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Authors: Uehara, Yoshitoshi, Kume, Atsushi, Chiwa, Masaaki, Honoki, Hideharu, Zhang, Jing, et al.

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Atmospheric deposition and interactions with *Pinus pumila* Regal canopy on Mount Tateyama in the Northern Japanese Alps

Yoshitoshi Uehara[1]
Atsushi Kume[1,5]
Masaaki Chiwa[1]
Hideharu Honoki[2]
Jing Zhang[3] *and*
Koichi Watanabe[4]

[1]Kyushu University Forest, Faculty of Agriculture, Kyushu University, 394 Tsubakuro, Sasaguri, Fukuoka 811-2415, Japan
[2]Toyama Science Museum, Nishinakano 1-8-31, Toyama 939-8084, Japan
[3]Faculty of Science, University of Toyama, Gofuku 3190, Toyama 930-8555, Japan
[4]Faculty of Engineering, Toyama Prefectural University, 5180 Kurokawa, Imizu, Toyama 939-0398, Japan
[5]Corresponding author: glacier_foreland@yahoo.co.jp

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Abstract

To estimate the impact of atmospheric deposition on alpine ecosystems, we evaluated the interactions between atmospheric deposition and the *Pinus pumila* canopy close to the summit of Mount Tateyama, central Japan. We analyzed the chemical characteristics of rain, fog, and throughfall under the canopy in the summertime for 5 years. The concentrations of inorganic nitrogen and sulfate in precipitation at the summit (2839 m a.s.l.) were lower than at the base (13 m a.s.l.), but the total amounts deposited tended to be larger, because the precipitation was greater at the summit and highly concentrated fog water was deposited on the pine canopy. Large fluctuations in the quality and quantity of deposition were due to interannual variations in the meteorological conditions. About 80% of NO_3^--N and NH_4^+-N (0.40 and 0.47 kg ha^{-1} month^{-1}) from rain and fog were retained in the *P. pumila* canopy, and contributions from fog were twice those from rain. Considerable amounts of K^+, Mg^{2+}, and Ca^{2+} ions (0.97, 0.13, and 0.13 kg ha^{-1} month^{-1}) were leached out from the canopy to the soil as throughfall. The leaching did not increase with rainfall, but increased as the duration of the fog and dew increased.

Introduction

Microorganism activity and the rate of soil biomass decomposition are restricted at high elevations because of low soil temperatures and a long period of snow cover (Holzmann and Haselwandter, 1988; Walker et al., 1993), both of which limit the mineralization of organic matter. Atmospheric deposition is one of the major pathways of nutrient delivery to the alpine environment (Mladenov et al., 2012). The main source of nutrients near the ridge line in high mountain regions is atmospheric deposition, that is, wet and dry deposition, although litter accumulation may have an important role as a pool of mineral nutrients. To understand the atmosphere-plant-soil continuum in alpine nutrient cycling, it is important to determine the quantity and quality of atmospheric deposition (Bowman, 2000; Burns, 2004). Studies have shown that the total amount of atmospheric deposition on mountains is equal to that in the lower areas (Annen et al., 1995; Lovett and Kinsman, 1990).

Most of the rainfall and fog precipitation on the canopy will come into contact with the needle surfaces. Some of the rain and fog precipitation falling on the tree canopy adheres to the needles and branches, and returns into the atmosphere by evaporation. While passing through the canopy, some elements are added to the throughfall. Chiwa et al. (2003) showed that there was a linear relationship between the amount of dry deposition per unit surface area of pine needles and nonprecipitation hours for all ions except for K^+ and H^+. The accumulated dry deposition is washed out by rain and fog, and several water-soluble plant nutrients are leached out from the needle surfaces. These are added to the throughfall (and stemflow), whereas certain fractions of nitrogen (N) may be absorbed by foliage (Parker, 1983; Lovett and Lindberg, 1984; Chiwa et al., 2004; Adriaenssens et al., 2011; Fenn et al., 2013). To quantify the amount of canopy exchange and atmospheric deposition, information is needed on the accumulation of atmospheric deposition, dry deposition wash-off, and leaching from plant tissues by precipitation.

Pinus pumila Regal dominates the vegetation growing in the Japanese alpine zone above the tree line (Yanagimachi and Ohmori, 1991). This dwarf evergreen coniferous shrub forms a dense canopy with needle leaves; the amount of needle biomass in the closed canopy is approximately 15–25 t ha^{-1} (Kajimoto, 1994), and the leaf area index (LAI) is >5 m^2 m^{-2}. *P. pumila* forms dense canopy patches on the mountain slopes and its presence has a significant influence on the alpine plant community (Okitsu and Ito, 1983; Wada, 2007). Uehara and Kume (2012) showed that the dense *P. pumila* canopy intercepted a large proportion of the bulk precipitation and also efficiently captured fog precipitation, and supplied it to the floor as throughfall water, even in the absence of rainfall.

In addition to the unique material cycling processes of the alpine ecosystem, the Northern Japanese Alps, because of their remote position far from anthropogenic atmospheric sources and their geographical situation, provide a suitable environment for detecting the influence of long-range transboundary air pollution. Increases in anthropogenic nitrogen deposition have been linked to several terrestrial ecological changes, including net primary production and the nitrogen cycle (Bowman et al., 2006). Japan is a recipient of long-range N transport from the Asian continent, where NO_x emissions have increased 2.8 times from 1980–2003 (Ohara et al., 2007). Although the risk of harmful effects on the alpine ecosystem of long-range transboundary air pollutants from the Asian

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continent has been reported (Kume et al., 2009, 2011), most of the previous studies have been conducted below the tree line (e.g., Muramoto et al., 2007), and there have been no systematic observations above the line, especially for *P. pumila* vegetation.

The first objective of this study was to examine the chemical characteristics of incident rain and fog precipitation on the summit of Mount Tateyama. In the high mountain regions, most of the precipitated water is supplied to the lower altitudinal areas by regional air mass transport (Wehren et al., 2010) and is strongly influenced by the conditions of lower altitudinal areas. Therefore, we considered the altitudinal profiles of water and ionic depositions of the whole mountain in our evaluation of the hydrological cycle on the mountain summits. The second objective was to clarify how the canopy of *P. pumila*, which is patchily distributed on the slopes compared to other vegetation cover, leads to increases in the net throughfall fluxes. The deposition rates of aerosol particles and cloud droplets on the forest canopy differ from those on shorter vegetation (Fowler et al., 1989), so the *P. pumila* canopy may make a large contribution to annual inputs at the mountain summits. The final objective was to clarify the nature of the interactions between atmospheric deposition and *P. pumila* vegetation. We hypothesized that the process of canopy exchange has an important role in the nutrient mass balance of *P. pumila* vegetation during the snow-free period. The chemical quantity and quality of the throughfall and bulk precipitation (including fog precipitation) were evaluated in the snow-free period on the top of mountains. Results from this study will provide information on the effects of increases in long-range transboundary air pollution on the alpine ecosystem.

Materials and Methods

STUDY SITES

This study was conducted at Jodo-daira (36.566°N, 137.606°E, 2839 m a.s.l.; Fig. 1), on the summit of Mount Tateyama. Mount Tateyama is one of the peaks in the Northern Japan Alps in the Chubu-Sangaku National Park in Honshu, which contains a high mountain range that includes several peaks of approximately 3000 m in height. The mean annual air temperature at the summit is –2.6 °C (2004–2013) and the soil is frozen and covered with snow for more than 8 months of the year. The air mass to the study area usually has come from the Pacific Ocean through central and western Japan in summer, and in some cases has come from Asia continent. Moreover, the air mass has also come from Asia continent and western part of Japan in September to October (Watanabe et al., 2011). This study site was also used in a previous study by Uehara and Kume (2012). The Toyama University research facility has a monitoring tower (about 6 m in height) at the top of Jodo-daira to monitor the environment. Jodo-daira comprises three main slopes, two of which are covered with a *P. pumila* canopy that is approximately 0.6 m in height.

METEOROLOGICAL OBSERVATION

Air temperature and relative humidity sensors (Pt1000 and capacitance type sensor, CVS-50; Climatec Inc., Tokyo, Japan), a 0.2-mm tipping bucket rain gauge (Rain Collector II; Davis Instruments Co., USA), and two capacitive grid sensors (S-LWA-M003, Onset Computer Co., Massachusetts, USA) were set up at the top of the university monitoring tower. The drenched period was measured by the capacitive grid sensors, and was defined as the time when the wetness value was >90% and during which no rain fell.

