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Leaching of Dissolved Organic Carbon and Cations and the Buffering Capacity of Litters from Forest Stands in Southwestern China

ABSTRACT

Forest soil can buffer acidification and neutralize acidic airborne pollutants, but for acid rainwater, it makes contact with forest litter in the forest ecosystem first before reaching the soil. However, leachate chemistry of forest litter treated with different acid load rates is rarely studied. A leaching experiment was performed on forest litter from mixed conifer-broadleaf (MCB) and evergreen broadleaf (EB) forests in Jinyun Mountain, Three Gorges area, Southwestern China with simulated acid rain (SAR) of pH=2.7, 3.5 and 4.5. Dissolved organic carbon (DOC) and cations were exported from MCB and EB when treated with various acid load rates. The rainwater deacidification of forest litter was enhanced by considerable leachate concentrations of DOC, Ca^{2+} and Mg^{2+}. The acid buffering capacity of EB was stronger than MCB with different composition of forest litter. Leaching of cations increased with decreasing pH of SAR. Although more easily decomposed, EB released greater Al^{3+} than MCB, leachate Ca/Al ratios of EB did not reach the critical value of 1.0. Thus, in the study area, EB forest may be a better choice for afforestation and reforestation with better forest litter, showing good buffering capacity, keeping soil from acidification and being a greater nutrient pool for soil under it.

Keywords: acid deposition, forest litter composition, DOC, cations, acid buffering

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INTRODUCTION

Forest ecosystems provide many hydrological functions, including regulating runoff, improving water quality, controlling soil erosion and conserving water resources (*Brauman et al. 2007; Liu 2019; Osborne and Kovacic 1993*). These functions are strongly related to spatial vegetation structure and species composition of the forest and their influence on rainfall-runoff processes (*Chang 2003; Liu et al. 2013*). Rainwater is intercepted and retained by forest canopy, crowns of shrubs and grasses, forest litter of dead needles, leaves and branches and soil (*Du et al. 2019; Naiman and Decamps 1997; Zhao et al. 2019*). Runoff occurs when the water holding capacities of canopy, depressions, forest litter and soil exceeded the maximum (*Chang 2003*). As precipitation is intercepted, water quality also changes by dissolving and leaching chemicals, particles or any other substances of canopy, depressions, forest litter and soil (*Hafner et al. 2005*) because nutrient cycling in forest ecosystems is tied to hydrological processes mentioned above (*Liu 2019; Tobón, Sevink and Verstraten 2004*). However, little is known about the effects of rainwater on leachate characteristics of forest litter. As acid rain pollution increases, the hydrochemistry of rainfall-runoff processes in forest ecosystems may be influenced by the input of acidic compounds.

The forest canopy, as the first level contacted by rainwater, has marked impacts on acid rainwater chemistry. Anion and cation concentrations are significantly changed in throughfall compared to concentrations in bulk precipitation collected outside the canopies (*Gandois et al. 2010; Jakovljevic et al. 2013; Shen et al. 2013*). Leaching dust and secretions on forest canopy could account for changing concentrations of these ions (*Clark et al. 1998; Liu et al. 2013*),hence, changing the pH of throughfall (*Cape et al. 2001; Liu et al. 2013*). The quality and quantity of dust and secretions on the canopy differ with tree species as well as forest canopy architectures (such as leaf area index, canopy density and gap). Mixed coniferous-broadleaf forest has different tree species composition and canopy architecture compared to broadleaf forest. Consequently, the pH and ions of throughfall varies with forest type. Such results, therefore, suggest that canopy architecture has important impacts on throughfall chemistry and that species-specific effects should not be overlooked when assessing acid deposition in forested areas (*Liu et al. 2013; Shen et al. 2013*).

