

Seasonal Variation of Tritium and Major Elements in Snow and Precipitation in Niigata, Japan

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Abstract: In order to evaluate seasonal and regional variations in precipitation in Niigata City, 65 hourly precipitation samples were collected from October 2009 to June 2011 including two snow events. In this work, major ions (Na^+ , K^+ , Ca^{2+} , Mg^{2+} , NH_4^+ , SO_4^{2-} , NO_3^- and Cl^-) were combined with tritium (^3H or T) concentration (T specific activity) in both precipitation and snow, and transformation patterns of polluted air mass in Niigata region were revealed. The low level tritium in precipitation was measured by a distillation process and an electrolytic enrichment process. Each tritium concentration in the precipitation sample thus obtained was measured by liquid scintillation counter. On the basis of the above measurement and analysis, it was found that the tritium and nss (non-sea-salt) calcium concentrations showed a seasonal variation with a highest value in spring over one year.

Key words: Precipitation chemistry, tritium tracer, backward trajectory analysis, Fukushima accident, Niigata City.

1. Introduction

Over last few decades, researches on precipitation chemistry in different areas have received some attention [1-4]. A precipitation generally includes important particles and soluble gases from the atmosphere. Variations of the chemical composition in precipitation mean significant changes in anthropogenic emissions, and in atmospheric chemical transformations [5], which also dynamically include the rapid change of the biogeochemical cycles in the earth atmosphere [6].

Tritium (^3H or T), a hydrogen radioisotope, is one of the most important transient tracers used in hydrological research [7-12]. T emits a weak β -ray ($E_{\text{max}} = 18.6$ keV) and has the half-life of 12.33 year [13]. It originates from both artificial (e.g., nuclear power plant and nuclear accident) and natural events. T-concentration in precipitation depends on latitude, sea, distance from artificial sources, and

thermonuclear test zones. Up to now, only a few studies can be documented to use T as naturally occurring tracer to observe the annual or seasonal variation of precipitation in some regions. In this study, both tritium as a tracer and other major ions present in precipitation (and snow) are tried to combine, and to reveal the transformation patterns of air mass in Niigata region.

Niigata City is located in the center of Honshu Island in Japan, and is surrounded by many mountains including the Echigo mountain range. It has a rainy and snowy season with strong north wind from the sea of Japan during November to February, while the climate is mild from spring to autumn (March-October) in Niigata [14]. Niigata City is a very peculiar place and acts as a receptor for the huge natural and anthropogenic emissions from adjacent continent, and also has long range transport with the influence of monsoon [15]. Thus, the place is a particularly interesting region to investigate variation of the concentration of chemical component in air mass. Considering the above-mentioned, precipitation

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and snow samples were collected and analyzed in Niigata University to monitor the variations of chemical component in snow and wet precipitation. This research was carried out on monthly precipitation samplers from 2004 to 2011 [16]. Such a long-term monitoring could provide a credible opportunity to understand the climate in the region. However, monthly sampling periods may not exactly reveal variations in elemental composition and concentration due to rapid changes in meteorological condition (e.g., wind speed/direction, humidity, and temperature, etc.). Short-term (hourly) precipitation was accordingly collected and measured in this works to record such rapid changes of air mass influenced by the local and ambient resource. It is expected that the results from short-term precipitation obtained will give a wide impact on characterizing the air mass. In addition, samples of snow were also collected and analyzed in the fixed sampling site.

2. Methods and Data

2.1 Standard Reagents and Materials

Tritiated water (its specific activity is 3.7×10^7 Bq·cm⁻³) purchased from Japan Radioisotope Association was used to determine the T detection efficiency.

The standard solutions of Na⁺, K⁺, Ca²⁺, Mg²⁺, NH₄⁺, SO₄²⁻, NO₃⁻ and Cl⁻ were prepared by diluting a 1,000 mg·dm⁻³ standard solution, respectively. Those standard solutions (1,000 mg·dm⁻³) were purchased from KANTO CHEMICAL in Japan. The distilled water was used for the preparation of each standard solution.

