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Deposition of Aerosols on Leaves in a Cool-temperate Larch Forest in Northern Hokkaido, Japan

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ABSTRACT

Aerosol concentrations at the CC-Lag site in the Teshio Experimental Forest increased from winter to spring and sometimes showed extremely high values associated with Kosa and/or forest-fire events. The range and mean of the mass concentrations of aerosol chemical species were as follows: total particulate mass, 1.2-29, 5.0; elemental carbon, 0.061-2.2, 0.43; organic carbon, 0.059-3.5, 0.79; and sulfate, 0.12-6.2, 1.8 $\mu g/m^3$. The total masses of the deposited particles on hybrid larch and on bamboo leaves were approximately 35 and 30 $\mu g/cm^2$, respectively. The amounts of soil particles on the leaves were 6 $\mu g/cm^2$ for the upper part of hybrid larch, 2 $\mu g/cm^2$ for the lower part of hybrid larch, and 1 $\mu g/cm^2$ for Sasa bamboo leaves. The amounts of deposited black carbon were 2.3 $\mu g/cm^2$ for the upper part of hybrid larch, 0.6 $\mu g/cm^2$ for the lower part of hybrid larch, and 0.2 $\mu g/cm^2$ for Sasa bamboo leaves. Half of the total deposited particular mass was attached on the hybrid larch; however, most of the total deposited mass was adhered on the Sasa bamboo leaves. Regardless of the species, there tend to be more deposited particles on the leaves in the upper part than in the lower part, with only a few meters height difference. Comparing the composition of the deposited particles to that of the atmospheric aerosols without any size cut, the fractions of water-soluble material sulfate and sea salt in the deposited aerosols were about one tenth and one hundredth lower than that in the aerosols, respectively. On the basis of the measured concentration and the deposited amount on leaves, the deposition velocity of black carbon was estimated to be approximately 0.5 cm/s.

Key words: Deposition, Aerosol, Leaf, Hybrid larch, Sasa bamboo, Black carbon

1. INTRODUCTION

Pollutants emitted into the atmosphere can easily be transported to remote areas. They can also be deposited in and affect areas that do not have any pollutant sources. Inoue *et al.* (2005) reported that 49% of annual sulfur deposition in Japan came from China in 1995. This contribution is believed to have increased in recent years (Kuribayashi *et al.*, 2011). Future air quality conditions will be strongly affected by growth in anthropogenic emissions, which are controlled by economic growth, environmental policy, and future implementation of emission controls. Asian total emissions of SO_2, NO_x, and black carbon were projected to increase substantially by 22%, 44%, and 1%, respectively, in the reference case scenario over 2000 levels (Ohara *et al.*, 2007). Given that Japan is located downwind from China, the amount of pollutant deposition in Japan is expected to increase in proportion. In addition to human health, the impact on vegetation, which is an important CO_2 sink, is a very urgent issue to be investigated. However, only a few studies on the effect of aerosols on vegetation have been conducted (U.S. EPA, 2004). In this paper, we first give an account of atmospheric aerosol concentrations and chemical compositions, before estimating the amount of aerosol depo-

sition on leaves at the CC-LaG site (Carbon Cycle and Larch Growth experiment site) in the Teshio Experimental Forest, Hokkaido University.

2. EXPERIMENTAL DESCRIPTION AND METHODOLOGY

2.1 Site Description

The study site is located on a flat terrace in the Teshio Experimental Forest, Hokkaido University (45° 03′N, 142°07′E), in northeastern Hokkaido, Japan. The site was a 9-year-old plantation (in 2010) of hybrid larch (*Larix gmelinii* × *L. Kaempferi*) with an area of 13.7 ha. A pristine forest at the site was clear-cut from January to March 2003 (during the snow-covered period). Sasa bamboos were left intact under the snowpack during clear-cutting; however, bamboos were strip-cut into 4-m-wide rows. Seven months after clear-cutting and just before, the 2-year-old hybrid larch saplings were planted (Takagi *et al*., 2009). There have been no significant local pollutant sources.

