

# The Chemistry of Precipitation and Throughfall of Three Forest Stands in Central Taiwan\*

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## [ Summary ]

The study objectives were to compare changes of ion concentrations and seasonal differences of throughfall for 3 types of forest stands, China-fir (*Cunninghamia lanceolata*) plantation, secondary hardwood stand, and natural hardwood stand in Guandaushi Forest, in central Taiwan from July 1995 to June 1996. Seasonal changes of concentrations of throughfall were higher in the winter especially in November, and the secondary hardwood stand had the highest concentrations. The standard deviation of enrichment factors of ion concentrations varied greatly among these 3 stands. Most cation concentrations from the natural and secondary hardwood stands were considerably higher than were precipitation concentrations except for  $H^+$  and  $Na^+$ . Most ion concentrations from China-fir also showed higher values than the precipitation concentrations except for  $Na^+$ ,  $NH_4^+$ ,  $SO_4^{2-}$  and  $HCO_3^-$ . The enrichment factor of  $H^+$  of China-fir was 1.08 which greatly exceeded the enrichment factors of 0.05 and 0.21 for natural and secondary hardwoods, respectively.

**Key words:** precipitation, throughfall, China-fir stand, secondary hardwood stand, natural hardwood stand.

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## 臺灣中部雨水與三種林分穿落水的化學組成

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### 摘 要

本試驗的目的在臺灣中部關刀溪試驗集水區內選定相鄰的杉木林、次生闊葉樹林及天然闊葉樹林三種不同林分，調查比較雨水流經此三種不同林分，即穿落水中離子濃度的變化及差異(1995年7月到1996年6月)。結果顯示此三個林分每次所收集之水樣的無機離子濃度差異甚大。其中天然及次生闊葉樹林穿落水的陽離子除 $H^+$ 和 $Na^+$ 外，其餘皆高出雨水甚多，杉木林則除 $Na^+$ 、 $NH_4^+$ 、 $SO_4^{2-}$ 及 $HCO_3^-$ 等離子外，其他離子之濃度亦較雨水高，唯其增加量較緩和，其中 $H^+$ 的增加係數在杉木林為1.08，高出闊葉樹林之增加係數0.05及0.21甚多。三林分之穿落水的離子濃度在一年當中時，以冬天的濃度最高，尤其是十一月，且以次生林為最明顯。

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## INTRODUCTION

There is general agreement that the chemistry of precipitation changes after it has passed through forest canopies. But the relative importance of processes (e.g., surface wash-off, leaching, absorption) which contribute to the changes of throughfall composition is still a matter of scientific debate (Cape *et al.*, 1987; Miller, 1984; Ulrich, 1983). Atmospheric pollution fluxes in throughfall vary in relation to the input of pollution, climate, the tree species, the distance from the forest edge, and the age of the stands. Crown leaching of ions is enhanced by deposition acidity and by the health status of the stand (Mahendrappa, 1989, 1990; Yawney *et al.*, 1978).

The potential of atmospheric pollutants to affect the canopy-mediated portion of the nutrient cycle was recognized in central Taiwan (King and Shiue, 1992; Liu and Sheu, 1996) but has not been systematically investigated. The objectives of this study were to discuss temporal variations of ion concentrations in precipitation and throughfall to understand if atmospheric pollutants promote leaching of substances from 3 adjacent forest canopies. We also report on ion enrichments which are ratios of throughfall to precipitation of volume-weighted mean ion concentrations for frequently measured solutes for 3 types of forest stands in central Taiwan. These 3 types of forests receive similar atmospheric loadings of anthropogenic strong acids and have similar time intervals between rain events.

