

CHEMICAL COMPOSITION OF FOG WATER AT MT. TATEYAMA NEAR THE COAST OF THE JAPAN SEA IN CENTRAL JAPAN

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With 6 figures, 2 tables and 2 photos

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Summary: Measurements of the chemical composition of fog water at Murododaira (altitude, 2,450 m), near the summit of Mt. Tateyama near the coast of the Japan Sea, were performed during the summers (late July and August) of 2004 and 2008 and the autumns (September and early October) of 2008 and 2009. Fog water was also sampled at Bijodaira (altitude, 977 m) and Midagahara (altitude, 1,930 m), on the western slope of Mt. Tateyama, in the autumn of 2009. Strong acidic fogs ($\text{pH} < 4$) were observed during the summer and autumn. The mean ionic concentration at Murododaira in the summer of 2008 was higher than that in the autumn. The air mass at Murododaira was derived primarily from the polluted regions of Asia in the summer of 2008, and the trans-boundary air pollution might have contributed to fog water acidification. On the other hand, acidic fogs might have been produced mainly by pollutants from central and western Japan in 2004. The non-sea-salt sulfate (nssSO_4^{2-}) concentration was usually higher than the NO_3^- concentration at Murododaira, and the chemical characteristics are different from that of fog water near the industrial regions of Japan where NO_x is the main air pollution. The average ratio of $\text{NO}_3^-/\text{nssSO}_4^{2-}$ in fog water was relatively high in the summer of 2004. The mean $\text{NO}_3^-/\text{nssSO}_4^{2-}$ ratio was significantly low during the autumn of 2008 when the air mass usually came from Asia. Relatively high concentrations of sea-salt components were detected at Murododaira during the autumn of 2009. The acidity of fog water was higher at Murododaira than at Bijodaira and Midagahara. The deposition flux of acidic species by fog water seemed to be higher at Murododaira than at the lower sites.

Zusammenfassung: Bei Murododaira (2.450 m N.N.) nahe der Westküste der Japanischen See, etwas unterhalb des Gipfels des Mt. Tateyama, wurden während der Sommermonate 2004 (Ende Juli und August) und im Herbst 2008 (September und Anfang Oktober) Messungen der chemischen Bestandteile von Nebelwasser durchgeführt. Beprobte wurde im Herbst 2009 ebenfalls das Nebelwasser bei Bijodaira (977 m N.N.) und bei Midagahara (1.930 m N.N.) auf der Westabdachung des Mt. Tateyama. Die Beobachtungen zeigten, dass die Nebel sowohl im Sommer als auch im Herbst einen ausgesprochen niedrigen pH-Wert (< 4) aufwiesen. Die mittlere ionische Konzentration lag bei Murododaira im Sommer 2008 höher als im Herbst. Im Sommer 2008 wurde der Raum Murododaira von Luftmassen beeinflusst, die mit südwestlicher Strömung Schadstoffe aus chinesischen und koreanischen Industriegebieten heranführten. Im Jahr 2004 hingegen müssen die sauren Nebel vermutlich als Resultat von Schadstoffeinträgen im zentralen und westlichen Japan angesehen werden. Bei Murododaira war die Konzentration der Sulfate (nssSO_4^{2-}) dabei in der Regel höher als die NO_3^- Konzentration und die chemischen Charakteristika wiesen deutliche Unterschiede zu jenen von Nebelwasser aus den japanischen Industriegebieten auf, in denen NO_x die Hauptbelastung darstellt. Das durchschnittliche Verhältnis von $\text{NO}_3^-/\text{nssSO}_4^{2-}$ im Nebelwasser war im Sommer 2004 relativ hoch, im Herbst 2008 aber, als die Luftmassen aus Richtung Asien anströmten, im Mittel ausgesprochen niedrig. Ein relativ hoher Anteil von Seesalzbestandteilen wurde im Herbst 2009 bei Murododaira gemessen. Insgesamt ließ sich eine höhere Acidität des Nebelwassers bei Murododaira im Vergleich zu den niedrigeren Stationen Bijodaira und Midagahara feststellen.

Keywords: Fog water, chemical composition, pollution, transport, Mt. Tateyama

1 Introduction

Acidic fogs or clouds have been observed not only near urban areas but also at mountainous sites far from industrial regions (OKITA 1968; MINAMI and ISHIZAKA 1996) and remote islands (GIODA et al. 2009). Acid fogs are thought to contribute to the de-

cline of forests at high elevations, where mountain slopes are frequently immersed in fog or cloud water (e.g., BLANK 1985). Many chemical measurements of fog water have been performed at high-elevation sites, especially in the United States, Canada and Europe (e.g. OGREN and RODHE 1986; MOHNEN and KADLECEK 1989; COLLETT et al. 1990, 2002; FUZZI et