2.2 Aerosol Sampling and Analysis

From May 30, 2002 to present, filter sampling of atmospheric aerosols was made every two weeks until December 2007, following which it was made every 3 weeks. The sampling was paused during clear-cutting from October 25, 2002 to April 25, 2003. We used two types of filters: Teflon filters (Sumitomo Poreflon WP-500-50) for the total particulate mass (TPM) and water-soluble matter and quartz fiber filters (Palluflex 2500 QAST-UP) for carbonaceous particles. The aerosols that had passed through a cyclone separator, whose 50% cut-off diameter is 2 μm, were collected on each filter using a pump at a flow rate of 20 L/min.

We first measured the TPM of the collected aerosols on Teflon filters by weighing them on an electric balance in a dry box with silica gel (Ohta *et al*., 1998). The collected aerosols were then extracted ultrasonically with distilled-deionized water. We analyzed the concentrations of sulfate (SO_4^{2-}), nitrate (NO_3^-), chloride (Cl^-), sodium (Na^+), calcium (Ca^{2+}), magnesium (Mg^{2+}), potassium (K^+), and ammonium (NH_4^+) using an ion chromatograph (Yokogawa Electric Works Inc., IC-100 or Dionex, DX-50). The amount of excess sulfate (ex. SO_4^{2-}) was calculated by subtracting the amount of SO_4^{2-} in sea salt from that of SO_4^{2-} measured in the atmosphere (Ohta and Okita, 1990), such that

$$\text{ex. } SO_4^{2-} = SO_4^{2-} - 0.251 \times Na^+.$$

The amount of sea-salt cations (s.s.C.) were calculated as follows (Ohta *et al*., 1998):

$$\text{s.s.C.} = (1/0.837) \times Na^+.$$

The carbonaceous components on the quartz filters were analyzed using an NC analyzer (Sumigraph NC-80) and GC-FID (Hitachi 164) (Ohta and Okita, 1990). Since December 2008, the carbonaceous components have been analyzed using a DRI OC/EC carbon analyzer (model 2001). Hasegawa cross-checked the results of carbonaceous matter using both methods and found that they fit closely with each other (Personal communication).

To measure metal concentrations, aerosols were digested at room temperature in a 1.5 mL HNO_3-0.5 mL HF mixture in a disposable test tube for two days (Jalkanen and Hansen, 1996). After digestion, metal concentrations were determined with ICP-MS (Agilent, HP-4500). We used the aluminum concentrations as a tracer material for the soil.

2.3 Leaf Sampling and Analysis

Since Sase *et al*. (1996) reported that the amounts of deposition tend to be higher in the upper part than in the lower part, hybrid larch leaves were collected from two heights, approximately 1.5 m and 3 m, at five points in the site on September 1, 2010. Current-year Sasa bamboo leaves were also collected from approximately 1 m height at the same points as the hybrid larch leaf collection. We took leaf samples from short shoots in plastic bags.

Approximately 10 g of leaves were cut off from branches and rinsed with 50 mL ultrapure water to find "attached particles" on leaves. Leaves rinsed with ultra pure water were then dried at room temperature and again rinsed with chloroform to find "adhered particles" on leaves. The rinsed waters were filtered with Teflon filters (Advantec PTFE pore size: 0.5 μm) and quartz fiber filters (Advantec QR-100). Suspended solids on Teflon filters were first weighed on an electric balance in a dry box in the same way for TPM. Black carbon on the quartz fiber filters was analyzed using the

Table 1. Average concentrations and ranges of aerosol components from 22 May 1993 to 19 November 2010 (μg/m^3).