## MATERIALS AND METHODS

### Study sites

This study was carried out in the Guandaushi Forest, central Taiwan (Fig. 1). The site is located at a 47-ha watershed with elevations ranging from 1100 to 1700 m. The mean annual temperature is about 25°C, and the annual rainfall is about 2700 mm with distinct rainy and dry seasons. Typhoons occur occasionally between June and September and bring a high intensity of precipitation and disturbance to the site. The site

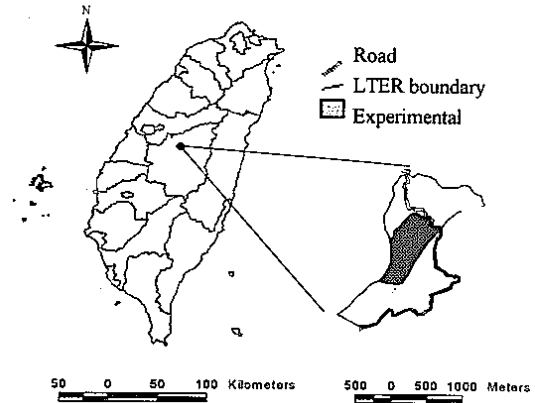


Fig. 1. Location of study site at Guandaushi Experimental Forest in central Taiwan.

is a typical subtropical mixed-hardwood forest in central Taiwan which is characterized by steep topographies, abundant riparian ferns, virgin hardwood forests, and abundant epiphytes. The forests on the ridges have been cut and planted with China-fir.

Three adjacent stands of China-fir (*Cunninghamia lanceolata*) plantation, secondary hardwood, and natural hardwood forests in the same 47-ha watershed were investigated. Both hardwood forests are the typical Lauro-Fagaceae association of Taiwan. Lauracea (15 species) and Fagaceae (14 species) are the major families in this study area and they occupy 4.60% and 4.29% of the total forest composition, respectively (Lu and Ou, 1996).

### Precipitation

Samples of precipitation were collected on a rain event base from July 1995 to June 1996 using 3 bulk precipitation collectors mounted on the top of a 24-m tower. Each collector has two 19-cm-diameter polyethylene funnels connected to a 30-L sampling bottle with black polypropylene tubes. Volumes were determined in the field and 500 ml was retrieved from each collector. All sample bottles were rinsed with distilled water immediately

after each collection, then samples were transported in a cooler to the analytical laboratory at the Department of Forestry, National Chung Hsing University, within 48 h of collection and stored at 4°C.

**Throughfall**

Each throughfall collector consisted of three 19-cm diameter polyethylene funnels mounted about 1 m above the ground and arranged in a triangular shape; the funnels were connected to a 30-L sampling bottle with black polypropylene tubes. In order to keep out leaves, small branches, and insects, 3-mm mesh plastic screening was used to cover the funnels. Six such throughfall collectors in each stand were installed randomly. Samples of throughfall were collected at the same time and in the same manner as samples of precipitation.

**Chemical analysis**

The pH and conductivity of samples were determined on unfiltered samples within a few days after arrival at the laboratory. After filtration (Gelman-science GN-6 grid 0.45-µm sterilized filter paper), samples were analyzed for F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, SO<sub>4</sub><sup>2-</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, and NH<sub>4</sub><sup>+</sup> by means of ion chromatography (Dionex 100, USA). Alkalinity was measured by titration with 0.005 N H<sub>2</sub>SO<sub>4</sub> solution to pH 4.52 (APHA, 1995).

**RESULTS AND DISCUSSION**

This study was design to compare changes of ion concentrations and seasonal differences of throughfall of 3 types of forest stands to infer controlling factors. First, we examined the temporal variation of ion concentrations in precipitation and throughfall of 3 types of forest stands to understand if atmospheric pollutants promote the leaching of substances from each type of forest stand canopy. Then we used ion enrichments which are ratios of throughfall to precipitation of volume-weighted mean ion concentrations to determine the influence of forest type.

