma tarn, so the value of these ion concentrations in the sample collected at Gishikinuma tarn were largely different from others, and the concentrations of the ions may be influenced by hot spring.

4.4 Conclusions

From the above-mentioned, the following two matters can be found:

- (1) The measured results show the specific of T was similar to that of the precipitation recently collected at Niigata city. Therefore, from the specific of T, the effect of the accident of Fukushima Dai-ichi Nuclear Power Plant on Niigata city may be no longer observed.
- (2) It seems that if the lake does not circulate, T cannot move, and be accumulated there.

Table and Figure

Table 4.1 Several sampling conditions

Sampling point	2013. May			2013. Sep.		
	Distance from shore / m	Lake water Temperature / °C	Depth /cm	Distance from shore/m	Lake water Temperature / °C	Depth /cm
Lake Inawashiro						
Reaction Park	5	16	30	5	21.2	10
Sakka beach	5	14	30	5	22.3	50
Tenjin beach	5	15.6	50	5	21.7	30
Lake Akimoto	3	17	10	5	23.8	20
Goshikinuma tarn	1	16	10	1	20.2	20
Lake Hayama	10	16	30	5	23.8	10
Lake Toyanogata	-	-	-	1	26	30
Sakata lagoon	1	23.1	20	1	23.6	30
Sampling point	2014. May			2014. Oct.		
	Distance from shore / m	Lake water Temperature / °C	Depth /cm	Distance from shore / m	Lake water Temperature / °C	Depth /cm
Lake Inawashiro						
Reaction Park	5	20.6	40	5	25.2	20
Sakka beach	5	18.6	30	5	20.9	20
Tenjin beach	5	19.4	40	5	21.6	30
Lake Akimoto	3	26.2	5	3	21.7	10
Goshikinuma tarn	1	20.6	10	1	17.2	5
Lake Hayama	10	26.1	10	10	20.6	20
Lake Toyanogata	1	29.4	15	1	18.6	20
Sakata lagoon	1	22.5	50	1	20.8	30
Sampling point	2015. Jun.					
	Distance from shore / m	Lake water Temperature / °C	Depth /cm			
Lake Inawashiro						
Reaction Park	5	24.3	30			
Sakka beach	5	20.6	30			
Tenjin beach	5	22.4	40			
Lake Akimoto	3	27.2	5			
Goshikinuma tarn	1	23.6	10			
Lake Hayama	10	27.1	30			
Lake Toyanogata	1	28.4	20			
Sakata lagoon	1	25.5	50			

Table 4.2 Geographic coordinates of sampling points

Sampling point	North latitude			East longitude		
Lake Inawashiro						
Reaction Park	37 °	30'	55"	140 °	1'	17"
Sakka beach	37 °	27'	45"	140 °	2'	48"
Tenjin beach	37 °	31'	18"	140 °	6'	57"
Lake Akimoto	37 °	39'	37"	140 °	7'	28"
Goshikinuma tarn	37 °	39'	10"	140 °	5'	22"
Lake Hayama	37 °	43'	42"	140 °	48'	27"
Lake Toyanogata	37 °	53'	49"	139 °	4'	1"
Sakata lagoon	37 °	49'	10"	138 °	52'	41"

Table 4.3 Comparing the specific activity of T [$\text{Bq} \cdot \text{kg}^{-1}$] in the monthly precipitation (Niigata city) with that in the lake water (Fukushima prefecture)

