Aerosol Deposition and Behavior on Leaves in Cool-temperate Deciduous Forests. Part 2: Characteristics of Fog Water Chemistry and Fog Deposition in Northern Japan

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ABSTRACT

The fog water chemistry and deposition in northern Japan were investigated by fog water and throughfall measurements in 2010. Fog water was sampled weekly by an active-string fog sampler at Lake Mashu from May to November. Throughfall measurements were conducted using rain gauges under three deciduous trees along the somma of the lake from August to October. The mean fog deposition rate (flux) was calculated using throughfall data to estimate the total fog water deposition amount for the entire sampling period. NH_4^+ and SO_4^{2-} were the most abundant cation and anion, respectively, in the fog water samples. A mean pH of 5.08 in the fog water, which is higher than those in rural areas in Japan, was observed. The $[NH_4^+]/[SO_4^{2-}]$ equivalent ratio in fog water was larger than 1.0 throughout the study period, indicating that NH_3 gas was the primary neutralizing agent for fog water acidity. The mean rate and total amount of fog water deposition were estimated as 0.15 mm h^{-1} and 164 mm, respectively. The amounts of nitrogen and sulfate deposition via fog water deposition were corresponded to those reported values of the annual deposition amounts via rainfall.

Key words: Aerosol, Fog chemistry, Fog deposition, Throughfall, Nitrogen deposition, Cool-temperate deciduous forest

1. INTRODUCTION

Lake Mashu, a caldera surrounded by a crater rim at a height of 500 m above sea level, is located in Akan National Park, Hokkaido, northern Japan. Because no surface streams flow into the lake, it has been studied as a representative baseline for the monitoring of inland water pollution on a global scale under the Global Environment Monitoring System (GEMS)/Water program (Atsutani and Minami, 2004; Fujinuma *et al.*, 2004). An injury in Japanese mountain birch (*Betula ermanii*) has been observed along the somma of the lake in recent years. Trees are generally subjected to many types of stresses, such as insect damage, microbial infection, nutrient deficiency, and air pollution. Although the key environmental factors that cause this injury are still unknown, acid deposition can be one of triggers of forest dieback (Eiden *et al.*, 1989).

Lake Mashu is frequently covered with fog or clouds during the growing season of trees. The ion concentrations of sulfuric and nitric acids are generally higher in fog water than in rainwater (Igawa *et al.*, 1998). Such acid fog deposits on vegetative surfaces and is harmful to plants (Schaberg *et al.*, 2000; Jacobson *et al.*, 1989; Schemenauer, 1986). In addition, fog water deposition significantly contributes to hydrological, nutrient and pollutant inputs to forest ecosystems (Ewing *et al.*, 2009; Fenn *et al.*, 2000; Dawson, 1998), and it may affect the woodland along the somma of Lake Mashu. Nevertheless, the fog water deposition on the woodland in the area, which can cause supply considerable amounts of pollutants and nutrients, has still not been investigated. Therefore, it is essential to quantify the deposition amount of ions such as sulfate and nitrate via fog water deposition in Lake Mashu.

In this study, we aimed to quantify the amount of fog water and ion deposition on the cool-temperate deciduous forests at Lake Mashu. The fog chemistry was measured during the growing season of trees. The physico-chemical characteristics of fog water sampled by

an active-string fog sampler were analyzed. Throughfall measurements under birch tree plots were applied to the area surrounding the somma of the lake. By combining the fog water sampling and throughfall data, the mean fog water deposition rate (flux) and total material inputs via fog water deposition were estimated. The results were used to evaluate the performance of the current parameterizations of the inferential method (Katata *et al*., 2013, the companion paper).

2. MATERIALS AND METHODS

2. 1 Site Description

Lake Mashu is a crater lake located in northeastern Japan (Fig. 1). The sampling site for fog water is the public observation deck on the somma of Lake Mashu (43°33′N, 144°30′E, 542 m above sea level). The weather station is located along the road approximately 300 m far from the fog water sampling site (Fig. 1b). The dominant plant species are *B. ermanii* Cham. (dark green areas in Fig. 1b, c) and *Sasa senanensis* (light green areas in Fig. 1b, c) (Fujinuma *et al*., 2004). The woodland areas spread widely in the eastern area of the throughfall sampling sites (Fig. 1c). Agriculture is the main land use in the areas within 50 km in radius (Fig. 1a), and there is no permanent air pollution source nearby. The nearest air pollutant source is local road traffic amounting to 500-600 vehicles a day. Thus, local air pollution sources are considered to contribute little to the ionic components of fog water. The period from early summer to autumn is usually the main foggy season at the site.

