

ORIGINAL ARTICLE

## Inorganic Nutrient Inputs from Precipitation, Throughfall, and Stemflow in *Pinus densiflora* and *Quercus mongolica* Stands in an Urban Forest Ecosystem

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### Abstract

We measured the amount of precipitation, stemflow, and throughfall and concentrations of nine major inorganic nutrients ( $H^+$ ,  $NH_4^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$ ,  $Na^+$ ,  $Cl^-$ ,  $NO_3^-$ , and  $SO_4^{2-}$ ) to investigate the nutrient inputs into soil from precipitation in *Pinus densiflora* and *Quercus mongolica* stands from September 2015 to August 2016. The precipitation inputs of  $H^+$ ,  $NH_4^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$ ,  $Na^+$ ,  $Cl^-$ ,  $NO_3^-$ , and  $SO_4^{2-}$  into soil were 0.170, 15.124, 42.227, 19.218, 14.050, 15.887, 22.391, 5.431, and 129.440  $kg \cdot ha^{-1} \cdot yr^{-1}$ , respectively. The *P. densiflora* stemflow inputs were 0.008, 0.784, 1.652, 1.044, 0.476, 0.651, 1.509, 0.278, and 9.098  $kg \cdot ha^{-1} \cdot yr^{-1}$ , and those for *Q. mongolica* were 0.008, 0.684, 2.429, 2.417, 2.941, 1.398, 2.407, 0.436, and 13.504  $kg \cdot ha^{-1} \cdot yr^{-1}$ , respectively. The *P. densiflora* throughfall inputs were 0.042, 21.518, 52.207, 27.694, 20.060, 24.049, 37.229, 10.241, and 153.790  $kg \cdot ha^{-1} \cdot yr^{-1}$ , and those for *Q. mongolica* were 0.032, 15.068, 42.834, 21.219, 20.294, 20.237, 24.288, 5.647, and 119.134  $kg \cdot ha^{-1} \cdot yr^{-1}$ , respectively. Of the total throughfall flux (i.e., stemflow + throughfall flux) of the nine ions for the two species,  $SO_4^{2-}$  had the greatest total throughfall flux and  $H^+$  had the lowest. The net throughfall fluxes of the ions for the two species had various correlations with the precedent dry period, rainfall intensity, rainfall amount, and pH of precipitation. The soil pH under the *Q. mongolica* canopy (4.88) was higher than that under the *P. densiflora* canopy (4.34). The difference in the soil pH between the two stands was significant ( $P < 0.01$ ), but the difference in soil pH by the distance from the stems of the two species was not ( $P > 0.01$ ). This study shows the enrichments of inorganic nutrients by two representative urban forests in temperate regions and the roles of urban forests during rainfall events in a year.

**Key words** : Inorganic nutrient, Net throughfall flux, pH, *Pinus densiflora*, Precedent dry period, *Quercus mongolica*, Rainfall intensity

### 1. Introduction

Precipitation supplies forest ecosystems with water and nutrients, and mediates nutrient cycles. The forest water balance is important to support the production, hydrology, and climate at regional scales (Esser and Overdieck, 1991). Under sustained precipitation, the

water-holding capacity of trees decreases and water is divided into stemflow, which drains through the crown and down the stem, throughfall, which passes through the canopy to the soil, and interception loss, which evaporates. Each route has a critical role in the water cycle in forests, and the distribution of water among the routes is affected by the forest canopy

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architecture (Parker, 1983; Pathak et al., 1985). Nutrient cycles are strongly associated with the hydrological cycle, and nutrient inputs and outputs are directly related to the amount of water that moves into and out of forest ecosystems (Bormann and Likens, 1967). For example, a study of nutrient transport from fallen dead leaves and rainfall revealed that rainfall had a 11 - 46% influence on the forest nutrient cycle (Mehra et al., 1985).

Nutrient inputs via precipitation include external inputs (e.g., atmospheric deposition) and internal inputs (e.g., leaching from plants) (Ivens, 1990). Therefore, stemflow and throughfall have greater nutrient contents than precipitation (Gosz, 1980) and fluctuate according to canopy conditions (Alenas and Skarby, 1988).

Atmospheric deposition, which is affected by natural and anthropogenic influences, can be subdivided into dry and wet deposition (Bidleman, 1988; Howells, 1990). The rough surface of trees is advantageous for dry deposition, which can be estimated to chemically analyze throughfall (Lindberg et al., 1986; Ivens, 1990; Lindberg and Lovett, 1992). Moreover, coniferous forests tend to capture more dry deposition than deciduous forests because of their durable canopies and wide leaf surface area (Reuss and Johnson, 1986).

Rainfall moves through branches and stems, interacting with plant surfaces and transporting minerals to soil, thereby changing the soil chemical characteristics (Parker, 1983). Both inner ions from plants and atmospheric fallout undergo such chemical eluviation on leaves via rainfall (Miller et al., 1975). Meanwhile, stemflow partially affects water and nutrients near the stem, which influences vegetation distribution (Zinke, 1962; Gersper and Holowaychuk, 1970; Falkengren-Grerup, 1989; Hazlett and Foster, 1989; Andersson, 1991).

In Korea, stemflow and throughfall have been measured in coniferous *Pinus taeda* forests and

deciduous forests predominated by *Alnus hirsuta* and *Quercus mongolica* (Kim and Woo, 1988). Additionally, runoff loss relative to total rainfall has been researched in forests of *Pinus densiflora* and *P. taeda* (Lee, 1992). Finally, scholars have investigated the mineral nutrient cycles of nitrogen, phosphorus, and potassium (Kim and Kwak, 1992), as well as the amounts of ammonium nitrogen and nitrate nitrogen in stemflow and throughfall (Seo, 1988).

As an extension of existing research, our first study objective is to quantify the inputs of nine inorganic nutrients into soil via stemflow and throughfall in *P. densiflora* and *Q. mongolica* trees in an urban forest of the representative temperate region. And second study objective is to show the roles of urban forest hydrology in urban forest ecosystem.

## 2. Methodology

### 2.1. Study sites

The two study sites were located on Gwanaksan Mountain, Seoul, South Korea (latitude: 37°26'N - 37°30'N, longitude: 126°56'E - 126°57'E) (Fig. 1). The slope, aspect and elevations of study site A and study site B were 2°, north-west, 260 m and 16°, north-east, 215 m, respectively. Study site A was predominated by *P. densiflora* (61% cover), with 51 individuals in a 20 × 20-m study plot, and *Q. mongolica* (56% cover), with 58 individuals in a 20 × 20-m study plot.

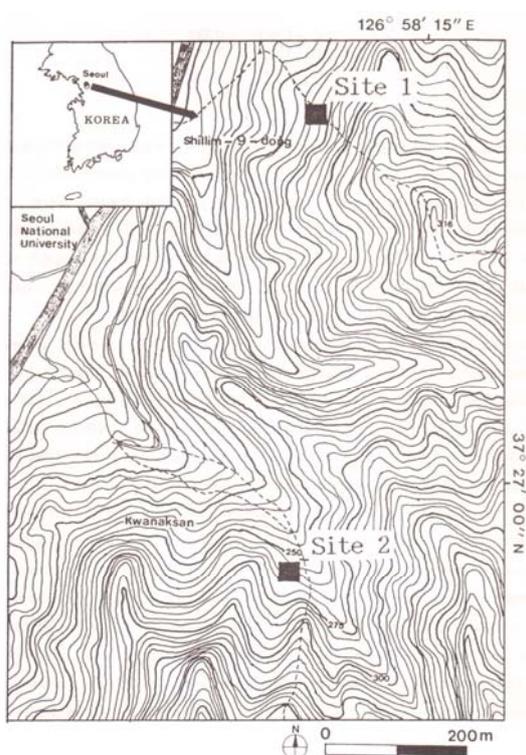
The plant vegetation of site A included *P. densiflora* in the tree layer, *Robinia pseudoacacia*, *Pinus koraiensis*, *A. hirsute*, *Pinus rigida*, *Sorbus alnifolia*, *Juniperus rigida*, *Rhus trichocarpa*, *Rhododendron schlippenbachii*, *Q. mongolica*, *Quercus serrata*, *Lindera obtusiloba*, *Zanthoxylum piperitum*, and *Lespedeza bicolor* in the shrub layer, and *Rubus crataegifolius*, *Spodiopogon cotulifer*, and *Carex lanceolata* in the herbaceous layer.