Sampling point	2013. May	2013. Sep	2014. May	2014. Oct	2015. Jun
Monthly Precipitation	0.47 ± 0.06	--	0.70 ± 0.07	0.48 ± 0.06	0.40 ± 0.05
Lake Inawashiro					
Reaction Park	0.48 ± 0.06	0.57 ± 0.06	0.40 ± 0.05	0.71 ± 0.07	0.33 ± 0.05
Sakka beach	0.18 ± 0.04	0.44 ± 0.05	0.45 ± 0.06	0.31 ± 0.05	0.35 ± 0.05
Tenjin beach	0.37 ± 0.05	0.57 ± 0.06	0.41 ± 0.05	0.30 ± 0.04	0.32 ± 0.05
Lake Akimoto	1.20 ± 0.09	0.53 ± 0.06	0.59 ± 0.06	0.39 ± 0.05	0.40 ± 0.05
Goshikinuma tarn	0.51 ± 0.06	0.70 ± 0.07	0.56 ± 0.06	0.74 ± 0.07	0.33 ± 0.05
Lake Hayama	0.70 ± 0.07	0.53 ± 0.06	0.65 ± 0.07	0.63 ± 0.06	0.40 ± 0.05
Lake Toyanogata	--	0.58 ± 0.06	0.56 ± 0.06	0.37 ± 0.05	0.28 ± 0.04
Sakata lagoon	0.52 ± 0.06	0.57 ± 0.06	0.45 ± 0.05	0.33 ± 0.05	0.39 ± 0.05

Table 4.4 Several date measured in 2013, 2014 and 2015 (mg / L)

Sampling point	2013. May			2013. Sep.		
	DO / mg · L ⁻¹	pH	ORP / mV	DO / mg · L ⁻¹	pH	ORP / mV
Lake Inawashiro						
Reaction Park	9.3	7.6	209	9.8	7.3	146
Sakka beach	10.3	8.0	220	8.3	8.6	156
Tenjin beach	9.8	7.6	206	9.3	9.0	148
Lake Akimoto	9.2	8.3	243	9.7	7.3	134
Goshikinuma tarn	9.2	7.3	209	7.9	7.0	156
Lake Hayama	9.6	7.9	230	7.9	7.6	155
Lake Toyanogata	-	-	-	6.5	6.4	72
Sakata lagoon	13.5	9.8	21.0	11.8	6.5	121
Sampling point	2014. May			2014. Oct.		
	DO / mg · L ⁻¹	pH	ORP / mV	DO / mg · L ⁻¹	pH	ORP / mV
Lake Inawashiro						
Reaction Park	9.1	7.8	557	9.7	6.8	130
Sakka beach	10.5	7.5	492	9.0	7.5	202
Tenjin beach	9.5	6.9	420	12.0	7.2	163
Lake Akimoto	8.0	7.3	398	10.0	7.4	216
Goshikinuma tarn	8.0	6.6	212	8.1	6.6	199
Lake Hayama	10.3	8.4	268	9.6	7.7	244
Lake Toyanogata	7.0	8.1	92	4.5	6.7	166
Sakata lagoon	11.0	8.2	591	11.3	7.9	221
Sampling point	2015. Jun.					
	DO / mg · L ⁻¹	pH	ORP / mV			
Lake Inawashiro						
Reaction Park	7.0	8.1	92			
Sakka beach	9.1	7.8	557			
Tenjin beach	10.5	7.5	492			
Lake Akimoto	9.5	6.9	420			
Goshikinuma tarn	8.0	7.3	398			
Lake Hayama	9.2	6.1	173			
Lake Toyanogata	10.3	8.4	268			
Sakata lagoon	11	8.2	591			