Chemical characteristics of soil solution are also influenced by acid rain. Solution of NO_3^- concentration in soil shows a rapid response to acid deposition that results in acidification of soil solution (*Jones et al. 2008; Liu et al. 2013*). Influent-effluent chemical comparisons suggest that the SO_4^{2-} concentration of soil solutions control the effluent pH (*Cai et al. 2011; Chen, Li and Gao 2010; Jung, Ok and Chang 2011*). Base cations (e.g., Ca^{2+}, Mg^{2+}, K^+ and Na^+), which usually buffer soil from the acid effects of deposited N and S, are released from soil to soil solution (*Cai et al. 2011; Helliwell et al. 2010*) and their concentrations in soil solution are increased with decreasing rainwater pH (*Nawaz et al. 2012*). In the long term, these mineral losses will adversely affect health and growth of undergrowth and the forest stands. The increased acidity of the environment will presumably have negative effects on the receiving aquatic systems (*Małek 2010*). Soil acidity could enhance the mobilization of metals (e.g., Al and Mn), leading to phytotoxic conditions and decreasing plant growth and development (*Jonard et al. 2011; Nawaz et al. 2012*).

Acid water from air or forest canopy first makes contact with forest litter before it reaches the soil. *Kammer and Hagedorn* (*2011*) and *Clarholm and Skyllberg* (*2013*) suggested that pH buffering and soil organic matter (SOM) turnover in organic soil are regulated by Ca and Mg cycling via the tree canopy, litter fall, and fungal translocation of Al from mineral soil. The deposited acid may change the process of litter decomposition (*Lv et al. 2014*) and influence the nutrient flux from litter fall to soil. The forest litter is a key point in the nutrient cycle, because of combined action of microbe and natural weathering during decomposition (*Guelland et al. 2013; Lv et al. 2014; Riaz et al. 2010*). Organic and inorganic nutrients are subsequently released to soil layer and provided to living plants (*Corrigan and Oelbermann 2013*). Consequently, forest productivity may be affected by acid rain. If forest productivity decreases, eventually logging is reduced; thus jobs will be lost and the national economy may deteriorate, at the same time as a key resource for industry diminishes (*Kikuchi 2004*).

The importance of forest litter in controlling nutrient cycling, effects on pH buffering ability and nutrient dynamics under acid rain are not well understood. This study will provide science-based input to forest management and future design of afforestation and reforestation in areas under heavy acidic deposition brought by intensive wood production and changes across landscape. This laboratory experiment investigated the buffering capacity of intact forest litters from leachates in the different forest stands with simulated acid rain (SAR) of varying pH values in Jinyun Mountain of Southwestern China. The hypotheses of this study were: pH of SAR was buffered by forest litter; release of dissolved organic carbon (DOC) and cations will be controlled by pH of SAR; and forest litter composition has relation with the release characteristics of DOC and cations.

MATERIALS AND METHODS

Description of Research Area and Forest Site Characteristics

The study was carried out at Jinyun Mountain, Three Gorges area, Southwestern China (29°41′-29°52′ N, 106°17′-106°24′ E,). The forest area is 1112.7 ha, which accounts for 96.6% of local land area, where subtropical forest species are abundant. The study area is bounded by the two major river systems of the region: Yangtze River and Jialingjiang River. The elevation of this area is between 350 and 952 masl. This region has a subtropical monsoon climate with long warm to hot humid summers and short cool to cold and cloudy winters with the lowest total number of sunshine days in China (about 1,000 hr yr^{-1}). The mean annual temperature is 13.6°C and the average annual precipitation is 1,611.8 mm.

Two typical forest stands were selected for the study: evergreen broadleaf (EB) forest with dominant tree species of *Gordonia acuminata* Chang and *Symplocos setchuensis* Brand and mixed conifer-broadleaf (MCB) forest with dominant tree species of *Pinus massoniana* Lamb and *Gordonia acuminata* Chang.

Sampling method

To test the forest litter chemical properties, three random samples (25 × 25 cm) of each forest type were collected and homogeneously mixed by forest type. The experiment was performed on 18 forest litter samples: 2 forest types (forest litter types) × 3 acid loading rates (SAR pH=2.7, 3.5 and 4.5) × 3 replications for each treatment. Samples of representative forest litters comprised of needle-broad leaves and twigs from MCB forest and broad leaves with twigs from EB forest. Samples were cut into a circle with 15 cm diameter to accommodate size of the leaching device for laboratory preparation.

Leaching Method

Deionized water and analytical-grade chemicals were used for laboratory simulation of acid precipitation. Concentrations of SO_4^{2-} and NO_3^- were increased with decreasing pH while SO_4^{2-}/NO_3^- ratio of SAR was consistent at 4.7: 1 following the average value of acid throughfall from two forests described by (*Liu et al. unpublished*). The pH of SAR was adjusted by adding HNO_3 and H_2SO_4 into solution (**Table 1**).