**Temporal variation of inorganic ion concent-**

**rations in precipitation and throughfall**

Mean monthly conductivity, pH, and solids concentrations of precipitation and throughfall for a full year from July 1995 to June 1996 are shown in Fig. 2. Fig. 3 illustrates Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, and NH<sub>4</sub><sup>+</sup> concentrations with respect to season. The distribution of Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and HCO<sub>3</sub><sup>-</sup> concentrations are shown in Fig. 4. Chemical concentrations were generally higher in throughfall than in precipitation, and higher chemical concentrations of throughfall appeared in winter, especially in November which had the highest chemical concentrations in all 3 stands, while the secondary forest had the highest concentration among these stands. These results show that canopy interactions with acidic precipitation may be influenced significantly by phenological status. Most ion concentrations in the growth season were

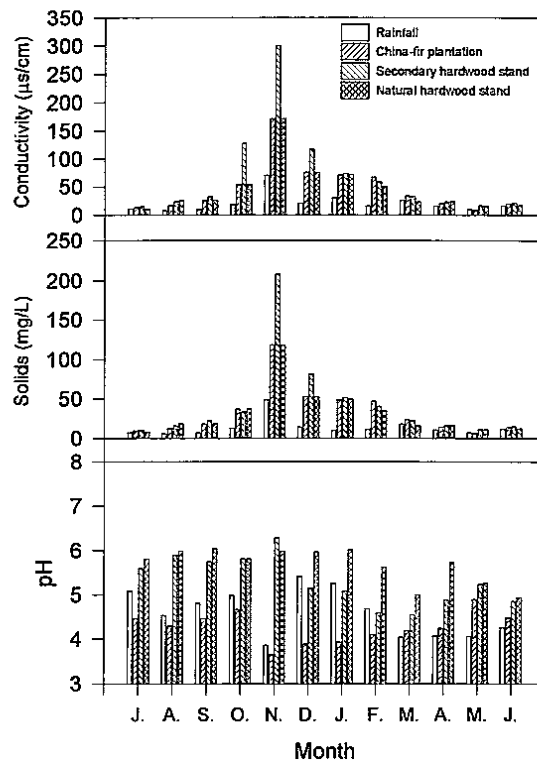


Fig. 2. Time course of conductivity, solids, and pH in rainfall and throughfall of China-fir plantation, secondary hardwood stand, and natural hardwood stand.

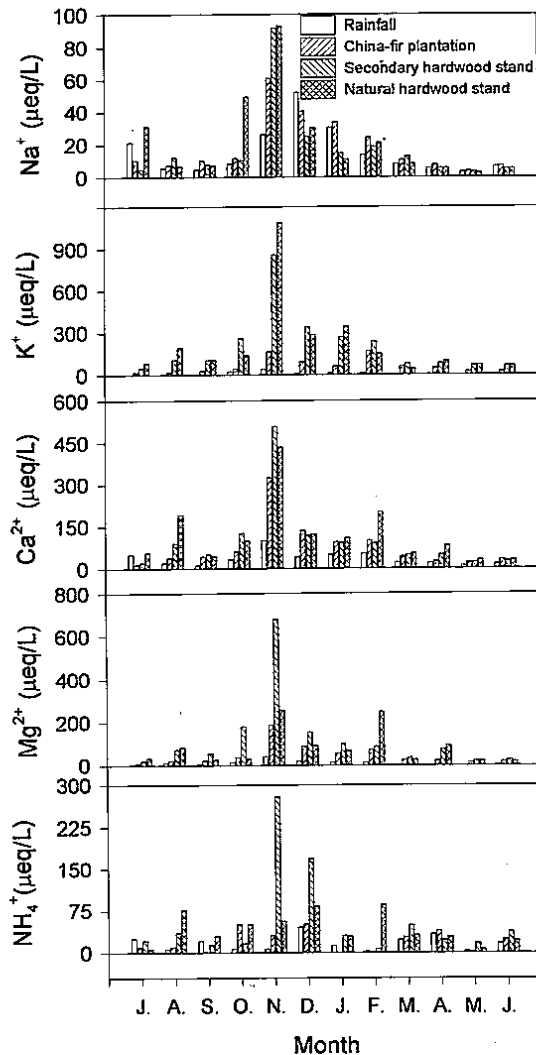


Fig. 3. Time course of  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{NH}_4^+$  in rainfall and throughfall of China-fir plantation, secondary hardwood stand, and natural hardwood stand.

lower than those in the dormant season. This phenomenon is very similar to the physiological pattern of the vegetation.