al. 1996; COLVILE et al. 1997; PLESSOW et al. 2001; KIM and ANEJA 1992; WOBROCK et al. 1994; SCHEMENAUER et al. 1995; ANDERSON et al. 1999, 2006; BRIDGES et al. 2002; BRÜGGEMANN et al. 2005; VAN PINXTEREN et al. 2005; KLEMM and WRZESINSKY 2007; ZAPLETAL et al. 2007; MÖLLER 2008; ALEKSIC et al. 2009; BLÁŠ et al. 2009; HUTCHINGS et al. 2009). After the 1990s, the chemical composition of cloud or fog water has also been measured in Asian countries, such as India (ALI et al. 2004; LAKHANI et al. 2007), China (LU and NIU 2009; LU et al. 2010; LIU et al. 2010; NIU et al. 2010; WANG et al. 2011), South Korea (CHOI et al. 1996; KIM et al. 2006), Taiwan (WANG et al. 2010) and Japan (IGAWA et al. 1998; WATANABE et al. 1999, 2006a; AIKAWA et al. 2001, 2005; OGAWA et al. 2004; OKOCHI et al. 2007; KIKUCHI et al. 2009). However, there is a shortage of fog water chemistry data from high-elevation sites, especially those higher than 2,000 m, in East Asian countries, including Japan, where the decline of forests has become a serious environmental problem. The transport of air pollution highly affects the chemical composition of fog water. Observations at mountainous sites higher than 2,000 m are useful to estimate the influence of the middle- or long-range transport of pollutants.

The rapid growth of industry in East Asia is associated with high anthropogenic emissions of pollutants (AKIMOTO and NARITA 1994; OHARA et al. 2007). Atmospheric pollutants from the Asian continent may be actively transported to the atmosphere over the Japan Sea (HATAKEYAMA et al. 2001; WATANABE et al. 2001a). The air pollutants promote cloud and precipitation water acidification and affect the microphysical properties of the clouds or fogs (WATANABE et al. 2001a). In addition to pollutants, Asian dust particles (called Kosa particles), mineral dust particles rich in CaCO_3 , are also transported from the Asian continent (TSURUTA 1991; WATANABE and HONOKI 2003; WATANABE et al. 2005a, 2006b). Recently, large amounts of acidic species have been deposited in the Hokuriku District (Fig. 1), along the Japan Sea coast in central Japan (HONOKI et al. 2001, 2007; WATANABE et al. 2011a). Mt. Tateyama, a part of the mountain range in the Hokuriku District, is near the Japan Sea coast, where a large amount of air pollution may be transported from the Asian continent as well as from industrial regions in Japan.

In previous studies at Mt. Tateyama, high concentrations of pollutants derived from the continent, such as O_3 , SO_2 and aerosols, were observed (KIDO et al. 2001; OSADA et al. 2003; WATANABE et al. 2006c). Preliminary measurements of the chemical constituents of several fog or cloud water samples

were also made at Mt. Tateyama in 2003 (WATANABE et al. 2005b). A strong acidic fog, probably affected by long-range transport of air pollution, was detected. Furthermore, a serious decline of the forest in the vicinity of Mt. Tateyama was observed (KUME et al. 2009). Air pollution and acidic fog may contribute to damage to vegetation. From 2004, we systematically performed fog chemistry observations at Mt. Tateyama, and the chemical compositions of fog water in the autumns (September and October) from 2004 to 2007 were described (WATANABE et al. 2010a). The chemical characteristics of fog water showed year-to-year variations. Highly acidic fogs containing high concentrations of SO_4^{2-} were observed under the influence of the air mass transported from polluted regions in the Asian continent (WATANABE et al. 2010a). To elucidate air quality at Mt. Tateyama, continuous measurements of fog chemistry for years are required.

The aim of this study is to explain the concentrations of fog water chemistry at high elevations near the coast of the Japan Sea in central Japan in the summer and autumn, when the vegetation surface is directly exposed to fogs at high elevations. The relationship between the mean ionic concentrations and the transport process of air mass (climatology) is of particular interest. According to Osada et al. (2009), the volume concentration of submicrometer aerosols near the summit of Mt. Tateyama was significantly higher in the summer than in the autumn. To evaluate vegetation damage by acidic fogs, it is important to understand the fog water chemistry during not only the autumn but also the summer. In this paper, the summer and autumn chemical characteristics (chemical climatology), especially the anthropogenic species, of the fog water sampled at the western slope of Mt. Tateyama, which faces the Asia continent, are discussed. The characteristics of sea-salt components in the fog water are also described

2 Methods

2.1 Sampling and analytical technique

Fog water collection was mainly performed at Murododaira (altitude, 2,450 m), which is located far from the heavy industrial areas of Japan on the western slope of Mt. Tateyama near the Japan Sea coast, from late July to October of 2004 and 2008 and in September and October of 2005, 2006, 2007 and 2009. Unfortunately, fog water sampling was not done during the summers from 2005 through 2007



Fig. 1: Map of Japan showing the locations of Mt. Tateyama and the Hokuriku District, and Murododaira (Mu), Midagahara (Mi) and Bijodaira (B) (the contour interval of elevation is 500 m)

and in 2009. A map of Japan with the location of Mt. Tateyama is shown in figure 1. Due to severe conditions, such as heavy snow cover from winter (or late autumn) to early summer, observations could usually be carried out only in the summer and autumn. Photo 1 presents a view of the sampling site, Murododaira. Because the chemical compositions of fog water during the autumns from 2004 to 2007 have already been reported (WATANABE et al. 2010a), the results of the observations during the summers (late July and August) of 2004 and 2008, and the autumns (September and early October) of 2008 and 2009 are discussed in this paper. Fog events are usually observed as extended clouds, not local upslope fogs, at Murododaira because local upslope fogs are mainly observed at altitudes below 2,000 m, which is lower than the altitude of Murododaira, at Mt. Tateyama.