The forest litter samples were sprinkled with SAR using vessels with perforated bottoms. The sprinkling lasted for 28 hr and pH values of forest litter leachates were recorded after 0.5 hr, 1 hr, 2 hr, 3 hr, 4 hr, 6 hr, 8 hr, 10 hr, 12 hr, 16 hr, 20 hr and 28 hr. The leaching rate was 1.2 mm hr^{-1} (21.21 mL h^{-1}). Forest litter leachates were immediately collected and analyzed within 24 h.

Analysis Methods

Forest litter samples were dried to constant weight (70°C, 24 hr) and ground using electronic grinder. Samples were digested with the sulphuric acid–hydrogen peroxide procedure (*Lu 2000*). Concentrations of Ca, Mg, K, Na and Al were determined using Inductively Coupled Plasma Optical Emission spectroscopy (ICP-OES, Agilent Technologies Inc., USA) while the total carbon (TC) was tested according to the dichromate acid wet oxidation method (**Table 2**).

Leachate pH was tested using a glass electrode (Tintometer GmbH, Germany). Filtered leachate samples (0.45 μm) were analyzed for base cations (Ca^{2+}, Mg^{2+}, K^+ and Na^+) and Al^{3+} using ICP-OES (Agilent Technologies Inc., USA) and tested for DOC with a Total Organic Carbon analyzer (Labtoc, Pollution and Process Monitoring Limited, UK).

Table 1. The pH gradient and concentrations of ions in simulated acid rain ($mmolL^{-1}$).

Ions	pH=2.7	pH=3.5	pH=4.5
Ca^{2+}	0.521	0.521	0.521
Mg^{2+}	0.191	0.191	0.191
K^+	0.042	0.042	0.042
Na^+	0.065	0.065	0.065
NH_4^+	0.532	0.532	0.532
SO_4^{2-}	0.933	0.148	0.015
NO_3^-	0.197	0.031	0.0031

Note: Concentrations of ions were mean value of all acid throughfall water from two forest types. pH gradients were also designed from acid throughfall of the two forests.

Statistical Analyses

Concentrations of DOC and cations were analyzed as net output by subtracting influent from effluent concentrations. For SAR treatments, differences between forest types were determined following a one-way ANOVA for pH, Ca/Al ratios, and concentrations of DOC, base cations and Al^{3+}. Within forest type, differences in DOC concentrations, as well as cation concentrations, among all levels of SAR were tested. The linear rate of change in leachate pH with leaching time was analyzed based on forest litter by type. Correlations between DOC concentrations and cation concentrations of leachates were quantified separately for each forest type. Data analysis was implemented using Microsoft Office Excel 2007 and IBM SPSS Statistics 20.

RESULTS AND DISCUSSION

Variation in pH of Leachates

Both pH values of leachates from EB (r=0.7595 for pH of SAR=2.7, r=0.8879 for pH of SAR=3.5, n=13) and MCB (r=0.8924 for pH of SAR=2.7, r=0.9707 for pH of SAR=3.5, n=13) were increased linearly when treated with pH of SAR≤3.5 (**Figure 1**). In contrast, leachate pH values of MCB treated with SAR at pH=4.5 decreased sharply at the beginning and then increased smoothly from 4 hr to the end. Leachate from EB also decreased, although less sharply, and then increased slowly after 8 hr in leaching period. The higher the SAR pH value, the slower the pH buffer factors (such as base cations) may release. Consequently, at the beginning of the test, leachate pH values of MCB changed conversely by treating with SAR of pH=4.5 or pH≤3.5. The same result was also observed in EB test process. Leachate pH values were significantly different between MCB and EB for all acid loading rates (p<0.001). The pH variations within influent-effluent process were greatly affected by forest litters from two forest types. The pH buffering ability of EB forest litter, with significant higher leachate pH (p<0.001), was better than MCB litter when leaching by SAR of all pH gradients. However, the pH buffering capacities of the two forest types were both insufficient while leaching by SAR with the largest pH because

Table 2. Chemical characteristics of forest litter (g kg^{-1}) in Jinyun Mountain, Three Gorges area, Southwestern China.