2. 2 Observations of Fog Water Chemistry and Deposition

Fog water was collected with an automated active-string fog sampler (FSK-01, Daisho Engineering Co. Ltd., Sapporo, Japan). The sampler operating duration was recorded by a data logger in the collection unit. The collecting air volume of the instrument was 1340 $m^3 h^{-1}$. The fog water collection unit was cleaned with deionized water on a weekly basis. The collection unit consists of a double screen with 0.8 mm diameter Teflon strands. A collection efficiency of 60%, which can be theoretically calculated assuming a fog droplet diameter of 10 μm (Langmuir and Blodgett, 1961), was used to calculate the liquid water content (LWC) of the fog water. The fog water collector is described in detail in Tago *et al*. (2001). The collection efficiency depends strongly on the fog droplet diameter; for example, when all the droplets are as small as 5 μm in diameter, the collection efficiency decreases to 39% (Tago *et al*., 2001). In this study, it is difficult to accurately
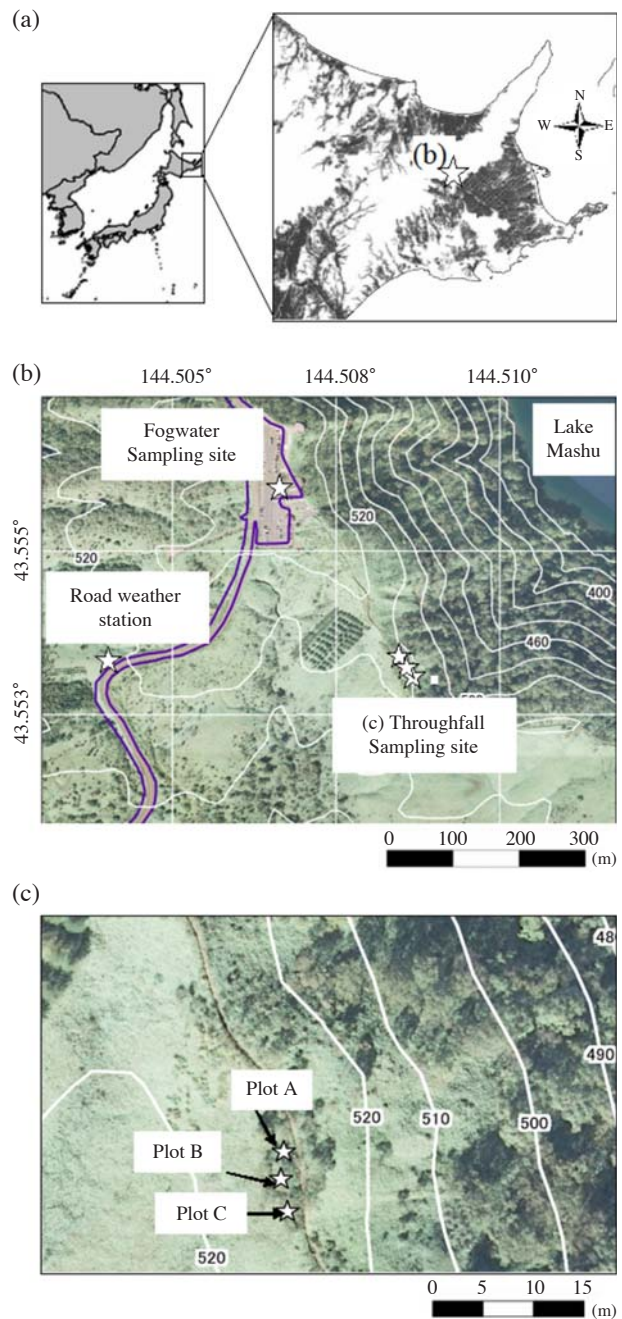

Fig. 1. (a) Land use map of eastern Hokkaido, Japan, and (b) locations of road weather station and sampling sites for fog water and (c) throughfall around Lake Mashu. Agricultural areas are shown as gray shaded areas in (a). Three plots for throughfall measurements, A, B, and C, are shown in (c).

determine the collection efficiency because several fog events with different droplet distributions contributed to the samples taken for one week. Therefore, the calculated LWC in this study contains the uncertainties derived from the fog droplet size distribution. However,

field experiments showed that the diameters of droplets making the greatest contribution to the LWC ranged from 9 to 15 μm (e.g., Klemm and Wrzesinsky, 2007). Considering that the collection efficiency of droplets larger than 10 μm in diameter does not increase very much (Tago *et al.*, 2001), it is reasonable to use the collection efficiency assuming droplets 10 μm in diameter to calculate the LWC. The fog water sampling period was 183 days, from May 11 to November 10, 2010. In winter, it is impossible to collect fog water because fog droplets freeze on the screen of the collection unit.