The plant vegetation of site B included *P. densiflora*

in the tree layer, *P. densiflora*, *S. alnifolia*, *Acer pseudosieboldianum*, *Rhododendron mucronulatum*, *Symplocos chinensis* f. *pilosa*, *stephanandra incisa*, *R. trichocarpa*, and *J. rigida* in the shrub layer, and *Pteridium aquilinum* var. *latiusculum*, *C. lanceolata*, *Artemisia keiskeana*, *Carex siderosticha*, *Viola mandshurica*, *Smilax sieboldii*, *Polygonatum odoratum* var. *pluriflorum*, and *Liparis kumokiri* in the herbaceous layer.

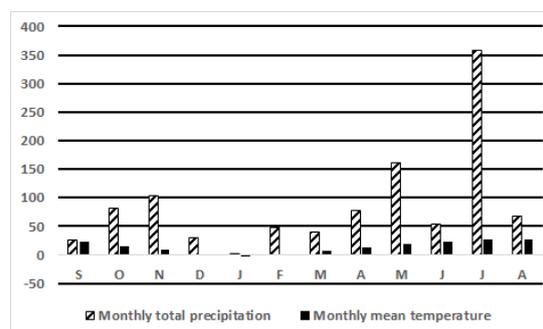
9.5 cm) and the stem diameter of *Q. mongolica* at site B had a maximum distribution at 0 - 2 cm (mean stem diameter: 6.9 cm). Stemflow and throughfall samplers were placed throughout the study plots.

The average annual temperature of Seoul during study days was 13.6°C (±10.8). The annual total precipitation of Seoul during study days was approximately 1047 mm (Fig. 2).



**Fig. 1.** Locations of *Pinus densiflora* stand (site 1) and *Quercus mongolica* stand (site 2).

To determine the stem thickness of the dominant vegetation (*P. densiflora* and *Q. mongolica*), the frequency distribution of the stem diameter at 120 cm above ground level was determined, and the diameter classes were grouped in 2.0-cm increments. The stem diameter of *P. densiflora* at site A had a maximum distribution at 4 - 6 and 6 - 8 cm (mean stem diameter:



**Fig. 2.** Monthly variation of amount of total precipitation (mm) and mean temperature (°C) during September 2015 to August 2016. The data are based on the climate data of meteorological auxiliary station in Seoul.

## 2.2. Precipitation water sampling

Rainfall, defined as precipitation above the canopy or in open sites (hereafter unintercepted precipitation), stemflow, and throughfall in the *P. densiflora* and *Q. mongolica* plots were sampled within 24 h of rainfall or snowfall from September 2015 to August 2016 (Stephen and Wigington, 1987).

### 2.2.1. Water sampling of unintercepted precipitation

A 30-L rain gauge with a 21.7-cm-diameter funnel (funnel cross-section area: 369.84 cm<sup>2</sup>) was fixed to a 1.5-m support stand on the rooftop of the biology building at Seoul National University. Water precipitation outside the forest was sampled in a cylinder (So and Lee, 1986). For the chemical analysis, precipitation was treated with 3 N hydrochloric acid, filtered through filter paper (Whatman No. 44) from a plastic bottle into a 220-mL

polyethylene bottle, and stored at 4 °C.

## 2.2.2. Water sampling of precipitation in trees

### 2.2.2.1. Stemflow sampling

Each stemflow collector was cleaned by soaking in hydrochloric acid for 24 h and glued with adhesive to the bark of a tree, which had been cut, wrapped with a rubber plate, and washed with distilled water (thickness: 6 cm; width: 7 cm). A piece of stainless steel was used to stabilize the rubber plate. An acrylic tube was placed between the bark and rubber plate and tilted to allow water to flow. The acrylic tube was connected to a plastic tube that drained into a 100-L plastic bottle. The apparatus was tested for leaks, and water gauges were attached to the plastic bottle and hose in July and August to prevent overflow. The water gauge accuracy was measured based on a comparative analysis of water flow measurements. Stemflow collectors were set up at diameter at breast height (DBH) on seven *P. densiflora* and seven *Q. mongolica* trees. Then, 100 L of water collected in the stemflow bottles was mixed ten times and transferred to a 220-mL polyethylene bottle that had been cleaned with hydrochloric acid. The remaining volume was measured using a cylinder. The acrylic tube entrance of the sampler was covered with nylon mesh (mesh size: 0.25 mm) to prevent leaf contamination. After each sample collection, leaves were removed from the rubber plate of the stemflow sampler. The remaining volume of stemflow was used to clean the sampler and the plastic bottle (Jorge and Lugo, 1985). Each 220-mL polyethylene bottle was transported to the lab, filtered through filter paper (Whatman No. 44), and stored at 4 °C.

### 2.2.2.2. Throughfall sampling

Throughfall was collected in a 20-L bottle connected to a plastic cone placed 1 m from the tree stem. To exclude water from the soil and nearby trees, the cone was positioned at a height of 60 cm (Feller, 1977). The cone was covered with nylon mesh (mesh

size: 0.25 mm) to exclude leaves and insects. To prevent algal and microorganism growth, the outer cone and tube were colored black. The throughfall collectors were set up near ten *P. densiflora* and ten *Q. mongolica* trees. Shrubs and bushes were removed within 7 m of the throughfall collectors. After each sample collection, newly grown shrubs and bushes were removed. During sampling, the throughfall volume was measured and 200 mL was transferred to a polyethylene bottle and stored at 4 °C for subsequent chemical analysis.

## 2.3. Precipitation measurements

### 2.3.1. Unintercepted precipitation

Unintercepted precipitation per area was measured after every rain event using a sampling funnel and converted into the volume per site area to determine the precipitation inflow into the forest.

### 2.3.2. Precipitation measurement in forests

#### 2.3.2.1. Stemflow

The total stemflow in forests is proportional to the density of trees and basal area at breast height (Killingbeck and Wali, 1978). The following formula was devised based on these two variables and the unintercepted precipitation in the projected area.

$$S = 1/2 \times [(D_1 + D_2) / D_1 + (B_1 + B_2) / B_1] \times V_c \quad (1)$$

$D_1$ : total tree density (No./m<sup>2</sup>)

$D_2$ : tree density of non-stemflow collection trees (No./m<sup>2</sup>)

$B_1$ : total tree basal area (m<sup>2</sup>/plot)

$B_2$ : basal area of non-stemflow collection trees (m<sup>2</sup>/plot)

$V_c$ : volume of stemflow collected (L/plot)

$S$ : stemflow (L/plot)

#### 2.3.2.2. Throughfall

Throughfall can be determined from the average throughfall measured with or without a water pipe

(Pathak et al., 1985).

$$T = (A_{wc} \times R + A_{uc} \times Ri) / A \quad (2)$$

$A_{wc}$ : area without overhead canopy ( $m^2$ )

R: unintercepted rainfall (mL)

$A_{uc}$ : area under the canopy ( $m^2$ )

$Ri$ : throughfall under the canopy (mL)

A: plot area ( $m^2$ )

T: throughfall (mL)

#### 2.4. Chemical analysis of precipitation

We measured the pH of unintercepted precipitation, stemflow, and throughfall filtered through filter paper using a suction pump with a pH meter (model 230A; Fisher) standardized with pH 4 and 7 buffer solutions. Conductivity was measured using a conductivity meter (DM 35; Kramer Electronics), and  $NH_4^+$  was assayed colorimetrically with phenate and measured with a spectrophotometer (model 24; Beckman Instruments) at 640 nm (APHA, 1989). For  $NO_3^-$ , samples were diluted by one-half, one-fifth, or one-tenth according to the concentration of the sample, and  $NO_3^-$  was detected colorimetrically with hydrazine using a reduction method and measured with a spectrophotometer (model 24; Beckman Instruments) at 540 nm (Cho et al., 1991).