The concentrations of inorganic species were measured by a DIONEX DX-120 Ion Chromatography having a conductivity detector (DIONEX, CO., SUN., CA 94086 USA). For analysis of anions (SO₄²⁻, NO₃⁻ and Cl⁻), separation column IonPac AS 12A and a suppressor IonPac AG 12A were used with a 2.7 mM Na₂CO₃/0.3 mM NaHCO₃ eluents at a flow rate of 1.0 mL·min⁻¹; and for cations

(Na⁺, K⁺, Ca²⁺, Mg²⁺ and NH₄⁺), a separation columns (IonPac CS 12A) and a suppressor column (IonPac CG 12A) were used with a 1.922 g·dm⁻³ CH₃SO₃H as eluents. In addition, concentrations of Na⁺ (and K⁺) were determined by flame spectrophotometry and Ca²⁺ (and Mg²⁺) by atomic absorption spectrophotometry (HITACHI, Japan). The authors' previous paper [16] presented the other methods.

2.2 Sampling

The study site (Fig. 1) is installed on the rooftop of the Faculty of Engineering, Niigata University, Niigata City of Japan (139°02' E, 37°55' N). At the fixed site, several open containers made of plastics were placed to collect precipitation. Thus, precipitations collected were placed in 2 dm⁻³ polyethylene bottles for the determination of each chemical component. For short-term precipitation, sampling times varied between one and several hours depending on each meteorological condition. Most of the samples were collected for 1 h. The study period of this paper was from October 2009 to June 2011.

During the above-mentioned period of collecting sample, short-term precipitation samples in three typhoon events were collected: the No. 20 Typhoon (October 26-27, 2009), the No. 4 Typhoon (August 4, 2010), and the No. 9 Typhoon (September 8, 2010), respectively [17]; two snow events as bulk precipitation were also collected (February 4, 2010 and January 2, 2011). The snow sample was melted after introducing in a 2 dm⁻³ polyethylene bottles. The precipitation accumulates over a period of time.

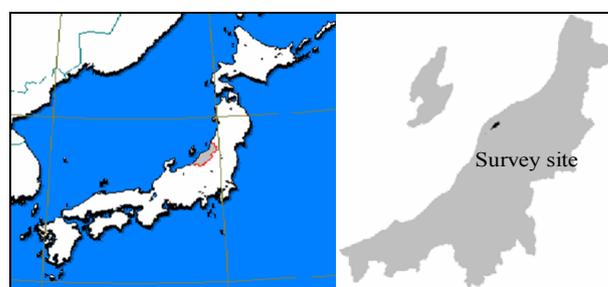


Fig. 1 Location of Niigata prefecture, Japan (139°02' E, 37°55' N).

2.3 Sample Pretreatment and Measurement

In this article, a cryogenic distillation process, a routine method for the determination of the low-level of T-concentration in natural water sample, was used as the first pretreatment step [18, 19] to prevent the concentration from other impurities and thereby minimize quenching [20]. This process was performed by adding 0.3 g of KMnO_4 and 0.3 g of Na_2O_2 to $1,380 \text{ cm}^3$ of each water sample. After being distilled, such samples were subsequently enriched by electrolytic enrichment method (SPE method) [21-23]. T-concentration of each enriched sample was detected by low-background liquid scintillation counter (LCS, Aloka Co., Ltd., Japan). Additionally, backward trajectory analysis has also been performed to estimate the passing route of air mass bringing the precipitation [24]. All these samples were stored for a week in Liquid Scintillation Counter (dark and cool

place) before measurement.

3. Results and Discussion

3.1 Seasonal Variation of Ionic Species

In general, the chemical composition of precipitation consists of a sea salt (denoted as “ss”) component generated from the ocean and a non sea salt (denoted as “nss”) one derived from the adjacent continents over the seas through medium-long range transport. It is assumed that Na^+ has no source other than marine origin, and all other ions originate from two major sources (“ss” and “nss”). The mass ratio of Ca^{2+} , Mg^{2+} , K^+ and SO_4^{2-} to that of Na^+ in seawater is 0.038, 0.12, 0.03 and 0.25, respectively. The analytical procedure was reported in detail by Prabha et al. [25, 26]. The concentrations of major ions in short-term precipitation are summarized in Table 1.