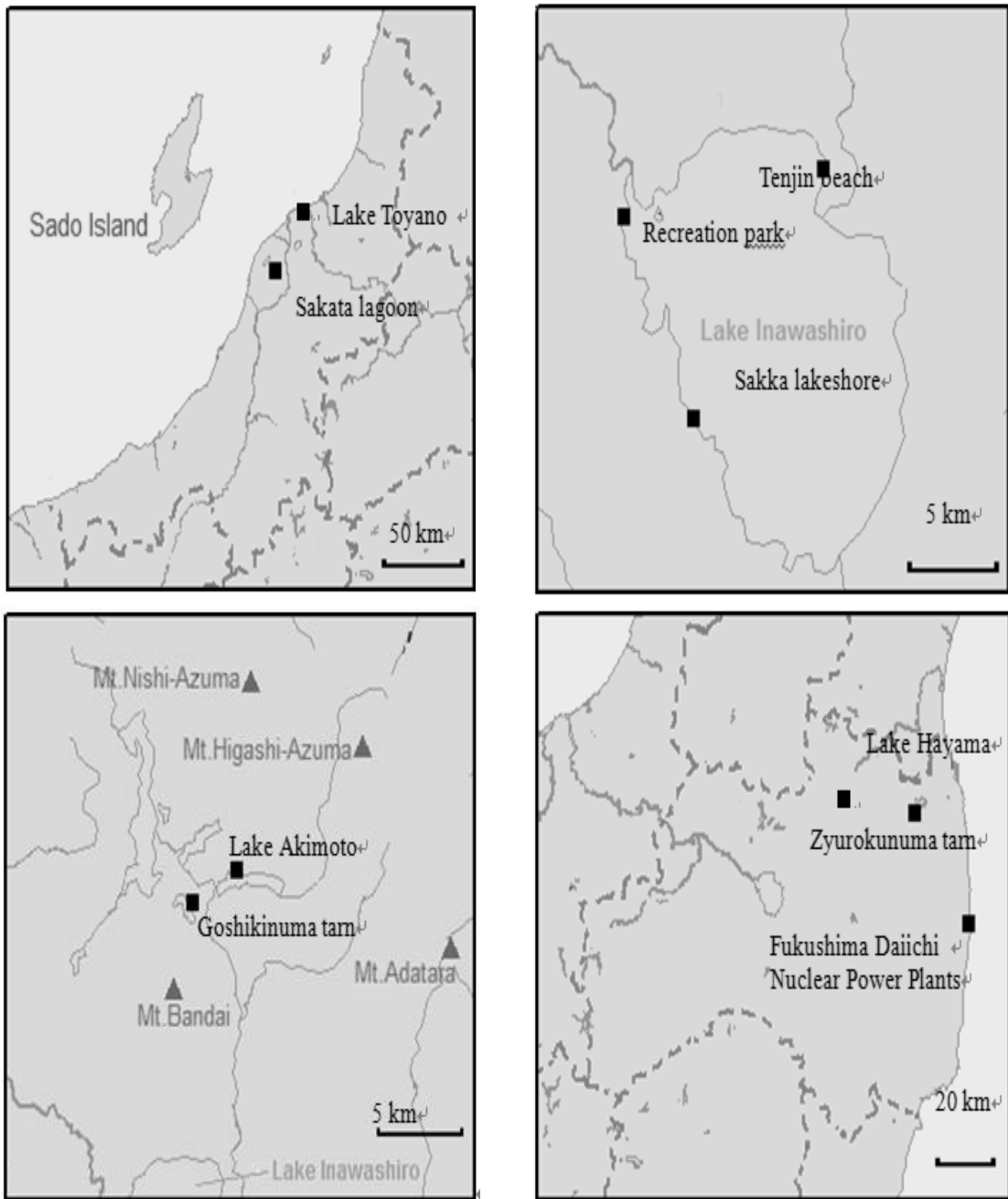


Fig.4.1 The sampling points at Lake Toyanogata and sakata lagoon (in Niigata city) and at Lake Inawashiro, Lake Akimoto, Goshikinuma and Lake Hayama (in Fukushima prefecture).