RAIN AND FOG WATER COLLECTION

Rainwater was collected at the top of the monitoring tower from 2006 to 2011, and fog water was collected at the base of the tower during the snow-free period (August to September) from 2007 to 2011. Samples were not collected during 2010 because of repair work to the research facility. Rainwater was collected using a bulk collector that had a 15-cm-diameter polyethylene funnel draining to a 10 L polyethylene bottle. Fog water was collected using a passive thin string fog sampler (Model FWP-500; Usui Kogyo Kenkyusho Inc., Tokyo, Japan) with a polyethylene tank volume of 10 L. The fog sampler was set up at a height of 1.5 m, and a large hood was mounted on the sampler to keep rain out. The water samples were collected every other week. Details of these methods have been described in Uehara and Kume (2012).

THROUGHFALL COLLECTION

With the exception of 2009, throughfall from the *P. pumila* canopy was collected by gutter-type bulk precipitation collectors from 2006 to 2011. Each of these collectors was 10 × 40 cm. The surface of the collectors was specially coated with fluoropolymer film (ASF-110; Chukoh Chemical Industries Ltd., Tokyo, Japan). The reservoirs were made of thick polyethylene film and were completely covered with aluminum foil and black plastic sheets. Six rain collectors were placed as low as possible on the vegetation floor (~30 cm above the ground) from late July to early October. The water samples were collected every other week. Details of these methods have been described in Uehara and Kume (2012). The water samples from 2006 to 2008 are the same as those reported by Uehara and Kume (2012).

CHEMICAL ANALYSIS

The total amount of collected water was quantified in a measuring cylinder, and the pH and electrical conductivity were measured by a portable pH/conductivity meter (D-54; Horiba Ltd., Kyoto, Japan) in the field. A portion of the collected water was brought back to the laboratory. The water samples were analyzed using an ion chromatograph after filtration with a 0.2 μm disc filter. Major cations (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) were analyzed using an ion chromatography system (Tosoh IC-8010, separation column TSK IC-Cation I/II HR, eluent 1.6 mM HNO_3), and major anions (Cl^-, NO_3^-, and SO_4^{2-}) were analyzed by another chromatography system (Metrohm 761 Compact IC, separation column Shodex SI-90 4E, eluent 1.8 mM Na_2CO_3, 1.7 mM $NaHCO_3$).

DATA ANALYSIS

The amount of precipitation was calculated by dividing the collected water volume (L) by the funnel area (m^2), while the amount of throughfall was calculated by dividing the collected water volume (L) by the gutter surface area (m^2). The detailed calculation method for fog water deposition is described in Uehara and Kume (2012). The amount of ion deposition in rainwater, throughfall, and fog water was calculated by multiplying the amount of incident precipitation (mm) by the concentration of each ion species (kg ha^{-1} or eq ha^{-1} for H^+).

To evaluate the contribution from the various ion species in the precipitation, the concentration of each of the ion components derived from sea salt should be subtracted. The concentration of nss-SO_4^{2-} was calculated from the molar ratio of SO_4^{2-}/

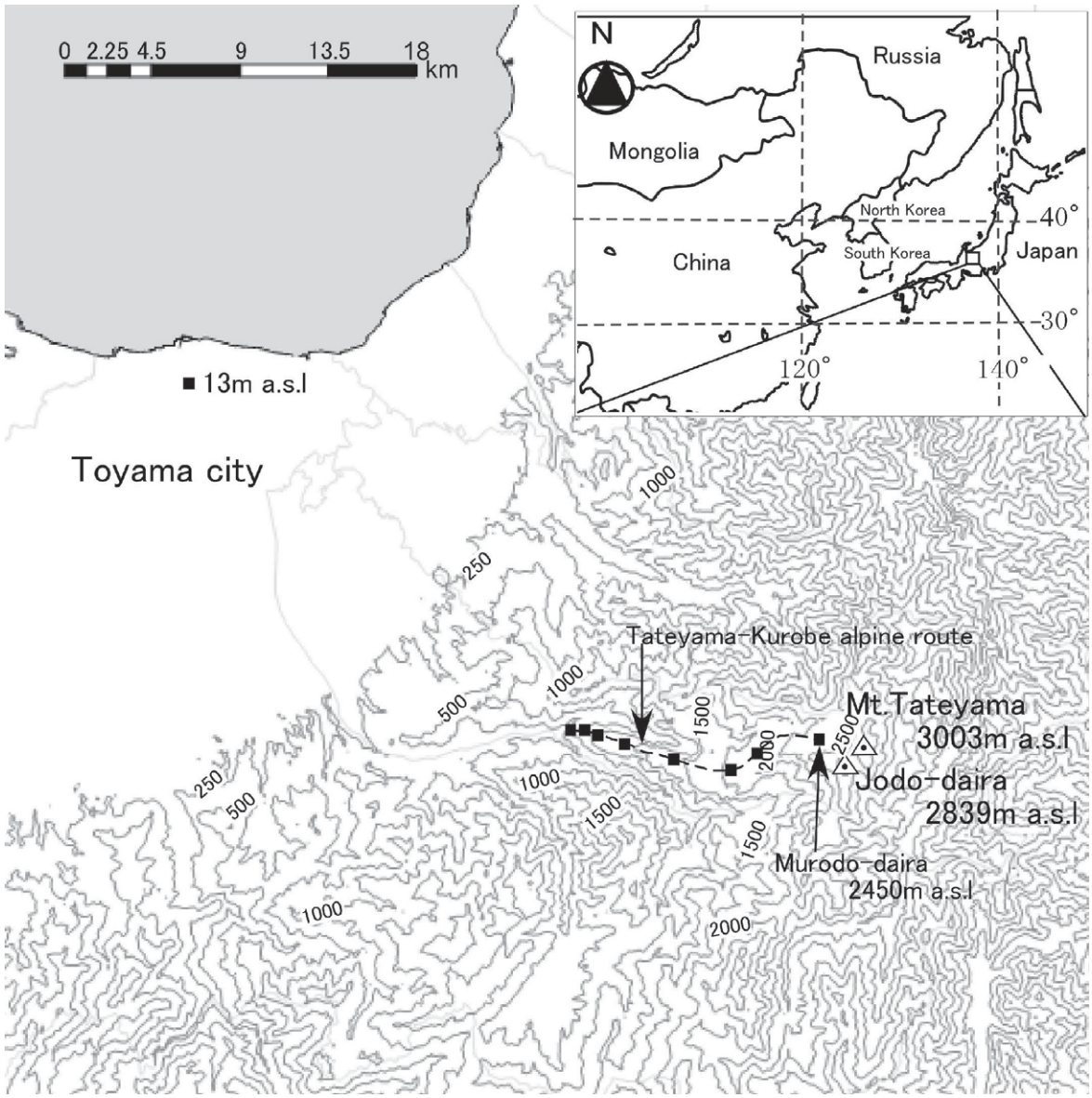

FIGURE 1. Location of the study site at Jodo-daira in Japan. Precipitation was sampled along the Tateyama-Kurobe alpine route (■).

Na^+ (0.121, μeq L^{-1} base) in sea water (Keene et al., 1986). The prefix nss- indicates that the concentration of the ionic species originated from non-sea salt (mainly anthropogenic).

The net throughfall ion deposition (NTD) was calculated with the following formula:

$$NTD = TD - BD \quad (1)$$

where TD is the throughfall ion deposition, and BD is the bulk ion deposition. The amount of deposition of each ion species is calculated by multiplying the concentration of each ion species by the volume of each precipitation event. In this study, the stemflow of *P. pumila* was not measured, because this dwarf shrub has a creeping growth form and does not have erect stems. The main stem generally extends in a downslope or leeward direction (Kajimoto, 1992; Okitsu and Ito, 1984), so stem flow to the base of the tree does not occur. BD consisted of rain and fog deposition and is described as follows:

$$BD = RD + FD \quad (2)$$

where RD is the ion deposition in rain, and FD is the ion deposition in fog. Based on previous studies (3 years' observation) by Uehara and Kume (2012), it was assumed that the amount of deposited fog water was 30% of the amount of rainwater, and therefore FD can be defined as:

$$FD = 0.3 \times n \times [X]_{fog}, \quad (3)$$

where n is the amount of rain precipitation and $[X]_{fog}$ is the ion concentration of fog water.