	TPM	E.C.	O.C.	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}	Al
Average	5.0	0.43	0.79	1.8	0.022	0.019	0.51	0.06	0.19	0.015	0.016	0.0078
Max.	29	2.2	3.5	6.2	0.58	0.16	1.5	0.33	1.1	0.056	0.15	0.030
Min.	1.2	0.061	0.059	0.12	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	0.00047

DRI OC/EC carbon analyzer.

We collected the adhered particles and adhered black carbon on leaves by using chloroform to Teflon filters and quartz filters, respectively, in the same way as Takamatsu *et al.* (2001).

We also calculate the deposition velocity of black carbon [cm/s] in the universal way (Seifeld and Pandis, 1998). It was assumed that black carbon deposition flux onto the leaves was directly proportional to the local atmospheric concentration at the site.

3. RESULTS AND DISCUSSION

3. 1 Chemical Composition of Atmospheric Aerosols at CC-Lag Site

Table 1 shows the average concentrations of the TPM and chemical constituents at the CC-Lag site from May

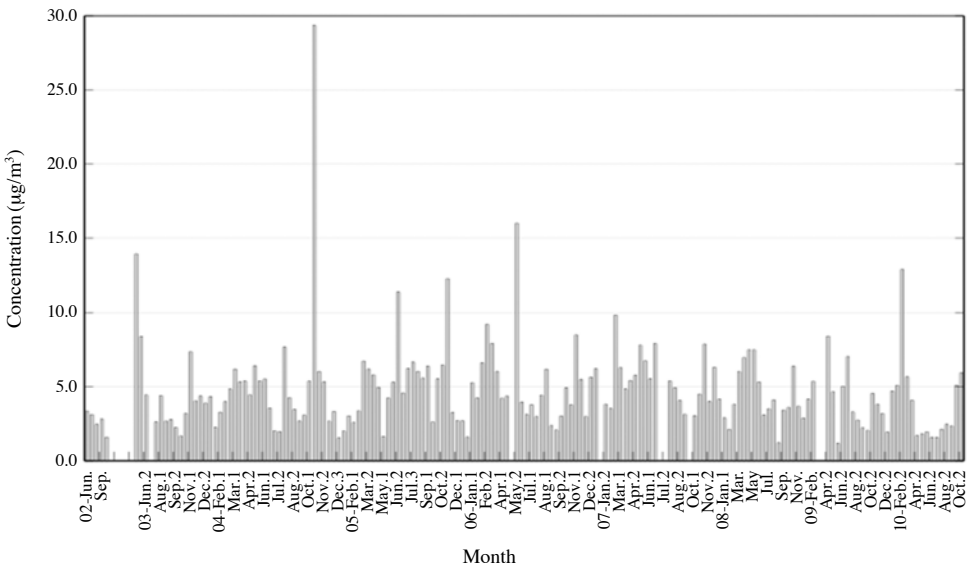

Fig. 1. Total particulate mass of atmospheric fine aerosols at CC-Lag site in the Teshio Experimental Forest from May 22, 2002 to November 19, 2010.