The concentrations of non-sea-salt components

Table 1 Tritium concentrations ($\text{Bq}\cdot\text{dm}^{-3}$) and chemical concentrations ($\text{mg}\cdot\text{dm}^{-3}$) in each short-term precipitation from October 2009 to July 2011 in Niigata City.

Time	T	Ca^{2+}	Mg^{2+}	K^+	Na^+	NH_4^+	SO_4^{2-}	NO_3^-	Cl ⁻	ss Ca^{2+}	ss K^+	ss Mg^{2+}	ss SO_4^{2-}
*2009.10.26 17:30-18:30	0.770	0.677	0.337	0.221	3.037	-	1.001	1.441	5.032	0.115	0.091	0.364	0.759
*2009.10.26 18:30-19:30	0.443	0.607	0.085	0.141	0.243	0.778	0.300	0.177	0.600	0.009	0.007	0.029	0.061
*2009.10.26 19:30-20:30	0.325	0.756	0.070	0.395	0.217	1.452	0.334	0.172	0.507	0.008	0.007	0.026	0.054
*2009.10.26 20:30-21:30	0.309	0.957	-	0.095	0.160	0.496	0.233	0.174	0.374	0.006	0.005	0.019	0.040
*2009.10.26 21:30-22:30	0.456	0.486	0.071	0.175	0.145	0.562	0.329	0.372	0.351	0.006	0.004	0.017	0.036
*2009.10.26 22:30-23:30	0.222	0.905	-	0.092	0.096	0.496	0.417	0.327	0.187	0.004	0.003	0.012	0.024
*2009.10.26 23:30-00:30	0.726	0.456	0.027	0.163	0.166	1.221	0.334	0.184	0.399	0.006	0.005	0.020	0.042
*2009.10.27 00:30-01:30	0.433	0.453	0.025	0.138	0.199	0.559	0.715	0.006	0.480	0.008	0.006	0.024	0.050
*2009.10.27 01:30-02:30	-	0.844	0.051	0.087	0.225	1.619	0.746	1.348	0.662	0.009	0.007	0.027	0.056
*2009.10.27 02:30-03:30	0.341	0.661	-	-	0.121	1.729	0.598	1.130	0.298	0.005	0.004	0.015	0.030
*2009.10.27 03:30-04:30	0.518	0.432	0.041	-	0.237	1.342	0.429	0.421	0.601	0.009	0.007	0.028	0.059
*2009.10.27 04:30-05:30	-	0.781	0.216	0.091	1.106	0.361	0.873	1.069	2.531	0.042	0.033	0.133	0.277
2010.06.15 12:30-14:30	0.428	1.091	0.093	1.528	0.773	4.847	1.179	1.988	1.887	0.029	0.023	0.093	0.193
2010.06.21 09:30-10:30	1.200	1.085	0.017	1.449	0.346	4.210	1.538	1.471	0.785	0.013	0.010	0.042	0.087
2010.06.21 10:30-11:30	1.156	0.396	0.018	0.724	0.056	1.182	1.374	0.731	0.151	0.002	0.002	0.007	0.014
2010.06.21 11:30-12:30	1.065	0.619	-	1.774	0.284	5.188	2.676	2.651	0.851	0.011	0.009	0.034	0.071
2010.06.23 09:30-10:30	1.153	0.799	0.030	2.274	0.398	3.205	1.492	2.000	0.916	0.015	0.012	0.048	0.100
2010.06.23 10:30-11:30	-	1.008	0.014	2.128	0.579	5.063	1.143	2.647	1.318	0.022	0.017	0.069	0.145
2010.07.12 11:30-12:30	3.858	0.768	0.128	0.124	1.101	-	0.635	0.322	1.555	0.042	0.033	0.132	0.275
2010.07.12 12:30-13:30	1.079	0.370	0.179	0.143	0.847	0.194	0.737	0.588	1.685	0.032	0.025	0.102	0.212
2010.07.14 14:30-15:30	0.472	0.272	-	0.091	0.112	0.316	0.741	1.053	0.323	0.004	0.003	0.013	0.028
2010.07.14 15:30-16:30	0.538	0.171	0.027	0.277	0.083	0.509	0.495	0.730	0.116	0.003	0.002	0.010	0.021
2010.07.14 16:30-17:30	0.461	0.377	-	0.373	0.097	0.775	0.516	0.728	0.196	0.004	0.003	0.012	0.024
2010.07.15 15:30-16:30	0.428	0.334	-	0.252	0.139	0.734	0.851	1.707	0.796	0.005	0.004	0.017	0.035