In this study, as rainfall was sampled by bulk collectors, the sample contained dry deposition. However, the dry period at Jodo-daira (Uehara and Kume, 2012) is short, so the amount of dry deposition should be negligible.

Results

PH AND ION CONCENTRATIONS OF INCIDENT WET DEPOSITION

Table 1 shows the weighted mean of the pH and major ion species contained in rain, fog, and throughfall at Jodo-daira in August and September from 2006 to 2011. The mean pH values for rainwater, fog water, and throughfall were 4.8 (3.8–5.9), 4.6 (3.8–6.1), and 4.8 (3.9–6.5), respectively. The pH of fog water tended to be lower than that of rainwater, but the coefficient of variation (CV) of fog water was similar to the CV values of rainwater and throughfall. The weighted mean acidity input (Ai = [nss-SO_4^{2-}] + [NO_3^-]) of fog water was 67.9 μeq L^{-1}, while that of the precipitation was 30.1 μeq L^{-1}. The acidity of fog water was about twice that of precipitation, but the cation concentrations were also high.

The volume-weighted mean concentration of major ion species fluctuated significantly each year during the observation period (Table 1). The amount of rain, fog, and throughfall also showed large interannual variation; their CVs were 0.23, 0.31, and 0.42, respectively.

The major cations in rainwater were H^+ and NH_4^+ and the anions were SO_4^{2-} and NO_3^-. The mean concentrations of each ion species in fog water were about twice as high as in rainwater. Concentrations of the sea salt components, Na^+ and Cl^-, were low; the volume-weighted mean concentrations of Na^+ and Cl in rainwater were 5.5 μeq L^{-1} and 6.5 μeq L^{-1}, respectively.

The percentage and concentration of ion species in throughfall also fluctuated each year (Table 1). The concentrations of Cl^-, Na^+, and SO_4^{2-} in throughfall were similar to those in rainwater, but the weighted mean concentrations of Ca^{2+}, Mg^{2+}, and K^+ in throughfall were 2.0, 7.8, and 9.2 times higher than those in rainwater, respectively. The interannual variations of Ca^{2+} and K^+ in rainwater and throughfall were relatively large and their CVs were in the range of 0.8–1.5 (Table 1).

Conversely, the concentrations of NH_4^+ and NO_3^- in throughfall were 42% and 30% lower than those in rainwater, respectively, and were also lower than in fog water. The concentration of NO_3^- in throughfall was frequently <0.15 μeq L^{-1} (61 samples/167 samples).

The cation-anion balances of incident precipitation were nearly equal, but there was an anion deficit in throughfall (Table 1).

BULK, THROUGHFALL, AND NET THROUGHFALL DEPOSITION

Table 2 shows the monthly mean wet deposition of each ion species on the *P. pumila* vegetation during the observation period (August to September) from 2006 to 2011. In 2007, 2008, and 2011, some of the throughfall collection tanks overflowed during typhoons, so the data for these periods were eliminated from the calculation of NTDs. The trends in mean rain deposition and throughfall deposition were different depending on the ion species. The 5-year mean throughfall depositions of H^+, Na^+, NH_4^+-N, Cl^-, NO_3^--N, and SO_4^{2-}-S were smaller than those of the rain depositions. The amounts of NH_4^+-N and NO_3^--N in throughfall were 35.4% and 25.6% less than those in rainfall. Conversely, deposition of K^+, Mg^{2+}, and Ca^{2+} from throughfall was greater than that of the rain deposition. There was about seven times more K^+ and Mg^{2+} in throughfall than in rainwater.

WET DEPOSITION ON MOUNT TATEYAMA

The spatial and temporal distribution of wet and dry deposition on Mount Tateyama is important when considering the hydrological cycle of the summit, because most of the deposition is supplied from the lower altitudinal areas. As shown in Figure 2, the amount of precipitation and its ionic concentration changed with year and altitude. The amount of precipitation increased with altitude, and was about twice as much at the summit as at the lowest altitudinal site (Toyama city, 13 m a.s.l.: Fig. 2, part a). The effects of sea salt decreased with altitude; with the exception of 2007, the concentrations of Na^+ and Cl^- at Jodo-daira were 15% of those at the lowest altitude site (Fig. 2, parts e and f), and the amounts of deposition were 33% and 31% of those at the lowest site (Fig. 2, parts j and k). In 2007, typhoon No. 9 (Fitow) struck Toyama and sea salt particles were blown up over the mountain. The concentrations of nss-SO_4^{2-}, NO_3^-, and NH_4^+ also decreased with altitude (Fig. 2, parts b, c, and d). The 5-year mean concentrations of nss-SO_4^{2-}, NO_3^-, and NH_4^+ in precipitation at Jodo-daira were approximately 76%, 35%, and 45%, respectively, of those at the lowest site, but the total deposition at Jodo-daira was comparable to that at the lower sites; in particular the SO_4^{2-} deposition tended to increase with altitude.

NET THROUGHFALL DEPOSITION

The NTDs of the canopy were calculated using Equation 1 based on the wet deposition of each ion species in rainwater, fog water, and throughfall (Table 2). The mean NTD varied from year to year, and showed different trends depending on the ion species. The fluctuation may have been due to interannual differences in the BD, but the CV of the NTD of each ion species was smaller than that of the amount of precipitation. The NTDs of H^+, NH_4^+, NO_3^-, SO_4^{2-}, Na^+, and Cl^- were negative, while those of K^+, Mg^{2+}, and Ca^{2+} were positive (Table 2, Fig. 3). The NTDs of Na^+ and Cl^- decreased by 68% (0.47 kg ha^{-1} $month^{-1}$) and 57% (0.67 kg ha^{-1} $month^{-1}$), respectively, in comparison with the bulk deposition (Fig. 3). However, the neutral (zero) values were included within the 95% confidence limits of the means of Na^+ and Cl^-. The NTDs of K^+, Mg^{2+}, and Ca^{2+} were 78%, 76%, and 33% of those of the throughfall deposition.

RELATIONSHIPS BETWEEN NET THROUGHFALL DEPOSITION AND SURFACE DRENCHED DURATION

Figure 4 shows the relationships between NTDs and the duration of time that the leaf surface area was drenched with fog and dew (the period when the wetness value of the captive grid sensors was >90% and when no rainfall fell) and rain duration. The NTDs of K^+, Mg^{2+}, Ca^{2+}, Cl^-, and SO_4^{2-} were positively correlated with the drenched duration. Conversely, the NTD of Na^+ was not correlated with the drenched duration. There were no clear correlations between the NTDs and the rain duration.

Discussion

THE EFFECTS OF REGIONAL AND LONG-RANGE TRANSBOUNDARY AIR POLLUTION

Comparison of the acidic deposition showed that the pH and pAi of rain precipitation (pAi = log([nss-SO_4^{2-}] + [NO_3^-])) at Jodo-daira were 4.81 ± 0.11 and 4.51 ± 0.08, respectively, and were slightly acidic when compared with sites at other altitudes on Mount Tateyama (Honoki et al., 2009) and the other mountains (Annen et al., 1995), but less acidic than mean concentrations for Japan (EANET,

TABLE 1

Volume-weighted mean concentrations during the snow-free period (August and September).