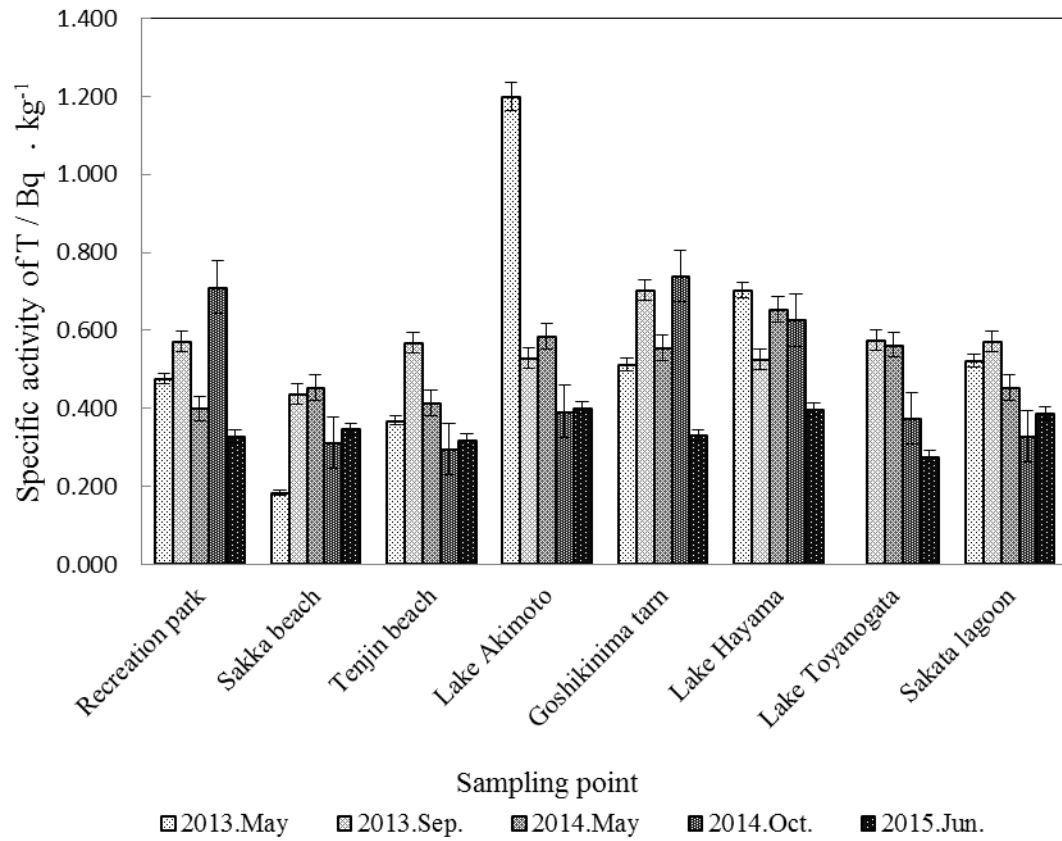
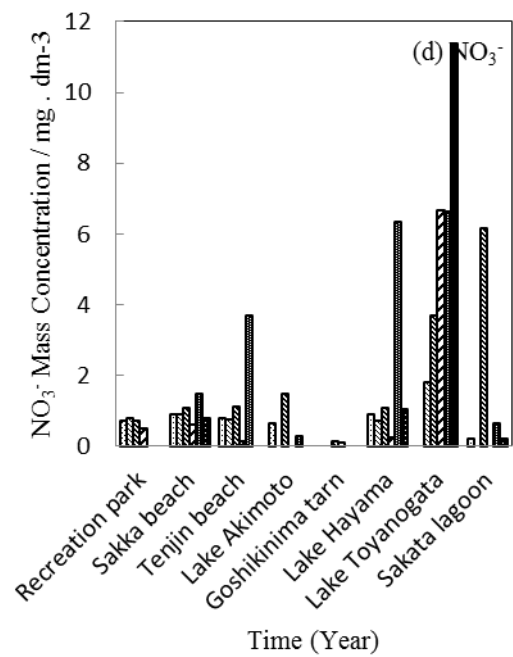
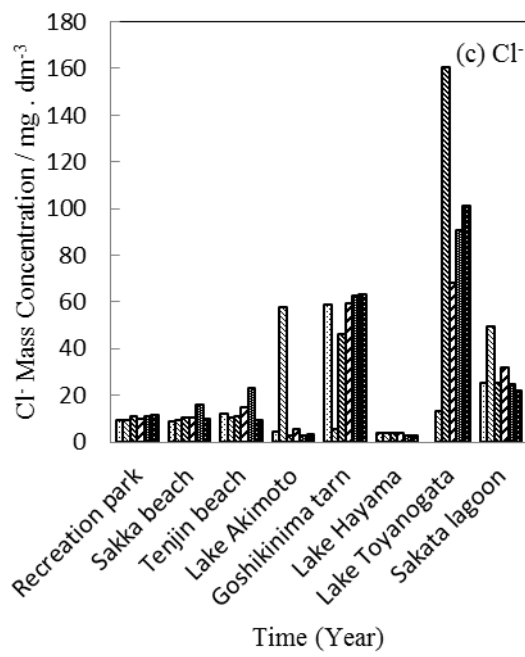