We measured  $SO_4^{2-}$  turbidity using a spectrophotometer (model 24; Beckman Instruments) at 420 nm after precipitation with  $BaCl_2$  (APHA, 1989). To block interference by  $K^+$  and  $Na^+$ ,  $CsCl_2$  was added in an appropriate quantity based on atomic absorption spectrophotometry (AAS; model 901; GBC Scientific Equipment). To block interference by  $Mg^{2+}$  and  $Ca^{2+}$ ,  $LaCl_2$  was added in an appropriate quantity based on AAS (Moore and Chapman 1986).  $Cl^-$  was measured using an ion analyzer with an ion-selective electrode (Model No 94-17; Orion Research Inc.) (Allen et al., 1974; Orion Research Inc., 1986). Because of freezing,

the samples from January and February were pooled and the monthly average was used.

#### 2.5. Mineral nutrient concentrations

Unintercepted precipitation, stemflow, and throughfall in quadrats were multiplied by the amount of nutrients per volume to determine the mineral nutrients per area (kg/ha).

#### 2.6. Soil pH measurement

In both *P. densiflora* and *Q. mongolica* plots, soil samples were collected near two trees located on flat ground and isolated from other trees. At each tree, one sample was collected where the stem of the tree met the ground, and the other samples were collected at 10-cm increments horizontally from the first sample. Soil was sampled using a soil sleeve (diameter: 4.5 cm) at a depth of 0–5 cm after leaf removal. To reduce errors related to differences in the soils at both sites, soil was also collected at a distance of 150 cm from each *P. densiflora* tree and 130 cm from each *Q. mongolica* tree. The sampled soil was transported to the lab and dried in the shade for one week. The dried soil was filtered through a 2-mm sieve, mixed with distilled water at a 1:5 ratio of soil:distilled water for 30 min, filtered through filter paper (Whatman No. 44), and measured using a pH meter (model 230A; Fisher) standardized with pH 4 and 7 buffer solutions.

#### 2.7. Statistical analysis

We performed a correlation analysis of the unintercepted precipitation, stemflow from the *P. densiflora* and *Q. mongolica* stands, and ion concentration, and completed a stepwise regression analysis of the net throughfall (NTF) input with five precipitation characteristics and a soil pH comparison by distance from the stem using a two-way analysis of variance (ANOVA) with no repetitions due to differences among trees. The

**Table 1.** Volume-weighted mean concentrations of ions in precipitation from September 1, 2015 to August 31, 2016

	Ion	mg/L	μeq/L
Cation	H <sup>+</sup>	0.02	17 ( 3%)
	NH <sub>4</sub> <sup>+</sup>	1.51	84 (15%)
	Ca <sup>2+</sup>	4.23	211(37%)
	Mg <sup>2+</sup>	1.92	158(27%)
	K <sup>+</sup>	1.41	36 (6%)
	Na <sup>+</sup>	1.59	69 (12%)
	Sum		
Anion	SO <sub>4</sub> <sup>2-</sup>	12.95	270 (79%)
	NO <sub>3</sub> <sup>-</sup>	0.54	9 ( 3%)
	Cl <sup>-</sup>	2.24	63 (18%)
Sum			-342 (100%)

analyses were performed using SAS software ver. 9.4 (SAS Institute Inc., 2015).

### 3. Results

#### 3.1. Mineral nutrient inflow

##### 3.1.1. Unintercepted precipitation

The pH of precipitation ranged from 4.2 to 7.13, and the H<sup>+</sup> concentration was at least 0.074–63 μg·L<sup>-1</sup>. From the frequency distribution of 41 precipitation events divided into pH increments of 0.2, the maximum number of precipitation events (11) was distributed within a pH of 4.9–5.2. We found that pH values of 4.1–5.2 and 5.2–7.2 accounted for 76% and 24%, respectively and rains of < 5.2 pH were predominant.

The monthly difference in H<sup>+</sup> based on the amount of precipitation was high in May–July, low in August–November (9 μmho), and increased to an average of 31 μmho in September–December.

The volume-weighted mean concentration of positive ions in 2015 was 211 (37%), 158 (27%), 84 (15%), 69 (12%), 36 (6%), and 17 (3%) μeq·L<sup>-1</sup> for Ca<sup>2+</sup>, Mg<sup>2+</sup>, NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, and H<sup>+</sup>, respectively, where Ca<sup>2+</sup> and Mg<sup>2+</sup> accounted for a total of 64% of positive ions. The volume-weighted

mean concentration of the negative ions SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, and NO<sub>3</sub><sup>-</sup> was 270 (79%), 63 (18%), and 9 (3%) μeq·L<sup>-1</sup>, respectively. The concentrations of both positive and negative ions followed the order SO<sub>4</sub><sup>2-</sup> > Ca<sup>2+</sup> > Cl<sup>-</sup> > Mg<sup>2+</sup> > Na<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > K<sup>+</sup> > NO<sub>3</sub><sup>-</sup> > H<sup>+</sup>. Positive ion concentrations followed the order Ca<sup>2+</sup> > Mg<sup>2+</sup> > Na<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > K<sup>+</sup> > H<sup>+</sup>, and negative ion concentrations followed the order SO<sub>4</sub><sup>2-</sup> > Cl<sup>-</sup> > NO<sub>3</sub><sup>-</sup> (Table 1).

The volume-weighted mean concentrations of the nine ions in precipitation revealed correlations between Ca<sup>2+</sup> and Mg<sup>2+</sup>, NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup>, K<sup>+</sup> and SO<sub>4</sub><sup>2-</sup>, and NH<sub>4</sub><sup>+</sup> and Cl<sup>-</sup> (P < 0.01), as well as between H<sup>+</sup> and Mg<sup>2+</sup>, NH<sub>4</sub><sup>+</sup> and K<sup>+</sup>, Mg<sup>2+</sup> and Na<sup>+</sup>, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup> and Cl<sup>-</sup> (P < 0.05).

##### 3.1.2. Forest precipitation ion contents

Annual stemflow at the *Pinus densiflora* and *Quercus mongolica* stand was 145×10<sup>3</sup> L/ha and 430×10<sup>3</sup> L/ha (Fig. 3). *Pinus densiflora* stemflow transported 0.042 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of H<sup>+</sup>, 21.518 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of NH<sub>4</sub><sup>+</sup>, 52.207 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Ca<sup>2+</sup>, 27.694 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Mg<sup>2+</sup>, 20.060 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of K<sup>+</sup>, 24.049 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Na<sup>+</sup>, 153.79 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of SO<sub>4</sub><sup>2-</sup>, 10.241 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of NO<sub>3</sub><sup>-</sup>, and 37.229 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Cl<sup>-</sup> (Table 2). The total annual inflow of positive ions followed the order Ca<sup>2+</sup> > Mg<sup>2+</sup> > NH<sub>4</sub><sup>+</sup> > Na<sup>+</sup> > K<sup>+</sup> >

H<sup>+</sup> and that of negative ions followed the order SO<sub>4</sub><sup>2-</sup> > Cl<sup>-</sup> > NO<sub>3</sub><sup>-</sup>.

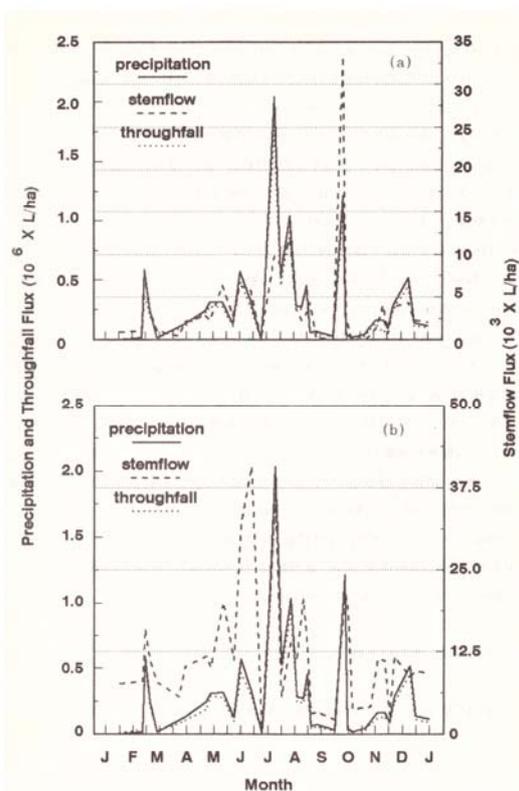


Fig. 3. Precipitation, throughfall and stemflow flux in *Pinus densiflora* stand (a) and *Quercus mongolica* stand (b).