		Precipitation		μeq L^{-1}									ion balance (μeq L^{-1})		
		(mm $month^{-1}$)	pH	H^+	Na^+	NH4$^+$	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	cation	anion	total
2006	Rain	256.4	5.0	22.1	4.0	11.4	0.8	0.5	7.6	7.3	13.9	30.7	46.3	51.8	98.1
	Fog	—	—	—	—	—	—	—	—	—	—	—	—	—	—
	Throughfall	73.8	5.1	5.3	5.1	1.6	38.3	30.5	55.1	8.6	2.0	26.2	135.8	36.7	172.6
2007	Rain	373.5	5.0	10.2	13.0	7.4	5.8	1.0	8.7	9.7	7.3	26.2	46.1	43.2	89.3
	Fog	115.3	4.7	18.2	13.2	23.4	6.3	0.0	9.3	14.2	23.1	62.8	70.4	100.0	170.3
	Throughfall	260.2	5.1	7.8	9.4	3.1	15.2	3.3	15.2	8.8	3.0	22.7	54.0	34.5	88.5
2008	Rain	353.1	4.6	27.9	2.9	8.9	0.2	0.9	2.5	4.2	7.7	21.3	43.4	33.3	76.7
	Fog	80.1	4.3	54.3	7.7	31.2	1.3	3.4	13.0	8.2	23.4	89.2	110.8	120.8	231.6
	Throughfall	264.7	4.5	29.9	3.1	2.5	15.8	9.1	7.1	5.8	2.6	23.3	67.5	31.8	99.2
2009	Rain	239.9	5.0	10.7	3.5	13.4	0.9	0.6	1.7	5.3	13.6	25.4	30.8	44.4	75.3
	Fog	168.5	5.4	3.7	5.6	10.1	1.4	1.4	20.5	7.6	19.7	30.8	42.7	58.1	100.8
	Throughfall	—	—	—	—	—	—	—	—	—	—	—	—	—	—
2010	Rain	209.7	5.3	4.7	1.4	21.3	0.5	1.1	11.6	3.1	11.2	2.0	40.6	16.3	56.9
	Fog	—	—	—	—	—	—	—	—	—	—	—	—	—	—
	Throughfall	282.9	5.1	7.8	1.0	2.5	10.7	2.9	3.9	4.2	4.7	24.3	28.9	33.2	62.1
2011	Rain	349.8	4.9	13.4	5.0	12.1	0.3	0.6	0.7	7.4	6.4	22.7	32.1	31.5	63.6
	Fog	112.2	4.4	39.0	7.5	32.4	1.8	2.3	8.7	11.2	17.2	39.6	91.6	54.7	146.3
	Throughfall	350.1	4.8	15.6	2.3	10.6	11.6	3.5	4.2	3.8	2.0	15.9	47.9	20.1	68.0
VWM	Rain	297.0	4.8	15.5	5.5	11.6	1.6	0.8	5.2	6.5	8.5	22.2	40.1	37.2	77.3
	Fog	119.0	4.6	24.0	8.2	22.1	2.7	1.6	13.7	10.1	17.4	50.5	72.4	78.0	150.4
	Throughfall	246.3	4.8	14.6	3.8	4.9	14.7	6.1	10.1	5.7	2.5	21.5	54.3	29.7	83.9
CV	Rain	0.23	0.05	0.58	0.83	0.39	1.53	0.33	0.81	0.39	0.51	0.47	0.17	0.34	0.20
	Fog	0.31	0.11	0.78	0.39	0.42	0.89	0.82	0.42	0.30	0.53	0.47	0.37	0.39	0.34
	Throughfall	0.42	0.05	0.76	0.79	0.91	0.62	1.20	1.27	0.38	0.60	0.17	0.61	0.21	0.45

Notes: VWM = volume weighted mean concentration (2006 to 2011). CV = coefficient of variation.

2011). However, during the observation period, high concentrations of nss-SO_4^{2-} (>200 μeq L^{-1}) in fog water were frequently observed at Jodo-daira. High concentrations of nss-SO_4^{2-} in fog water were also observed at Mount Norikura (2770 m a.s.l.) (Watanabe et al., 1999) and at Murodo-daira (2450 m a.s.l.) on Mount Tateyama (Watanabe et al., 2010). As a result, the Ai of rain precipitation was different from that of fog water. At Jodo-daira, the fog water was about twice as acidic as the precipitation. When the Ai values of fog water were included in the total precipitation, the values were comparable to the mean concentration of Japan. Therefore, fog water plays an important role in controlling acidity inputs at Jodo-daira.

Honoki et al. (2007) reported that the concentrations of sodium ions in rain decreased with distance from the coastline and with elevation. The different trends in the altitudinal profile of deposition between sea salt components (Na^+, Cl^-) and NH_4^+, nss-SO_4^{2-}, and NO_3^- (Fig. 2) suggest that the NH_4^+, nss-SO_4^{2-}, and NO_3^- were not from the sea, but from other nonpoint (regional) sources. Previous studies of Mount Tateyama have reported high concentrations of pollutants transported from the Asian continent, such as O_3, SO_2, and acidic aerosols (Kido et al., 2001; Osada et al., 2003; Watanabe et al., 2010). Watanabe et al. (2010), however, suggested that the nitric acid gas, which mainly affects the NO_3^- in fog water, comes from industrial regions that are relatively close to Mount Tateyama. As a result, the effects of regional and transboundary air pollution are considerably different each year (Tables 1 and 2, Fig. 2). Watanabe et al. (2010) reported that the NO_3^-/nss-SO_4^{2-} ratio and the concentrations of SO_4^{2-}, NH_4^+, and H^+ of fog water at Murodo-daira fluctuated widely year by year depending on the air mass distribution of the particular year. When the air mass came from Asia, especially from the coasts of the Yellow Sea, the NO_3^-/nss-SO_4^{2-} ratio decreased. This indicates that the vegetation canopy at Mount Tateyama is exposed to different chemical deposition conditions each year.

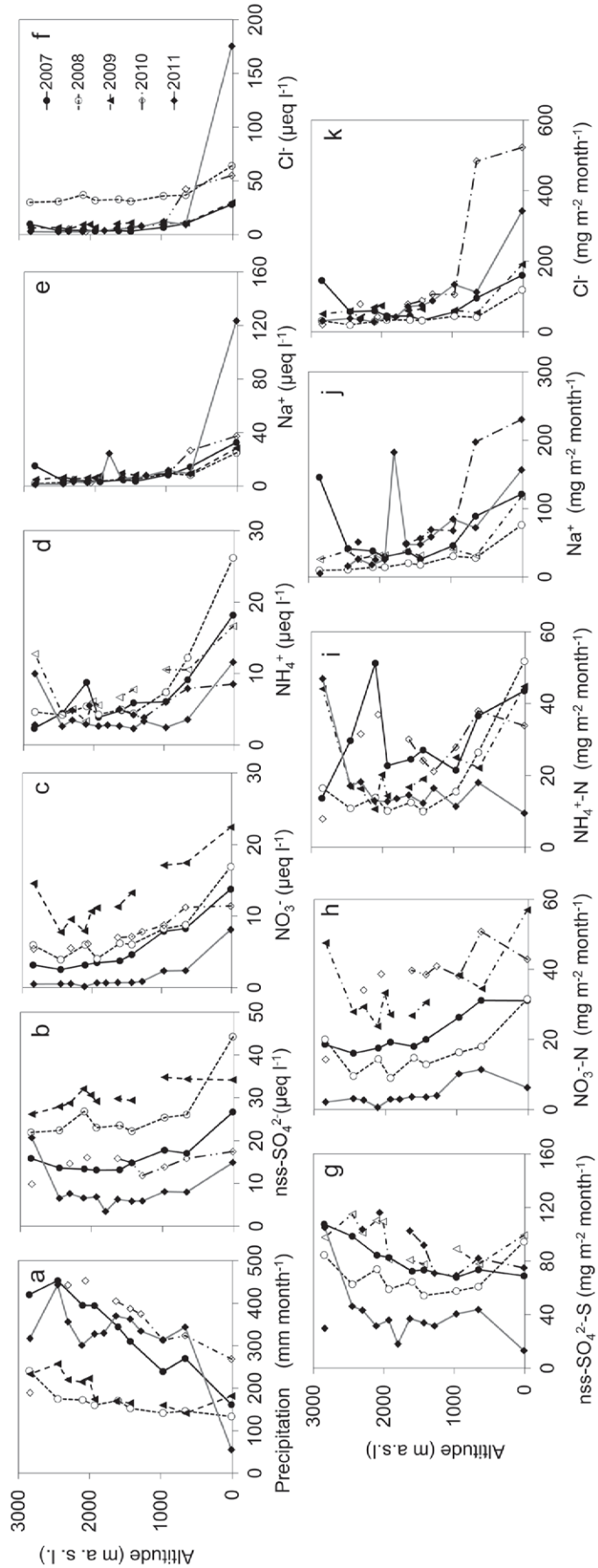

FIGURE 2. Relationships between altitude and the amounts of precipitation and their ion concentrations at Mount Tateyama in September from 2007 to 2011. (a) precipitation (mm), (b) nss-SO_4^{2-} concentration, (c) NO_3^- concentration, (d) NH_4^+ concentration, (e) Na^+ concentration, (f) Cl^- concentration, (g) nss-SO_4^{2-}-S deposition, (h) NO_3^--N deposition, (i) NH_4^+-N deposition, (j) Na^+ deposition, and (k) Cl^- deposition. ● 2007, ○ 2008, ▲ 2009, ◊ 2010, and ♦ 2011. These data are from Honoki et al. (2009) and Honoki and Watanabe (2010, 2011, 2012), except for Jododaira (2839 m a.s.l.).