(Table 1 continued)

Time	T	Ca ²⁺	Mg ²⁺	K ⁺	Na ⁺	NH ₄ ⁺	SO ₄ ²⁻	NO ₃ ⁻	Cl ⁻	ssCa ²⁺	ssK ⁺	ssMg ²⁺	ssSO ₄ ²⁻	
*2010.08.12	11:30-12:30	1.028	0.794	-	0.134	0.098	1.266	0.755	1.346	0.387	0.003	0.003	0.012	0.025
*2010.08.12	12:30-13:30	1.377	0.418	0.095	0.461	0.438	2.264	0.730	0.860	1.154	0.017	0.011	0.052	0.110
*2010.08.12	13:30-14:30	1.445	1.728	2.316	0.801	16.810	-	5.870	4.269	28.00	0.639	0.504	2.017	4.202
2010.09.07	17:30-18:30	0.749	0.244	0.266	0.079	1.396	0.094	0.817	0.006	2.277	0.053	0.042	0.168	0.340
2010.09.07	18:30-19:30	0.675	0.110	0.171	0.060	0.861	-	0.357	0.026	1.744	0.033	0.026	0.103	0.215
2010.09.07	19:30-20:30	0.749	0.102	0.086	-	0.408	-	0.178	-	0.784	0.016	0.012	0.049	0.102
2010.09.07	20:30-21:30	0.697	0.066	0.058	-	0.349	-	0.198	0.126	0.720	0.013	0.007	0.042	0.087
2010.09.07	21:30-22:30	0.554	0.302	-	0.024	0.094	-	0.075	0.022	0.121	0.004	0.003	0.011	0.024
2010.09.07	22:30-23:30	1.531	0.179	-	0.111	0.081	0.267	0.018	0.047	0.159	0.003	0.002	0.010	0.020
2010.09.07	23:30-00:30	2.204	0.118	0.051	0.068	0.294	0.054	0.265	0.163	0.573	0.011	0.009	0.035	0.074
2010.09.07	00:30-01:30	1.719	0.173	0.066	0.083	0.427	0.078	0.279	0.011	0.864	0.016	0.013	0.051	0.107
*2010.09.08	05:30-06:30	1.102	0.207	0.051	0.266	0.237	0.210	0.165	0.324	0.881	0.009	0.007	0.028	0.059
*2010.09.08	10:30-11:30	0.968	0.125	0.194	0.214	0.887	0.373	0.885	0.856	1.460	0.034	0.027	0.106	0.222
*2010.09.08	11:30-12:30	0.863	0.108	4.396	0.279	0.389	0.478	1.256	0.665	0.611	0.015	0.012	0.047	0.097
2010.10.04	10:30-11:30	3.723	0.164	0.078	0.078	0.551	1.145	1.102	0.453	1.517	0.021	0.017	0.066	0.137
2010.10.05	10:30-11:30	1.108	0.388	0.378	1.847	2.381	9.565	6.117	-	5.546	0.090	0.071	0.286	0.595
2010.11.15	09:30-10:30	1.463	1.108	0.716	0.308	6.755	0.132	5.597	3.343	9.166	0.257	0.203	0.811	1.689
2010.11.15	10:30-11:30	1.550	0.669	0.541	0.594	3.989	0.539	4.868	2.357	6.153	0.152	0.203	0.479	0.997
2010.11.15	11:30-12:30	0.896	0.675	0.714	0.196	5.280	0.116	3.721	1.405	7.288	0.201	0.158	0.634	1.320
2010.11.15	12:30-13:30	-	0.199	0.482	0.188	4.106	0.067	2.230	0.851	5.599	0.156	0.123	0.493	1.027