Based on the volume-weighted mean concentration of ions in *P. densiflora* stemflow, SO<sub>4</sub><sup>2-</sup> had the highest concentration (110.9 mg·L<sup>-1</sup>) and H<sup>+</sup> had the lowest concentration (0.09 mg·L<sup>-1</sup>). Overall, the concentrations of ions followed the order SO<sub>4</sub><sup>2-</sup> > Ca<sup>2+</sup> > Cl<sup>-</sup> > Mg<sup>2+</sup> > NH<sub>4</sub><sup>+</sup> > Na<sup>+</sup> > K<sup>+</sup> > NO<sub>3</sub><sup>-</sup> > H<sup>+</sup>. The highest and lowest maximum volume-weighted mean concentrations were observed for SO<sub>4</sub><sup>2-</sup> (321.34 mg·L<sup>-1</sup>) and H<sup>+</sup> (0.3 mg·L<sup>-1</sup>), respectively. The maximum values followed the order SO<sub>4</sub><sup>2-</sup> > Cl<sup>-</sup> > Ca<sup>2+</sup> > Mg<sup>2+</sup> > NH<sub>4</sub><sup>+</sup> > K<sup>+</sup> > Na<sup>+</sup> > NO<sub>3</sub><sup>-</sup> > H<sup>+</sup>, while the minimum values followed the order SO<sub>4</sub><sup>2-</sup> > Ca<sup>2+</sup> > Na<sup>+</sup>

> K<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > Mg<sup>2+</sup> > Cl<sup>-</sup> > NO<sub>3</sub><sup>-</sup> > H<sup>+</sup>. The greatest difference between the minimum and maximum was observed for SO<sub>4</sub><sup>2-</sup> (313.79 mg·L<sup>-1</sup>), while H<sup>+</sup> had the least difference (0.29 mg·L<sup>-1</sup>).

*Quercus mongolica* stemflow transported 0.03 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of H<sup>+</sup>, 15.07 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of NH<sub>4</sub><sup>+</sup>, 42.83 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Ca<sup>2+</sup>, 21.22 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Mg<sup>2+</sup>, 20.29 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of K<sup>+</sup>, 20.24 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Na<sup>+</sup>, 119.13 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of SO<sub>4</sub><sup>2-</sup>, 5.65 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of NO<sub>3</sub><sup>-</sup>, and 24.29 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Cl<sup>-</sup> (Table 3). The total inflow of positive ions followed the order K<sup>+</sup> > Ca<sup>2+</sup> > Mg<sup>2+</sup> > Na<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > H<sup>+</sup> and that of negative ions followed the order SO<sub>4</sub><sup>2-</sup> > Cl<sup>-</sup> > NO<sub>3</sub><sup>-</sup>.

The maximum and minimum volume-weighted mean concentrations of ions in *Q. Mongolica* stemflow were observed for SO<sub>4</sub><sup>2-</sup> (36.78 mg·L<sup>-1</sup>) and H<sup>+</sup> (0.02 mg·L<sup>-1</sup>), respectively. Overall, the volume-weighted mean concentration of ions followed the order SO<sub>4</sub><sup>2-</sup> > K<sup>+</sup> > Mg<sup>2+</sup> > Cl<sup>-</sup> > Ca<sup>2+</sup> > Na<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > NO<sub>3</sub><sup>-</sup> > H<sup>+</sup>. The highest and lowest maximum volume-weighted mean concentrations were observed for SO<sub>4</sub><sup>2-</sup> (126.91 mg·L<sup>-1</sup>) and H<sup>+</sup> (0.13 mg·L<sup>-1</sup>), respectively. Overall, the maximum volume-weighted mean concentration followed the order SO<sub>4</sub><sup>2-</sup> > K<sup>+</sup> > Cl<sup>-</sup> > Mg<sup>2+</sup> > Ca<sup>2+</sup> > NH<sub>4</sub><sup>+</sup> > Na<sup>+</sup> > NO<sub>3</sub><sup>-</sup> > H<sup>+</sup> and the minimum volume-weighted mean concentration followed the order K<sup>+</sup> > SO<sub>4</sub><sup>2-</sup> > Na<sup>+</sup> > Ca<sup>2+</sup> = Cl<sup>-</sup> > NH<sub>4</sub><sup>+</sup> > NO<sub>3</sub><sup>-</sup> > Mg<sup>2+</sup> > H<sup>+</sup>. The greatest difference between the minimum and maximum was observed for SO<sub>4</sub><sup>2-</sup> (125.58 mg·L<sup>-1</sup>), while H<sup>+</sup> had the least difference (0.13 mg·L<sup>-1</sup>).

The total inflow of H<sup>+</sup> via stemflow was nearly the same for *P. densiflora* and *Q. mongolica*. For *Q. mongolica*, the total inflows of SO<sub>4</sub><sup>2-</sup> (4.406 kg·ha<sup>-1</sup>·yr<sup>-1</sup>), NO<sub>3</sub><sup>-</sup> (0.158 kg·ha<sup>-1</sup>·yr<sup>-1</sup>), Ca<sup>2+</sup> (0.777 kg·ha<sup>-1</sup>·yr<sup>-1</sup>), Mg<sup>2+</sup> (1.373 kg·ha<sup>-1</sup>·yr<sup>-1</sup>), K<sup>+</sup> (2.465 kg·ha<sup>-1</sup>·yr<sup>-1</sup>), and Na<sup>+</sup> (0.747 kg·ha<sup>-1</sup>·yr<sup>-1</sup>) were greater than those for *P. densiflora*. For *P. densiflora*, the total inflows of NH<sub>4</sub><sup>+</sup> (0.1 kg·ha<sup>-1</sup>·yr<sup>-1</sup>) and Cl<sup>-</sup> (0.898 kg·ha<sup>-1</sup>·yr<sup>-1</sup>) were greater than those for *Q. mongolica*

**Table 2.** Annual inputs (kg/ha/yr) of 9 major ions in precipitation, stemflow, and throughfall at *Pinus densifolia* stand

	H <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	K <sup>+</sup>	Na <sup>+</sup>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>
Precipitation	0.17	15.12	42.23	19.22	14.05	15.89	129.44	5.43	22.39
Stemflow	0.04	21.52	52.21	27.69	20.06	24.05	153.79	10.24	37.23
Throughfall	0.01	0.78	1.65	1.04	0.48	0.65	9.10	0.28	1.51

**Table 3.** Annual inputs (kg/ha/yr) of 9 major ions in precipitation, stemflow, and throughfall at *Quercus mongolica* stand

	H <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	K <sup>+</sup>	Na <sup>+</sup>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>
Precipitation	0.17	15.12	42.23	19.22	14.05	15.89	129.44	5.43	22.39
Stemflow	0.03	15.07	42.83	21.22	20.29	20.24	119.13	5.65	24.29
Throughfall	0.01	0.68	2.43	2.42	2.94	1.40	13.50	0.44	2.41

(Tables 2, 3). In both *P. densiflora* and *Q. mongolica*, SO<sub>4</sub><sup>2-</sup> had the maximum inflow and H<sup>+</sup> had the minimum.

The volume-weighted mean concentration of SO<sub>4</sub><sup>2-</sup> in *P. densiflora* stemflow was 110.94 mg·L<sup>-1</sup>, and that in *Q. mongolica* stemflow was 36.78 mg·L<sup>-1</sup>, about three times lower than the concentration in *P. densiflora* stemflow. The volume-weighted mean concentrations of NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, Cl<sup>-</sup>, and H<sup>+</sup> were 2.12, 4.25, 3.37, 1.88, 1.91, 2.59, and 3.91 times higher in *P. densiflora* stemflow than in *Q. mongolica*, respectively. Meanwhile, that of K<sup>+</sup> was 1.61 times higher in *Q. mongolica* stemflow than in *P. densiflora*.