TABLE 2

Monthly mean ionic deposition (kg ha^{-1} month^{-1}) for the *Pinus pumila* stands.

		eq ha^{-1}	Kg ha^{-1} month^{-1}							
		H^+	Na^+	NH_4^+-N	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^--N	SO_4^{2-}-S
2006	Rain	56.6	0.24	0.41	0.08	0.01	0.39	0.66	0.50	1.26
	BD	—	—	—	—	—	—	—	—	—
	TD	3.9	0.09	0.02	1.11	0.27	0.81	0.22	0.02	0.31
	NTD	—	—	—	—	—	—	—	—	—
2007	Rain	50.4(38.1)	0.97(1.12)	0.33(0.39)	0.66(0.84)	0.04(0.04)	0.23(0.65)	1.09(1.29)	0.28(0.38)	1.34(1.57)
	BD	60.4(58.5)	1.19(1.46)	0.60(0.75)	0.81(1.12)	0.04(0.04)	0.27(0.86)	1.48(1.85)	0.48(0.74)	2.02(2.69)
	TD	14.0(20.2)	0.3(0.56)	0.08(0.11)	0.75(1.55)	0.06(0.10)	0.35(0.79)	0.51(0.81)	0.10(0.11)	0.66(0.95)
	NTD	–46.4	–0.89	–0.52	–0.07	0.01	0.08	–0.97	–0.38	–1.36
2008	Rain	62.7(98.6)	0.08(0.23)	0.24(0.44)	0.03(0.02)	0.02(0.04)	0.09(0.18)	0.25(0.53)	0.19(0.38)	0.67(1.21)
	BD	103.3(156.2)	0.19(0.42)	0.57(0.91)	0.07(0.08)	0.05(0.08)	0.28(0.46)	0.41(0.83)	0.43(0.73)	1.87(2.72)
	TD	43.8(79.1)	0.18(0.19)	0.10(0.09)	1.48(1.64)	0.28(0.29)	0.33(0.38)	0.49(0.55)	0.06(0.10)	0.90(0.99)
	NTD	–59.5	–0.00	–0.47	1.41	0.23	0.05	0.08	–0.38	–0.97
2009	Rain	25.7	0.19	0.45	0.09	0.02	0.08	0.45	0.46	0.98
	BD	9.0	0.28	0.55	0.13	0.03	0.38	0.65	0.66	1.33
	TD	—	—	—	—	—	—	—	—	—
	NTD	—	—	—	—	—	—	—	—	—
2010	Rain	9.8	0.07	0.62	0.04	0.03	0.49	0.23	0.33	1.13
	BD	—	—	—	—	—	—	—	—	—
	TD	22.1	0.07	0.10	1.19	0.10	0.22	0.42	0.19	1.10
	NTD	—	—	—	—	—	—	—	—	—
2011	Rain	29.9(46.7)	0.11(0.40)	0.44(0.59)	0.04(0.05)	0.02(0.03)	0.05(0.05)	0.27(0.91)	0.19(0.31)	1.02(1.27)
	BD	59.0(87.6)	0.25(0.58)	0.76(1.07)	0.09(0.12)	0.04(0.05)	0.18(0.23)	0.06(1.33)	0.38(0.56)	1.48(1.64)
	TD	42.2(54.8)	0.17(0.18)	0.47(0.52)	1.68(1.59)	0.16(0.15)	0.30(0.30)	0.47(0.47)	0.07(0.10)	0.93(0.89)
	NTD	–16.8	–0.09	–0.29	1.59	0.12	0.13	–0.13	–0.31	–0.55
Average	Rain	39.2(45.9)	0.28(0.38)	0.41(0.48)	0.16(.019)	0.02(0.03)	0.22(0.31)	0.49(0.68)	0.32(0.39)	1.07(1.24)
	BD	57.9(77.8)	0.48(0.69)	0.62(0.82)	0.28(0.36)	0.04(0.05)	0.28(0.48)	0.79(1.17)	0.49(0.67)	1.68(2.17)
	TD	25.2(36.0)	0.16(0.22)	0.15(0.17)	1.24(1.41)	0.17(0.18)	0.40(0.50)	0.42(0.49)	0.09(0.10)	0.78(0.85)
	NTD	–32.7	–0.32	–0.47	0.97	0.13	0.13	–0.36	–0.40	–0.90

Notes: BD = bulk ion deposition; TD = throughfall ion deposition; NTD = net throughfall ion deposition.

INTERACTIONS BETWEEN THE P. PUMILA CANOPY AND PRECIPITATION

Canopy Interception and Climate Conditions

Uehara and Kume (2012) reported that approximately 48% of rainfall was intercepted by the pine canopy from 2006 to 2008, but that from 2006 to 2011, the mean interception was approximately 17%; this indicates that the amount of rainfall intercepted in 2010 and 2011 was small. Uehara and Kume (2012) showed that fog deposition on the *P. pumila* canopy made a large contribution to the amount of throughfall during the snow-free period; they demonstrated that the canopy efficiently captured fog and supplied it as throughfall water. However, the canopy interception of *P. pumila* is also high because of its large LAI. Therefore, the amount of throughfall is not simply determined by inputs of precipitation and fog, but also affected by evaporation from the canopy, and it fluctuates widely depending on the meteorological conditions. For example, in August 2006, the average rainfall per rain event was only 2.7 mm day^{-1} and the average solar radiation exceeded 180 W m^{-2}. However in August 2010, the average rainfall per rain event and average solar radiation were 13.1 mm d^{-1} and 82 W m^{-2}, respectively (Kume, unpublished data). As a result,

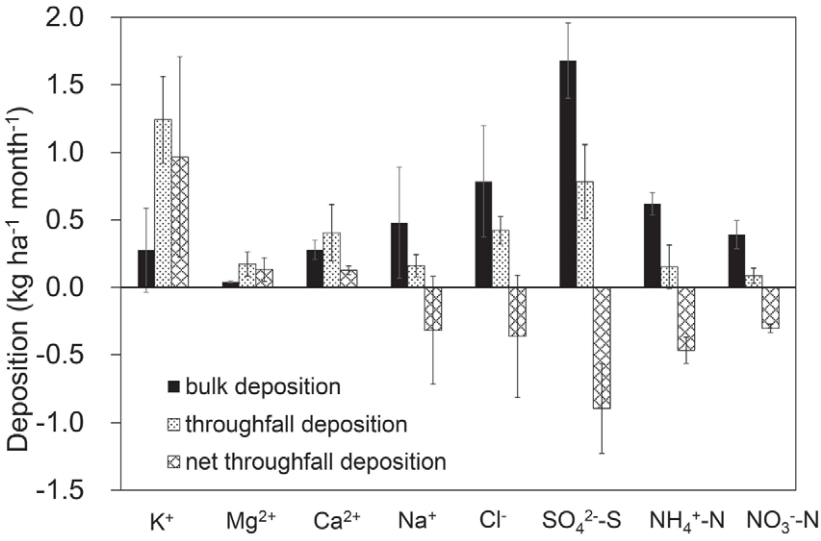

FIGURE 3. Monthly mean bulk deposition, throughfall deposition, and net throughfall deposition of the *Pinus pumila* canopy at Jododaira during the snow-free period (August and September) from 2006 to 2011. Error bars represent standard deviation for a year. The data of overflowed periods during typhoons were removed.

when compared with other years, the ion concentrations of the throughfall in 2006 were significantly condensed and the concentrations tended to be lower in 2010. The year by year differences in the meteorological conditions caused the large CV values for the amount of throughfall deposition (Table 1).