2010.11.15	13:30-14:30	1.367	0.209	0.455	0.182	3.995	0.073	2.366	0.010	5.188	0.152	0.120	0.479	0.999
2010.11.15	14:30-15:30	2.405	0.469	1.260	0.382	10.97	0.156	5.364	2.611	16.909	0.417	0.329	1.316	2.743
2010.11.15	15:30-16:30	1.000	0.618	0.903	0.479	7.430	0.129	3.222	1.925	12.959	0.282	0.223	0.892	1.858
2010.11.15	16:30-17:30	0.637	1.471	0.640	0.301	5.453	0.111	2.624	1.234	7.569	0.207	0.164	0.654	1.363
2010.12.03	10:30-17:30	1.762	3.513	1.900	0.587	13.76	-	5.802	0.014	24.744	0.523	0.413	1.651	3.440
2011.03.15	13:30-14:30	-	3.828	1.184	1.546	4.739	2.311	18.860	21.10	11.500	0.180	0.142	0.569	1.185
2011.03.15	14:30-15:30	2.039	1.920	0.767	1.204	3.263	1.831	12.600	12.49	7.718	0.124	0.098	0.392	0.816
2011.03.15	15:30-16:30	2.628	1.189	0.458	0.620	1.718	1.136	6.810	7.559	3.021	0.065	0.052	0.206	0.430
2011.03.15	16:30-17:30	2.130	0.594	0.283	0.456	1.233	0.673	3.069	3.739	2.141	0.047	0.037	0.148	0.308
2011.03.15	17:30-18:30	11.554	0.293	0.188	0.505	1.061	0.422	1.504	2.394	1.726	0.040	0.032	0.127	0.265
2011.04.19	11:30-12:30	1.118	0.436	0.045	0.295	0.220	0.144	0.646	0.841	0.322	0.008	0.007	0.026	0.055
2011.04.19	12:30-13:30	2.048	0.329	0.028	0.109	0.104	0.083	0.301	0.409	0.188	0.004	0.003	0.012	0.026
2011.04.19	13:30-14:30	5.464	0.215	0.028	0.154	0.059	0.100	0.525	0.372	1.388	0.002	0.002	0.007	0.015
2011.04.19	14:30-15:30	5.674	0.201	0.027	0.123	0.054	0.084	0.56	0.464	0.928	0.002	0.002	0.006	0.014
2011.04.19	15:30-16:30	5.363	0.143	0.026	0.115	0.053	0.116	1.525	0.700	0.789	0.002	0.002	0.006	0.013
2011.04.19	16:30-17:30	6.715	0.365	0.013	0.117	0.076	0.080	0.347	0.235	0.280	0.003	0.002	0.009	0.019
2011.04.19	17:30-18:30	8.228	0.561	0.018	0.100	0.091	0.096	0.605	0.953	0.824	0.003	0.003	0.001	0.023
2011.06.23	09:00-14:30	8.408	1.950	0.533	0.893	4.030	-	5.493	3.383	3.803	0.153	0.121	0.484	1.001
2011.06.24	09:30-10:30	0.486	0.908	0.405	0.706	3.100	-	4.489	0.763	1.648	0.118	0.093	0.372	0.775
2011.06.24	12:30-13:30	0.218	0.264	0.320	0.368	2.330	-	3.581	0.658	1.536	0.089	0.070	0.280	0.583
2011.06.24	13:30-14:30	0.716	0.048	0.105	0.250	1.098	-	1.430	0.100	0.508	0.042	0.033	0.132	0.275