Photo 1: View of the sampling site, Murododaira (altitude, 2,450 m)

To compare altitudinal characteristics of chemical composition of fog water on the western slope of Mt. Tateyama, fog water was also sampled at Bijodaira (altitude, 977 m) and Midagahara (altitude, 1,930 m), on the western slope of Mt. Tateyama in the autumn of 2009. Measurements of aerosol particles and trace gases were also performed at Bijodaira (WATANABE et al. 2006c, 2010b, 2011b). Fog events are mainly seen as upslope and radiation fogs at Bijodaira and Midagahara.

Fog water was collected using a passive thin-string sampler (Model FWP-500) from Usui Kogyo Kenkyusho, Inc., as described in detail by WATANABE et al. (2005b) and AOKI and WATANABE (2009). The passive sampler, which does not require electricity, is advantageous for use at high elevations and has been used at Mt. Fuji (DOKIYA et al. 2001; WATANABE et al. 2006a). A hood with a diameter of 1 m to keep out rain droplets was also mounted (AOKI and WATANABE 2009). However, perfect sampling of fog water without contamination of rain or drizzle may be impossible. In this report, water collected using the passive sampler is regarded as a fog water sample. Photo 2 shows the fog water sampler mounted at Murododaira. The fog water collected with the sampler was stored in a 5-liter tank and recovered every 3–14 days. The weight (volume) of the fog water samples was measured *in situ*. The samples were carried back to Toyama Prefectural University and stored in a refrigerator. The fog water sampler as well as the tank was washed using pure water every 3–14 days after recovery of the fog water samples. In addition to the regular sampling, intensive fog water sampling (every 2–4 hours) was also conducted at Murododaira during



Photo 2: Passive thin-string fog water sampler mounted at Murododaira

the autumns of 2004, 2005 and 2006. The concentrations of peroxides (mainly, H_2O_2) as well as those of major ions were measured during the intensive observation. The results of the intensive observation were also reported by WATANABE et al. (2009, 2010a); not only strong acidic fogs but also high concentrations of peroxides were observed at Murododaira.

Unfortunately, the liquid water content (LWC) was not directly measured by an LWC analyzer, which requires electricity. The collection efficiency of the passive sampler is affected by the wind speed. In particular, the volume of fog water samples is small when the wind velocity is low. The wind speed is usually high during a fog occurrence near the summit of Mt. Tateyama, therefore, fog water could be collected effectively even with the passive sampler at Murododaira. However, wind speed is relatively low during fog events at Bijodaira and Midagahara, the samples collected by the passive sampler at the two sites were usually smaller than those at Murododaira. In this study, the mean chemical concentrations in fog water were calculated by the concentrations and the weight (volume) of the samples, and the chemical climatology is mainly discussed because the relationships between the chemical composition of fog water and transport processes during individual fog events (the intensive sampling) have already been reported (WATANABE et al. 2010a). Sampling with a passive sampler may be useful to evaluate indirectly chemi-

cal deposition by fog water, which has important effects on the vegetation surface at high elevations.

The pH of the fog water was measured with an electrode using a pH meter. The analysis of the dissolved ionic species was performed using an ion chromatograph, as described in detail by WATANABE et al. (2005b).

2.2 Backward trajectory analysis

Backward trajectory analysis was employed to examine the transport process of the air mass to Murododaira, Mt. Tateyama. The analysis was based on the HYSPLIT4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) Model, 1997 (Web address:

<http://www.arl.noaa.gov/ready/hysplit4.html>, NOAA Air Resources Laboratory, Silver Spring, MD), including the vertical motion mode, with the reanalyzed meteorological data available from the National Centers for Environmental Prediction/National Centers for Atmospheric Research (NCEP-NCAR) (Web address: <http://www.cpc.ncep.noaa.gov/products/wesley/reanalysis.html>). The reanalysis datasets have a spatial resolution of $2.5^\circ \times 2.5^\circ$. Backward trajectories were calculated for 5 days, arriving at altitudes of 2,500 m.