Canopy Absorption

The monthly mean NTDs of Na^+, NH_4^+-N, Cl^-, NO_3-N, and SO_4^{2-}-S of the *P. pumila* canopy were negative (Table 2, Fig. 3). However, the NTDs of Na^+ and Cl^- were not significantly different from zero (Fig. 3), which suggests they could be regarded as being inert within the canopy (Parker, 1983; Tukey, 1970; Ulrich, 1983). Some canopy exchange processes, however, seemed to occur in Cl^- (Fig. 4, part h). SO_4^{2-}-S is also regarded as an inert element; however, the NTD of SO_4^{2-}-S was negative (Fig. 3) and the absorption of SO_4^{2-} decreased with drenched duration (Fig. 4, part i). There was no significant correlation between the SO_4^{2-} concentration in fog water and the drenched duration ($r = 0.18$). It is noted that trees can directly absorb SO_2 and accumulate sulfur in their needles via stomatal openings (Olszyk and Tingey, 1985; Rautio and Huttunen, 2003). Marty et al. (2012) estimated that a significant fraction of dry deposition (~1.2 kg S ha^{-1} yr^{-1}) would be taken up by the canopy of boreal forests in spring and summer. Precise physiological mechanisms are still not known, but it is thought that SO_4^{2-} is taken up by plant cells via co-transport of H^+/SO_4^{2-} (Scherer, 2001). Further research is needed to clarify the interaction mechanism between SO_4^{2-} absorption and drenched duration.

In this study, approximately 79.0% (0.65 kg ha^{-1} mo^{-1}) of NH_4^+-N and 85% (0.57 kg ha^{-1} mo^{-1}) of NO_3^--N was retained in the *P. pumila* canopy from the bulk deposition, which suggests that a high percentage of deposited inorganic N compounds were absorbed by the *P. pumila* canopy. It is well known that the canopy takes up some N from precipitation (Tomaszewski et al., 2003; Klopatek et al., 2006; Sparks, 2009; Fenn et al., 2013). In their review of studies of throughfall in North America and Europe, Lovett and Lindberg (1993) showed that a maximum of 40% of N was retained in the canopy. Ignatova and Dambrine (2000) suggested that coniferous trees absorb a significant amount of N from dry and occult deposition through their canopy. Klopatek et al. (2006) showed that about 75% of NO_3^--N and 90% of NH_4^+-N were retained in the canopy of young and old-growth Douglas fir stands in the Cascade Mountains of southern Washington State during the summer. Chiwa et al. (2012) showed that, in 1 year, needles of a *Pinus densiflora* canopy assimilated approximately 60% of the N dose sprayed on the canopy as artificial mist; the absorbed N was directly assimilated by the needles and was used in photosynthetic apparatus.

It has been suggested that uptake of N compounds was strongly related to leaf wettability (Sase et al., 2008; Adriaenssens et al., 2011). However, we did not detect any significant relationship between canopy N absorption and canopy wetness; that is, inorganic N was well absorbed by the canopy under all conditions. One possibility is the high needle surface area density of the *P. pumila* canopy with long needle longevity (ca. 4 years). The *P. pumila* canopy efficiently captured fog water (Uehara and Kume, 2012) and the long wet period and relatively high sustained concentrations of NO_3^- and NH_4^+ on the needle surface would result in efficient nutrient retention by the canopy. The observed absorption rates seemed to be saturated regardless of the surface condition of the needles.

LEACHING AND CANOPY WETNESS

The cation-anion balances of the incident precipitation were nearly equal, but an anion deficit was observed for the throughfall (Table 1). Anion deficits in throughfall have been reported in previous studies (e.g., Cronan et al., 1983; De Walle et al., 1985; Currie et al., 1999). Leaching out of weak Brønsted acid (an organic acid) from the foliar surface (Cronan et al., 1983; Fillion et al., 1998) that is not detected by ion chromatography or deprotonated ions may be the cause of the anion deficit.

Concentrations of K^+, Ca^{2+}, and Mg^{2+} in the throughfall were high compared to those in the BD with NTDs of 0.97, 0.13, and 0.13 kg ha^{-1} mo^{-1} (3-year average), respectively. This result is consistent with those from other studies that have shown that, in various forest types, a large proportion of the ions in the throughfall originated from canopy leaching (Cronan et al., 1983; Parker, 1983, Okochi et al., 1995; Houle et al., 1999; Muramoto et al., 2007). As shown in Figure 4, NTDs of most ions except Na^+ increased as the drenched duration of the canopy increased. On the other hand, H^+ deposition of fog and rain precipitation and NTDs were not significantly correlated (data not shown). Therefore, the

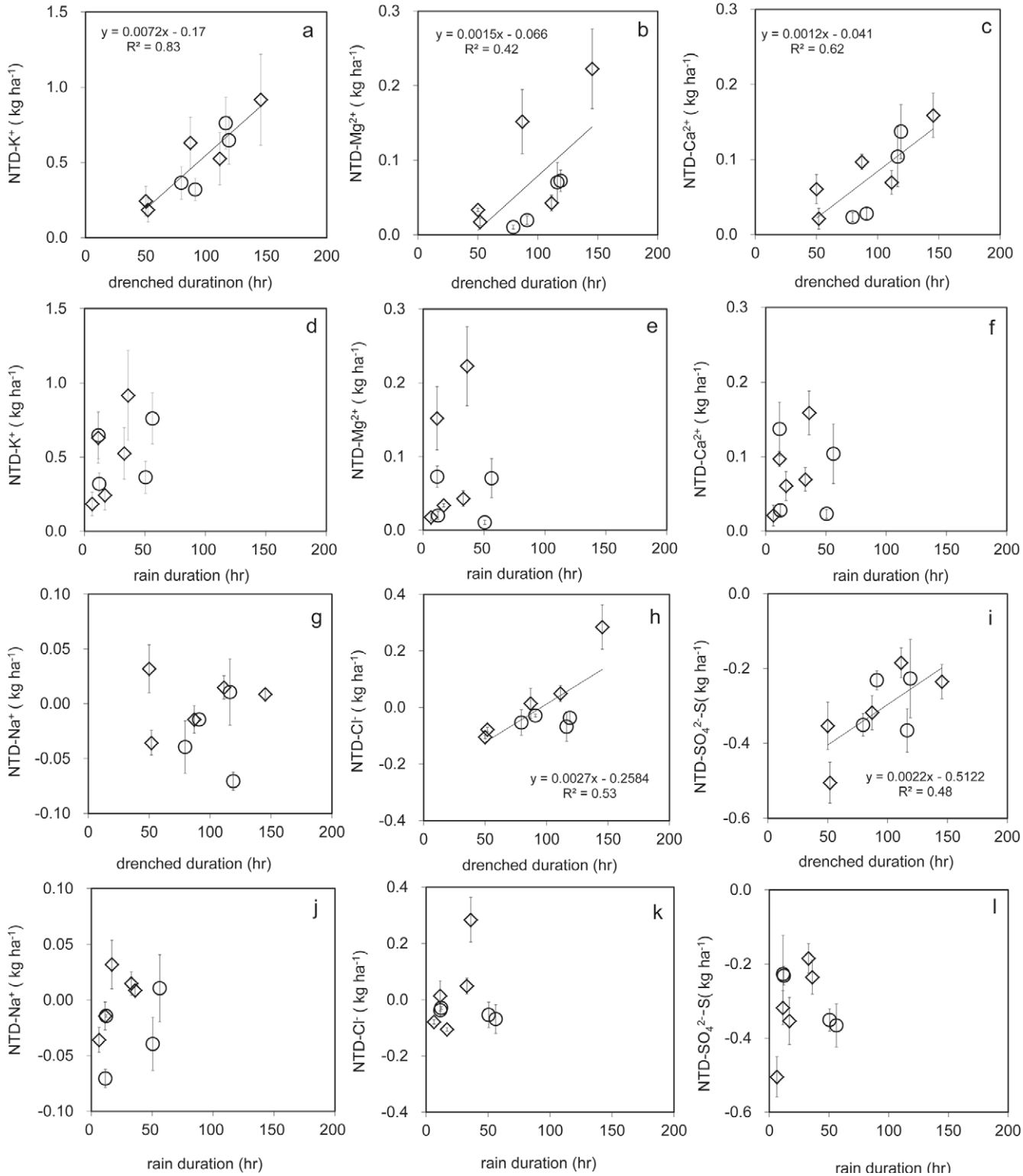

FIGURE 4. Relationships between drenched duration of the canopy and net throughfall deposition (NTD) of various ions. (a) NTD-K^+, (b) NTD-Mg^{2+}, (c) NTD-Ca^{2+}, (g) NTD-Na^+, (h) NTD-Cl, and (i) NTD-SO_4^{2-}-S. Relationships between rain period and (d) NTD-K^+, (e) NTD-Mg^{2+}, (f) NTD-Ca^{2+}, (j) NTD-Na^+, (k) NTD-Cl^-, and (l) NTD-SO_4^{2-}-S. Regression lines are drawn for significant relationships ($P < 0.05$). Error bars represent the standard error of six throughfall collectors. ◊ 2008 and ○ 2011. The drenched duration was defined as the proportion when the wetness value was >90% and when no rain fell. One point represents the value of the sampling period. The data of the overflow periods during typhoons were removed.

amount of leaching was mainly determined by the period when the needles were drenched with fog and dew. Uehara and Kume (2012) reported that, even in the driest year, the canopy surface was drenched for one-third of the day in August. During the drenched period, leached ions were concentrated on the needle surface and dripped into the collectors. In contrast, the rain duration was not so long and the needle surface condition was unstable. The rain drops disturbed the accumulation of leached ions and diluted them. Such microclimatological differences may affect the leaching process.