Annual throughfall at the *Pinus densiflora* and *Quercus mongolica* stand was 9,587×10<sup>3</sup> L/ha and 9,687×10<sup>3</sup> L/ha (Fig. 3). *Pinus densiflora* throughfall transported 0.008 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of H<sup>+</sup>, 0.784 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of NH<sub>4</sub><sup>+</sup>, 1.652 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Ca<sup>2+</sup>, 1.044 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Mg<sup>2+</sup>, 0.476 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of K<sup>+</sup>, 0.651 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Na<sup>+</sup>, 9.098 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of SO<sub>4</sub><sup>2-</sup>, 0.278 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of NO<sub>3</sub><sup>-</sup>, and 1.509 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Cl<sup>-</sup> (Table 2). The inflow of ions followed the order SO<sub>4</sub><sup>2-</sup> > Ca<sup>2+</sup> > Cl<sup>-</sup> > Mg<sup>2+</sup> > Na<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > K<sup>+</sup> > NO<sub>3</sub><sup>-</sup> > H<sup>+</sup>.

*Quercus mongolica* throughfall contained 0.008 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of H<sup>+</sup>, 0.684 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of NH<sub>4</sub><sup>+</sup>, 2.429 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Ca<sup>2+</sup>, 2.417 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Mg<sup>2+</sup>, 2.941

kg·ha<sup>-1</sup>·yr<sup>-1</sup> of K<sup>+</sup>, 1.398 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Na<sup>+</sup>, 13.504 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of SO<sub>4</sub><sup>2-</sup>, 0.436 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of NO<sub>3</sub><sup>-</sup>, and 2.407 kg·ha<sup>-1</sup>·yr<sup>-1</sup> of Cl<sup>-</sup> (Table 3). The ion inflows followed the order SO<sub>4</sub><sup>2-</sup> > Ca<sup>2+</sup> > Cl<sup>-</sup> > Mg<sup>2+</sup> > K<sup>+</sup> > Na<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > NO<sub>3</sub><sup>-</sup> > H<sup>+</sup> (Table 3).

The sums of the ions inflowing via stemflow and throughfall differed significantly between *Q. mongolica* and *P. densiflora* (P < 0.05; Tables 2, 3). With the exception of K<sup>+</sup>, all ion inflows were higher in *P. densiflora* than in *Q. mongolica*. Ion inflows via throughfall divided by the inflow from rainfall (i.e., the enrichment ratio) revealed that NO<sub>3</sub><sup>-</sup> had the highest enrichment ratio (1.89) and H<sup>+</sup> had the lowest (0.25) in *P. densiflora* (Tables 2, 3). The enrichment ratios of the other ions followed the order Cl<sup>-</sup> > Na<sup>+</sup> > Mg<sup>2+</sup> > K<sup>+</sup> > NH<sub>4</sub><sup>+</sup> > Ca<sup>2+</sup> > SO<sub>4</sub><sup>2-</sup> (Fig. 4). In *Q. mongolica*, K<sup>+</sup> had the highest enrichment ratio (1.44) and H<sup>+</sup> the lowest (0.19), while the other ions followed the order Na<sup>+</sup> > Mg<sup>2+</sup> > Cl<sup>-</sup> > NO<sub>3</sub><sup>-</sup> > Ca<sup>2+</sup> > NH<sub>4</sub><sup>+</sup> > SO<sub>4</sub><sup>2-</sup> (Fig. 4).

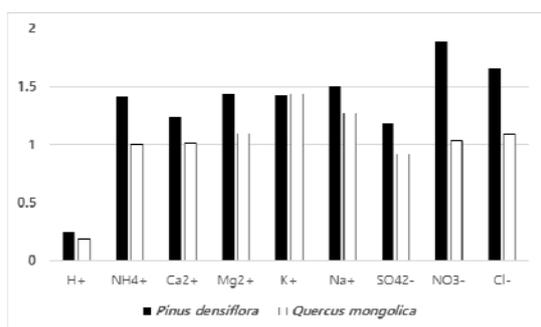
Comparison of the pH of rainfall and *Q. mongolica* and *P. densiflora* throughfall revealed that the pH of the rainwater increased from rainfall to throughfall. For *P. densiflora*, when rainfall had a pH of 4.20 - 5.71, the pH of the rainwater increased in throughfall; however, when the pH of rainfall was above 5.71, the pH of the rainwater decreased in throughfall. For *Q. mongolica*, when the pH of rainfall 4.20 and 5.50, the

**Table 4.** Result of stepwise regression between net throughfall flux of 9 ions and 5 environmental variables at *Pinus* stand and *Quercus* stand. All regressions were based on data from 10-29 rainfall events

Ion	<i>Pinus</i> stand (RE; R)	<i>Quercus</i> stand (RE; R)
NH <sub>4</sub> <sup>+</sup>	12.97-1.91×pH-0.75×INT; 0.54**	0.06+0.01×PD; 0.62
Ca <sup>2+</sup>	8.44-2.11×INT; 0.41*	None
Mg <sup>2+</sup>	9.86-0.23×pH-0.03PRE; 0.70**	2.21+0.02×PD-0.05×PRE; 0.64**
K <sup>+</sup>	3.48-0.02×PRE; 0.28	6.83-3.40×INT; 0.50*
Na <sup>+</sup>	2.60+0.02×PD-0.05×PRE; 0.71**	1.89-0.01×PRE; 0.45
SO <sub>4</sub> <sup>2-</sup>	14.38+0.11×PD-0.61×PRE+7.90×INT; 0.72**	-7.78+7.78×pH; 0.53
NO <sub>3</sub> <sup>-</sup>	3.24-0.03×PRE; 0.41	None
Cl <sup>-</sup>	3.30+0.03×PD; 0.48*	1.49-0.46×INT; 0.4

RE; regression equation, R; Square of coefficient of determination, PRE; amount of precipitation(mm), INT; intensity of rainfall (mm/hr), PD; precedent dry period, \* P < 0.05, \*\* P < 0.01.

pH of the rainwater typically increased in throughfall, and when the pH of rainfall was above 5.5, the pH of the rainwater decreased in throughfall.



**Fig. 4.** Enrichment ratio (the ratio of amount of ion input by throughfall to that by bulk precipitation) of 9 ions in *Pinus densiflora* stand and *Quercus mongolica* stand.

The correlation analysis of the volume-weighted mean ion concentrations in *P. densiflora* throughfall revealed the following correlations at a 1% significance level: NH<sub>4</sub><sup>+</sup> and Mg<sup>2+</sup>; Ca<sup>2+</sup> and Mg<sup>2+</sup>; K<sup>+</sup> and H<sup>+</sup>; Ca<sup>2+</sup> and K<sup>+</sup>; Mg<sup>2+</sup> and K<sup>+</sup>; Na<sup>+</sup> and H<sup>+</sup>; Na<sup>+</sup> and Ca<sup>2+</sup>; Na<sup>+</sup> and Mg<sup>2+</sup>; Na<sup>+</sup> and K<sup>+</sup>; SO<sub>4</sub><sup>2-</sup> and H<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>, and Na<sup>+</sup>; NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup>; and Cl<sup>-</sup> and Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>, Na<sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>. Additionally, the following correlations at the 5%

significance level were observed: Mg<sup>2+</sup> and H<sup>+</sup>; and NH<sub>4</sub><sup>+</sup> and K<sup>+</sup>, Na<sup>+</sup>, and SO<sub>4</sub><sup>2-</sup>. The following correlations were identified in *Q. mongolica* throughfall at the 1% significance level: Ca<sup>2+</sup> and H<sup>+</sup>, Mg<sup>2+</sup>, and SO<sub>4</sub><sup>2-</sup>; Mg<sup>2+</sup> and K<sup>+</sup>, Na<sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and Cl<sup>-</sup>; Na<sup>+</sup> and H<sup>+</sup>; NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup>; SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>; SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup>; and NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup>. Finally, the following correlations were identified in *Q. mongolica* throughfall at the 5% significance level: SO<sub>4</sub><sup>2-</sup> and H<sup>+</sup>; Ca<sup>2+</sup> and NO<sub>3</sub><sup>-</sup>; and Na<sup>+</sup> and Cl<sup>-</sup>.

