

ATMOSPHERIC DEPOSITION AND FOLIAR LEACHING IN A REGENERATING SOUTHERN APPALACHIAN FOREST CANOPY

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SUMMARY

(1) Incident precipitation, throughfall and stemflow were collected to examine the importance of factors potentially determining net canopy element fluxes, and to quantify canopy exchange and dry deposition rates in a regenerating southern Appalachian forest.

(2) Net throughfall fluxes (throughfall minus precipitation transfers) showed consistent canopy effects on rainfall chemistry, with SO_4^{2-} , PO_4^{3-} , Cl^- , K^+ , Ca^{2+} and Mg^{2+} added to rainfall by foliage, whereas NO_3^- -N, NH_4^+ -N and H^+ ions were absorbed from precipitation. Storm characteristics (event amount and duration) accounted for the largest portion of the variability in growing season net throughfall fluxes, suggesting that canopy exchange was the major mechanism of throughfall transfer.

(3) Stemflow fluxes increased canopy exchange rates of SO_4^{2-} , PO_4^{3-} , Cl^- , K^+ , and Mg^{2+} by greater than 20% in a regression model of total below-canopy element fluxes.

(4) Cation leaching fluxes were highly variable (C.V. > 50%) over spatial scales of several m^2 , but could be explained largely by heterogeneity in canopy cover.

(5) Foliar cation leaching losses in the early successional forest accounted for 4–13% of leaf nutrient reserves. As cation throughfall transfers were highest during storms with the greatest hydrogen ion uptake from rainwater, it is hypothesized that acid precipitation is causing accelerated foliar nutrient leaching in south-eastern hardwood forests.

INTRODUCTION

Precipitation chemistry is dramatically altered upon passage through forest canopies (Voigt 1960; Attiwill 1966; Eaton, Likens & Bormann 1973). Elements such as sulphur and potassium are added to throughfall and stemflow as a result of plant-atmosphere interactions, whereas inorganic nitrogen may be absorbed from rainfall by foliage (Lovett & Lindberg 1984). The two primary mechanisms influencing throughfall chemistry are (i) washoff of dry-deposited elements from leaf surfaces, and (ii) canopy exchange, through leaching of plant nutrients and absorption of ions

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from rainfall. The origin of mobile elements in the atmosphere–plant–soil continuum is largely unresolved, although several attempts have been made to separate sources of throughfall nutrient fluxes (Miller, Cooper & Miller 1976; Bache 1977; Mayer & Ulrich 1978; Lakhani & Miller 1980; Lovett & Lindberg 1984; Lindberg & Garten 1988).

The need for analysis of natural surfaces for accumulated dry deposition and wash-off by precipitation is often cited as a major research goal in assessing acid-rain impacts on forest ecosystems. Forest trees can effectively filter pollutants from the atmosphere (Mayer & Ulrich 1978), and dry deposition wash-off by precipitation may contribute significant amounts of certain elements to the forest floor (Swank & Henderson 1976; Lovett & Lindberg 1984).

Leaching, which is defined as the movement of substances derived exclusively from plant tissues to an aqueous solution in direct contact with vegetation, has taken on added importance as precipitation acidification has increased in recent decades. Leaching may represent the largest transfer pathway from the canopy to the forest floor for mobile elements (Parker 1983). One indirect effect of acid precipitation on forest vegetation may be the acceleration of foliar nutrient leaching (Tamm & Cowling 1977; Abrahamsen 1984). Leaching may indirectly affect plant energetics by increasing rates of root uptake and translocation to replace ions leached from foliar tissues, effectively accelerating plant nutrient cycling (Tukey 1970; Fairfax & Lepp 1975; Wood & Bormann 1975; Hornvedt, Dollard & Joranger 1980). Physiological stress resulting from increased foliar cation leaching may be occurring in forests in the eastern United States (Lovett *et al.* 1985), and characterization of the role of acidification in foliar leaching remains an important research task (Johnson *et al.* 1983; Lefohn & Brocksen 1984; Amthor 1986; Pitelka & Raynal 1989).

The objectives of this study were (i) to examine the importance of factors potentially determining net throughfall fluxes in a regenerating southern Appalachian forest canopy, (ii) to quantify canopy exchange and dry deposition rates by regression analysis of net throughfall and net stemflow, and (iii) to compare throughfall and stemflow transfers in an early successional forest with transfers in more mature forest stands.

Throughfall composition is determined by a complex interaction of atmospheric, hydrological and biochemical processes. Several factors, either temporal or spatial in nature, affect net throughfall chemistry in a given forest ecosystem (Table 1). These factors, in turn, are associated with certain biotic and abiotic mechanisms which influence biogeochemical cycling in forests. Human disturbance of forest canopy structure brought about by clear-cut logging may substantially alter throughfall and stemflow fluxes, thereby providing a framework for testing hypothesized mechanisms of canopy nutrient transfers.

METHODS

Study area

The study area was Watershed 7 (WS7) at the United States Department of Agriculture, Forest Service Coweeta Hydrologic Laboratory (35°N, 83°W), near Franklin, North Carolina. The 59-ha catchment has a south-facing aspect, with altitude ranging from 720 to 1065 m a.s.l. The watershed was clear-cut and harvested in 1977 by cable-logging, a technique which minimized disturbance to the forest

TABLE 1. Hypothesized factors accounting for temporal and spatial variability in nutrient throughfall fluxes and probable associated mechanisms of element transfer.

Temporal factor	Probable mechanism	Spatial factor	Probable mechanism
Antecedent dry period (h)	Dry deposition	Relative canopy cover (%)	Dry deposition + canopy exchange (leaching/uptake)
Dry deposition rate ($\mu\text{equiv. m}^{-2}\text{h}^{-1}$)	Dry deposition	Tree species frequency (%)	Dry deposition + canopy exchange (leaching/uptake)
Event amount (mm)	Canopy exchange (leaching/uptake)	Event amount (mm)	Canopy exchange (leaching/uptake)
Event duration (h)	Canopy exchange (leaching/uptake)	Event duration (h)	Canopy exchange (leaching/uptake)
Event intensity (mm h^{-1})	Canopy exchange (leaching/uptake)	Event intensity (mm h^{-1})	Canopy exchange (leaching/uptake)
Storm date (days/months)	Leaf damage (leaching/uptake)		

floor. Recovery of the vegetation and foliar nutrient contents one year after clear-cutting were studied by Boring, Monk & Swank (1981). Leaf area index and net primary productivity on WS7 had nearly returned to pre-clearcut levels by 1985 (Boring & Swank 1986). The accessible (5–10-m-high), well-developed canopy offered a unique opportunity for study of precipitation–canopy interactions.