Forest Litter	TC	Ca	Mg	K	Na	Al
MBC	423.602±48.243	3.530±0.468	0.675±0.243	0.629±0.336	0.043±0.010	0.332±0.097
EB	443.843±15.433	3.824±0.157	0.561±0.048	0.465±0.208	0.038±0.005	0.355±0.180

Note: Values are performed as average ± standard error; MCB- Mixed Conifer-Broadleaf; EB- Evergreen Broadleaf

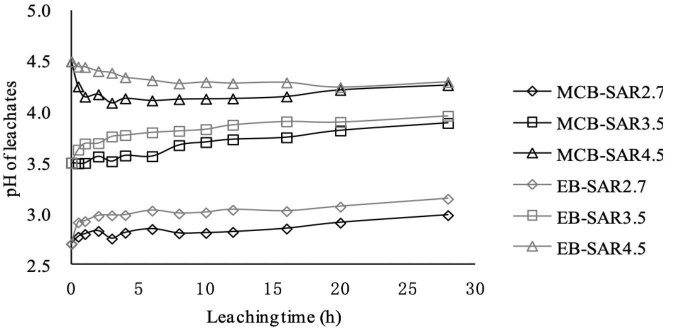

Figure 1. Change in pH of leachates from evergreen broadleaf (EB)and mixed conifer-broadleaf (MCB) forests in Jinyun Mountain, Three Gorges area, Southwestern China treated with different SARs.

of the decreasing-increasing trend of leachate pH values rather than always increasing trend (**Figure 1**). In that case, leachate pH values finally increased as test-time increased with much higher pH for EB ($p<0.001$).

Forest litter can buffer rainwater from acidification (*Department 2000*). However, more studies on various forests are still necessary to expand the deacidification data for forest litter from all over the world. In Jinyun Mountain, litter from EB and MCB forests had favorable deacidification effect on acid rainwater. Although the leachate pH of EB and MCB decreased in the first few hours of test process while treating with SAR pH=4.5, but both of them kept increasing in the rest hours of test. Both EB and MCB had pH buffer ability leached by SAR of pH 4.5. And the pH buffer effects were respectively observed from 4 hr to 28 hr for MCB and from 8 hr to 28 hr for EB. Such deacidification as shown in other forest types (*Kikuchi 2004; Skiba and Cresser 1986*), but *Shen et al.* (*2013*) and *Liu et al.* (*2013*) suggest that species-specific effects should not be overlooked while assessing the acid deposition in forested areas. This study found significantly lower pH values ($p<0.001$) in leachates of MCB treated with the three types of SAR, suggesting that leachates released from MCB led to further acidification of soil than those of EB.

Hydrochemical studies of effects of canopy and soil (mineral soil and litter fall were combined as an individual level) in the same EB and MCB forests found that soil pH was 4.10 for MCB and was 4.20 for EB (*Liu et al. 2013*). The difference is due to the release of acidic compounds from decomposition of coniferous litter, which especially is a source of soil acidification (*Lv et al. 2014*), combined with more acidic dust falling to the surface of forest litter through more lacunose canopy of MCB forest (*Wei et al. 2005*).

Variation in Concentrations of Released Base Cations

Concentrations of released Mg^{2+} were higher in EB than in MCB when treated with SAR of pH=3.5 ($p<0.05$, **Table 3**). No other significant differences were found for concentrations of released Mg^{2+} or Ca^{2+}, although concentrations released from EB were usually higher than those released from MCB. The results may support the opinion that Ca and Mg of forest litter will be more easily released under large acid load (pH of rainwater ≤ 3.5).

Table 3. Changes in net released concentrations of dissolved organic carbon (DOC) and cations and Ca/Al ratios in leachates of EB and MCB treated with various acid loading rates ($mmol \cdot L^{-1}$).