Time is referring to local time.

“-” represents the defect of data due to small amount of sample; “*” represents typhoon events.

(Na⁺, Mg²⁺ and Cl⁻) are influenced by marine sources. During typhoon, they have higher values (16.81 mg·dm⁻³, 2.316 mg·dm⁻³ and 28 mg·dm⁻³, respectively) than other days due to an increase in sea salt aerosols.

In all of precipitation samples, NO₃⁻ was the most abundant ion with concentration of 21.10 (mg·dm⁻³) (Table 1). The precipitation event was caused by continental air mass. However, the backward

trajectories are useful to understand the path route of air mass.

3.2 Tritium Concentration in Precipitation

Measured T-concentrations in each sample are listed in Table 1, ranging from $0.218 \text{ Bq}\cdot\text{cm}^{-3}$ to $11.554 \text{ Bq}\cdot\text{cm}^{-3}$. The seasonal variations could be estimated from a series of T data for hourly precipitation. Overview one year, a significant difference appears among the four seasons. The T-concentration is higher in spring than in other seasons. In general, natural T-concentration reported in precipitation depends on latitude, sea, distance from artificial sources, and thermonuclear test zones. A relatively high T-concentration in spring is attributed to the downward movement of natural T from stratosphere to troposphere (named “spring peak”). This phenomenon generally appears in the northern hemisphere between 30° N and 60° N in spring each year. The sampling site locates at $37^\circ 55' \text{ N}$, and is warm due to the land mass effect, what is called

“continental effect”.

Under normal conditions, the significant difference of the T-concentration in precipitation within one air mass would not be expected because the T-concentration in precipitation is not influenced by temperature variation. It seems that a different air mass contributes to the precipitation on the site in a short period. Precipitations collected during a severe convective storm showed no significant difference within hourly periods in our work. Only a quick change of T-concentration has been observed within a typhoon event due to the complexity of meteorological conditions. It should be pointed out that the precipitation collected in 2011 after Tohoku Earthquake Japan occurred in March 11, 2011 showed high T-concentrations. It caused the Pacific tsunami that led to the disastrous Fukushima nuclear accident in the following days. Up to the present, the nuclear accident was therefore clarified in this work by the presence of high T-concentration in precipitation (Fig. 2).

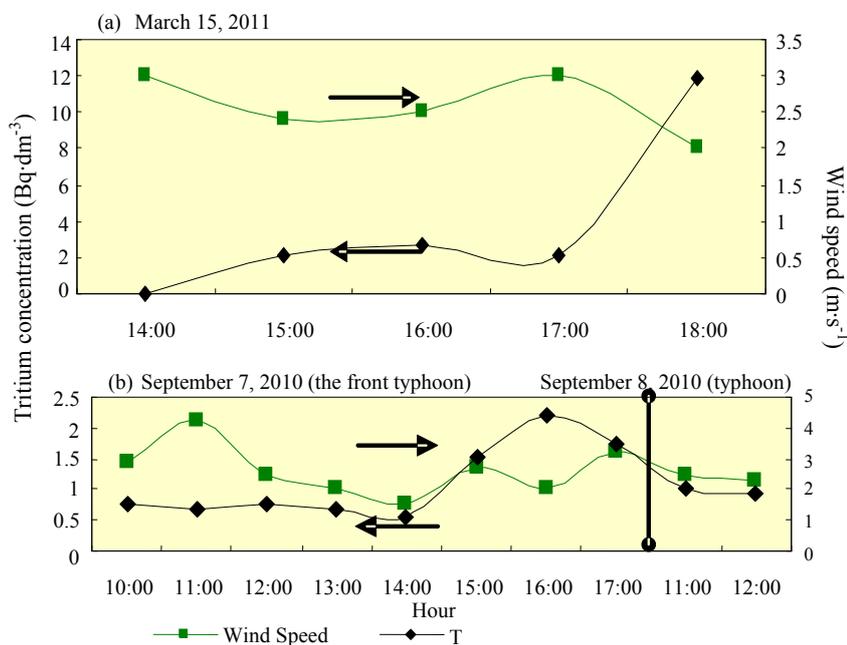


Fig. 2 Time dependences of tritium concentration and wind speed in short-term precipitation: (a) air mass derived from continent; (b) air mass came from ocean.

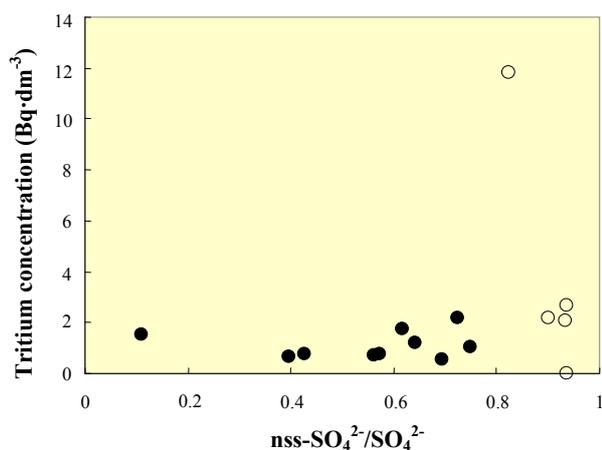


Fig. 3 The ratio of nss-SO₄²⁻ to SO₄²⁻ in precipitation vs. tritium concentration: ○: precipitation coming from continent; ●: precipitation in a typhoon.