For each collected fog sample, the LWC of the fog water (mg m^{-3}) was calculated using the equation

$$LWC = F_v / (S_D R_C C_E), \qquad (1)$$

where F_v is the collected fog water weight (g), S_D is the sampler operating duration (h), R_C is the air volume collecting rate (m^3 h^{-1}), and C_E is the collection efficiency. The fog frequency was calculated by dividing the sampler operating duration by the total sampling period for each month.

The wind velocity and direction, and rainfall data were obtained from a road weather station near Lake Mashu (Fig. 1b). Rainfall was measured with a tipping-bucket rain gauge with a resolution of 1.0 mm (WB0015, Yokogawa Denshikiki Co., Ltd., Tokyo, Japan) equipped with a logger.

After the fog water samples were transported to the laboratory, the sample volumes, pH (HM-30R, DKK-TOA, Tokyo, Japan), and electrical conductivity (CM-30V, DKK-TOA, Tokyo, Japan) were measured. Before the inorganic ion compounds were measured, the samples were filtered by a membrane filter (0.2 μm pore size, DISMIC-13C, ADVANTEC-Toyo, Tokyo, Japan) and stored in a refrigerator. The major ions (Cl$^-$, NO$_3^-$, SO$_4^{2-}$, Na$^+$, NH$_4^+$, K$^+$, Mg^{2+}, and Ca^{2+}) were measured by ion chromatography (Dionex ICS-1500 and ICS-2000, Nippon Dionex Co., Ltd., Osaka, Japan). The volume-weighted mean values were computed for all inorganic ion concentrations in each fog water sample. The pH value was calculated from the mean H$^+$ concentration in fog water (Table 2).

To calculate the fog water deposition rate and total deposition amount, the throughfall was measured using tipping-bucket rain gauges (RS-102, OGASAWARA KEIKI Co., Ltd., Tokyo, Japan) under three tree plots labeled A, B, and C (Fig. 1c). The results of these rain gauges were recorded with a resolution of 0.5 mm by data loggers. Three rain gauges were set under the *B. ermanii* canopy approximately 300 m far from the fog water sampling site (Fig. 1b, c). The throughfall sampling period was August 10 to October 15, 2010. The mean canopy height of deciduous trees was approximately 10 m. The leaf area index (LAI) was measured as 1.5 ± 0.16 m^2 m^{-3} by the plant canopy analyzer (LAI-2000, Li-cor Inc., Nebraska, USA) on August 10, 2010.

Fog water deposition can generally be calculated from throughfall measurements on the basis of water balance equation at the canopy top as follows:

$$A_{FG}(i) = A_{TF}(i) + A_{SF}(i) + A_{CI}(i) - A_{RF}(i), \qquad (2)$$

where $A_{FG}(i)$, $A_{TF}(i)$, $A_{SF}(i)$, $A_{CI}(i)$, and $A_{RF}(i)$ are the amounts of fog water deposition, throughfall, stemflow, canopy interception loss, and rainfall (mm), respectively, for each throughfall event i. Here, a throughfall event was counted when zero values of the measured throughfall data continued for 3 h.

In this study, data for the canopy interception loss and stemflow are unfortunately unavailable. Hence the variables $A_{SF}(i)$ and $A_{CI}(i)$ were both assumed to be zero. On the basis of this simplification, Eq. (2) can be rewritten as

$$A_{FG}(i) \cong A_{TF}(i) - A_{RF}(i). \qquad (3)$$

In several throughfall events (Nos. 3, 7, 9, and 13-15 in Table 3), the throughfall amounts (A_{TF}) were smaller than the rainfall amounts (A_{RF}), and thus A_{FG} became negative. These negative values are due mainly to the assumption of zero canopy interception loss and stemflow in the simplified water balance equation of Eq. (3). The uncertainties of fog water deposition estimation using Eq. (3) are discussed in subsection 3.3.