The acidity of precipitation did not show a seasonal pattern in this study. The pH in precipitation was lower than 5.6 through a entire year. Undoubtedly, this site is under threat by acid rain. However, the pH tended to increase when the rain passed through both hardwood canopies. Neutralization of acid precipitation in the 2 hardwood stands appears occur through 2 major processes: (1) ion exchange removal of free  $\text{H}^+$  by the

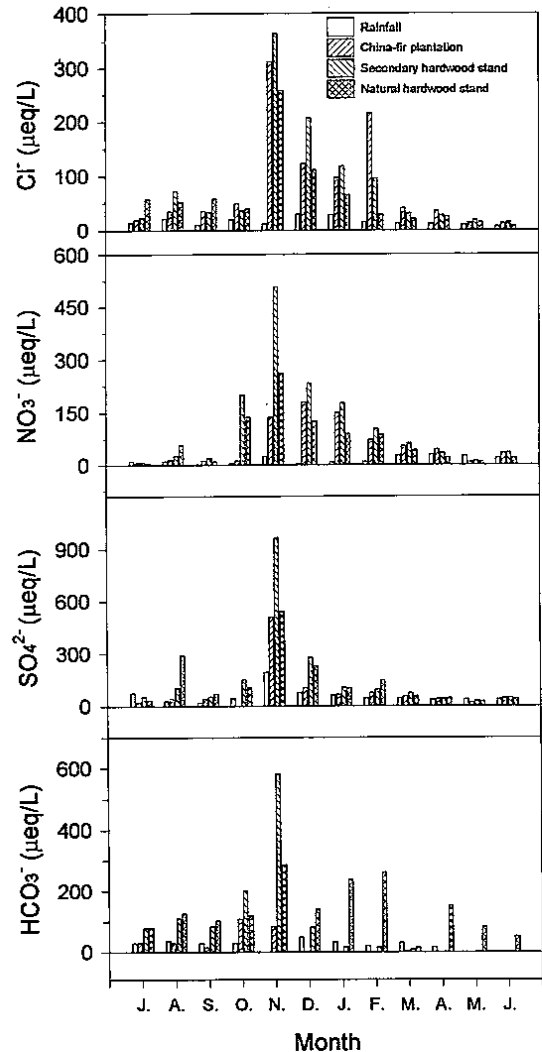


Fig. 4. Time course of  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{HCO}_3^-$  in rainfall and throughfall of China-fir plantation, secondary hardwood stand, and natural hardwood stand.

canopy (Miller, 1984; Parker, 1983) which is consistent with uptake by surface cation exchange, or association with organic anions (Edmons *et al.*, 1991); and (2) Bønsted base leaching from the plant canopy (Cronan and Reiners, 1983).

The pH values of throughfall of both hardwood stands were higher pH than precipitation throughout the year, but pH values of the China-fir stand were higher only in the period of March to July. Potter *et al.* (1991) statistically quantified the effect of event type in throughfall chemistry in a southern Appalachian forest and found precipitat-

ion intensity to be the most important factor. Analyzing sequential water samples is a potential subject to be studied in the future.

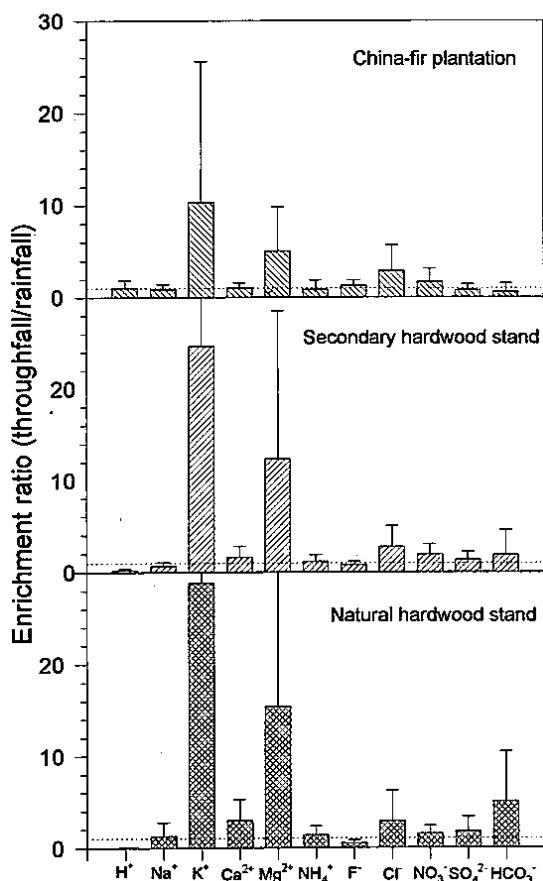
**Ion enrichments in throughfall chemistry**