### 3.2. Net throughfall input and rainfall

The net throughfall (NTF) input of the nine analyzed ions was defined as the difference between the ions in rainfall (mg·m<sup>-2</sup>) and those in throughfall (mg·m<sup>-2</sup>), and was set as a dependent variable. Five variables (precipitation, rainfall duration, rainfall intensity, rainfall pH, and dry days before rainfall or precipitation period) were set as explanatory variables for the stepwise regression analysis, where the best-suited model was chosen based on the correlation coefficients (Table 4). H<sup>+</sup> frequently had a negative NTF input due to absorption by *P. densiflora* (20/35 cases), and *Q. mongolica* (24/35 cases); thus, it was not included in the analysis. NH<sub>4</sub><sup>+</sup> was significantly correlated with pH and rainfall intensity in *P. densiflora* and with dry days before rainfall or

**Table 5.** Results of two-way analysis of variance for soil pH ratio along distance at intervals of 10 cm from a stem of *Pinus densiflora* and *Quercus mongolica*

Source	Soil pH ratio	
	Direction 1	Direction 2
Species	**	**
Distance	n. s.	n. s.

Direction 1 indicates the direction connected to soil which vertical groove of stem reached. Direction 2 indicates the direction perpendicular to direction 1. Ratio means soil pH of direction 1 or 2 to that of peripheral area.

\*\* :  $P < 0.01$ , n. s.: not significant

precipitation period in *Q. mongolica*.  $\text{Ca}^{2+}$  was correlated with rainfall intensity in *P. densiflora* but was not significantly correlated in *Q. mongolica*.  $\text{Mg}^{2+}$  was significantly correlated with rainfall pH in *P. densiflora*. In *Q. mongolica*, dry days before rainfall or precipitation period was correlated with precipitation.  $\text{K}^+$  in *P. densiflora* was correlated with rainfall intensity in *Q. mongolica*.  $\text{Na}^+$  was correlated with dry days before rainfall or precipitation period and precipitation in *P. densiflora* and with precipitation in *Q. mongolica*.  $\text{SO}_4^{2-}$  was correlated with dry days before rainfall or precipitation period, precipitation, and rainfall intensity in *P. densiflora* and with rainfall pH in *Q. mongolica*.  $\text{NO}_3^-$  was correlated with precipitation in *P. densiflora* but was not significantly correlated in *Q. mongolica*.  $\text{Cl}^-$  was correlated with dry days before rainfall or precipitation period in *P. densiflora* and with rainfall intensity in *Q. mongolica*. In *P. densiflora*,  $\text{NH}_4^+$  and  $\text{Mg}^{2+}$  were positively correlated with dry days before rainfall or precipitation period and negatively correlated with rainfall pH. Moreover, the regression coefficient of  $\text{NH}_4^+$  was 1.91, which was higher than that of  $\text{Mg}^{2+}$  (0.23).  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{SO}_4^{2-}$ , and  $\text{NO}_3^-$  were negatively correlated with the amount of precipitation, of which  $\text{SO}_4^{2-}$  had the highest regression coefficient (0.61). In *P. densiflora*,  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  were negatively correlated with rainfall intensity, where  $\text{NH}_4^+$  had a lower regression coefficient than  $\text{Ca}^{2+}$  (0.75 vs. 2.11, respectively). In *P. densiflora*, dry days before rainfall or precipitation

period and  $\text{Na}^+$ ,  $\text{SO}_4^{2-}$ , and  $\text{Cl}^-$  were positively correlated, where  $\text{SO}_4^{2-}$  had the highest regression coefficient (0.11). In *Q. mongolica*, the net inflow amounts of  $\text{NH}_4^+$  and  $\text{Mg}^{2+}$  via throughfall were positively correlated with dry days before rainfall or precipitation period, where the regression coefficient of  $\text{Mg}^{2+}$  was higher than that of  $\text{NH}_4^+$  (0.02 vs. 0.01, respectively).  $\text{Mg}^{2+}$  and  $\text{Na}^+$  were negatively correlated with the amount of precipitation.  $\text{SO}_4^{2-}$  and pH were positively correlated. Finally,  $\text{K}^+$  and  $\text{Cl}^-$  and rainfall intensity were negatively correlated.

### 3.3. Influence of *Quercus mongolica* and *Pinus densiflora* on soil pH

The soil pH at various distances from the two tested *P. densiflora* trees ranged from 3.99 to 6.05. The soil pH at distances from the trunk of 0 - 30, 40 - 70, 80 - 110, and > 150 cm averaged 4.32, 4.30, 4.32, and 4.40, respectively. Although pH appeared to increase slightly with distance, the average pH showed no linear regression with distance ( $R = 0.06$ ,  $n = 12$ ).

The soil pH at various distances from the two tested *Q. mongolica* trees ranged from 4.42 to 5.84. The soil pH at distances from the trunk of 0 - 30, 40 - 70, 80 - 110, and > 130 cm averaged 4.81, 4.85, 4.96, and 4.86, respectively. The average pH increased with distance ( $R = 0.34$ ,  $n = 13$ ).

The average soil pH near *Q. mongolica* and *P. densiflora* trees was 4.32 and 4.88, respectively. Based on a two-way ANOVA of the pH of the two *Q.*

*mongolica* and *P. densiflora* individuals by distance and species (the species differed in terms of soil and location, so the pH of the soil from these locations was divided by the pH of the soil collected at the farthest distance from the trees), soil pH was not associated with distance ( $P > 0.01$ ), although there was a significant difference between the two species ( $P < 0.01$ , Table 5). Based on Tukey's studentized range test of the pH by direction from the trees that exhibited differences between species, the soil pH of *Q. mongolica* was higher than that of *P. densiflora*. However, neither *Q. mongolica* nor *P. densiflora* had differences in soil pH in both directions ( $P > 0.01$ ).

#### 4. Discussion

##### 4.1. Precipitation, stemflow, throughfall, and interception loss change

Although fog and frost contributed to indirect precipitation during the study period, they were not considered in this study, and only direct precipitation was measured. Precipitation was concentrated in June, July, August, and September, consistent with the summer and fall rainy season in Korea. Ice inhibited the measurement of stemflow and throughfall in January and February, so the data for these months were averaged, and unintercepted precipitation was measured after ice melted. Precipitation likely differed somewhat due to the distance between the study sites (640 m). Moreover, precipitation type can differ significantly by location. Because plants, terrain, and wind could have introduced experimental errors when measuring forest precipitation, measurements were taken on the roof of the biodiversity institute 500 m from the *P. densiflora* site and 850 m from the *Q. mongolica* site.