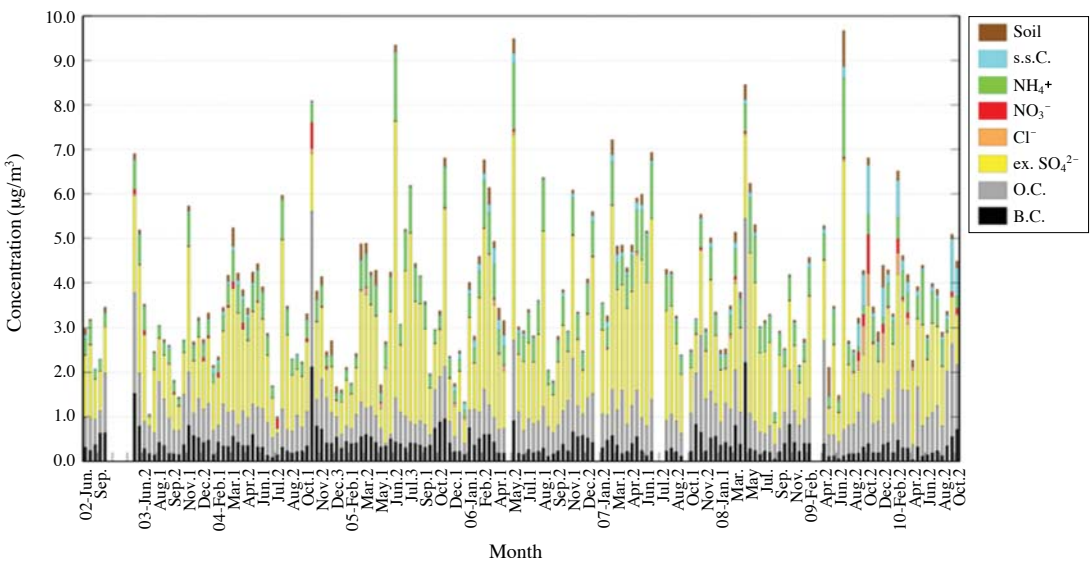

Fig. 2. Concentrations of aerosol chemical species at the CC-Lag site in the Teshio Experimental Forest from May 22, 2002 to November 19, 2010. B.C., O.C., ex. SO_4^{2-}, and s.s.C show black carbon, organic carbon, excess sulfate, and sea salt cations, respectively.

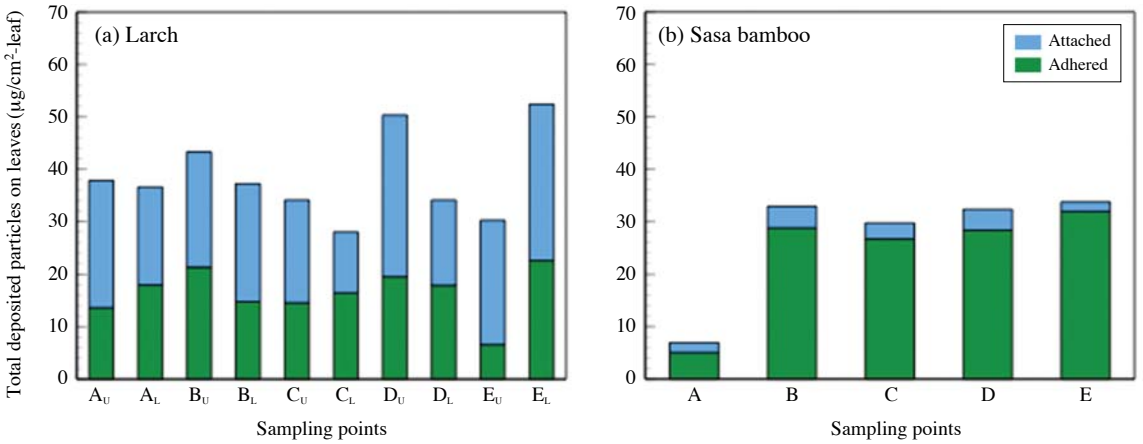

Fig. 3. Total deposited particles on hybrid larch leaves (a) and Sasa bamboo leaves (b) at the five sampling points at the CC-Lag site in the Teshio Experimental Forest. The subscript notations "U" and "L" show data from upper and lower parts, respectively.