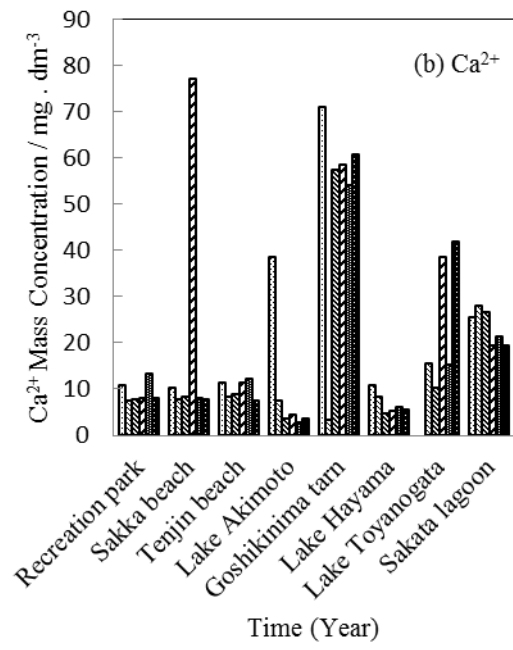
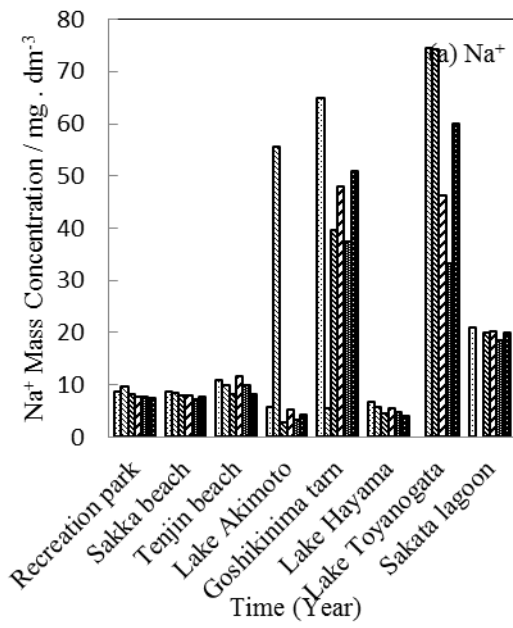
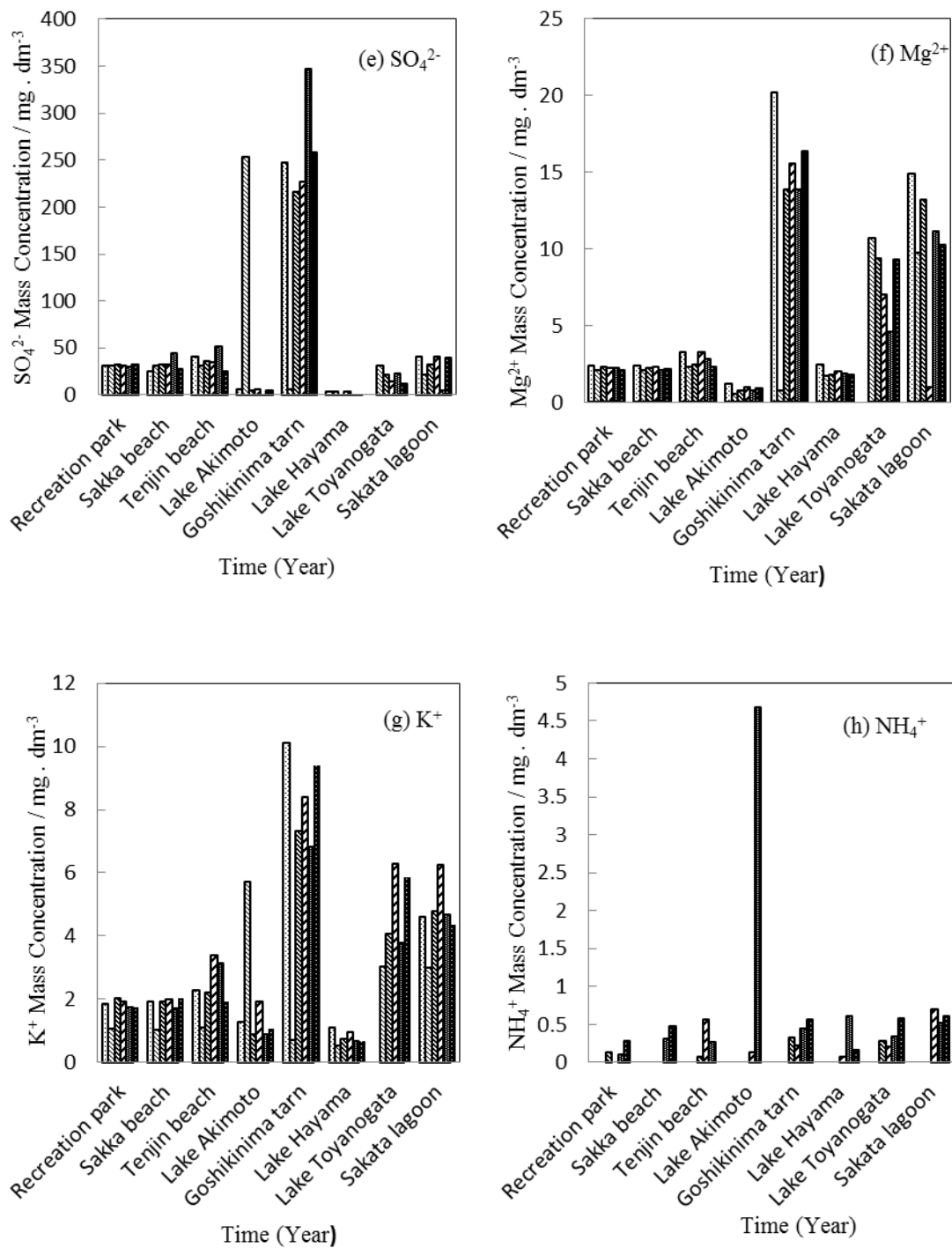