Stemflow volume is influenced by tree size, branch angle, presence/absence of sempervirent leaves, tree smoothness, season, and precipitation amount (Falkengren-Grerup, 1989), as well as precipitation

duration, rainfall intensity, and tree density (Esser and Overdieck, 1991). Additionally, individual trees have unique branching patterns (Choi, 1984), and the angle between the main branch and twigs has been shown to affect stemflow in *Quercus petraea* and *Quercus cerris* (Jakucs, 1985). Regression analysis of *Q. mongolica* and *P. densiflora* stemflow with the cross-section at the upper trunk, DBH, cover, and tree height revealed that the cross-section at the upper trunk and stemflow followed a linear relationship. The proportion of stemflow to total precipitation in *P. densiflora* and *Q. mongolica* was 0.013 and 0.039, respectively. The higher proportion in *Q. mongolica* can be explained by the fact that the angles between the main branch and twigs in *Q. mongolica* are between 45° and 90°, which are greater than those of *P. densiflora*. Moreover, its bark is smooth and has vertical gaps, which act as waterways when precipitation exceeds the storage capacity, whereas *P. densiflora* has rough bark and no vertical gaps. Although the proportion of water from stemflow was lower than that of unintercepted precipitation, it has a major effect on the inflow of nutrients and water in soil (Parker, 1983). The gradients of the linear regressions of stemflow and rainfall were the same at all locations (within 0.01), so the rate of increase for stemflow was the same as the increase in rainfall. Meanwhile, the y-intercept was higher in *Q. mongolica*, indicating that the amount of stemflow was greater in *Q. mongolica* than in *P. densiflora*. For example, when rainfall was 0.46 mm, only stemflow occurred for *P. densiflora* trees with a diameter at the upper trunk lower than 11.46 cm, suggesting that *P. densiflora* intercepted all precipitation below 0.46 mm. In previous studies, no stemflow was measured when precipitation was below 2.2 or 11 mm, possibly due to the difference in the diameter at the upper trunk (Pathak et al., 1985; Kim and Woo, 1988). In this study, there were no instances when the measured *Q. mongolica* stemflow was zero, likely because rainfall events less than 16 mm were

excluded from the study. Assuming that the *Q. mongolica* forest density and diameter at the upper trunk area were the same as those of *P. densiflora*, the total annual stemflow was  $431.958 \text{ kL}\cdot\text{ha}^{-1}$ , which was  $1.64 \text{ kL}\cdot\text{ha}^{-1}$  higher than the value calculated without this assumption ( $430.320 \text{ kL}\cdot\text{ha}^{-1}$ ). Killingbeck and Wali (1978) speculated that two study sites had similar densities and diameters at the upper trunk area when the total stemflow did not differ significantly, even when the coefficient was the same.

We quantified throughfall by measuring the volume in the rainfall samplers in areas with and without crown (i.e., unintercepted precipitation) (Pathak et al., 1985). However, areas with no crown could be influenced by nearby crowns, which would result in underestimation. Additionally, the funnel gauges used in samplers had a lower surface area than the trough gauges used to measure throughfall, which could have contributed to measurement errors (Kostelnik et al., 1989). To reduce this spatial variability, we molded the funnel gauges to expand the surface area. Interference effects at the edge of the funnel gauge were also reduced, because circles have a greater relative surface area. The coefficient of variation is a measure of variation regardless of the average (Sokal and Rohlf, 1981); therefore, as throughfall increased, the coefficient of variation decreased. However, in *P. densiflora*, when throughfall exceeded 50 mm, the coefficient of variation did not show any changes. The coefficient of variation for *Q. mongolica* was 0.1 - 0.9, while that for *P. densiflora* was 0.1 - 1.2. The higher coefficient of variation in *Q. mongolica* could indicate that the stemflow samplers on *Q. mongolica* had a significant influence on the results. The total annual *P. densiflora* and *Q. mongolica* throughfall was 86.8% and 87.8% from unintercepted precipitation, respectively. For comparison, an area where *Rigitaeda* pine (*Pinus rigitaeda*) was the main species had a total annual throughfall of 76.7%, and that of an area predominated by *Q. mongolica* and teal tree was

81.8%. As rainfall accrued, the differences in the amount of throughfall among the locations were not significant (paired *t*-test,  $P < 0.01$ ). The gradient of the linear regression of rainfall and throughfall was greater for *P. densiflora* than for *Q. mongolica*, indicating that *P. densiflora* crown intercepted more rain, which could cause interception loss (i.e., the difference between throughfall and stemflow) to be greater in *P. densiflora* than in *Q. mongolica*.

#### 4.2. Ion transport in unintercepted precipitation

Kim et al. (1994) measured unintercepted precipitation and found an annual average pH of 4.2 - 4.8 and monthly average pH of 4.00 - 4.82. Moreover, pH was high in spring and summer but low in winter, and 0.5 and  $85.8 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  of  $\text{H}^+$  and  $\text{SO}_4^{2-}$ , respectively, were transported to the surface via rainfall (Kim et al., 1994).

The total inflows of  $\text{H}^+$  and  $\text{SO}_4^{2-}$  via rainfall were 0.17 and  $129.44 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ , respectively, where  $\text{H}^+$  was 0.34 times lower and  $\text{SO}_4^{2-}$  was 1.5 times higher than the results of Kim et al. (1994).  $\text{SO}_4^{2-}$  had the highest total ion transport via rainfall, followed by  $\text{Ca}^{2+}$  ( $42.227 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ), similar to experimental results involving holm oak (Roda et al., 1990). Overall, the rainfall at the study sites contained dissolved  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ , and  $\text{SO}_4^{2-}$  salts. Based on the volume-weighted mean concentration,  $\text{SO}_4^{2-}$  had the highest inflow ( $12.95 \text{ mg}\cdot\text{L}^{-1}$ ), followed by  $\text{Ca}^{2+}$  ( $4.23 \text{ mg}\cdot\text{L}^{-1}$ ). The volume equivalent value ( $\mu\text{eq}\cdot\text{L}^{-1}$ ), based on the sum of all positive and negative ions, revealed that the concentration of positive ions was  $233 \mu\text{eq}\cdot\text{L}^{-1}$  higher than that of negative ions, which were presumed to include  $\text{CO}_3^{2-}$ , organic anions, and  $\text{PO}_4^{2-}$  (Fahey et al., 1988). The concentration of  $\text{NO}_3^-$  was 0.03 times lower than that of  $\text{SO}_4^{2-}$ ; therefore, its contribution to  $\text{H}^+$  was considered to be small.  $\text{NO}_3^-$  transport by area had no relationship to the transport of  $\text{H}^+$  by area but was related to the transport of  $\text{SO}_4^{2-}$  by area.

The correlations between positive and negative ions

(i.e. omitting positive-positive and negative-negative correlations) revealed correlations between  $K^+$  and  $SO_4^{2-}$ ,  $NH_4^+$  and  $SO_4^{2-}$ ,  $Mg^{2+}$  and  $Cl^-$ , and  $Ca^{2+}$  and  $Cl^-$ ; therefore, these ions likely formed ionic bonds that dissolved in rain.  $(NH_4)_2SO_4$  particularly fits this trend: it is found mostly as an aerosol, has strong correlations, and is known to neutralize acidity (Summer and Whelpdalf, 1976). Although ion concentrations are typically high when rainfall is low (Madgwick and Ovington, 1959), ion concentration and rainfall amount were not negatively correlated; however, they could be considered to be related because the total amount of ions transported is the product of the average ion concentration multiplied by the rainfall amount (Likens et al., 1980). Therefore, in summer, when Korea is subject to substantial rainfall, the average ion concentration was low, but ion transport was high (Veneklaas, 1990).

#### 4.3. Comparison of ion inflow between rainfall and *Quercus mongolica* and *Pinus densiflora* stands

Assuming that the ion inflow via rainfall was the same in the *Q. mongolica* and *P. densiflora* stands to study throughfall and stemflow, we collected rainfall samples at a site 500 and 800 m from sites A and B, respectively. The total annual ion inflows into the soil via stemflow and throughfall were higher in *P. densiflora* than *Q. mongolica*, except for  $K^+$  (Table 2). Meanwhile, the ratio of  $K^+$  supply was higher in *Q. mongolica* (Fig. 4); therefore, leaching and dry or wet deposition of  $K^+$  were greater in *Q. mongolica* than *P. densiflora*. Ultimately, the throughfall at each stand did not differ substantially from the rainfall ( $P < 0.01$ ), and the ion inflow via the crown in *P. densiflora* was higher than that in *Q. mongolica*. Because more dry deposition occurred in the *P. densiflora* stand than in the *Q. mongolica* stand, and defoliation occurs more often in *P. densiflora*, we assumed that *P. densiflora* would transport more ions to the soil. Six major factors influenced throughfall and stemflow: evaporation rate,

dry deposition rate, wet deposition rate, plantation activity, leaf leaching, and leaf absorption. Among these, dry deposition, plantation activity, and leaching and absorption in leaves differ among species, and are thought to affect the spatial distribution of ions.