The fog water deposition rate (flux) of each plot and throughfall event, $F(i)$ (mm h^{-1}), and its time-weighted mean value for all throughfall events, F_{mean} (mm h^{-1}), were calculated as follows:

$$F(i) = \overline{A_{FG}(i)} / T_{FD}(i), \qquad (4)$$

$$F_{mean} = \sum_i [F(i) T_{FD}(i)] / \sum_i T_{FD}(i), \qquad (5)$$

where $\overline{A_{FG}(i)}$ (mm h^{-1}) is the mean value of $A_{FG}(i)$ for plots A, B, and C, and $T_{FD}(i)$ (h) is the duration of each throughfall event. Because the throughfall data were available only from August 10 to October 15, the amount of fog water deposition for the entire fog water sampling period from May 11 to November 10, $A_{FGtotal}$ (mm), was estimated using the mean fog water deposition rate (F_{mean}) as

$$A_{FGtotal} = F_{mean} S_{Dtotal}, \qquad (6)$$

where S_{Dtotal} (h) is the sum of the fog water sampler operating duration for the entire fog water sampling period. The deposition of inorganic ions was calculated as the product of the fog water deposition and the ion concentration for each fog water sample.

3. RESULTS AND DISCUSSION

3. 1 Meteorological Conditions in Fog Water Sampling Period

Table 1 summarizes the rainfall and meteorological conditions during the fog water measurement period. Note that the period for each month was defined by the weekly fog water sampling times. The mean wind velocity and most frequent wind direction were the derived from those when the fog water sampler was activated. The mean wind velocity, which ranged from 3.7 to 5.7 m s^{-1}, decreased gradually from May to August and increased afterwards. During all months in 2010 except for May, southerly winds were dominant. The cumulative rainfall amount in the fog water sampling period from May 11 to November 10, 2010 was 719 mm.

3. 2 Fog Characteristics

Monthly summaries of the physical and chemical characteristics of the fog water are shown in Table 2. A large monthly variation appeared in the LWC; e.g., the lowest LWC was in September (39.8 mg m^{-3}), and

the highest one was in June (69.7 mg m^{-3}). The mean fog frequency was 24.2% in entire fog sampling period. The monthly fog water duration was highest in June (267.6 hours).

The mean concentrations of NH_4^+ (165.5 µeq L^{-1}) and nss-SO_4^{2-} (126.6 µeq L^{-1}) (Table 2) were approximately eight times higher than those in rainfall in northern Japan, which were reported as 20.6 µeq L^{-1} and 17.7 µeq L^{-1}, respectively (Tomoyose *et al.*, 2009). These were the most abundant cation and anion, accounting for more than half of the ion constituents of fog water (Fig. 2). Past studies also reported similar results in ion constituent (Aleksic *et al.*, 2009; Klemm and Wrzesinsky, 2007; Mohnen and Kadlecek, 1989) because sulfates act as cloud condensation nuclei (Boucher and Lohmann, 1995).

The mean pH value of fog water was 5.08 in the fog water sampling period (Table 2). In Japan, fog and/or cloud chemistry were measured in several mountainous sites in near-urban and rural areas. On Mts. Oyama and Rokko, which are located near urban areas, mean fog water pH values of 3.66 (Igawa *et al.*, 1998) and 3.80 (Aikawa *et al.*, 2001) were observed, respectively.

Table 1. Fog water sampling time, number of samples (N), accumulated rainfall (P_r), mean air temperature (T_{mean}) and wind velocity (U_{mean}), and most frequent wind direction at the study site during the fog water sampling period from May 11 to November 10, 2010. The statistics for the wind velocity and wind direction were calculated when the fog water sampler was activated.

| Month | Sampling time | | N | P_r (mm) | T_{mean} (°C) | U_{mean} (m s^{-1}) | Most frequent wind direction[1] |
	Start	End					
May	11-May 10:45	01-Jun 11:45	3	66	5.3	5.7	NNE
June	01-Jun 11:45	05-Jul 11:34	5	58	14.3	4.2	S
July	05-Jul 11:34	02-Aug 14:29	4	146	16.9	4.0	S
Aug.	02-Aug 14:29	30-Aug 11:00	4	238	20.2	3.7	S
Sep.	30-Aug 11:00	04-Oct 13:16	5	113	14.6	4.8	S
Oct.	04-Oct 13:16	10-Nov 10:11	5	98	7.3	5.7	SSW
Total/mean			26	719	13.2	4.6	S

[1]Most frequent wind direction was determined from the data observed by a wind vane with 16 sectors.