3 Results and discussion

3.1 Acidity of fog water

Table 1 shows the range of the concentrations of major ions and pH in fog water at Murododaira (altitude, 2,450 m) during the regular (3-14 days) sampling in the summers (late July and August) of 2004 and 2008 and the autumns from 2004 through 2009. The data shown for September and October from 2004 through 2007 are from WATANABE et al. (2010a). The fog water samples in the autumns of 2008 and 2009 were taken during September and early October. The levels of non-sea-salt sulfate (nssSO_4^{2-}) and non-sea-salt calcium (nssCa^{2+}) were calculated using the following equations:

$$\text{nssSO}_4^{2-} = \text{SO}_4^{2-} - (\text{SO}_4^{2-}/\text{Na}^+)_{\text{seawater}} \cdot \text{Na}^+,$$

$$\text{nssCa}^{2+} = \text{Ca}^{2+} - (\text{Ca}^{2+}/\text{Na}^+)_{\text{seawater}} \cdot \text{Na}^+,$$

where $(\text{SO}_4^{2-}/\text{Na}^+)_{\text{seawater}}$ and $(\text{Ca}^{2+}/\text{Na}^+)_{\text{seawater}}$ are the

Tab. 1: Summary of the chemistry of fog water obtained by regular sampling at Murododaira (altitude, 2,450 m) during the summers (July and August) and autumns (September and October) from 2004 through 2009. Data in September and October from 2004 through 2007 are from WATANABE et al. (2010). Units of ionic concentrations are micro-equivalents per liter

Period	pH	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	nssSO ₄ ²⁻	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	nssCa ²⁺
Summer of 2004	4.4-5.1	8-16	15-83	38-69	35-65	12-30	15-82	1-5	3-14	11-59	10-57
Summer of 2008	3.7-4.2	23-35	63-146	146-296	138-293	27-31	77-181	0-4	8-14	26-58	25-56
Autumn of 2004	4.0-5.4	1-125	0-140	15-254	14-234	2-171	1-101	0-13	0-50	14-55	14-48
Autumn of 2005	3.5-5.5	6-59	2-52	24-311	22-310	4-53	28-143	3-18	2-23	5-32	4-29
Autumn of 2006	4.5-6.3	21-171	5-156	40-273	35-263	23-89	8-60	5-14	6-48	24-377	23-374
Autumn of 2007	4.3-5.5	3-34	5-166	26-123	25-111	3-92	8-65	0-26	0-28	5-217	5-212
Autumn of 2008	4.1-4.7	13-134	25-123	100-392	99-382	9-194	40-302	2-19	2-37	7-130	7-126
Autumn of 2009	3.8-5.7	6-41	15-93	28-199	24-197	14-108	18-120	1-7	0-11	8-29	7-27

concentration ratio of SO₄²⁻ to Na⁺ and that of Ca²⁺ to Na⁺ in seawater, which are 0.12 and 0.044 (equivalent ratio), respectively (Keene et al. 1986). According to the Na⁺ concentrations, SO₄²⁻ and Ca²⁺ were mainly nssSO₄²⁻ and nssCa²⁺, respectively.

The pH of fog water at Murododaira ranged from 3.7 to 5.1 during the summer. The acidity of fog water in the summer was comparable with that in the autumn. The pH of fog water ranged from about 3.5 to 6.3 in September and October from 2004 through 2009. Highly acidic fogs (pH < 4) were sometimes observed during the regular (3-14 days) sampling at Murododaira. Moreover, strong acidic fogs with pH values lower than 3.5 were also sometimes taken during the intensive sampling when short-interval (2-4 hours) collection was performed (WATANABE et al. 2010a). On the other hand, the pH of fog water was relatively high in the autumn of 2006, and the concentration of nssCa²⁺ was also high (Tab. 1). Kosa particles (background Kosa) rich in CaCO₃ might have been abundant over the Japan Sea side during the autumn of 2006, and acidic fogs seemed to be neutralized (WATANABE et al. 2010a). Kosa particles were also detected at Mt. Tateyama after late October in 2009 (WATANABE, pers. comm.). However, the chemistry of fog water sampled at Murododaira during September and early October in 2009 was scarcely affected by Kosa particles.

Table 2 shows the range of the concentrations of major ions and pH in fog water at Bijodaira and Murododaira during the autumn (September and early October) of 2009. The pH of fog water ranged from 3.9 to 6.0 and from 3.8 to 6.2 at Bijodaira and Midagahara, respectively. Highly acidic fogs were also observed at both sites. The chemical composition of fog water at Bijodaira seems to be highly affected by local pollution (WATANABE et al. 2005b).

3.2 Chemical characteristics in the summer

The chemical characteristics of fog water at Murododaira were dramatically different during the summers of 2004 and 2008. Highly acidic fogs were frequently observed in 2008, on the other hand, the pH of fog water was relatively high in 2004. The concentrations of nssSO₄²⁻ were high during the summer of 2008. The volume-weighted mean concentrations of major ions in fog water during the summers of 2004 and 2008 are shown in figure 2. The mean pH values of fog water in 2004 and 2008 were 4.5 and 4.0 (3.95), respectively. The concentrations of chemical constituents in 2008 were much higher than those in 2004. The mean ionic concentrations in the summer of 2008 were also higher than those in the autumn (WATANABE et al. 2010a).

First of all, the chemical concentrations in cloud water are highly dependent on the liquid water content (LWC) or fog duration. Unfortunately, the LWC was not directly measured with an LWC analyzer in this study. The total weight of all samples which correlates with the LWC and cloud duration was about 1.5 times higher in 2004 than in 2008 (about 6,000 g/month in 2004 and about 4,000 g/month in 2008). The concentrations of total ions in 2008 were three times higher than those in 2004 (Fig. 2). The chemical characteristics in the 2 years cannot be explained by the sample amounts alone. According to OSADA et al. (2009), submicrometer aerosols in the summers (August) at Murododaira were much higher in 2008 than in 2004. Air pollution might have been easily transported in the summer of 2008.