Incident deposition, throughfall and stemflow

A 10-m tower was erected near the middle of WS7, and an Aerochem Metrics Model 301 wetfall–dryfall collector was placed just above canopy level on the tower to collect incident deposition. Rainfall was collected in the wet bucket by five plastic funnel-bottle collectors. The dry side of the collector was modified to hold five 100-mm x 10-mm polystyrene Petri plates for estimating coarse dry particulate inputs to the canopy (Lindberg & Lovett 1985). Petri plates were placed inside shallow (15-mm-deep) plastic holders mounted at the top rim of the dry bucket. Sets of five replicate plates were exposed to dry conditions for various time periods between storms, ranging from 24 h to 3 weeks. Dry-deposited elements were extracted from plates using distilled water, following the procedures established by Lindberg & Lovett (1985).

Plastic troughs (1.0 m x 0.1 m) were used to collect throughfall. Troughs were randomly placed, ten per plot on each of three 20-m x 20-m plots — a total of thirty collectors. Study plots were located near the middle of WS7 in an area characterized as a xeric *Quercus prinus* (chestnut oak) community (Boring & Swank 1986). (See Radford, Ahles & Bell (1968) for authorities of species names.) Troughs were fitted with fibreglass screen inserts to minimize throughfall splashing and to trap coarse debris. All troughs and plastic collection jugs (23 litre) were rinsed thoroughly with

distilled water between storms. Phenyl mercuric acetate (PMA) was sprayed into jugs to inhibit bacterial growth. Troughs were covered until the initiation of a storm event to prevent contamination by dry deposition.

Stemflow was collected from all living stems greater than 1 cm in diameter (at 40 cm from stem base) in nine randomly located 1-m x 2-m areas. Three stemflow areas were established on each of the larger study plots. Two to six total stems of various tree species and size classes in each of the nine areas were fitted with collars. A funnel under a hole in each collar was used to drain stemflow through plastic tubing into a common stemflow jug collector. Collars were rinsed thoroughly with distilled water just before storms.

Foliar sampling

Five individuals from each of four tree species were randomly chosen on the study plots for foliar sampling. The species were *Acer rubrum*, *Quercus prinus*, *Cornus florida* and *Rhododendron maximum*. Together, these species made up approximately 48% of the total basal area on the study plots (Table 2). Extensive sampling of WS7 8 years after clear-cutting indicated that these four species formed approximately 39% of above-ground biomass in regenerating chestnut oak communities (Boring & Swank 1986).

Leaves were collected throughout the 1985 growing season for foliar nutrient analysis. Ten mid-crown leaves were randomly sampled at each collection period. Plastic gloves were worn while placing leaves in paper sacks for transport to the laboratory. Leaves were photocopied and leaf areas were measured using a Microplan II image analysis system (Laboratory Computer Systems, Inc. 1983). Leaves were wiped clean of coarse deposited surface material with a moist tissue, and leaf petioles were removed before processing in order to obtain an accurate estimate of foliar nutrient contents per unit area of photosynthetic tissue. Samples were dried at

TABLE 2. Basal area and stem numbers for tree species on study plots in 1985, using stem diameters measured at 40 cm from the base of trees.

Species	Basal area (cm ² m ⁻²)	% Basal area	Stems (0.12 ha ⁻¹)	% Stems
<i>Quercus prinus</i>	2.2	18.7	155	5.3
<i>Acer rubrum</i>	1.8	15.0	313	10.7
<i>Cornus</i> spp.	1.5	12.8	342	11.7
<i>Kalmia latifolia</i>	1.6	13.2	1200	40.9
<i>Robinia pseudo-acacia</i>	1.3	10.5	79	2.7
<i>Quercus coccinea</i>	1.0	8.4	154	5.2
<i>Oxydendrum arboreum</i>	0.9	7.5	178	6.1
<i>Castanea dentata</i>	0.4	3.6	62	2.1
<i>Castanea prinus</i>	0.3	2.4	52	1.8
<i>Nyssa sylvatica</i>	0.2	1.9	71	2.4
<i>Carya</i> spp.	0.2	1.7	43	1.5
<i>Rhododendron maximum</i>	0.2	1.6	143	4.9
<i>Sassafras albidum</i>	0.1	1.1	65	2.2
<i>Liriodendron tulipifera</i>	< 0.1	0.2	9	0.3
Others	0.2	1.2	71	2.4
Total	12.0	100.0	2937	100.0

80°C for 48 h and weighed. Leaves were ground through a 20-mesh screen in a Wiley mill and composited by individual tree. Samples were stored in plastic bags at 4°C until digestion.

Sample collection and analysis

A storm was defined as a period of greater than 0.6 mm of rainfall, occurring between dry periods of at least 6 h duration (see Lovett & Lindberg 1984 for a similar definition). Twenty storms were sampled throughout the period July 1984–August 1986. Six dormant-season (November–April) storms and fourteen growing-season (May–October) storms were sampled. As nutrient cycling processes in forests may be misinterpreted if flux data are based solely on long-term bulk samplers (Richter & Lindberg 1988), precipitation, throughfall and stemflow samples were collected within 24 h after a storm event. Precipitation amounts (mm) were measured by three replicate rain gauges located at the top of the tower. Throughfall amounts were measured from thirty trough collectors in the field using plastic graduated cylinders. Subsamples (150 ml, whenever possible) were taken from all collectors for chemical analysis.

Sulphate, nitrate–N, ammonium–N, total ortho-phosphate and chloride were determined by Autoanalyzer (Technicon Industrial Systems 1970; McSwain 1973). Sample pH was measured using a Corning model 120pH meter and a calomel combination electrode. Total leaf nitrogen was assayed by Kjeldahl digestion and ammonium analysis using standard Autoanalyzer techniques (Technicon Industrial Systems 1970). Cation analyses were performed on a Perkin–Elmer 306 Atomic Absorption Spectrophotometer. Potassium, sodium, calcium and magnesium were determined with flame analysis under optimized conditions. Foliar cation concentrations were measured by ashing leaf tissue in a muffle furnace for 5 h at 400°C, followed by digestion in hot nitric acid.

As a quality control measure, all elemental concentrations were determined for Environmental Protection Agency (EPA) aqueous reference samples (EPA 1984), and for National Bureau of Standards (NBS) plant tissue reference material (SRM-1573; NBS 1981). All standard reference material concentrations were within the 10% range of accepted values. Elemental data were tested for significant regression coefficients using SYSTAT Multiple General Linear Hypothesis programs (Wilkinson 1986).