Table 2. Monthly accumulated sampler operating duration (S_D), mean fog frequency, liquid water content (LWC), pH, electrical conductivity (EC), and ion concentration and balance from May 11 to November 10, 2010.[2]

	S_D (hour)	Fog frequency (%)	LWC (mg m^{-3})	pH	EC (mS m^{-1})	H^+	NH_4^+	Na^+	K^+	Mg^{2+}	Ca^{2+} (µeq L^{-1})	Cl^-	NO_3^-	SO_4^{2-}	nss-SO_4^{2-}	Ion balance
May	151.4	30.0	61.0	5.69	3.6	2.0	176.7	44.1	49.8	10.9	17.6	49.7	40.3	139.9	134.6	1.31
June	267.6	32.8	69.7	5.10	2.7	8.0	146.7	6.4	6.0	2.7	6.1	13.6	36.7	106.0	105.3	1.12
July	202.8	30.0	54.1	5.50	2.6	3.1	145.2	9.9	6.0	2.4	3.9	14.7	36.6	99.6	98.4	1.13
Aug.	134.5	20.1	63.2	5.98	2.7	1.0	146.5	20.5	5.0	4.1	3.9	27.2	41.1	96.1	93.6	1.10
Sep.	131.4	15.6	39.8	5.11	10.3	7.8	217.8	410.2	17.1	84.9	30.8	462.5	87.1	204.2	154.8	1.02
Oct.	175.8	19.9	50.4	4.52	7.0	29.9	209.2	152.4	7.6	38.3	17.3	174.6	93.0	176.9	158.6	1.02
Mean		24.2	58.3	5.08	4.1	8.4	165.5	68.4	13.4	15.8	10.7	80.7	49.9	126.6	118.4	1.10
S.D.		21.4	1.57		4.8	15.6	96.5	228.2	26.8	48.7	18.0	252.6	62.7	87.5	73.7	

[2]The pH, EC, and each ion concentration were calculated as the volume-weighted average
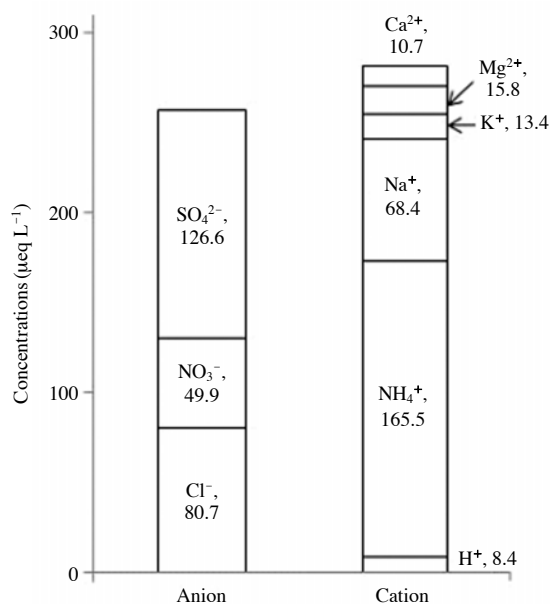
Fig. 2. Volume-weighted mean concentrations of major inorganic ions in fog water measured from May 11 to November 10, 2010.

In rural areas, higher mean pH values of 4.45 (Adzuhata *et al.*, 2001) and 4.00-4.50 (Watanabe *et al.*, 2011) were reported in northern and central Japan, respectively. The result of our previous study at Lake Mashu (mean pH=4.29-4.60; Yamaguchi *et al.*, 2010) was close to the above results for rural areas. The last three results showed that the mean pH values of fog water ranged from 4.00 to 4.60 in rural areas. The mean pH value of 5.08 from this study was even higher than the above values.

The difference in mean pH between this and past studies in rural areas can be explained by the neutralization of fog water by NH_3 gas and ammonium particles. One of prior studies, the equivalent ratio of $[NH_4^+]/[SO_4^{2-}]$ in fog water was overall less than 1.0 in summer and autumn (Watanabe *et al.*, 1999). In contrast, this study shows that a $[NH_4^+]/[SO_4^{2-}]$ ratio of more than 1.0 continued throughout the same season (Table 2). This result indicates that ammonium ions contributed greatly to the neutralization of fog water at Lake Mashu in 2010. Eastern Hokkaido has widespread agricultural areas that are a large source of NH_3, emitting 3 g m^{-2} y^{-1} (Kannari *et al.*, 2007). Fog was carried mainly by southerly winds, which were predominant in the observational period (subsection 3.1), and passed over these areas located south of the fog water sampling site (Fig. 1). Thus, it is considered that the fog water has taken up a large amount of NH_3 emitted from these areas and/or secondary ammonium particles. With

regard to the high mean pH value at Lake Mashu in 2010, very high air temperatures were recorded in Hokkaido in summer 2010 compared with those of 2006 to 2008. Higher temperatures may cause a larger NH_3 gas emission from agricultural areas, resulting in the higher mean pH of fog water in 2010.