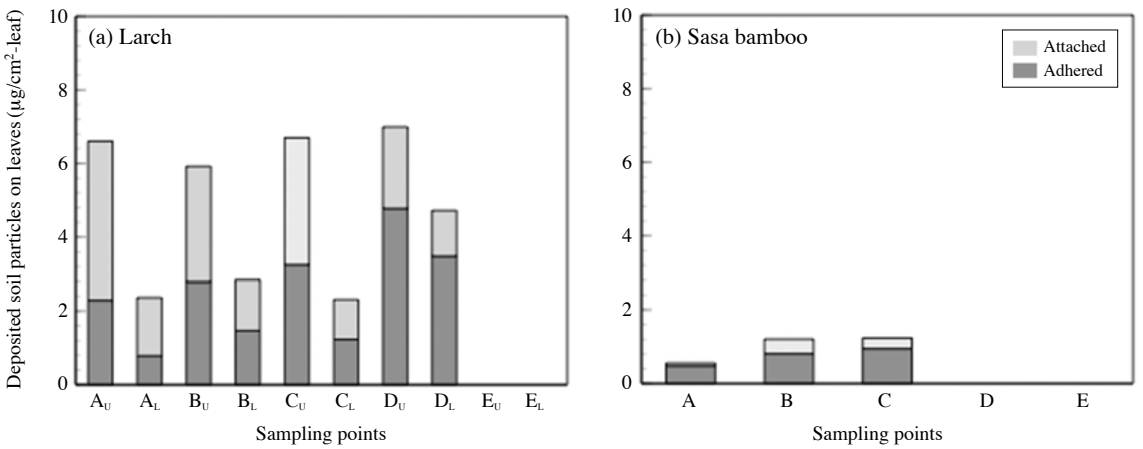

Fig. 4. Deposited soil particles on hybrid larch leaves (a) and Sasa bamboo leaves (b) at the five sampling points at the CC-Lag site in the Teshio Experimental Forest. The subscript notations "U" and "L" show data from the upper and lower parts, respectively.

22, 1993 to November 19, 2010. At this site, the range and the mean of the mass concentrations of aerosol chemical species were as follows: TPM, 1.2-29, 5.0; elemental carbon, 0.061-2.2, 0.43; organic carbon, 0.059-3.5, 0.79; and sulfate, 0.12-6.2, 1.8 μg/m^3. Figs. 1 and 2 show the variations of the mass concentrations of the TPM and of each aerosol chemical species. The fine aerosols mainly consisted of sulfate, organic carbon, and black carbon. The aerosol concentration usually increased from winter to spring, mainly owing to an increase in the fossil fuel combustion products and sea salts, and sometimes showed extremely high values owing to Kosa and/or forest-fire events. MODIS images suggest that plumes of massive forest fires in Russia and China largely covered the Hokkaido area in the latter half of October 2004 and April 2008. In those periods, black carbon concentrations reached more than 2 μg/m^3.

3.2 Depositions of Aerosols on Leaves

As shown in Fig. 3, the total masses of the deposited particles on hybrid larch and bamboo leaves were approximately 35 and 30 μg/cm^2, respectively. Half of the total deposited particular mass was attached to the hybrid larch; however, most of the total deposited mass was adhered to the Sasa bamboo leaves. The amounts of epicuticular wax on Sasa bamboo leaves and hybrid larch leaves were 0.135 mg/cm^2 and 0.075 mg/cm^2, respectively. This might influence the attached/adhered ratio of Sasa bamboo leaves and hybrid larch leaves. As shown in Fig. 4, the amounts of soil particles on the leaves were 6 μg/cm^2 for the upper part of hybrid larch,