Fig.4.2 Specific activity of T in each lake in Fukushima or Niigata.





□ 2013. May ▨ 2013. Sep ▩ 2014. May ▪ 2014. Oct ▣ 2015. Jun ▤ 2015. Oct.

Fig.4.3 The concentrations of several ions in each location.

CHAPTER 5

CONCLUSIONS

In order to clarify the effect of the accident of Fukushima Dai-ichi Nuclear Power Plant on environment, the specific activity of T and major ion concentrations in precipitation, spring water and lake water were investigated, and the following main conclusions have been obtained:

- (1) The specific activity of T in monthly precipitation in March, 2011 was 5 times higher than in March in annual years. It is considered that the thus high level specific activity of T attributes the accident of the accident of Fukushima Dai-ichi Nuclear Power Plant.
- (2) The specific activity of T in precipitation having a strong dependence on season, is a maximum value in later winter and spring, and minimum value in autumn and early winter.
- (3) From the previous reports, it is obtained that the concentration of Ca^{2+} mainly changes with specific activity of T in monthly precipitation in Niigata city. In this study, the increases of the Ca^{2+} concentration was not found as significant change on March, 2011. Consequently, it shows that the increase of the specific of T was affected by the accident of Fukushima Dai-ichi Nuclear Power Plant.
- (4) All of the specific activity of T in spring water are less than 1.0 Bq / kg.
- (5) The specific activity of T in spring water was not greatly increased after 1 year, after the accident of Fukushima Dai-ichi Nuclear

Power Plant.

- (6) The specific activity of T in lake water was similar to the level of monthly precipitation in Niigata city.
- (7) Based on the specific activity of T, Niigata city is not so affected by the accident of Fukushima Dai-ichi Nuclear Power Plant after 4 months.
- (8) It can be concluded that the effect of the accident of Fukushima Dai-ichi Nuclear Power Plant on Niigata city is small.

From this thesis, it is found that the method used in this study is useful to obtain the reference value of specific activity of T and ion concentrations in precipitation, in spring water, and in lake water after any accident of Nuclear Power Plant, and also useful to obtain the reference value of seasonal variation of atmosphere. In addition, these data thus obtained is useful to evaluate the effect of the accident of Fukushima Dai-ichi Nuclear Power Plant on the environment.

Moreover, the results in this research can be useful to obtain a reference of other aspects of a nuclear accident.

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