The Na^+ and Cl^- concentrations of fog water in September 2010 were as high as 410.2 and 462.5 µeq L^{-1}, respectively, in the fog water sampling period (Table 2), which were close to the reported values of Na^+ and Cl^- concentrations for sea fog over the North Pacific Ocean in late summer (Yoshida *et al.*, 2007). This suggests that fog occurring in September 2010 was transported by southerly winds from the North Pacific Ocean. In contrast, the NH_4^+ proportion in this month, 28% of the total cations, was clearly higher than the 6% reported in the above study (Yoshida *et al.*, 2007). As discussed above, these larger NH_3 concentrations in this study also indicate that ammonia gas and ammonium particles were scavenged by fog when it was transported from the ocean and flowed over the agricultural areas.

The mean concentration of SO_4^{2-} in fog water (127 µeq L^{-1}) (Table 2) was close to the value of 108 µeq L^{-1} observed in a rural area in northern Japan (Adzuhata *et al.*, 2001) and was relatively low compared with those of 273 and 138 µeq L^{-1} measured at near-urban (Igawa *et al.*, 1998) and rural areas in central Japan (Watanabe *et al.*, 2010), respectively. More than 90% of the SO_4^{2-} in fog water was observed as nss-SO_4^{2-} throughout the observational period (Table 2). However, the study site has few local emission sources of SO_2. Prior studies of fog water chemistry in rural areas suggested that long-range transport of air pollutants from China increased the sulfate ions in fog water in rural areas of northern and central Japan (Watanabe *et al.*, 2011; Adzuhata *et al.*, 2001). Yamaguchi *et al.* (2010) also indicated that fog water acidification was triggered by air pollutants transported from Kanto area in Japan to Lake Mashu. A similar long-range transport of air pollutants is considered as a source of sulfate ions in fog water in this study. The NO_3^- concentration remained about 40 µeq L^{-1} from May to August and increased in September and October, synchronized with the concentrations of Na^+ and Cl^- (Table 2). This tendency also can be explained by air pollutants transported from Kanto area (Yamaguchi *et al.*, 2010).

3. 3 Estimation of Fog Water Deposition Rate

From August 10 to October 15, 21 throughfall events were observed. The throughfall amount A_{TF}, fog deposition rate of each event $F(i)$, and mean fog deposition rate for the entire sampling period F_{mean}, are shown in Table 3.

The fog water deposition rate $F(i)$ exhibited temporal changes. $F(i)$ ranged from 0.1 to 0.6 mm h^{-1} until the end of September, while it decreased to 0-0.1 mm h^{-1} in October despite almost same fog frequency as other two months (Table 2). Because the dominant species at Lake Mashu are deciduous trees, most of the leaves dropped from the canopy to the floor below at the beginning of October. The results supported by the fact that the change in the canopy structure caused the variability in the deposition rate (Keim *et al.*, 2005; Nadkarni and Sumera, 2004). The temporal change in the fog water deposition amount is discussed in the companion paper (Katata *et al.*, 2013).

The calculated mean fog deposition rate for the entire sampling period (F_{mean}) was 0.15 mm h^{-1} (Table 3). The reported fog deposition rates are 0.17 mm h^{-1} (Chang and Lai, 2002), estimated by weighing epiphytic bryophytes; 0.11-0.24 (Holwerda *et al.*, 2006) and 0.074-0.68 mm h^{-1} (Igawa *et al.*, 1998) by throughfall measurements; and 0.24-0.69 mm h^{-1} (Walmsley *et al.*, 1996) by a combination of fog water measurement using passive fog collectors and model calculation. The estimated fog deposition rate in this study was in the range of the above values.