3.3 Relation between Tritium and Major Ions in Precipitation

Short-term precipitation consists of two parts: one is normal precipitation; the other is typhoon. The wind speed and T-concentration in precipitation were shown in Fig. 2. It is clear that the T-concentration is proportional to the wind speed, which means that the higher T-concentration in precipitation was closely related to the air mass from the sea.

To further clarify the origin of air mass, T-concentration was compared with the ratio of nss-sulfate to all sulfate in two events (Fig. 3). T-concentrations and the ratios in typhoon precipitation are always lower than that in normal precipitation, that is, typhoon precipitation has notable character of marine air mass.

In order to further clarify the origin of air mass and the relationship of each ion, the variations of NH₄⁺ with NO₃⁻ (or nss-SO₄²⁻) in two events were also compared with each other. The results were shown in Figs. 4a and 4b, respectively. The origin of air mass can be determined by comparing the relationship between NH₄⁺ and nss-SO₄²⁻ (or NO₃⁻). Each correlation coefficient in continental air mass (0.958 or 0.9571) is higher than that in the marine air mass (0.0239 or 0.4871).

3.4 Variations of Ionic Species in Snow Sample

The measured result of snow sample was shown in Table 2. Any relation between the concentrations of ionic species in melting snow is not systematically clear. However, these data provide a unique result to understand the air mass variation in winter.

Groundwater generally varies with a little heavy water in evaporation without significant transpiration by vegetations. Therefore, T-concentration in snow is relatively low. The high concentrations of ions are attributed to the air mass origin from the vicinal continent. This aspect may be found using the backward trajectory in winter (see Section 3.5).

3.5 Tracing the Source of Air Mass Using Backward Trajectory

Backward (or simply “back”) trajectory analysis is a flexible tool to understand the variations of air mass for short-term precipitation. It could provide additional data of deducing some source-receptor relation. Herein the authors try to estimate the pollution sources for each collection event using the METE (meteorological data explorer) model, which was introduced by Zeng [27] in more detail. To enter the starting time (refer to UTC) in this model, the approximate travel path of air mass can be derived back for several days. This method covered a 120-hour span and 1,000 meters high of air mass. The other analysis data were based on “Meteorological Data explorer” [28] by the Center of Global Environmental Research, National Institute for Environmental Studies. The backward trajectory of rain event in each month was shown in Figs. 5a-5j.

It is noted that backward trajectory analysis is also a good tool to estimate the most possible travel path of air mass. On the basis of backward trajectory analysis of short-term precipitation at different time in one day, a variation of air mass behavior was clearly revealed in one day, which was consistent with one experiment result.

Table 2 Tritium concentrations ($\text{Bq}\cdot\text{dm}^{-3}$) and chemical concentrations ($\text{mg}\cdot\text{dm}^{-3}$) in snow in Niigata City.

Time	T	Ca^{2+}	Mg^{2+}	K^+	Na^+	NH_4^+	SO_4^{2-}	NO_3^-	Cl ⁻	ssCa ²⁺	ssK ⁺	ssMg ²⁺	ssSO ₄ ²⁻
2010.02.04	0.719	0.401	0.527	0.140	4.369	-	2.340	0.681	6.984	0.166	0.131	0.524	1.092
2011.01.12	1.369	0.279	0.431	0.215	3.701	0.091	1.338	0.515	4.887	0.141	0.111	0.444	0.925

Time is referring to local time.

“-” represents the defect of data due to small amount of sample.