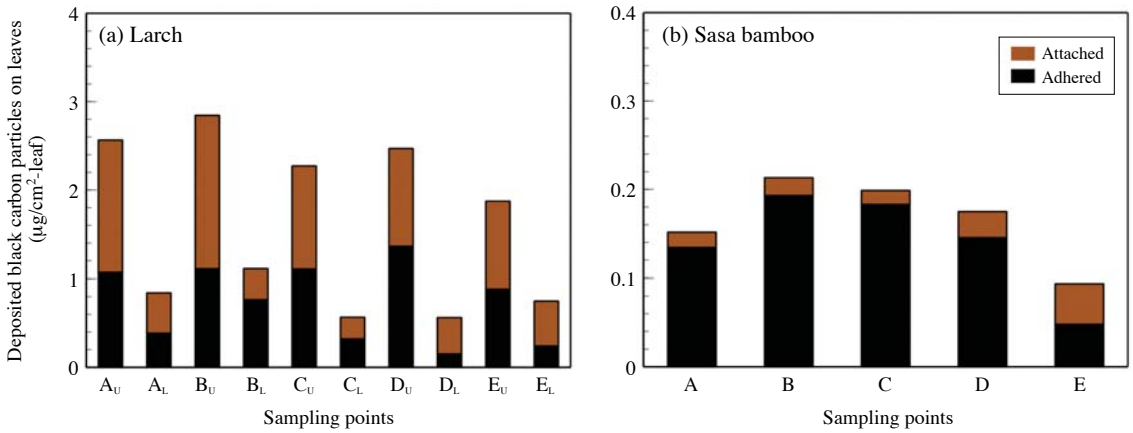

Fig. 5. Deposited black carbon particles on hybrid larch leaves (a) and Sasa bamboo leaves (b) at the five sampling points at the CC-Lag site in the Teshio Experimental Forest. The subscript notations "U" and "L" show data from upper and lower parts, respectively.

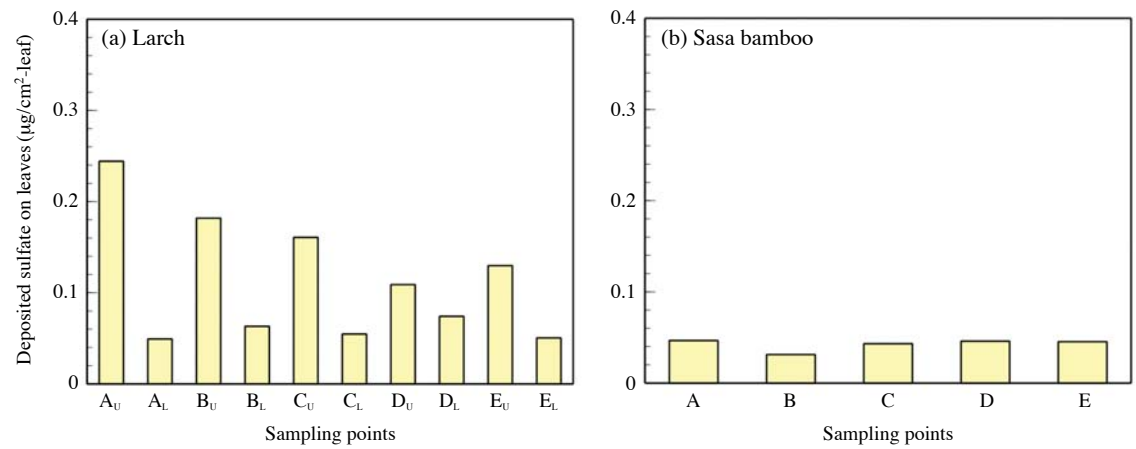

Fig. 6. Deposited sulfate on hybrid larch leaves (a) and Sasa bamboo leaves (b) at the five sampling points at the CC-Lag site in the Teshio Experimental Forest. The subscript notations "U" and "L" show data from upper and lower parts, respectively.

2 μg/cm^2 for the lower part of hybrid larch, and 1 μg/cm^2 for Sasa bamboo leaves. The amounts of deposited black carbon were 2.3 μg/cm^2 for the upper part of hybrid larch, 0.6 μg/cm^2 for the lower part of hybrid larch, and 0.2 μg/cm^2 for Sasa bamboo leaves (Fig. 5). The results for sulfate and sea salt are shown in Figs. 6 and 7, respectively. Regardless of the species, there tend to be more deposited particles on the leaves in the upper part than in the lower part, particularly for soil-derived species that may be supplied from the ground surface. We compared the composition of the deposited particles to that of atmospheric aerosols, without any size cut. The fraction of water-soluble material sulfate and sea salt in the deposited aerosols were about one tenth and one hundredth lower than that in the aerosols, respectively. These results indicate that attached water-soluble or hydrophilic aerosols on leaves may easily wash out with rain. On the other hand, hydrophobic aerosols strongly adhered on the leaves. Soil particles were hydrophilic, and black carbon was hydrophobic. This is why the composition of deposited matter was different from that of the atmospheric aerosols.