RESULTS

Temporal variability in net throughfall fluxes

Net throughfall element fluxes (NTF = throughfall minus precipitation fluxes; mequiv. m^{-2}) showed consistent canopy effects on rainfall chemistry (Table 3). In general, SO_4^{2-} , PO_4^{3-} , Cl^- , K^+ , Ca^{2+} and Mg^{2+} ions were added to rainfall by foliage, whereas the canopy absorbed NO_3^- –N, NH_4^+ –N and H^+ ions from precipitation.

Comparison of growing to dormant season fluxes indicates the influence of full canopy cover on the temporal variability in NTF. The ratio of growing-season to dormant-season (G/D) NTF was greater than unity for all ions except sodium (Table 3), although temporal variability in net throughfall chemistry was high in both

TABLE 3. Net throughfall transfers (throughfall-precipitation: mequiv. m^{-2} ; $n = 30$ collectors) for storms during the dormant season (November-April) and the growing season (May-October).

Season	SO ₄ ²⁻	NO ₃ ⁻ -N	NH ₄ ⁺ -N	PO ₄ ³⁻	Cl ⁻	K ⁺	Ca ²⁺	Mg ²⁺	Na ⁺	H ⁺
Dormant (D) ($n = 6$ storms)										
Mean	0.16	0.00	-0.02	0.01	0.07	0.13	0.08	0.05	0.01	-0.04
S.E.	0.16	0.02	0.02	0.01	0.04	0.09	0.02	0.03	0.01	0.04
C.V.(%)	232	> 1000	187	118	142	159	53	122	172	209
Growing (G) ($n = 14$ storms)										
Mean	0.67	-0.02	-0.05	0.03	0.10	0.19	0.18	0.08	0.00	-0.19
S.E.	0.46	0.03	0.02	0.01	0.05	0.04	0.04	0.02	0.02	0.09
C.V.(%)	237	433	159	104	173	78	87	84	> 1000	174
G/D quotient	4.5	11.5	2.6	2.8	1.4	1.5	2.2	1.7	0.0	4.5

dormant and growing seasons, indicated by coefficients of variation (C.V.) greater than 100% for most ions. Cation transfers (K⁺, Ca²⁺, Mg²⁺) showed the lowest temporal variability (C.V. 75-90%).

The importance of each of the hypothesized factors (Table 1) potentially accounting for the temporal variability between the net throughfall element fluxes in growing-season storms was evaluated independently by simple linear regression. The most important factors explaining temporal variability were related to storm event characteristics. Storm event amount (EA) alone explained a significant portion (> 60%) of NTF variability for all ions, and was the factor accounting for the greatest proportion of the between-storm variability in SO₄²⁻, Cl⁻, NO₃⁻-N, NH₄⁺-N, K⁺ and H⁺ ion fluxes (Table 4). Storm intensity (INT) was the most important factor explaining the NTF variability (> 80%) in PO₄³⁻ and Mg²⁺. Storm event duration (ED) accounted for the greatest part of the variability in Ca²⁺ fluxes. None of the factors accounted for a significant portion ($P < 0.05$) of the variability in Na⁺ net throughfall transfers.

It was also hypothesized that storm date (the number of days after 1 May) during the growing season influences the chemical composition of net throughfall (Table 1). Simple linear regression showed that a substantial portion of the temporal NTF variability (from 30% to 40%) of K⁺, Ca²⁺ and Mg²⁺ was explained by the storm date (Table 4).

In addition to storm characteristics and date, two factors related to canopy dry deposition fluxes explained a significant part of NTF temporal variability. Antecedent dry period (ADP) independently accounted for a significant portion (30-40%) of the variability in NTF of PO₄³⁻, K⁺, Ca²⁺ and Mg²⁺ ions (Table 4). Net throughfall fluxes of two elements with an important gaseous phase, S and N, were unrelated to ADP duration.

Surrogate surface dry deposition rates (DDR) provide an index of particle input to forest canopies (Hosker & Lindberg 1982). DDR was estimated for both growing and dormant seasons from Petri-dish extractions (Table 5). Sulphate showed the highest growing season DDR, followed by relatively high rates for NH₄⁺-N and Ca²⁺.

TABLE 4. Coefficients of determination (R^2) for simple linear regressions through the origin of mean net throughfall fluxes vs. temporal sources of variability for growing-season storms. All regressions based on data from fourteen storms, except for chloride and hydrogen with thirteen storms (each with an outlier in the data set).

Ion	EA	ED	INT	DATE	ADP	DDR
SO ₄ ²⁻	0.89**	0.23	0.43**	0.04	0.01	0.10
NO ₃ ⁻ -N	0.61**	0.05	0.17	0.02	0.00	0.04
NH ₄ ⁺ -N	0.81**	0.34*	0.34*	0.19	0.05	0.29*
PO ₄ ³⁻	0.66**	0.42**	0.83**	0.18	0.37*	0.58*
Ce ⁻	0.92**	0.44**	0.55**	0.18	0.10	0.13
K ⁺	0.76**	0.62**	0.74**	0.43**	0.34*	0.34*
Ca ²⁺	0.69**	0.72**	0.61**	0.28*	0.38*	0.51**
Mg ²⁺	0.78**	0.54**	0.81**	0.34*	0.38*	0.68**
H ⁺	0.88**	0.40*	0.58**	0.24		

* $P < 0.05$; ** $P < 0.01$.

EA = event amount; ED = event duration; INT = rainfall intensity; DATE = storm date; ADP = antecedent dry period; DDR = dry deposition rate.

Growing-season DDR preceding storms explained a substantial portion (30-70%) of the variability in growing-season NTF of PO₄³⁻, NH₄⁺-N, K⁺, Ca²⁺ and Mg²⁺ (Table 4).

Spatial variability in net throughfall fluxes

It appeared from the analysis of factors independently explaining the temporal variability in NTF that acid deposition interactions with the regenerating canopy could be modelled using multiple linear regression. Lovett & Lindberg (1984) described a method for determining canopy exchange and dry deposition rates in a

TABLE 5. Mean dry deposition rates ($\mu\text{equiv. m}^{-2} \text{h}^{-1}$) to inert surfaces during the dormant season (November–April) and the growing season (May–October), excluding periods during which no significant deposition to inert surfaces was measured.