From the correlation analysis, no correlation between the average  $SO_4^{2-}$  concentration with  $Ca^{2+}$  or  $Mg^{2+}$  was observed in rainfall for *P. densiflora*, indicating that  $Ca^{2+}$  and  $Mg^{2+}$  were leached or distributed into throughfall via dry deposition. Moreover, the correlations between  $NO_3^-$  with  $Ca^{2+}$  and  $Mg^{2+}$  indicated that  $NO_3^-$  was transported via throughfall. *Q. mongolica* throughfall exhibited different characteristics from rainfall, where  $SO_4^{2-}$  and  $Na^+$ ,  $Ca^{2+}$ , and  $Mg^{2+}$  and  $NO_3^-$  and  $Ca^{2+}$ ,  $Mg^{2+}$  and  $Cl^-$ ,  $K^+$  and  $Na^+$  were correlated.

#### 4.4. Soil pH in the area of influence of stemflow and throughfall

Gwanaksan Mountain is composed of granite, and the soil is typically shallow with undeveloped A, B, and C horizons in the soil profile. Shallow soil is typically composed of sandy soil and deeper soil is composed of sandy loam (Lee, 1984). Soil pH is an indicator of the acidity of soil moisture and is an important factor influencing nutrient availability (Courtney and Trudgil, 1984).  $H^+$  circulation is an important indicator of acidification in ecosystems and watersheds (Verstraten et al., 1990). Rainfall containing dissolved air pollutants has an important role in producing  $H^+$  (Bache, 1980; Verstraten et al., 1990), and acid precipitation in turn drives soil acidification (Simmons and Kelly, 1989; Wolt, 1990). For example,  $H^+$  deposition via throughfall in sugar maple and yellow birch accounted for 100% of the influence on soil pH in the defoliation season and 40% in the growth period (Hazlett and Foster, 1989). Soil acidification is more prevalent in coniferous forests than in broadleaf forests (Bredemeier, 1988). In this study, the soil under *P. densiflora* had a lower pH than that under *Q.*

*mongolica*.

Soil is heterogeneous, even at small spatial scales, because of the diversity of organic materials and minerals from which it originates, their reaction, and environmental conditions (e.g., altitude, gradient, slope, and rainfall amount and ion content) (Howells, 1990). Microbial nitrogen fixation produces  $H^+$ , which affects acidity (Paul and Clark, 1989). Therefore, to decrease the error associated with pH measurement, we collected soil samples at various distances from the stem. Soil pH was not correlated with distance from the stem ( $P < 0.01$ ), but the difference between the two species was significant ( $P < 0.01$ ). Verification with Tukey's test revealed that soil pH within the area of influence of *Q. mongolica* was higher than that of *P. densiflora* ( $\alpha = 0.05$ ). The total annual inflow of  $H^+$  was  $0.01 \text{ kg}\cdot\text{ha}^{-1}$ , and three positive ions ( $Ca^{2+}$ ,  $Mg^{2+}$ , and  $Na^+$ ) had high enrichment ratios after passing through the crown of *P. densiflora* (Fig. 4, Tables 2, 3). This indicated that as positive ion transmission from leaf cells to buffer pH increased, the soil pH increased, because plants must absorb more positive ions to maintain a balance (Mecklenburg and Tukey, 1963; Reuss and Johnson, 1986; Bredemeier, 1988; Percy, 1989). The NTFs of  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$ , and  $Na^+$  were mostly positive, indicative of leaching. The proportions of the NTFs of  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$ , and  $Na^+$  to the frequency of rainfall were 0.83, 0.91, 0.91, and 0.86 in *P. densiflora* and 0.7, 0.88, 0.88, and 0.68 in *Q. mongolica*, respectively. *Q. mongolica* experienced a defoliation season from the end of August to March, when it lacked a crown, resulting in a lower total ion movement than *P. densiflora*, which maintained its crown throughout the year.

Previous research revealed that soil pH was lowest near *Pinus contorta* stems and increased with distance from the stem, and that stemflow near the plants resulted in chemical reactions in the soil, resulting in high exchangeable  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$ ,  $Na^+$ , and organic carbon concentrations (Zinke, 1959; Gersper and

Holowaychuk, 1970). We also observed low pH levels near *P. densiflora*, differing from *Q. mongolica*, likely because stemflow infiltration caused the stemflow to disperse widely throughout the soil, and because we sampled the 0 - 10-cm soil layer, where the horizontal sampling method possibly decreased the effects of stemflow. Gersper and Holowaychuk (1970) recommended the use of species with high stemflow volumes in such studies. In the present study, the pH of stemflow was lower than that of throughfall, and the pH of *P. densiflora* stemflow had about four times the annual average concentration of *Q. mongolica* stemflow. However, stemflow is localized to only a small area around the stem (Parker, 1983). Kil and Yim (1983) observed a soil pH of 4.5 around *P. densiflora*, while that of the surrounding forest was 6.0, and when rainfall passed through *P. densiflora*, its leaves leached allelopathic substances; therefore, this species might have the capacity to lower soil pH.

#### 4.5. Influence of *Quercus mongolica* and *Pinus densiflora* on net throughfall

Net throughfall (NTF) can be described by rainfall intensity and the amount and duration of rainfall (Potter et al., 1991). The stepwise regression analysis of NTF revealed that the ion NTF values were correlated with chemical factors (e.g., rainfall pH), physical factors (e.g., amount and intensity of rainfall), and temporal factors (e.g., dry period before rainfall or the dry season, and rainfall duration) (Table 4). With the exception of  $H^+$ , most of which appeared to be absorbed by the trees, the NTF values were positive; therefore, leaching and dry deposition likely influenced the movement of the other ions. The positive and negative coefficients for a given factor indicated the condition and state of each ion. For example,  $NH_4^+$ ,  $Na^+$ ,  $Cl^-$ , and  $SO_4^{2-}$  were positively correlated with dry periods before rainfall or the dry season in *P. densiflora*, suggestive of dry deposition. Meanwhile, rainfall intensity and  $SO_4^{2-}$  were positively

correlated, indicating that increased rainfall intensity could leach  $\text{SO}_4^{2-}$  from the leaves and wash off dry deposits, increasing the inflow of  $\text{SO}_4^{2-}$ . Rainfall intensity and dry period before rainfall or dry season had coefficients of 7.9 and 0.11, respectively (Table 4). In *Q. mongolica*,  $\text{NH}_4^+$  and dry period before rainfall or dry season were positively correlated. Further, more  $\text{NH}_4^+$  was deposited on *Q. mongolica* than *P. densiflora* leaves. Rainfall duration was not correlated with any ion NTF; however, it is thought to be related to ion mobility. Meanwhile, rainfall amount and ion NTF were negatively correlated due to dilution; therefore, most ions had negative coefficients with this factor (Table 4).

In *Q. mongolica*, rainfall pH and  $\text{SO}_4^{2-}$  were positively correlated, indicating that  $\text{H}^+$  concentration and  $\text{SO}_4^{2-}$  were inversely correlated. This finding suggests that  $\text{H}^+$  from  $\text{SO}_4^{2-}$  was buffered by *Q. mongolica* leaves. Finally, in *P. densiflora*, the correlation between  $\text{Mg}^{2+}$  and  $\text{NH}_4^+$  indicated that positive ion leaching via throughfall increased with increasing  $\text{H}^+$  concentrations.

#### 4.6. Implications for the relationships between dry and wet deposition

Ion transport via rainfall is referred to as wet deposition, where falling rain dissolves ions dispersed in the atmosphere and transports them downward. With the exception of internal circulation in plants, ions transported via dry deposition are predominantly washed off surfaces by rainfall or scoured off by floating solids on the leaf interface layer near the stem. Such deposits can be influenced by humans, although some ions such as  $\text{NH}_4^+$  can be influenced by animals. For example,  $\text{SO}_2$ ,  $\text{NO}_2$ , and other sulfur-containing molecules from industrial and automotive pollution are dissolved and altered in rainfall, and likely affected the results of this study. Overall, there are little precise data available on dry deposition rates in forests in Korea; therefore, further research is needed.

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