Enrichment ratios of ion concentrations in throughfall by precipitation are shown in Fig. 5. It is not difficult to see that mean enrichment factors vary greatly among these 3 stands. Cations of throughfall increased considerably in both natural and secondary hardwood stands, except for H<sup>+</sup> and Na<sup>+</sup> ions. Studies of bulk and wet-only precipitation have indicated an in-canopy removal of 30-40% of the free H<sup>+</sup> in precipitation (Hoffman *et al.*, 1980; Cronan and Reiners, 1983), whereas our study suggests that the hardwood

canopy apparently removed more than 70% of free H<sup>+</sup> of total precipitation. This removal is assumed to involve both ion exchange and weak base-buffering reactions (Lovett *et al.*, 1985). Hoffman *et al.* (1980) and Cronan and Reiners (1983) also reported that the absorption of H<sup>+</sup> from wet and dry deposition could result in a reduction of nutrient pools in the canopy through exchange of H<sup>+</sup> by other cations, most of which are K<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup>, etc. Hoffman *et al.* (1980) also reported that leaching 40% to 60% of K<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup> from the forest canopy at Walker Branch Watershed could be attributed to exchange for deposited airborne acids. The effects of the loss of nutrient cations from the canopy through exchange for H<sup>+</sup> could be either positive or negative depending on the ability of the tree to replenish these pools and on the soil nutrient status (Lovett *et al.*, 1989).

Throughfall of the China-fir stand was not as much enriched with solutes leached from the canopy as was throughfall of hardwoods (Fig. 5). In comparison with their respective values of concentration with precipitation, throughfall concentration of H<sup>+</sup> was multiplied by the factor of 1.08. The positive H<sup>+</sup> flux in this study does not imply the absence of proton buffering processes in the canopy. Although the causes are not understood clearly, recent experiments have proved that throughfall contains significant concentrations of short-chain organics, such as carbohydrates, phenolics, aldehydes, and carboxylic acids (Lovett *et al.*, 1985; McDowell and Likens, 1988; Schaefer *et al.*, 1989; Cappellato *et al.*, 1993). The kinds and amounts of organic acids in the throughfall of these 3 stands are unknown, and this is definitely an important subject remaining to be studied in the near future.

It is also evident that the concentration of chloride was much higher than that of sodium in each stand (Fig. 5). These results suggest that not all the chloride in throughfall results from NaCl of canopy interception. Neary and Gizyn (1994) suggested that the forest canopy may intercept HCl and perhaps small amounts of NH<sub>4</sub>Cl, and then absorbs NH<sub>4</sub><sup>+</sup> and releases chloride. In the same process, chloride is probably combined with K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, and small amounts of Na<sup>+</sup> and



**Fig. 5. Enrichment ratios of ions of throughfall in China-fir plantation, secondary hardwood stand, and natural hardwood stand. Balance ratios are showed as dotted lines.**

then leached out. This may be the reason to interpret chloride concentrations being much higher than  $\text{Na}^+$  concentrations.

Most concentrations of ions increased in throughfall in the 3 types of stands. However, in China-fir stands, canopy absorption resulted in decreases of concentrations of atmospheric  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$  and  $\text{HCO}_3^-$  by factors of 0.90, 0.85, 0.82, and 0.58 compared with throughfall to precipitation concentrations, respectively. Coniferous forest canopies have been found to be more efficient collectors of atmospheric deposition than are broad-leaved canopies (Likens *et al.*, 1977; Mayer and Ulrich, 1978; Gosz, 1980). Coniferous needles have smaller effective diameters than broad-leaved foliage. A small effective diameter means that particles in a moving air stream are close to the underlying surface when they enter the laminar boundary layer which thus can drive them to the surface. A thinner boundary layer may also increase the capture of gases and submicron particles by diffusion (Chamberlain, 1975), therefore, China-fir canopies are likely to remove particles and gases from the atmosphere more efficiently.