There are several uncertainties in the throughfall measurements, as follows. First, the number of rain gauges for the throughfall observations was limited to three in this study. As a result, it is hard to evaluate the spatial and temporal variability in the throughfall measurement (ICP Forests, 2010; Konishi *et al.*, 2006; Keim *et al.*, 2005). Second, the contribution of the canopy interception loss and stemflow was neglected in this result. Past studies of broadleaf forests reviewed by Carlyle-Moses (2004) were summarized as the canopy interception loss ranged from 9.7% to 19.5% of the throughfall at several sites worldwide. Stemflow values of 3.7% (Price and Carlyle-Moses, 2003), 8.5% (Carlyle-Moses, 2004), 6% (Holwerda *et al.*, 2006), and 1.1% (Marin *et al.*, 2000) were measured in Canada, Mexico, Puerto Rico, and Colombia, respectively. If the sum of the canopy interception loss and stemflow is assumed to be 10% of the throughfall in estimating the fog water deposition using Eq. (2), the mean fog deposition rate, F_{mean} in Eq. (5), is computed as 0.23 mm h^{-1}, which is 153% of that calculated without the canopy interception loss and stemflow [i.e., Eq. (3)]. However, these previous studies also showed wide variations in the observed interception loss and stemflow. Further investigation at the study site is clearly necessary for an accurate estimation of the fog deposi-

Table 3. Event number of throughfall (i), throughfall duration [$TF_D(i)$], amounts of rainfall (A_{RF}), throughfall (A_{TF}), and fog water deposition (A_{FG}), rate of each throughfall event [$F(i)$], and time-weighted mean fog water deposition rate for the entire throughfall measurement period (F_{mean}).[3]

i	Start time	End time	$TF_D(i)$ (hour)	A_{RF} (mm)	A_{TF} (mm)			A_{FG} (mm)			$F(i)$ (mm h^{-1})	
					Plot A	Plot B	Plot C	Plot A	Plot B	Plot C	Mean	S.D.
1	10-Aug 18h	13-Aug 00h	54	114	124	116.5	145	0.19	0.05	0.57	0.27 ± 0.27
2	15-Aug 16h	16-Aug 07h	15	3	6	6	7	0.20	0.20	0.27	0.22 ± 0.04
3	23-Aug 19h	24-Aug 10h	15	27	34	25.5	47	0.47	0*	1.33	0.60 ± 0.72
4	30-Aug 18h	31-Aug 01h	7	0	0.5	1	0.5	0.07	0.14	0.07	0.10 ± 0.04
5	03-Sep 13h	03-Sep 21h	8	15	18	15.5	20	0.38	0.06	0.63	0.35 ± 0.28
6	04-Sep 01h	04-Sep 06h	5	1	1.5	1.5	1	0.10	0.10	0.00	0.07 ± 0.06
7	07-Sep 08h	07-Sep 13h	5	3	3.5	2.5	4	0.10	0*	0.20	0.10 ± 0.15
8	14-Sep 13h	14-Sep 15h	2	0	1	1	1	0.50	0.50	0.50	0.50 ± 0.00
9	17-Sep 02h	17-Sep 03h	1	5	4	3.5	6.5	0*	0*	1.50	0.50 ± 1.61
10	17-Sep 23h	18-Sep 10h	11	1	2.5	3.5	4.5	0.14	0.23	0.32	0.23 ± 0.09
11	20-Sep 17h	21-Sep 07h	14	1	2	2.5	2	0.07	0.11	0.07	0.08 ± 0.02
12	25-Sep 18h	26-Sep 09h	15	11	14.5	12	11.5	0.23	0.07	0.03	0.11 ± 0.11
13	27-Sep 17h	28-Sep 15h	22	7	6.5	5	6	0*	0*	0*	0.00 ± 0.03
14	28-Sep 20h	29-Sep 00h	4	7	6	4.5	5.5	0*	0*	0*	0.00 ± 0.19
15	03-Oct 20h	05-Oct 03h	31	29	32	27	28.5	0.10	0*	0*	0.03 ± 0.08
16	07-Oct 17h	08-Oct 09h	16	0	1	3	0	0.06	0.19	0.00	0.08 ± 0.10
17	08-Oct 18h	09-Oct 08h	14	0	1	1	1	0.07	0.07	0.07	0.07 ± 0.00
18	09-Oct 18h	10-Oct 10h	16	5	5	5.5	5.5	0.00	0.03	0.03	0.02 ± 0.02
19	11-Oct 00h	11-Oct 03h	3	0	0.5	0	0.5	0.17	0.00	0.17	0.11 ± 0.10
20	12-Oct 15h	13-Oct 08h	17	0	0.5	1	0.5	0.03	0.06	0.03	0.04 ± 0.02
21	14-Oct 00h	14-Oct 13h	13	5	6.5	6	7.5	0.12	0.08	0.19	0.13 ± 0.06
	Sum		288	234	271	244	305		F_{mean}		0.154	

[3] *The values of A_{FG} were set to zero when they became negative (see subsection 2.2).