Season	SO ₄ ²⁻	NO ₃ ⁻ -N	NH ₄ ⁺ -N	PO ₄ ³⁻	Cl ⁻	K ⁺	Ca ²⁺	Mg ²⁺	Na ⁺
Dormant (D)									
1985-86									
(n = 6)									
Mean	0.44	0.18	0.06	0.12	0.11	0.14	0.63	0.17	0.13
S.E.	0.03	0.06	0.04	0.05	0.01	0.04	0.25	0.07	0.17
C.V.(%)	17	72	128	84	23	66	88	85	134
Growing (G)									
1985 and 1986									
(n = 16)									
Mean	0.37	0.08	0.23	0.02	0.05	0.04	0.21	0.06	0.08
S.E.	0.06	0.02	0.04	0.00	0.02	0.01	0.05	0.01	0.02
C.V.(%)	65	97	68	42	130	69	84	64	129
G/D quotient	0.8	0.4	3.8	0.2	0.5	0.3	0.3	0.4	0.6

deciduous forest, using regression analysis of net throughfall fluxes. Assuming that canopy exchange fluxes are correlated with storm-event amount (EA; mm rain), and dry deposition fluxes are correlated with length of the antecedent dry period (ADP; h), the model

$$\text{NTF} = b_1\text{EA} + b_2\text{ADP} \quad (1)$$

can be used to separate canopy exchange and dry deposition inputs to net throughfall element fluxes (NTF; mequiv. m^{-2}). The rate coefficients, b_1 and b_2 , represent canopy exchange and dry deposition rates, respectively. Field studies (Schaefer & Reiniers 1990) empirically support the assumption that dry deposition wash-off is proportional to ADP and canopy exchange is proportional to EA. Model results are most reliable for water-soluble aerosol components deposited to leaf surfaces.

Lovett & Lindberg (1984) reported that the intercept term was not significantly different from zero for most ions ($P < 0.05$), and therefore was not included in the model. Consequently, R^2 represents the fraction of the sum of squares deviations about zero that can be explained by the model.

Numerous assumptions must be made in the application of regression analysis to mean storm net throughfall transfers. Among the most important of these assumptions are (i) storm amounts are sufficient to saturate the canopy, producing measurable throughfall amounts, and (ii) mean net throughfall transfers for individual storm events are normally distributed.

Verification of these two assumptions was attempted using data from fourteen growing-season storms (Table 3). With regard to canopy saturation, the relationship of throughfall amount (TA; mm throughfall) vs. storm EA (mm rain) was linear, even at low precipitation amounts. The equation for the relationship was

$$\text{TA} = 0.12 + 0.77\text{EA} \quad (2)$$

with $R^2 = 0.99$ ($n = 14$ storms). The intercept term was not significantly different from zero ($P < 0.05$). It was therefore assumed that all storms in the data set were of sufficient amount to saturate the canopy.

With regard to the second assumption, within-storm net throughfall transfers for thirty collectors were not, as a rule, normally distributed. Some degree of skewness or kurtosis, or both, was observed in the distributions of most net throughfall element transfers. Consequently, the regression model (Eqn 1) was applied to net throughfall transfers for each individual trough collector, to examine overall spatial patterns in net throughfall chemistry, and to separate canopy exchange and dry deposition transfers. Linear regression analysis using log-normally distributed data was thereby avoided for making inferences concerning the significance of rate constants (b_1 and b_2 , Eqn 1).

Having verified these major assumptions, the NTF model (Eqn 1) was applied to data from fourteen growing-season storms. Coefficients of determination (R^2) for storm net throughfall means were significant ($P < 0.01$) for all ions (Table 6; regressions using sodium transfers were not significant, and are not reported). Mean canopy exchange rates (b_1) were significant ($P < 0.05$; $rc = 30$ throughfall collectors) for all ions. Sulphate leaching rates appeared to be the most variable over space (C.V. = 187%), whereas calcium leaching and ammonium uptake rates were the least variable (C.V. = 48%). Canopy dry deposition rates (b_2) were significant ($P < 0.05$, $n = 30$ collectors) for PO_4^{3-} , Cl^- , NO_3^- -N, K^+ , Ca^{2+} , and Mg^{2+} ions (Table 6).

TABLE 6. Canopy exchange and dry deposition rate estimated by regression analysis* of net throughfall for individual collectors ($n=30$) during the growing season (May–October).

	SO ₄ ²⁻	NO ₃ ⁻ -N	NH ₄ ⁺ -N	PO ₄ ³⁻	Cl ⁻	K ⁺	Ca ²⁺	Mg ²⁺	H ⁺
Coefficients of determination (R^2)†	0.92	0.68	0.82	0.92	0.93	0.88	0.85	0.93	0.92
Canopy exchange rates ($\mu\text{equiv. m}^{-2} \text{ mm rain}^{-1}$)									
Mean	81.47	-4.11	-4.19	1.59	8.24	9.15	9.69	4.08	-16.51
S.E.	28.45	0.61	0.37	0.27	0.91	1.26	0.84	0.38	2.76
C.V. (%)	187	80	48	92	59	74	47	50	90
Dry deposition rates ($\mu\text{equiv. m}^{-2} \text{ h}^{-1}$)									
Mean	N.S.	0.26	N.S.	0.21	0.13	0.85	0.98	0.44	
SE	-	0.04	-	0.02	0.05	0.11	0.07	0.04	
C.V. (%)	-	75	-	54	184	69	39	44	
Dry quotient (C/IS)‡	-	3.7	-	9.0	2.5	17.5	4.6	9.0	

* Based on the model of Lovett & Lindberg (1984), and data from fourteen storms, except for chloride and hydrogen with thirteen storms (each with an outlier in the data set). N.S. = not significant at $P < 0.05$.

† R^2 values are based on mean storm net throughfall transfers. All regressions significant at $P < 0.01$.

‡ Ratio of canopy to inert surface (Table 5) dry deposition rates.

Among these ions, spatial variability in dry deposition was greatest for chloride (C.V. = 184%), and lowest for calcium (C.V. = 39%).

The ratio of canopy to inert surface (C/IS) dry deposition rates provides an index for scaling surrogate surface to foliar surface deposition estimates (Hosker & Lindberg 1982; Lindberg & Lovett 1985). The C/IS index was greater than unity for all ions except NH₄⁻-N (Table 6). Potassium showed the highest C/IS quotient, with dry deposition rates to the canopy about eighteen times greater than deposition to inert surfaces. Leaching during initial stages of a storm event may account for this large difference (Lovett & Lindberg 1984). The scaling factor (C/IS) for calcium (4.6) reported in this study was in the range of those previously reported for *Pinus*, *Quercus* and *Picea* stands in the south-eastern U.S.A. (1.3, 2.8 and 5.0, respectively; Lindberg *et al.* 1988).