The nssSO₄²⁻ concentration was usually higher than the NO₃⁻ concentration at Murododaira, and the equivalent ratio of NO₃⁻/nssSO₄²⁻ in fog water was quite different from that in fog water at the mountainous sites near the industrial regions of

Tab. 2: Summary of the chemistry of fog water obtained by regular sampling at Bijodaira (altitude, 977 m) and Midagahara (altitude, 1,930 m) during the autumn (September and October) of 2009. Units of ionic concentrations are microequivalents per liter

Site	pH	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	nssSO ₄ ²⁻	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	nssCa ²⁺
Bijodaira	3.9-6.0	41-140	61-323	60-383	48-364	98-250	26-187	8-25	18-92	35-162	30-151
Midagahara	3.8-6.2	12-246	41-619	108-627	104-580	32-394	46-432	6-73	9-170	44-270	42-253

Japan. The NO₃⁻ concentration is usually higher than the nssSO₄²⁻ concentration (the NO₃⁻/nssSO₄²⁻ ratio is higher than 1) in fog water highly affected by polluted air from urban areas in Japan where the emission of SO₂ is suppressed strictly and NO_x is main air pollution (MURANO 1993; HOSONO et al. 1994; IGAWA et al. 1998; WATANABE et al. 2001b; TAGO et al. 2006). The NO₃⁻/nssSO₄²⁻ ratio at Murododaira seems to show year-to-year variations. The ratio of NO₃⁻/nssSO₄²⁻ was relatively high in 2004. Some samples showed that the concentrations of NO₃⁻ were higher than those of nssSO₄²⁻ in 2004. The NO₃⁻/nssSO₄²⁻ ratio in the summer of 2004 was also higher than that in the autumn (WATANABE et al. 2010a). The most predominant cation other than H⁺ was NH₄⁺ (Fig. 2). The concentrations of NH₄⁺ were also high in 2008. However, the ratio of (NH₄⁺ + nssCa²⁺)/(NO₃⁻ + nssSO₄²⁻), a useful indicator of the amount of bases needed to neutralize the acids, was similar during both years.

The chemical characteristics in the 2 years might have been due to the difference in the transport processes of air pollution. The results of a 5-day backward trajectory analysis arriving at altitudes of 2,500 m during the sampling periods in the summers of 2004 and 2008 are shown in figure 3. The trajectories were plotted once daily. Figure 3 presents predominant transport processes of air mass during both years. The air mass usually came from central

and western Japan in the summer of 2004, whereas that at Murododaira in 2008 was mainly derived from Asia, especially from the coasts of the Yellow Sea and East China Sea, where SO₂ (and NH₃) emissions are large (AKIMOTO and NARITA 1994; OHARA et al. 2007). In the summer, central Japan is usually under the influence of a maritime air mass; however, the continental air mass was predominant in 2008. We experienced typically a hot summer and a relatively cool summer in 2004 and 2008, respectively. Climate of monsoon summer in Japan may be significantly affected by global warming as well as El Niño and Southern Oscillation (ENSO) (e.g. KIMOTO 2005; KUSUNOKI et al. 2006). According to the results of calculation by the Earth Simulator, the withdrawal of the rainy season called the Baiu in Japan will be delayed until August by global warming (KUSUNOKI et al. 2006); as a result, central Japan will be easy to be influenced by a continental air mass during the summer. Perhaps global warming or climate change will control the fog water chemistry at Murododaira.

The predominant air mass significantly influences the chemical composition of fog water at Murododaira, Mt. Tateyama. WATANABE et al. (2010a) examined the relationship between the chemistry of fog water collected intensively (every 2–4 hours of sampling) at Murododaira in the autumn and the transport processes of the air mass. High concentrations of nssSO₄²⁻ and low pH were observed under the influence of the air mass transported from the polluted regions in Asia (WATANABE et al. 2005b, 2010a). Large amounts of air pollutants, such as sulfate aerosols (e.g. (NH₄)₂SO₄), might have been easily transported from the Asian continent to Mt. Tateyama in the summer of 2008. The acidification of fog water is accelerated by trans-boundary pollution. According to the results of the backward trajectory analysis done in 2008, the fog water chemistry might have also been affected by the air pollution from western Japan (mixed pollution). Therefore, the ionic concentrations were high in the summer of 2008. On the other hand, chemical constituents in fog water in 2004 might have been mainly affected by the air pollution from the industrial regions of Japan, where SO₂ emission is restricted and nitrogen