It has been suggested that the leaf surface properties are important regulatory factors for the leaching of K^+ (Sase et al., 2008; Kume et al., 2010). In the preliminary study, we found that the K^+ leakage from needle surface is tightly linked with the needle development stage, and the newly developing needles in August showed the highest leakage rate and decreased to the minimum in the end of October (Kume, unpublished data). Therefore, the cation leaching occurs regardless of the winter damage of needle surface.

Conclusion

To obtain information on the interactions between atmospheric deposition and *P. pumila* vegetation above the tree line, the chemical characteristics of rain and fog precipitation and NTDs of the canopy were investigated on a summit of Mount Tateyama (2839 m a.s.l.), central Japan. Although the concentrations of inorganic nitrogen and sulfur in the rain precipitation were low at the summit, the total amount of deposition tended to be larger than at the lower sites because of the large amount of precipitation and the highly concentrated fog deposition. Concentrations of sulfate, nitrate, and ammonium ions in fog water were about twice those in rain. Regional and long-range transboundary air pollution were the main sources of atmospheric nitrogen and sulfur. About 80% of NH_4^+-N and NO_3^--N deposition was retained in the canopy; these may be important N sources for the pine vegetation. With the exception of Na^+, the drenched duration, approximately the fog period, determined the amount of ion leaching. The pine canopy significantly leached K^+ and Mg^{2+}. These results indicate that canopy ion exchange processes and microclimatic conditions are important when evaluating the nutrient budgets of alpine ecosystems during snow-free period.

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References Cited

Adriaenssens, S., Staelens, J., Wuyts, K., de Schrijver, A., Van Wittenberghe, S., Wuytack, T., Kardel, F., Verheyen, K., Samson, R., and Boeckx, P., 2011: Foliar nitrogen uptake from wet deposition and the relation with leaf wettability and water storage capacity. *Water Air and Soil Pollution,* 219: 43–57.

Annen, K., Ohnishi, M., Fujitani, R., Hayakari, S., Fukuzaki, N., Sasaki, K., Shimizu, G., Koyama, I., Kume, K., Dokiya, Y., Maruta, E., Hatakeyama, I., and Murano, K., 1995: Ion concentration and acid deposition in mountainous region of Japan. *Nippon Kagaku Kaishi*, 11: 916–921.

Bowman, W. D., 2000: Biotic controls over ecosystem response to environmental change in alpine tundra of the Rocky Mountains. AMBIO: A *Journal of the Human Environment,* 29: 396–400.

Bowman, W. D., Gartner, J. R., Holland, K., and Wiedermann, M., 2006: Nitrogen critical loads for alpine vegetation and terrestrial ecosystem response: are we there yet? *Ecological Applications*, 16: 1183–1193.

Burns, D. A., 2004: The effects of atmospheric nitrogen deposition in the Rocky Mountains of Colorado and southern Wyoming, USA- a critical review. *Environmental Pollution,* 127: 257–269.

Chiwa, M., Oshiro, N., Miyake, T., Nakatani, N., Yuhara, T., Hashimoto, N., and Sakugawa, H., 2003: Dry deposition washoff and dew on the surfaces of pine foliage on the urban- and mountain-facing sides of Mt. Gokurakuji, western Japan. *Atmospheric Environment,* 37: 327–337.

Chiwa, M., Crossley, A., Sheppard, L. J, Sakugawa, H., and Cape, J. N., 2004: Throughfall chemistry and canopy interactions in a Sitka spruce plantation sprayed with six different simulated polluted mist treatments. *Environment Pollution,* 127: 57–64.

Chiwa, M., Matsuda, T., Nakatani, N., Kobayashi, T., Kume, A., and Sakugawa, H., 2012: Effects of canopy N uptake on foliar CO_2 assimilation rates and biomass production and allocation in Japanese red pine seedlings. *Canadian Journal of Forest Research,* 42: 1395–1403.

Cronan, C. S., and Reiners, W. A., 1983: Canopy processing of acid precipitation by coniferous and hardwood forest in New England. *Oecologia,* 59: 216–223.

Currie, W. S., Aber, J. D., and Driscoll, C. T., 1999: Leaching of nutrient cations from the forest floor; effects of nitrogen saturation in two long-term manipulations. *Canadian Journal of Forest Research,* 29: 609–620.

De Walle, R. D., Ribblet, G. C., Helvey, J. D., and Kochenderfer, J., 1985: Laboratory investigation of leachate chemistry from six Appalachian forest floor types subjected to simulated acid rain. *Journal of Environmental Quality*, 14: 234–240.

EANET, 2011: *Second Periodic Report on the State of Acid Deposition in East Asia. Part II: National Assessments.* Niigata, Japan: Acid Deposition Monitoring Network in East Asia (EANET), 305 pp.

Fenn, M. E., Ross, C. S., Schilling, S. L., Baccus, W. D., Larrabee, M. A., and Lofgren, R. A., 2013: Atmospheric deposition of nitrogen and sulfur and preferential canopy consumption of nitrate in forests of the Pacific Northwest, USA. *Forest Ecology and Management*, 302: 240–253.

Fillion, N., Probst, A., and Probst, J. L., 1998: Natural organic matter contribution to throughfall acidity in French forests. *Environment International*, 24: 547–558.

Fowler, D., Cape, J. N., Unsworth, M. H., Mayer, H., Crowther, J. M., Jarvis, P. G., Gardiner, B., and Shuttleworth, W. J., 1989: Deposition of atmospheric pollutants on forests. *Philosophical Transactions of the Royal Society B,* 324: 247–265.

Holzmann, H. P., and Haselwandter, K., 1988: Contribution of nitrogen fixation to nitrogen nutrition in an alpine sedge community (*Caricetum curvulae*). *Oecologia*, 76: 298–302.

Honoki, H., and Watanabe, K., 2010: Acid rain observation report at different altitudes on the Mt. Tateyama (2008). *Bulletin of the Toyama Science Museum*, 33: 113–120.

Honoki, H., and Watanabe, K., 2011: Acid rain observation report at different altitudes on the Mt. Tateyama (2009). *Bulletin of the Toyama Science Museum*, 34: 151–158.

Honoki, H., and Watanabe, K., 2012a: Acid rain observation report at different altitudes on the Mt. Tateyama (2010). *Bulletin of the Toyama Science Museum*, 35: 119–128.

Honoki, H., and Watanabe, K., 2012b: Acid rain observation report at different altitudes on the Mt. Tateyama (2011). *Bulletin of the Toyama Science Museum*, 36: 13–26.

Honoki, H., Watanabe, K., Iida, H., Kawada, K., and Hayakawa, K., 2007: Deposition analysis of non sea-salt sulfate and nitrate along to the northwest winter monsoon in Hokuriku district by a snow boring core and bulk samples. *Bulletin of the Glaciological Research*, 24: 23–28.

Honoki, H., Watanabe, K., and Kometani, M., 2009: Acid rain observation report at different altitudes on the Mt. Tateyama (2007). *Bulletin of the Toyama Science Museum,* 32: 125–131.

Houle, D., Ouimet, R., Paquin, R., and Laflamme, J., 1999: Interactions of atmospheric deposition with a mixed hardwood and a coniferous forest canopy at the Lake Clair Watershed (Duchesnay, Quebec). *Canadian Journal of Forest Research*, 29: 1944–1957.