Changes in the C.V. of canopy exchange and dry deposition rates as a function of additional number of throughfall collectors used were examined in an effort to separate the effect of sampling design from the true spatial variability in net throughfall fluxes. The C.V. for base cation (K⁺, Ca²⁺ and Mg²⁺) canopy exchange and dry deposition rates appeared to become relatively stable using a minimum of twelve randomly selected throughfall collectors (Fig. 1). Although variability in K⁺ transfers was consistently higher than variability in Mg²⁺ and Ca²⁺ transfers for a given number of collectors, all C.V. values stabilized at about the same level of sampling. Fluctuations in C.V. values for NO₃⁻-N and PO₄³⁻ stabilized at between thirteen and fifteen collectors, whereas the more highly variable SO₄²⁻ fluxes did not become relatively constant until twenty collectors were included.

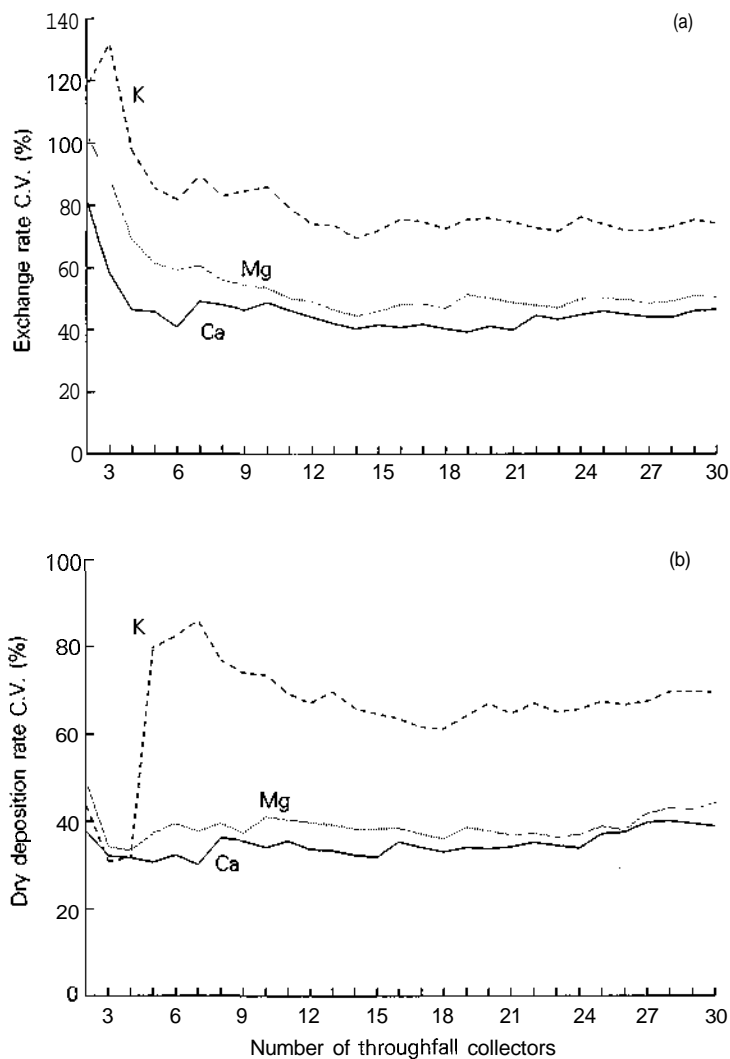


FIG. 1. Changes in (a) cation canopy exchange and (b) dry deposition rate coefficients of variation (C.V.) with the number of throughfall collectors used.

It was hypothesized that the spatial variability in both dry deposition and canopy exchange rates was chiefly related to structural characteristics of the vegetation, such as relative canopy cover or tree species frequency (Table 1). To test this hypothesis, relative canopy cover over each throughfall collector was estimated by collecting leaf litter in open troughs during November 1986. Litter turnover in troughs was checked by tagging three leaves with plastic tape and placing them, one on top of the other, in each collector. Twenty-three of thirty troughs had at least two of the three tagged leaves remaining at the end of the collection period. The overall retention efficiency for thirty troughs was 73% (66 of 90 leaves). In mid-November, litter was collected by trough, sorted by species, dried at 80°C for 24 h, and weighed.

The effect of relative canopy cover on the spatial variability of canopy exchange and dry deposition rates for twenty-three throughfall collector locations was investigated by simple linear regression through the origin, because, by definition, zero canopy cover implies zero net throughfall flux. Canopy cover was significantly correlated ($P < 0.05$) with canopy exchange for all ions (Table 7) and accounted for 50-80% of the variability in exchange rates of all ions except sulphate, which was more weakly related to canopy cover ($R^2 = 0.19$). Throughfall amount was negatively correlated with canopy cover ($P < 0.01$), suggesting that the litterfall collection method accurately reflected relative cover above each trough. Dry deposition rates at individual sampling locations were significantly correlated with relative canopy cover for PO_4^{3-} , $\text{NO}_3^- - \text{N}$, K^+ , Ca^{2+} and Mg^{2+} ions ($P < 0.01$; Table 7). Canopy cover accounted for 60-70% of the spatial variability in dry deposition of K^+ , Ca^{2+} and Mg^{2+} .

The average contribution of tree species to trough litter totals ($n = 23$ collectors) was highest for *Quercus prinus* (28%), followed by *Acer rubrum* (22%), *Quercus coccinea* (15%) and *Kalmia latifolia* (9%). These results are in general agreement with relative basal area estimates for tree species on the study plots (Table 2), and suggest that the litter collection method accurately reflected the influence of tree species on throughfall transfers.

Stepwise regression analysis was used to evaluate the influence of twelve tree species canopy exchange rates. Those species occurring most frequently in litter collections (*Quercus prinus*, *Acer rubrum* and *Quercus coccinea*) accounted for more than 50% of the spatial variability in canopy exchange rates (Table 8). The presence of these three species explained over 80% of the variability in Ca^{2+} and Mg^{2+} leaching rates within the canopy. *Kalmia latifolia* appeared to be important in foliar nitrogen absorption from rainwater.

Stemflow fluxes

Stemflow amounts comprised about 4.6% of incident rainfall amounts on an area basis for eleven growing-season storms. The influence of stemflow on total below-

TABLE 7. Coefficients of determination (R^2) for regressions of canopy exchange and dry deposition rates versus relative canopy cover ($n = 23$ collectors).