On the other hand, sulfate concentrations increased in throughfall of both hardwood stands. Lindberg and Garten (1988) and Lovett and Lindberg (1984) reported that sulfate concentration increase in throughfall of natural stands is attributed to deposition of particulate sulfate and gaseous  $\text{SO}_2$  and also by the diffusion from the foliar apoplast. In any particular forested ecosystem, the proportional contributions of these mechanisms depend on both atmospheric  $\text{SO}_2$  concentrations and the availability of sulfate to the plants from soil solution. When dry deposition gaseous  $\text{SO}_2$  is absorbed by the canopy and assimilated into canopy tissues, it may undergo subsequent release in throughfall as sulfate in solution. This sulfate is not easily distinguished from apoplastic sulfate derived from root uptake (Parker, 1983).

In comparison with both secondary and natural hardwood stands, the China-fir stand retained larger amounts of ammonium, but showed on increasing proton flux in throughfall caused by the replacement of  $\text{NH}_4^+$  and  $\text{H}^+$ . This implies

that the China-fir canopy favors ammonia -N uptake.

Nitrate was also released in these 3 adjacent stands, and the amount of  $\text{NO}_3^-$  released was much higher than that of earlier reports (Cronan, 1984; Lovett and Lindberg, 1986; Mahendrapa, 1989). Our data may over-estimate the enrichment of  $\text{NO}_3^-$  because dry deposition and cloud deposition as inputs were not taken into consideration. This enrichment of  $\text{NO}_3^-$  might also come from effects of age of these stands. Foster *et al.* (1989) reported that  $\text{N}_2$  uptake rates were generally low, and any disturbance of the stand structure with mortality might actually accelerate  $\text{N}_2$  mineralization and nitrification rates in mature to over-mature stands. Increase in N inputs to such systems would lead to concomitant increases in  $\text{NO}_3^-$  leaching. Mitchell *et al.* (1992) also suggested that the capacity for older ecosystems to retain  $\text{N}_2$  may be reduced. On the other hand, concentrations of phosphate were mostly low or near the detection limit of 0.01 mg/L in this study.

## CONCLUSIONS

This study supports the following conclusions for 3 adjacent stands in Guandaushi Forest. (1) The pH values of all precipitation samples were lower than 5.6 throughout the year. The acidity of throughfall varied among forest types, being higher in the China-fir stand and lower in the 2 hardwood stands than values of precipitation. (2) Chemical concentrations were generally higher in throughfall than in precipitation and the highest chemical concentrations of throughfall appeared in winter. Most of the ion concentration of throughfall in the growth season were lower than in the dormant season. (3) The standard deviations of enrichment factors of ion concentrations vary greatly among these 3 stands. Most of the base cation concentrations from the natural and secondary hardwood stands were considerably higher than those of precipitation concentrations except for  $\text{H}^+$  and  $\text{Na}^+$ . Most of the anion concentrations from China-fir also showed higher values than the precipitation concentrations except for  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$ , and  $\text{HCO}_3^-$ . The enrichment factor of  $\text{H}^+$  of China-fir was 1.08 and it greatly exceeded the enrichment factors of 0.05 and 0.21

of natural and secondary hardwoods, respectively. (4) The positive  $H^+$  flux caused by organic materials definitely is an important subject to be studied in the future. (5) The concentration of chloride was much higher than that of sodium in each stand. These results suggest that not all the chloride in throughfall results from NaCl of canopy interception. It could result from HCl and  $NH_4Cl$  of canopy interception. (6) The concentration of nitrate was higher in throughfall of these 3 adjacent stands than in precipitation. This could be the effect of concentration, or because these 3 adjacent stands are mature stands.

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