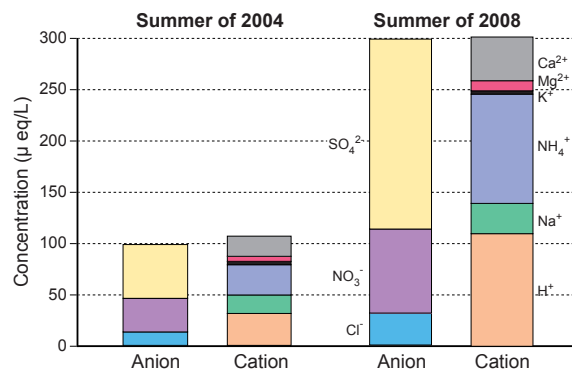


Fig. 2: Weighted mean concentrations of major ions in fog water at Murododaira (altitude, 2,450 m) obtained by the regular sampling in late July and August of 2004 and 2008

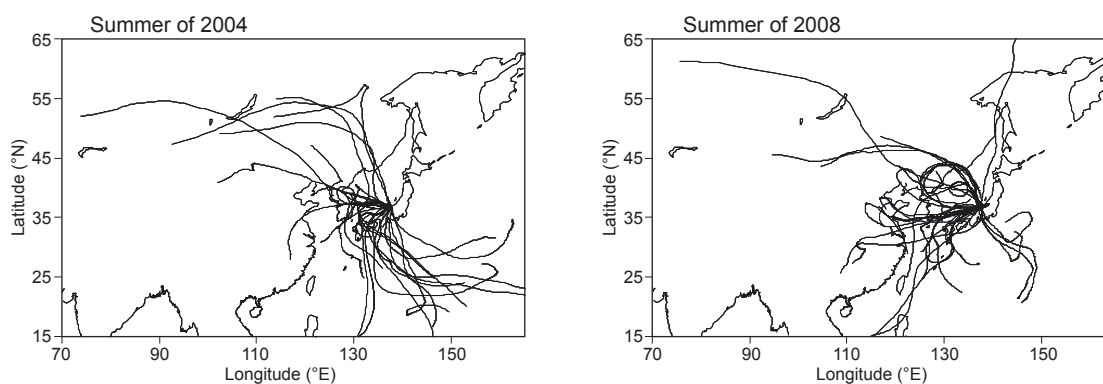


Fig. 3: Results of the 5-day backward trajectory analysis at an altitude of 2,500 m during the regular sampling periods in late July and August of 2004 to 2008. The trajectories were plotted once daily, and the starting time was 9:00 JST (0:00 UTC)

oxides (NO_x) are the main air pollution. As a result, a relatively high $\text{NO}_3^-/\text{nssSO}_4^{2-}$ ratio was observed in 2004. Some trajectories in 2004 were derived from the Asian continent; however, very little of the air mass was transported from the highly polluted areas.

3.3 Chemical characteristics in the autumn

The volume-weighted mean concentrations of major ions in fog water during the autumns of 2008 and 2009 are shown in figure 4. The mean pH values of fog water in 2008 and 2009 were 4.1 (4.11) and 4.2 (4.15), respectively. While the acidity (H^+) was slightly higher in 2008, the ionic concentrations in 2009 were higher than those in 2008. The total weight of all samples was about 1.3 times higher in 2008 than in 2009; about 4,000 g/month in the autumn of 2008 and about 3,000 g/month in the autumn of 2009. Perhaps the chemical concentrations in the 2 years were due to the difference in the sample amounts. The mean ionic constituents in the autumns of 2008 and 2009 were higher than those in the summer of 2004 and lower than those in the summer of 2008.

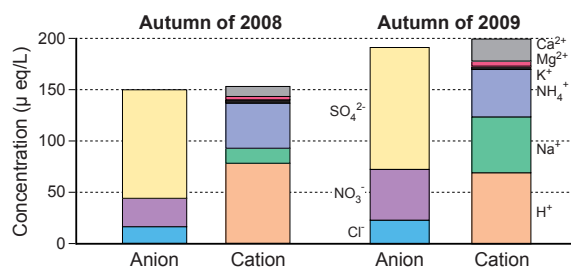


Fig. 4: Weighted mean concentrations of major ions in fog water at Murododaira (altitude, 2,450 m) obtained by the regular sampling in September and early October of 2008 and 2009

Therefore, the deposition of the chemical composition, such as acidic species, onto the sampler by fog water, which is difficult to estimate quantitatively, might have been significantly high during the summer of 2008.

The SO_4^{2-} (or nssSO_4^{2-}) concentration was also higher than the NO_3^- concentration in the autumn. The mean $\text{NO}_3^-/\text{nssSO}_4^{2-}$ ratio in 2008 was low, and the mean ratio of $\text{NO}_3^-/\text{nssSO}_4^{2-}$ was relatively high in 2009. The results of a 5-day backward trajectory analysis arriving at altitudes of 2,500 m during the sampling periods in the autumns of 2008 and 2009 are shown in figure 5. The air mass usually came from Asia in the autumn of 2008, whereas that at Murododaira was derived from not only Asia but also the western part of Japan in the autumn of 2009. According to the difference in the trajectories in both years, the fog water chemistry was mainly affected by the trans-boundary pollution from the Asian continent in the autumn of 2008 and was influenced by not only trans-boundary pollution but also air pollution from the industrial regions of Japan in 2009. The relatively high $\text{NO}_3^-/\text{nssSO}_4^{2-}$ ratio in 2009 might have been due to the influence of domestic pollution. The ratio of $(\text{NH}_4^+ + \text{nssCa}^{2+})/(\text{NO}_3^- + \text{nssSO}_4^{2-})$ was similar during both years (Fig. 4). The result was similar to those for the summers of 2004 and 2008.