Ion	Relative canopy cover vs.	
	Canopy exchange rates	Dry deposition rates
SO_4^{2-}	0.19*	0.08
$\text{NO}_3^- - \text{N}$	0.65**	0.44**
$\text{NH}_4^+ - \text{N}$	0.80**	0.00
PO_4^{3-}	0.56**	0.62**
Cl^-	0.68**	0.25*
K^+	0.70**	0.62**
Ca^{2+}	0.77**	0.70**
Mg^{2+}	0.72**	0.67**
H^+	0.70**	
Throughfall amount	0.68**	

* $P < 0.05$; ** $P < 0.01$.

TABLE 8. Stepwise regressions of canopy exchange vs. percentage canopy cover by tree species. Models required a significance level of $P < 0.05$ to enter or remove a species ($n = 23$ collectors).

Ion	Steps	Species in model	Step R^2	P value
SO_4^{2+}	1.	<i>Robinia pseudo-acacia</i>	0.18	0.041
$\text{NO}_3^- - \text{N}$	1.	<i>Kalmia latifolia</i>	0.44	< 0.001
	2.	<i>Quercus coccinea</i>	0.72	< 0.001
	3.	<i>Sassafras albidum</i>	0.78	0.031
$\text{NH}_4^+ - \text{N}$	1.	<i>Acer rubrum</i>	0.45	< 0.001
	2.	<i>Kalmia latifolia</i>	0.68	< 0.001
	3.	<i>Quercus coccinea</i>	0.79	0.005
PO_4^{3-}	1.	<i>Acer rubrum</i>	0.45	< 0.001
	1.	<i>Quercus prinus</i>	0.53	0.004
Cl^-	2.	<i>Castanea dentata</i>	0.66	0.017
	3.	<i>Acer rubrum</i>	0.74	0.003
	4.	<i>Castanea prinus</i>	0.84	0.003
	1.	<i>Acer rubrum</i>	0.56	0.002
K^+	2.	<i>Quercus prinus</i>	0.67	0.012
	1.	<i>Acer rubrum</i>	0.56	< 0.001
Ca^{2+}	2.	<i>Quercus coccinea</i>	0.70	0.003
	3.	<i>Quercus prinus</i>	0.81	0.002
	1.	<i>Acer rubrum</i>	0.64	< 0.001
Mg^{2+}	2.	<i>Quercus coccinea</i>	0.79	0.033
	3.	<i>Quercus prinus</i>	0.83	< 0.001
	1.	<i>Quercus coccinea</i>	0.45	< 0.001
H^+	2.	<i>Quercus prinus</i>	0.72	0.001
	3.	<i>Oxydendrum arboreum</i>	0.79	0.006
	4.	<i>Kalmia latifolia</i>	0.84	0.025

All regressions based on data from fourteen storms except for chloride and hydrogen with thirteen storms (each with an outlier in the data set).

canopy element fluxes was estimated by regression analysis of net throughfall plus stemflow, using the model shown in Eqn 1. Inclusion of stemflow fluxes in the dependent variable of the model increased canopy exchange rates (b_1) of PO_4^{3-} and K^+ by more than 30% over exchange rates derived from net throughfall alone (Table 9). Canopy absorption rates of $\text{NH}_4^+ - \text{N}$ and H^+ decreased by more than 10% when stemflow fluxes were included in the model. Dry deposition rates (b_2) for all elements changed less than 10% with the inclusion of stemflow.

Magnitude of nutrient leaching

Autumn foliar nutrient burdens on the study plots were calculated from an average 1985 leaf biomass estimate of $314.8 \text{ g leaf m}^{-2}$ (derived from basal area plot surveys: Table 2, and biomass regression equations from Boring & Swank 1986). Leaf biomass was multiplied by maximum seasonal nutrient concentrations to calculate canopy nutrient pools (Table 10). Projected growing season leaching transfers were calculated from canopy exchange rates (Table 6) and total 1985 summer rainfall (748 mm). Comparisons of foliar leaching transfers to leaf nutrient burdens showed that K^+ was leached in greatest quantities from foliage, followed by Mg^{2+} , Ca^{2+} and PO_4^{3-} (Table 10). Foliage appeared to serve as a sink for N absorption from precipitation.

TABLE 9. Total below-canopy exchange rates (net throughfall and stemflow) during the growing season (May–October), estimated by regression analysis* of mean storm values ($n = 11$ storms).

	SO ₄ ²⁻	NO ₃ ⁻ -N	NH ₄ ⁺ -N	PO ₄ ³⁻	Cl ⁻	K ⁺	Ca ²⁺	Mg ²⁺	H ⁺
Coefficients of determination (R^2)	0.93	0.65	0.84	0.86	0.93	0.91	0.86	0.93	0.58
Exchange rates (uequiv. m ⁻² mm rain ⁻¹)									
Mean	102.40	-4.43	-3.58	2.34	10.32	12.74	10.41	5.07	-12.61
S.E.	9.16	1.09	0.54	0.39	0.96	1.65	1.87	0.58	4.18
% change†	26.7	7.8	-14.6	47.2	25.2	39.2	7.4	24.3	-23.6

All flux values significant at $P < 0.05$.

* Based on the model of Lovett & Lindberg (1984).

† Percentage change in exchange rates, compared to analysis of net throughfall only (Table 6).

TABLE 10. Projected growing season foliar nutrient burdens and leaching transfers from a regenerating forest canopy. All values are in units of g m⁻².

	N	P	K	Ca	Mg
Foliar burden*	3.75	0.38	1.98	3.78	0.64
Foliar leaching†	-0.09	0.04	0.26	0.14	0.04
% leached‡	-2.5	2.5	13.1	3.7	6.1

* Based on maximum 1985 foliar nutrient concentrations (3 November 1985).

† Based on canopy exchange rates from Table 6 and a growing season rainfall total of 748 mm for 1985. Nitrogen is expressed as the sum of NO₃⁻-N and NH₄⁺-N absorption rates.

‡ Projected percentage of the foliar nutrient burden leached during the growing season on Coweeta WS7.

DISCUSSION

Explaining net throughfall variability

~~Net throughfall fluxes in the regenerating forest were two to twelve times higher~~ during the growing season than during the dormant season, suggesting the importance of deciduous foliage in modifying precipitation chemistry (Mahendrappa 1974; Lovett & Lindberg 1984). Although dry deposition rates to inert surfaces were generally highest during the dormant season (Table 5), increased leaf area for dryfall interception probably balanced lower growing-season dry deposition rates measured to inert surfaces, and resulted in greater relative dry inputs to the full forest canopy during the summer months (Hosker & Lindberg 1982).