Indices	Forest Litter	pH of SAR		
			3.5	4.5
Ca^{2+}	MCB	0.0247±0.0059	0.0146±0.0056	0.0166±0.0012
	EB	0.0403±0.0148 (a)	0.0204±0.0022 (b)	0.0153±0.0036 (c)
Mg^{2+}	MCB	0.0042±0.0003	0.0036±0.0004 (A)	0.0039±0.0004
	EB	0.0060±0.0013	0.0049±0.0006 (B)	0.0047±0.0009
K^+	MCB	0.00030±0	0.00027±0.00006	0.00023±0.00006
	EB	0.00033±0.00012	0.00027±0.00006	0.00030±0.00010
Na^+	MCB	0.00037±0.00006	0.00030±0.00010	0.00033±0.00006
	EB	0.00040±0.00010	0.00037±0.00006	0.00037±0.00015
Al^{3+}	MCB	0.0046±0.0042	0.0013±0.0014	0.0014±0.0016
	EB	0.0104±0.0069	0.0020±0.0011	0.0013±0.0006
Ca/Al	MCB	9.6446±8.3562	17.1043±8.9494	28.7797±26.8034
	EB	4.5436±1.8608	11.4702±4.3262	13.1493±4.6050
DOC	MCB	0.3247±0.1690	0.2360±0.0166 (A)	0.2296±0.0402 (A)
	EB	0.6276±0.4013	0.6282±0.0913 (B)	0.6502±0.2227 (B)

EB- evergreen broadleaf; MCB- mixed conifer-broadleaf

Note: values are average ± standard error (n=3). Capital letters indicate significant differences ($p<0.05$) between values of EB and MCB. Different lowercase letters present significant differences ($p<0.05$) among values of each single forest type treated with various acid loading rates.

The Ca^{2+} concentrations discharged from EB increased with decreasing SAR pH ($p<0.05$); those from MCB differed less ($p=0.0848$) (**Table 3**). No differences among SAR rates were found for other cations ($p>0.2$), although concentrations also increased with decreasing SAR pH. In this case, changes in pH of rainwater were thought to be more effective on efflux of Ca of forest litter under evergreen broadleaf forests (**Table 3**).

Exchange of hydrogen ions (H^+) with base cations (e.g. Ca^{2+}, Mg^{2+}, K^+ and Na^+) is important from the viewpoint of deacidification mechanisms in soil (*Liu et al. 2013*). Deducting the cations added through the supply of SAR, both of the two forest litters released base cations while treating with SAR of different acidity. However, the significant differences were only found between Ca^{2+} and Mg^{2+} of EB and MCB rather than K+ and Na+. Concentrations of Ca^{2+} and Mg^{2+} were considerably greater than those of other base cations. Given the remarkably different leachate pH between EB and MCB, Ca^{2+} and Mg^{2+} were thought to be main factors for exchanging H^+. However, significantly positive correlations between DOC and Ca^{2+} ($r=0.9804$, $n=3$) and Mg^{2+} ($r=0.9399$, $n=3$) were only found in leachates of EB treated with SAR of pH=2.7. Thus, EB, with higher Ca^{2+} and Mg^{2+}, had a larger capacity to buffer acidification than MCB with different litter composition. Increasingly acidic input will prevent Ca^{2+} and Mg^{2+} from being captured by DOC, which could be important in controlling Ca^{2+} and Mg^{2+} transport and availability, at least on a micro site scale (*Hafner et al. 2005*). Enhanced Ca^{2+} and Mg^{2+} was also reported when treated with artificial rain acidified to pH=3 with H_2SO_4. *Kikuchi* (*2004*) suggested that litter fall of beech mainly discharged Ca^{2+} and Mg^{2+}, although the concentrations of Ca^{2+} and Mg^{2+} in SAR were considerably lower than those of our study. Net output of base cations was probably related to forest litter chemistry (**Table 2**). The output concentrations of base cations were increased with decreasing SAR acidity. However, a significant difference was only found in Ca^{2+} concentrations of EB, with more activity under changing SAR, possibly enhancing the ability of EB to buffer rainwater pH.

Variation in Concentrations of Released Dissolved Organic Carbon (DOC)

Organic matter released by EB was significantly larger than MCB while treating with SAR of pH≥3.5 ($p<0.05$, **Table 3**). Effluent DOC concentrations of EB and MCB under the greatest acid loading rate (SAR=2.7) were not different ($p=0.2947$). Within forest type, DOC concentration was similar for all levels of SAR pH. Thus, organic matter of the two forest litters was stable suffering changing acid loading rate.