The most predominant cation other than H^+ was NH_4^+ in the autumn of 2008 (Fig. 4). The result was similar to those in the summers of 2004 and 2008 (Fig. 2). However, the concentration of Na^+ , which is mainly derived from sea-salt particles, was higher than NH_4^+ in the autumn of 2009 (Fig. 4). Mt. Tateyama is located near the Japan Sea; however, high concentrations of sea-salt particles are not easily transported from the Japan Sea at high elevations

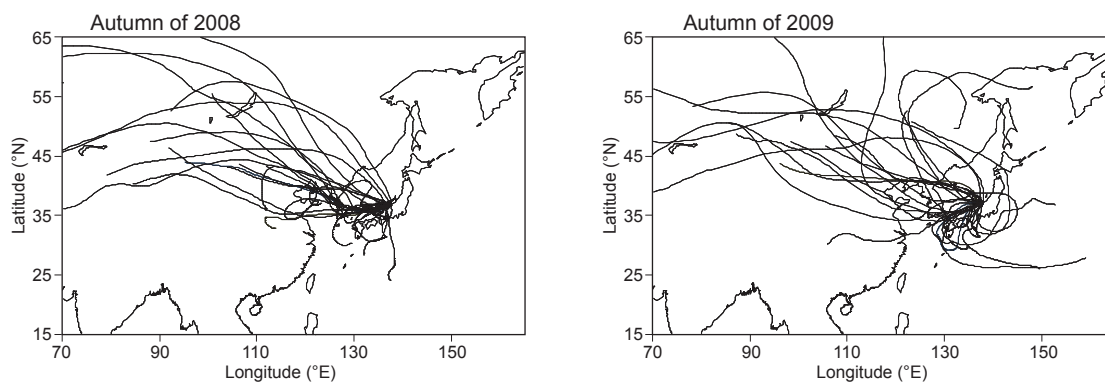


Fig. 5: Results of the 5-day backward trajectory analysis at an altitude of 2,500 m during the regular sampling periods in September and early October of 2008 and 2009. The trajectories were plotted once daily, and the starting time was 9:00 JST (0:00 UTC)

at Mt. Tateyama, such as Murododaira, during the summer and early autumn (WATANABE et al. 2005b). During the autumn of 2009, typhoons sometimes approached the south of Japan (Pacific Ocean). Sea-salt particles carried into the upper atmosphere by the typhoons might have been transported to Murododaira. High concentrations of sea-salt components were also observed during the intensive (every 2–4 hours of sampling) observation during 14–16 October 2005 at Murododaira. These concentrations were transported from Pacific Ocean, where there was a strong typhoon (WATANABE et al. 2010a). The vertical and long-range transport of sea-salt particles would influence the cloud microphysical properties in the middle troposphere. Relatively high sea-salt components have sometimes been detected at high mountainous sites in central Japan, such as at Mt. Fuji, in the autumn (WATANABE et al. 2006a).

3.4 Altitudinal characteristics of chemical composition

The volume-weighted mean concentrations of major ions in fog water at Bijodaira (altitude, 977 m) and Midagahara (altitude, 1,930 m) during the autumn of 2009 are shown in figure 6. The total ionic concentrations at Bijodaira were similar to or slightly higher than those at Midagahara. While the mean concentrations of total major ions at both sites were also higher than those at Murododaira, the acidity was higher at Murododaira (Figs. 4 and 6). The mean pH values were 4.2, 4.4 and 4.7 at Murododaira, Midagahara and Bijodaira, respectively. The mean nssSO_4^{2-} concentrations at all sites were similar, with the mean concentration of nssSO_4^{2-} at Murododaira slightly higher than that at Bijodaira and slightly low-

er than that at Midagahara (Fig. 6). The mean concentration of NH_4^+ at Murododaira was slightly lower than those at Midagahara and Bijodaira. Whereas the sample weights at Murododaira were about 3,000 g/month (described in 3.3), they were much lower at Bijodaira (about 1,000 g/month) and Midagahara (about 500 g/month), where the prevailing wind speeds are lower. Therefore, the deposition flux of acidic species, such as H^+ and nssSO_4^{2-} , by fog water seemed to be significantly higher at Murododaira than at the lower sites. Perhaps acidic deposition onto the vegetation surface by fogs is significantly large at the high-elevation sites.

The NO_3^- concentration was higher than the nssSO_4^{2-} concentration at Bijodaira; that is, the ratio of $\text{NO}_3^-/\text{nssSO}_4^{2-}$ was high. As mentioned above, the NO_3^- concentration is usually higher at a lower altitude than at a higher altitude in Japan. High NO_3^- has also been detected at mountainous sites affected by polluted air from industrial regions in Japan (e.g. MURANO 1993; IGAWA et al. 1998; WATANABE et al. 2001a). Unlike that at Murododaira, air pollution at Bijodaira is mainly transported from lower sites, for example, Toyama city areas, the foot of Mt. Tateyama (KUME et al. 2009). The $\text{NO}_3^-/\text{nssSO}_4^{2-}$ ratio at Midagahara was lower than that at Bijodaira but higher than that at Murododaira. The same tendency was seen in the autumn of 2003 (WATANABE et al. 2005b) and at other mountains in Japan (WATANABE et al. 2006a). The equivalent ratio of $\text{NO}_3^-/\text{nssSO}_4^{2-}$ seems to decrease with height.