Storm characteristics (EA, ED and INT) accounted for the largest portion of the variability in growing season net throughfall fluxes, suggesting that canopy exchange was the major mechanism of throughfall transfer in the regenerating forest. Annual

canopy exchange, estimated by multiple linear regression of net throughfall element fluxes (Eqn 1) for the 1985 growing season, was greater than dry deposition wash-off for all ions (Table 11). Leaching contributed about 2.5 more K^+ and Cl^- , and about twice as much Ca^{2+} and Mg^{2+} , to forest-floor inputs than dry deposition wash-off.

Base cation (K^+ , Ca^{2+} and Mg^{2+}) net throughfall fluxes increased significantly over the course of the growing season. One explanation for this pattern is that increasing leaf damage, through weathering (Tukey 1966), herbivory (Seastedt, Crossley & Hargrove 1983; Boerner 1984), or disease, induced cation leaching from foliage. Alternately, higher dry deposition rates in early autumn (Swank & Henderson 1976) may contribute to increasing net throughfall fluxes over the growing season.

Hydrogen ion exchange generally accounts for about 40-60% of base cation leaching from deciduous canopies in aggrading forest ecosystems (Lovett *et al.* 1985). Regression of mean net throughfall transfers for thirteen growing-season storms on WS7 showed that hydrogen ion uptake accounted for 77% of the between-storm, temporal variability in K^+ net throughfall transfers, 69% of the variability in Ca^{2+} transfers, and 79% of the variability in Mg^{2+} transfers. Based on net throughfall data, about 2 mequiv. of H^+ ion is absorbed by the canopy for each mequiv. of K^+ and each mequiv. of Ca^{2+} added to throughfall, whereas about 4 mequiv. of H^+ ion is absorbed for each mequiv. of Mg^{2+} added to throughfall. The sum of base cation canopy exchange rates, estimated by multiple linear regression (Eqn 1) for thirty throughfall collectors, was $22.9 \text{ mequiv. m}^{-2} (\text{mm rain})^{-1}$, compared to $16.5 \text{ mequiv. m}^{-2} (\text{mm rain})^{-1}$ of hydrogen ion absorbed from precipitation (Table 6). It appears that hydrogen ion exchange accounts for about 70% of combined base cation leaching from the regenerating deciduous canopy at Coweeta, and that passive diffusion of K^+ and Mg^{2+} from leaf tissue to rainwater probably accounts for the remaining cation leaching fluxes.

Sulphate net throughfall transfers were highly correlated with EA, but not with ADP. Results from other studies in the south-eastern United States, however, showed that dry deposition provides more than 85% of the enrichment of sulphate in throughfall fluxes at low-altitude sites, whereas foliar leaching contributed only a small percentage (5%) of the sulphate throughfall enhancement (Lindberg & Garten 1988). One explanation for the sulphate flux patterns on WS7 is the leaching of previously absorbed sulphur dioxide gas (Bache 1977) resulting in unanticipated dry deposition contributions to canopy exchange rates and misinterpretation of the model coefficients b_1 and b_2 (Eqn 1). Although the regression model of Lovett & Lindberg (1984) can be used directly for estimation of some element fluxes, uncertainty in the behaviour of dry-deposited SO_2 suggests that tracer methods may be more accurate (Lindberg *et al.* 1988; Lindberg & Garten 1988).

An additional explanation is that sulphate dry deposition at Coweeta originates from a remote source, and is highly episodic in nature, leading to a lack of correlation between ADP and net throughfall fluxes during a given storm. Ambient levels of SO_4^{2-} appear to vary considerably at Coweeta, whereas SO_2 concentrations remain fairly constant (Swank & Reynolds 1987). Episodes of high SO_4^{2-} dry inputs during short dry periods (< 48 h) would therefore result in an inverse relationship between ADP and SO_4^{2-} net throughfall fluxes (Table 6), because actual dry deposition rates would often be highest during relatively short dry periods. This hypothesis was supported by a negative correlation between 1985 inert surface sulphate dry deposition

rates and the duration of the antecedent dry period ($R^2 = 0.42$; $n = 8$). Evidence from several forests in the eastern U.S. indicate that total atmospheric deposition of sulphate is strongly reflected in the measured flux of throughfall (Lindberg & Garten 1988).

In contrast to sulphate NTF, the positive correlations of dry condition characteristics (ADP and DDR) with net throughfall fluxes of PO_4^{3-} , $\text{NH}_4^+ - \text{N}$, K^+ , Ca^{2+} and Mg^{2+} suggest a local source of large particulate dry deposition to the canopy. Internal canopy redistribution of vegetation or soil-derived elements may explain dry deposition of these ions (Gosz 1980; Lovett & Lindberg 1984).

Spatial variability in canopy exchange and dry deposition rates were high in the regenerating canopy. Most of this variability (60-80%), however, could be explained by relative foliage cover at sampling locations. Although recovery of leaf area index on WS7 has been rapid (Boring & Swank 1986), relative canopy cover was still patchy (C.V. = 55% for twenty-three leaf collectors). The frequency of *Quercus* spp. and *Acer rubrum* leaves above throughfall collectors accounted for a significant portion of the spatial variability of net throughfall transfers for all ions except sulphate (Table 8). These two species alone comprised over 30% of the total basal area on our study plots. Because leaf biomass is highly correlated with stem diameter on WS7 (Boring & Swank 1986), heterogeneity in foliage cover of abundant tree species appeared to account for most of the spatial variability in net throughfall fluxes.

Changes in spatial canopy exchange and dry deposition C.V.s with collector number (Fig. 1) suggested that the true spatial variability in net throughfall fluxes exceeded our sampling error when using more than about twelve throughfall collectors. Canopy exchange C.V. for Ca^{2+} and Mg^{2+} stabilized at values close to the canopy cover C.V. of 55%, whereas K^+ canopy exchange C.V. stabilized at about 74%, implying that foliar K^+ leaching is more variable over space than would be predicted solely on the basis of leaf cover heterogeneity.