Ignatova, N., and Dambrine, É., 2000: Canopy uptake of N deposition in spruce (*Picea abies* L. Karst) stands. *Annals of Forest Science,* 57: 113–120.

Kajimoto, T., 1992: Dynamics and dry matter production of belowground woody organs of *Pinus pumila* trees growing on the Kiso mountain range in central Japan. *Ecological Research*, 7: 333–339.

Kajimoto, T., 1994: Aboveground net production and dry matter allocation of *Pinus pumila* forests in the Kiso mountain range, central Japan. *Ecological Research,* 9: 193–204.

Keene, W. C., Pszenny, A. A. P., Galloway, J. N., and Hawley, M. E., 1986: Sea-salt corrections and interpretation of constituent ratios in marine precipitation. *Journal of Geophysical Research,* 91(D6): 6647–6658.

Kido, M., Osada, K., Matsunaga, K., and Iwasaka, Y., 2001: Diurnal variation of ionic aerosol species and water-soluble gas concentrations at a high-elevation site in the Japanese Alps. *Journal of Geophysical Research*, 106: 17335–17345.

Klopatek, J. M., Barry, M. J., and Johnson, D. W., 2006: Potential canopy interception of nitrogen in the Pacific Northwest, USA. *Forest Ecology and Management*, 234: 344–354.

Kume, A., Numata, S., Watanabe, K., Honoki, H., Nakajima, H., and Ishida, M., 2009: Influence of air pollution on the mountain forests along the Tateyama-Kurobe Alpine route. *Ecological Research,* 24: 821–830.

Kume, A., Nagafuchi, O., Akune, S., Nakatani, N., Chiwa, M., and Tetsuka, K., 2010: Environmental factors influencing the load of long-range transported air pollutants on *Pinus amamiana* in Yakushima island Japan. *Ecological Research,* 25: 233–243.

Kume, A., Watanabe, K., Nagafuchi, O., and Honoki, H., 2011: Influence of the road of long-range transported air pollutants on forest ecosystem: some examples in the Yakushima Island and Mt. Tateyama in Japan. *Japanese Journal of Ecology,* 61: 97–106.

Lovett, G. M., and Kinsman J., D., 1990: Atmospheric pollutant deposition to high-elevation ecosystems. *Atmospheric Environment. Part A. General Topics*, 24: 2767–2786.

Lovett, G. M., and Lindberg, S. E., 1984: Dry deposition and canopy exchange in a mixed oak forest as determined by analysis of throughfall. *Journal of Applied Ecology,* 21: 1013–1027.

Lovett, G. M., and Lindberg, S. E., 1993: Atmospheric deposition and canopy interactions of nitrogen in forests. *Canadian Journal of Forest Research,* 23: 1603–1616.

Marty, C., Houle, D., Duchesne, L., and Gagnon, C., 2012: Canopy interaction with precipitation and sulphur deposition in two boreal forests of Quebec, Canada. *Environmental Pollution,* 162: 354–360.

Mladenov, N., Williams, M. W., Schmidt, S. K., and Cawley, K., 2012: Atmospheric deposition as a source of carbon and nutrients to an alpine catchment of the Colorado Rocky Mountains. *Biogeosciences,* 9: 3337–3355.

Muramoto, M., Nara, M., Asari, T., and Suzuki, K., 2007: Hydro-chemical cycle of forest ecosystem in the Norikura Highlands—1. Chemical characteristic and seasonal change of throughfall in forest ecosystem. *Journal of Japanese Association of Hydrological Sciences*, 37: 73–83.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T., 2007: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020. *Atmospheric Chemistry and Physics,* 7: 4419–4444.

Okitsu, S., and Ito, K., 1983: Dynamic ecology of the *Pinus pumila* community of Mts. Taisetsu, Hokkaido, Japan. *Journal of the Graduate School of Environmental Science, Hokkaido University,* 6: 151–184.

Okitsu, S., and Ito, K., 1984: Vegetation dynamics of the Siberian dwarf pine (*Pinus pumila* Regel) in the Taisetsu mountain range, Hokkaido, Japan. *Vegetatio,* 58: 105–113.

Okochi, H., Hosono, T., Maruyama, F., and Igawa, M., 1995: Interaction between acid deposition and the canopy of the Japanese cedar and fir in Mt. Oyama. *Environmental Science,* 8: 305–315.

Olszyk, D. M., and Tingey, D. T., 1985: Interspecific variation in SO_2 flux: leaf surface versus internal flux, and components of leaf conductance. *Plant Physiology*, 79: 949–956.

Osada, K., Kido, M., Iida, H., Matsunaga, K., Iwasaka, Y., Nagatani, M., and Nakada, H., 2003: Seasonal variation of free tropospheric aerosol particles at Mt. Tateyama, central Japan. *Journal of Geophysical Research,* 108(D23): 8667.

Parker, G. G., 1983: Throughfall and stemflow in the forest nutrient cycle. *Advances in Ecological Research,* 13: 57–133.

Rautio, P., and Huttunen, S., 2003: Total vs. internal element concentrations in Scots pine needles along a sulphur and metal pollution gradient. *Environmental Pollution,* 122: 273–289.

Sase, H., Takahashi, A., Sato, M., Kobayashi, H., Nakata, M., and Totsuka, T., 2008: Seasonal variation in the atmospheric deposition of inorganic constituents and canopy interactions in a Japanese cedar forest. *Environmental Pollution,* 152: 1–10.

Scherer, H. W., 2001: Sulphur in crop production—invited paper. *European Journal of Agronomy,* 14: 81–111.

Sparks, J. P., 2009: Ecological ramification of the direct foliar uptake of nitrogen. *Oecologia,* 159: 1–13.

Tomaszewski, T., Boyce, L. R., and Sievering, H., 2003: Canopy uptake of atmospheric nitrogen and new growth nitrogen requirement at a Colorado subalpine forest. *Canadian Journal of Forest Research,* 33: 2221–2227.

Tukey, H. B., Jr., 1970: The leaching of substances from plants. *Annual Review of Plant Physiology,* 21: 305–324.

Uehara, Y., and Kume, A., 2012: Canopy rainfall interception and fog capture by *Pinus pumila* Regal at Mt. Tateyama in the Northern Japan Alps, Japan. *Arctic, Antarctic, and Alpine Research,* 44: 143–150.

Ulrich, B., 1983: Interaction of forest canopies with atmospheric constituents: SO_2, alkali and earth alkali cations and chloride. *In* Ulrich, B., and Pankrath J. (eds.), *Effects of Accumulation of Air Pollutants in Forest Ecosystems.* Boston: Reidel Publishing Company, 33–45.

Wada, N., 2007: Fine-scale species richness of alpine fellfield plant communities on a middle-latitude mountain of central Japan: influences of alpine dwarf pine and substrate properties. *Far Eastern Studies,* 6: 31–34.

Walker, D. A., Halfpenny, J., C., Walker, M., D., and Wessman, C. A., 1993: Long-term studies of snow-vegetation interactions. *BioScience*, 43: 287–301.

Watanabe, K., Ishizaka, Y., and Takenaka, C., 1999: Chemical composition of fog water near the summit of Mt. Norikura in Japan. *Journal of the Meteorological Society of Japan,* 77: 997–1006.

Watanabe, K., Honoki, H., Iwai, A., Tomatsu, A., Noritake, K., Miyashita, N., Yamada, K., Yamada, H., Kawamura, H., and Aoki, K., 2010: Chemical characteristics of fog water at Mt. Tateyama, near the coast of the Japan sea in central Japan. *Water, Air, and Soil Pollution,* 211: 379–393.

Watanabe, K., Honoki, H., Iwama, S., Iwatake, K., Mori, S., Nishimoto, D., Komori, S., Saito, Y., Yamada, H., and Uehara, Y., 2011: Chemical composition of fog water at Mt. Tateyama near the coast of the Japan sea in central Japan. *Erdkunde*, 65: 233–245.

Wehren, B., Weingartner, R., Schädler, B., and Viviroli, D., 2010: General characteristics of alpine waters. *In* Bundi, U. (ed.), *Alpine Waters*. Heidelberg: Springer, *Handbook of Environmental Chemistry*, Volume 6, 17–58.

Yanagimachi, O., and Ohmori, H., 1991: Ecological status of *Pinus pumila* scrub and the lower boundary of the Japanese alpine zone. *Arctic and Alpine Research*, 23: 424–435.

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