The concentrations of sea-salt components, such as Na^+ , were high at Bijodaira. Unlike those at Murododaira, sea-salt particles are transported from the Japan Sea at Bijodaira, the lower site of Mt. Tateyama (WATANABE et al. 2010b). In the previous study at Bijodaira, high levels of Na^+ and Cl^- were

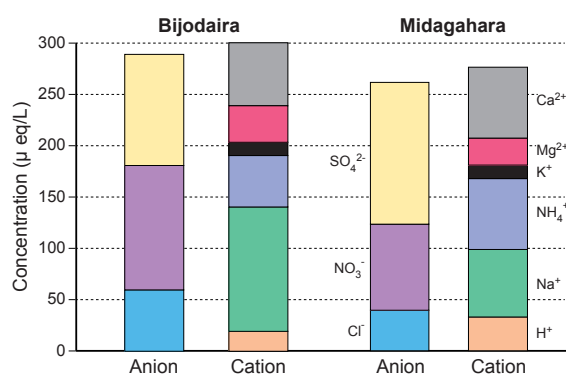


Fig. 6: Weighted mean concentrations of major ions in fog water at Bijodaira (altitude, 977 m) and Midagahara (altitude, 1,930 m) obtained by the regular sampling in September and early October of 2009

also detected in the autumn of 2003 (WATANABE et al. 2005b). The concentrations of Ca²⁺ (or nssCa²⁺), mainly derived from soil dust particles, were relatively high at Bijodaira and Midagahara (Fig. 6). The ratios of nssCa²⁺ to the total cation at both sites were higher than that at Murododaira. The ratio at Bijodaira was also higher in 2009 than in the autumn of 2003 (WATANABE et al. 2005b). Perhaps dust aerosols were relatively abundant at sites lower than 2,000 m during the autumn of 2009.

4 Conclusions

To investigate the atmospheric environment at mountainous sites near the coast of the Japan Sea, the fog water chemistry was measured at Murododaira (altitude, 2,450 m) on the western slope of Mt. Tateyama during the summers of 2004 and 2008 and the autumns of 2008 and 2009. Fog water was also sampled at Bijodaira (altitude, 977 m) and Midagahara (altitude, 1,930 m) in the autumn of 2009. Fog water collected using a passive string sampler was usually recovered every 3–14 days.

Strong acidic fogs (pH < 4) were observed at Murododaira during summer and autumn. The mean ionic concentrations in the summer of 2008 were higher than those in the autumn. According to the results of the backward trajectory analysis, the air mass at Murododaira was derived primarily from the polluted regions of Asia in 2008, and the transboundary air pollution might have contributed to fog water acidification at Murododaira. In the summer, central Japan is usually under the influence of a maritime air mass; however, the continental air mass seemed to be predominant in 2008. On the other hand, acidic fogs might have been produced by pol-

lutants from central and western Japan in 2004. The nssSO₄²⁻ concentration was usually higher than the NO₃⁻ concentration, but the ratio of NO₃⁻/nssSO₄²⁻ in fog water was relatively high in the summer of 2004. The concentrations of total ions in 2008 were much higher than those in 2004.

The nssSO₄²⁻ concentration was also higher than the NO₃⁻ concentration at Murododaira during the autumn. Especially, the mean NO₃⁻/nssSO₄²⁻ ratio was low in the autumn of 2008, when the air mass usually came from Asia. The ratio of NO₃⁻/nssSO₄²⁻ was relatively high in the autumn of 2009, when the air mass was derived from not only Asia but also from western Japan. The predominant air mass may significantly influence the chemical characteristics of fogs at Murododaira. The fog water chemistry was significantly affected by chemical substances derived from Asia as well as Japan. To evaluate the chemical deposition by fog water and its effects on ecosystems, further measurements of fog water chemistry will be required.

The acidity of fog water was higher at Murododaira than at Bijodaira and Midagahara. The weights of all samples of fog water at Murododaira were much greater than those at Bijodaira and Midagahara. The deposition flux of acidic species, such as H⁺ and nssSO₄²⁻, by fog water seemed to be significantly higher at Murododaira than at the lower sites. The NO₃⁻ concentration was higher than the nssSO₄²⁻ concentration at Bijodaira. The equivalent ratio of NO₃⁻/nssSO₄²⁻ seems to become lower with height.

Relatively high concentrations of sea-salt components, such as Na⁺, were observed at Murododaira during the autumn of 2009. The sea-salt particles might have been transported from the Pacific Ocean by strong typhoons. The vertical and long-range transport of sea-salt seems to affect the cloud microphysical properties in the middle troposphere.

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