Canopy N transformations

Nitrification of ammonium on leaf surfaces has been hypothesized to account for apparent $\text{NH}_4^+ - \text{N}$ uptake by forest canopies (Chen *et al.* 1983). To test this hypothesis, the relationship between ammonium uptake and nitrate enrichment of throughfall was examined. If nitrification occurs, net throughfall $\text{NO}_3^- - \text{N}$ fluxes should increase as $\text{NH}_4^+ - \text{N}$ fluxes decrease. Regression of canopy exchange rates for thirty collectors showed that $\text{NH}_4^+ - \text{N}$ uptake was positively correlated with $\text{NO}_3^- - \text{N}$ uptake ($R^2 = 0.80$; $P < 0.01$). These results do not support the nitrification hypothesis, and instead suggest that the forest canopy is absorbing both $\text{NH}_4^+ - \text{N}$ and $\text{NO}_3^- - \text{N}$ from precipitation.

Canopy uptake of inorganic nitrogen from rainwater appears to be a common phenomenon in deciduous forests (Verry & Timmons 1977; Olson, Reiners & Cronan 1981; Lovett & Lindberg 1984). In the context of ecosystem N fluxes, however, canopy N uptake (about 0.1 g N m^{-2} ; this study) is small in regenerating southern Appalachian forests compared, for example, to accretion by woody symbiotic N-fixing species (4.8 g N m^{-2} in 4-year-old forest stands; Boring & Swank 1984). In more-mature forest stands, however, canopy N uptake from rainwater may be greater.

Importance of stemflow fluxes

The addition of stemflow fluxes to net throughfall fluxes for regression analysis increased total below-canopy element transfers of SO_4^{2-} , PO_4^{3-} , Cl^- , K^+ and Mg^{2+} by more than 20%, suggesting that, for certain mobile ions, stemflow is an important pathway of element input to the soil in the regenerating forest. Inclusion of stemflow in the model decreased canopy absorption rates of NH_4^+ and H^+ ions per m^2 of forest, indicating that, unlike leaf surfaces, very little ion exchange occurs on bark surfaces.

Stemflow contributions to total below-canopy fluxes of water and dissolved nutrients average about 12%, depending on the element and stand type (Parker 1983). Although proportional stemflow nutrient inputs to the forest floor are low, stemflow effects on soils immediately surrounding tree boles can be substantial (Zinke 1962; Gersper & Hollowaychuk 1970).

Small-scale stemflow inputs may be especially important in an early successional forest, because the number of stems ha^{-1} on WS7 increased from about 3000 (>2.5 cm dbh) before clear-cutting (Boring, Monk & Swank 1981) to over 80000 stems ha^{-1} 2 years after disturbance (Boring & Swank 1986).

Throughfall in a regenerating forest

Annual growing season dry deposition transfers in the regenerating forest (Table 11) were about two to five times lower than dry deposition transfers (6.4 mequiv. $\text{NO}_3^- - \text{N m}^{-2}$, 25.1 mequiv. $\text{K}^+ \text{m}^{-2}$ and 14.0 mequiv. $\text{Ca}^{2+} \text{m}^{-2}$) to a more mature forest canopy at Oak Ridge, TN (Lovett & Lindberg 1984). This pattern may reflect the difference in proximity to air pollution sources between Coweeta and Oak Ridge (Swank & Henderson 1976), and higher LAI for dryfall interception in mature forests.

Annual growing season canopy exchange fluxes in the regenerating forest (Table 11) were about four times lower for K^+ , and two times lower for Ca^{2+} , than canopy exchange fluxes (25.1 mequiv. $\text{K}^+ \text{m}^{-2}$ and 14.0 mequiv. $\text{Ca}^{2+} \text{m}^{-2}$) in a more mature chestnut oak canopy (Lovett & Lindberg 1984). Similarly, foliar cation leaching losses in the early successional forest (Table 10) accounted for 4-13% of leaf nutrient reserves, and were more conservative estimates of the importance of leaching in forest nutrient cycles than reported in a review (Parker 1983), which found that cation leaching contributions to total yearly nutrient return in more mature forests generally range from 10 to 65%.

There are several possible reasons for the differences between leaching estimates from this study and those derived from previous studies.

(i) In this study, dry deposition was separated from leaching fluxes by statistical analysis. Conversely, commonly reported bulk throughfall measurements include dry deposition inputs (Parker 1983), and consequently, overestimate leaching transfers.

(ii) Results from this study predict leaching fluxes for a regenerating forest canopy, rather than for a more mature, aggrading stand. Leaf area index on WS7 had returned to 85% of pre-cut levels by 1985 (Boring & Swank 1986), but recovery of the canopy was incomplete, leading to lower area-based leaching rates (Lemee 1974; Parker 1983).

TABLE 11. Annual canopy exchange and dry deposition inputs to the forest floor of a regenerating forest during the growing season at Coweeta. All values are in units of mequiv. m^{-2} .

	SO ₄ ²⁻	NO ₃ ⁻ -N	NH ₄ ⁺ -N	PO ₄ ³⁻	Cl ⁻	K ⁺	Ca ²⁺	Mg ²⁺	H ⁺
Ion exchange†	60.9	-3.1	-3.1	1.2	0.6	6.8	7.2	3.1	-12.3
Dry deposition‡	**	1.1	**	0.9	0.5	3.5	4.1	1.8	

† Based on canopy exchange rates from Table 6, and a 1985 growing-season rainfall total of 748 mm.

‡ Based on dry deposition rates from Table 6 and a 1985 growing-season dry period total of 4160 h.

** Model coefficient was not significant at $P < 0.05$.

(iii) Nutrients are tightly cycled in regenerating forests, through high productivity (Marks & Bormann 1972; Boring, Monk & Swank 1981) and autumn resorption (Ryan & Bormann 1982; Potter, Ragsdale & Berish 1987). The readily leachable pool of elements in foliage of early successional plants may be lower than in leaves of more-mature forest trees, due to rapid nutrient uptake and assimilation into tissues of coppice vegetation (Tukey 1970). Further work is needed to elucidate specific metabolic processes involved in foliar nutrient retention under conditions of acid-rain leaching.

This study illustrates the important contribution of foliar leaching to throughfall nutrient fluxes in a regenerating hardwood forest. Rapid, patchy recovery of leaf cover on a clear-cut watershed has produced a mosaic pattern of throughfall and stemflow inputs to the forest floor. Cation leaching fluxes are highly variable over small spatial scales, but the variability can be explained largely by heterogeneity in canopy cover and variation in hydrogen ion uptake from precipitation. Because cation throughfall transfers were highest during storms with the greatest hydrogen ion uptake from rainwater, it is hypothesized that acid deposition is causing accelerated foliar nutrient leaching in southern Appalachian hardwood forests.

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