The solute with the highest concentration in both leachates of EB and MCB was DOC, but the concentrations measured in this study were less than those obtained in other studies (*Hafner et al. 2005; Skiba and Cresser 1986*). However, soil C can accumulate as a consequence of increasing acid deposition and this may seriously affect the balance of ecosystem C flux (*Lv et al. 2014*). In contrast, dissolved organic matter contained in the litter has a deacidification effect (*Kikuchi 2004; Riha et al. 1986*). In this study, the effect may be demonstrated by larger values of pH and DOC simultaneously occurring in leachates of EB. Furthermore, pH values and DOC concentrations of EB were significantly greater than those of MCB except for DOC under SAR of pH=2.7 ($p=0.2947$), suggesting that the deacidification capacities of EB and MCB were different because of their distinct litter composition. Among pH treatments within a single forest type, DOC concentrations were not significantly different. Increased acidity can reduce decomposition of forest litter (*Skiba and Cresser 1986*). In this study, litter from mixed conifer-broadleaf forest decomposed more slowly than that from EB due to more waxes covering needle surface (*Prescott et al. 2004*). Therefore, DOC released from MCB was much lower than EB even if TC contents of EB and MCB were similar with each other (**Table 2**).

Variation in Concentrations of Released Al^{3+} and Ca/Al

Leachates of EB treated with SAR pH≤3.5 released the most Al^{3+}, but no significant differences were found among pH levels for either forest type (EB: $p=0.0613$; MCB: p=0.3222; **Table 3**). Levels were also similar between the two forest types within each SAR acidity level ($p>0.5$). However, Al^{3+} discharged from both forest litters increased with decreasing pH.

Toxicity of Al to plants is usually presented by Ca/Al ratio. Plants are likely to be affected by aluminum toxicity in soil when Ca/Al<1.0 (*Riha et al. 1986*). Ca/Al of EB and MCB decreased with increasing acidity (**Table 3**), although the trend was not significant (EB: $p=0.0689$; MCB: $p=0.4334$). Ca/Al ratios of leachates discharged from EB were lower than those of MCB under leaching of SARs.

Special attention was given to Al leachate in this experiment because the fine roots of trees were damaged by the toxicity of labile Al, which, together with nutrient deficiency, causes a stress condition known as crown dieback: leaves or needles at the tree top turn yellow, then brown, and eventually drop off (*Kikuchi 2004*). As discussed above, EB was more active while leaching by SAR. Al^{3+} concentrations in leachates of EB was larger when treated with SAR of pH≤3.5 although without significant differences from MCB. Ca/Al ratios were also less than those of MCB. Toxicity of EB forest may be enhanced by larger net Al^{3+} input of forest litter. Exchangeable Al in mineral soil under EB forest was larger than MCB forest (*Liu et al. 2013*). Although both leachate Ca/Al ratios of EB and MCB did not fall to the critical value of 1.0, the same as *Kikuchi* (*2004*) suggested, the values of EB treated with SAR of pH=2.7 were near the critical value (**Table 3**) and Ca/Al ratios of EB decreased with increasing SAR acidity. Moreover, Al^{3+} concentrations of EB (r=0.8754, n=3) and MCB (r=0.9887, n=3) were both positively correlated with DOC when treated with the highest acid loading. The results indicated the possibility of temporal (*Kikuchi 2004*) and acidic (*Liu et al. 2013*) critical condition (namely, root damage of trees and/or alteration of soil properties) at an EB forest site. As with Ca^{2+} and Mg^{2+}, increasing acidity input will prevent Al^{3+} from being captured by DOC. Complexation of metal cations by dissolved organic matter is an important process in the movement and availability of both nutrient and toxic cations (*Hafner et al. 2005*).

CONCLUSION AND RECOMMENDATIONS

The considerably higher concentrations of DOC and other solutes from leachates of EB and MCB demonstrated that forest litter is a source of organic carbon and cations in the two studied forests. Concentrations of cations in leachates increased with increasing acid loading rates. Composition of EB and MCB litter influenced the leachate chemistry, especially for Ca^{2+}, Mg^{2+} and DOC, which are main factors for pH buffering capacity of forest litter. EB, releasing more DOC and base cations, can buffer leachate acidification better than MCB. However, the more easily decomposed EB litter will release greater Al^{3+} than MCB as time and acidity are increased, and Ca/Al of EB will drop to the critical value of 1.0 in future. Leaching from forest litter should be more closely studied for its contribution to nutrient cycling within forested ecosystems, especially where acid deposition is a larger component of the precipitation. The study will be useful in management of productivity and sustainability functions of the forest ecosystem.

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