Table 4. Estimated total deposition amounts of fog water ($A_{FGtotal}$) and inorganic ions from May 11 to November 10, 2010.

	$A_{FGtotal}$ (mm)	Ion deposition amounts (meq m^{-2})									
		H^+	NH_4^+	Na^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	nss-SO_4^{2-}
May	23.3	0.04	4.08	1.24	1.09	0.28	0.39	1.38	0.90	3.21	3.06
June	41.2	0.25	5.83	0.23	0.26	0.09	0.20	0.53	1.41	4.01	3.98
July	31.2	0.08	4.39	0.27	0.19	0.06	0.10	0.44	1.04	2.90	2.87
Aug.	20.7	0.02	3.43	0.84	0.13	0.17	0.15	1.00	0.98	2.35	2.24
Sep.	20.2	0.16	4.52	7.89	0.34	1.64	0.62	8.90	1.79	4.18	3.23
Oct.	27.1	0.82	6.25	6.59	0.32	1.59	0.70	7.42	3.15	5.27	4.47
Sum	163.7	1.4	28.5	17.1	2.3	3.8	2.2	19.7	9.3	21.9	19.9

tion rate.

3. 4 Estimation of Fog Water and Inorganic Ion Deposition

Table 4 summarizes the cumulative amounts of fog water and inorganic ion deposition. The total fog water deposition amount, 164 mm, corresponded to 23% of the rainfall amount (719 mm) measured in the same period. Kobayashi *et al*. (2002) reported that the annual fog water deposition amount ranged from 38% to 103% of the rainfall amount in coniferous forest in Japan, whereas Holwerda *et al*. (2006) showed a relatively small value of 18% in a broad-leaved forest in Puerto Rico. This result in a broad-leaved forest was close to the ratio of fog water deposition of 23% estimated in this study. This variety in the ratios of fog deposition to rainfall in the above studies is considered to originate from the differences in meteorological conditions (e.g., fog frequency and wind speed) and vegetative conditions (e.g., LAI and canopy height).

The total deposition amount of SO_4^{2-} from May 11 to November 10, 2010 due to fog water deposition was estimated as 21.9 meq m^{-2} (Table 4), and 90% of the SO_4^{2-} was nss-SO_4^{2-} (19.9 meq m^{-2}) (Table 4). This value corresponds to the annual mean wet deposition of nss-SO_4^{2-} via rainfall, 21.0 meq m^{-2}, in the rural area of western Hokkaido (Noguchi *et al*., 2007). The total deposition amount of inorganic nitrogen compounds (i.e., $NH_4^+ + NO_3^-$) in the fog water observation period was 37.8 meq m^{-2} (Table 4), which is also similar to the annual wet deposition amount of 28.8 meq m^{-2} y^{-1} in the same area of western Hokkaido (Hayashi *et al*., 2006). The above comparisons indicate that large amounts of sulfate and nitrogen deposit on the deciduous forests at Lake Mashu by fog water deposition as well as wet deposition due to precipitation. Prior studies have found that the amount of inorganic ions deposited by fog water is a large proportion of the total (Klemm and Wrzesinsky, 2007; Igawa *et al*., 2002; Fenn *et al*., 2000; Kobayashi *et al*., 1999).

Nitrogen deposition via fog water should be investigated by long-term observations (e.g., annual or inter-annual) to evaluate the nitrogen load at Lake Mashu. Comprehensive field experiments on the meteorological and hydrological conditions at Lake Mashu are expected to improve our understanding of the effect of fog water deposition on the nitrogen load.

4. CONCLUSIONS

The amounts of fog water and ion deposition on deciduous forests in northern Japan were estimated by fog water and throughfall measurements. A mean fog water pH of 5.08 was obtained in the growing season of trees in 2010; this was higher than those in prior studies in rural areas in Japan. The $[NH_4^+]/[SO_4^{2-}]$ ratio remained above 1.0 throughout the study period, indicating that ammonium ions contributed greatly to fog water neutralization at Lake Mashu in 2010. The result suggested that ammonia gas and ammonium particles were scavenged by fog when it passed over agricultural areas.

The fog water deposition amount calculated from throughfall data from May 11 to November 10, 2010, was 164 mm, which was small compared with the cumulative rainfall of 719 mm. Evaluation of the canopy interception loss and stemflow, which are uncertainties in the throughfall measurements in this study, is necessary for accurate estimations of the fog deposition rate and amount. The amounts of sulfate and nitrogen depositions via fog water deposition were thought to be on the same order as annual amounts via rainfall reported in western Hokkaido in the literature.

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