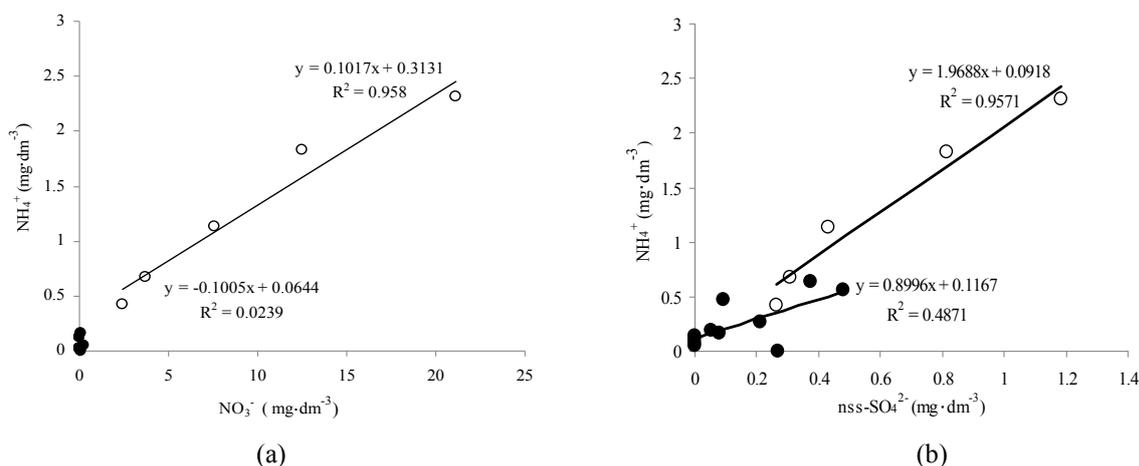


Fig. 4 Relationships between ionic concentrations in continental air mass (○) and marine air mass (●): (a) NH_4^+ - NO_3^- ; (b) NH_4^+ -nss- SO_4^{2-} .

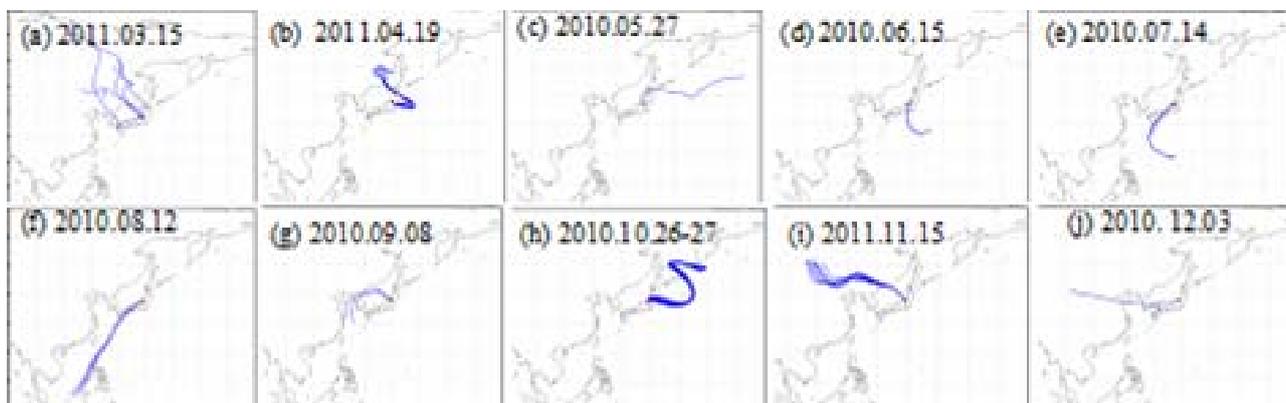


Fig. 5 5-days back trajectories arrived at Niigata University ($139^{\circ}02'$ E, $37^{\circ}55'$ N) with 1,000 m altitude in each month of year (for each picture, longitude from 90° to 180° and latitude from 0° to 80°).

4. Conclusions

T-concentration and concentrations of major ions in some short-term precipitations were investigated to understand the regional variation of air mass. The results indicate that: (1) Elemental compositions and concentrations in precipitations among different days were remarkably affected by variations of air masses caused by the changes of meteorological conditions; (2) T-concentration and other ionic concentrations could be

used to determine the origin of air mass. T-concentration in precipitation shows a seasonal variation with the highest value in spring, and a slow decrease from autumn to early winter over the whole year; (3) Each ionic concentration in precipitation depends on local atmosphere.

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