On the basis of the measured concentration and deposited amount on leaves, the deposition velocity of black carbon was estimated to be approximately 0.5 cm/s. This value is close to the deposition velocity of black carbon estimated for a tropical forest in Thailand (Matsuda *et al.*, 2012) and that of PM2.5 sulfate estimated for a deciduous forest in central Japan (Matsuda *et al.*, 2010). Since the value is significantly higher than that used in chemical transport model calcula-

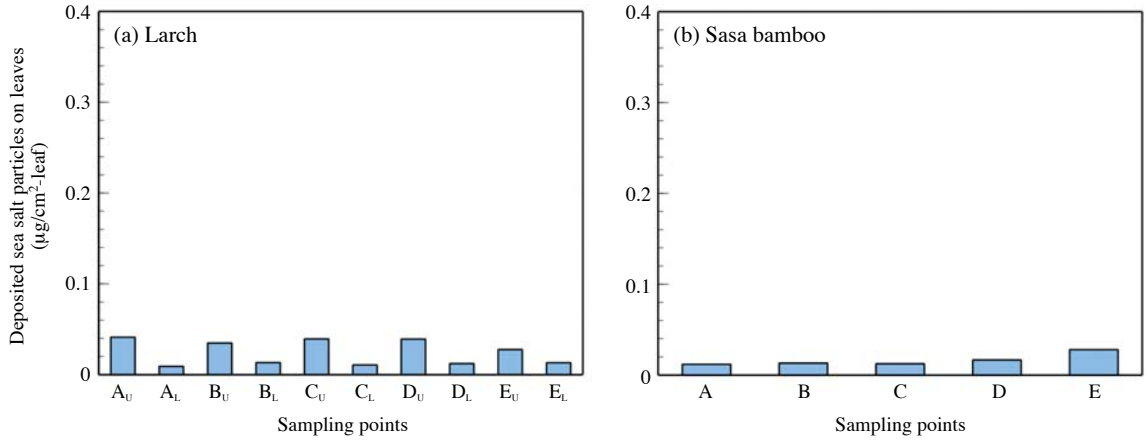

Fig. 7. Deposited sea salt particles on hybrid larch leaves (a) and Sasa bamboo leaves (b) at the five sampling points at the CC-Lag site in the Teshio Experimental Forest. The subscript notations "U" and "L" show data from upper and lower parts, respectively.

tions, model results for forest areas may have to be re-evaluated.

4. CONCLUSIONS

Aerosol concentrations at the CC-Lag site in the Teshio Experimental Forest were measured from May 22, 2002 to November 19, 2010. They showed a broad maximum from winter to spring, and Kosa and/or forest-fire events induced extremely high values. In particular, in the latter half of October 2004 and April 2008, plumes of massive forest fires in Russia and China largely covered the Hokkaido area. During that period, black carbon concentrations exceeded 2 μg/m^3.

The amount of deposited particles on hybrid larch and Sasa bamboo leaves including the total mass of all deposited particles, of soil particles, and of black carbon were also measured. Half of the total deposited particular mass was attached on hybrid larch, whereas most of the mass was adhered on Sasa bamboo leaves. The deposited aerosol species tend to be higher in the upper part than in the lower part. The fraction of material that was water soluble in the deposited aerosols was much lower than that in aerosols, indicating that attached water-soluble aerosols on leaves may easily wash out with rain.

The estimated deposition velocity of black carbon was approximately 0.5 cm/s, which is close to that for a tropical forest in Thailand (Matsuda *et al*., 2012).

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