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ELEVATIONAL TRENDS IN THE FLUXES OF SULPHUR AND NITROGEN IN THROUGHFALL IN THE SOUTHERN APPALACHIAN MOUNTAINS: SOME SURPRISING RESULTS

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Abstract. From 1986-1989, a team of scientists measured atmospheric concentrations and fluxes in precipitation and throughfall, and modeled dry and cloudwater deposition in a spruce-fir forest of the Great Smoky Mountains National Park which is located in the Southern Appalachian Region of the United States. The work was part of the Integrated Forest Study (IFS) conducted at 12 forests in N. America and Europe. The spruce-fir forest at 1740 m consistently received the highest total deposition rates (~2200, 1200, and 700 eq ha⁻¹ yr⁻¹ for SO₄²⁻, NO₃⁻, and NH₄⁺). During the summers of 1989 and 1990 we used multiple samplers to measure hydrologic, SO₄²⁻, and NO₃⁻ fluxes in rain and throughfall events beneath spruce forests above (1940 m) and below (1720 m) cloud base. Throughfall was used to estimate total deposition using relationships determined during the IFS. Although the SO₄²⁻ fluxes increased with elevation by a factor of ~2 due to higher cloudwater interception at 1940 m, the NO₃⁻ fluxes decreased with elevation by ~30%. To investigate further, we began year round measurements of fluxes of all major ions in throughfall below spruce-fir forests at 1740 m and at 1920 m in 1993-1994. The fluxes of most ions showed a 10-50% increase with elevation due to the ~70 cm yr⁻¹ cloudwater input at 1920 m. However, total inorganic nitrogen exhibited a 40% lower flux in throughfall at 1920 m than at 1740 m suggesting either higher dry deposition to trees at 1740 m or much higher canopy uptake of nitrogen by trees at 1920 m. Differential canopy absorption of N by trees at different elevations would have significant consequences for the use of throughfall N fluxes to estimate deposition. We used artificial trees to understand the foliar interactions of N.

Key Words: atmospheric deposition, high elevation forests, foliar uptake, cloudwater, nitrogen, sulphur

1. Introduction

The Integrated Forest Study (IFS) developed uniform protocols for sampling and analyzing acid deposition and nutrient cycling in over a dozen sites located in the United States, Canada, and Norway. The study showed that high-elevation southern Appalachians forest stands received some of the highest of sulphate and nitrate loadings relative to all of the other IFS study sites (Johnson and Lindberg, 1992). The Smoky Mountain Tower site (ST) located at 1740 m in the Noland Divide Watershed (NDW) received an average of 2200, 1200, and 700 eq ha⁻¹ yr⁻¹ for SO₄²⁻, NO₃⁻, and NH₄⁺, respectively, in total deposition (Johnson and Lindberg, 1992) (from 4/86 through 3/89).

Elevation plays a critical role in the amount of acid deposition measured in throughfall (TF) within the NDW (Lindberg and Owens, 1993). Lindberg and Owens (1993) found that an increase from ~1720 to ~1940m in elevation corresponded to a two-fold increase in water and SQ²⁻ fluxes measured in TF. This increase was attributed to the enhancement of cloudwater interception at the upper site. During the IFS, cloud base was observed to be at or above ~1800m within the watershed (Johnson and Lindberg, 1992). Because cloudwater can exert a great influence on total deposition fluxes over small elevational gradients (Lovett *et al.*, 1982), sampling site location and

elevation can greatly affect depositional flux estimates even within small catchments (Lindberg and Owens, 1993).

Here we report on the influence that elevation has on TF fluxes within the spruce-fir communities in the southern Appalachian Mountains. Using relationships formulated during the IFS, we estimate the annual cloud water input and compute annual net canopy exchange (NCE) of nitrogen (N) at two elevations and assess the importance of foliar uptake on nitrogen flux through the canopy.

2. Sites and Methods

Precipitation and TF sampling stations were located in the 17.4 ha. Noland Divide Watershed (NDW) (35°34'N, 83°28'W) in the Great Smoky Mountains National Park, North Carolina. The 1740-m elevation station (the lower site), was originally established in 1986 as part of the IFS to monitor ion fluxes in deposition. The upper site at 1920-m was established in the summer of 1993 to monitor the influence of cloud deposition to the upper portions of the catchment.

The overstory vegetation of the area is dominated by red spruce (*Picea rubens* Sarg) and interspersed patches of standing dead Fraser-fir *Abies fraseri* (Pursh) Poir.) which has been devastated by the infestation of the balsam woolly adelgid *Adelges piceae* Ratz.). A complete description of the sites can be found in Johnson and Lindberg (1992), Johnson *et al.* (1991), and Nodvin *et al.* (1995).

Precipitation and TF samples were collected from 3-Aug-93 to 3-Aug-94. Precipitation chemistry was sampled using an Aerochemetrics automatic wet-only collector located in natural gaps adjacent to canopy covered TF plots. During the freeze-free season (1-May until 31-Oct), TF was collected in 1 liter polyethylene bottles with polyethylene funnels of 3.5 cm diameter. Eight TF collectors were randomly placed beneath the canopy of mature red spruce and volume composited to account for the variability in TF. From November 1 to April 31, TF was collected at each site in 4 large-diameter plastic lined buckets located on platforms ~1 m from the ground and volume composited. Precipitation and TF volumes were measured using wedge-type rain gauges located adjacent to each TF collector in the freeze-free season or by weighing the collection buckets during the winter season. Sampling methodologies followed IFS protocols as given in Lindberg *et al.*, (1989).

Samples were collected twice weekly during the freeze-free period and were volume-composited for weekly totals. During winter, samples were collected weekly. The samples were analyzed for pH and conductivity immediately upon return from the field (within 24 hours), preserved with chloroform (10 μ l per 30 ml sample), and stored at 4°C until analysis for major ions (using ion chromatography).

In addition, from June 23 to October 25, 1993, samples were collected beneath artificial trees located in adjacent forest gaps at 1700m and 1900m within the watershed. The use of inert artificial surfaces such as 'Christmas trees' can give a good estimate of total N and S deposition (Joslin *et al.*, 1990).

3. Results and Discussion

3.1 Water Flux

During the collection period, the amount of wet precipitation entering both sites was similar (Table I). The upper site received a slightly lower volume of precipitation which we attribute to a lower catch efficiency of the rain sampler during snow periods due to its ridge-top location. Both sites received a significantly higher amount of rain compared to the three year average collected during the IFS (Table I). However, the average weighted mean concentrations at the lower site for all the major

Table I. Throughfall (TF) and precipitation deposition of SO_4^{2-} , NO_3^- , and NH_4^+ , to the Noland Divide Watershed expressed as eq ha yr^{-1} . The Lower Site and the IFS Site are the same.

	IFS Site (4/86-3/89)	Lower Site (8/93-7/94)	Upper Site (8/93-7/94)
Precipitation (cm)	203	298	281
SO_4^{2-}	596	740	770
Throughfall (cm)	215	288	342
SO_4^{2-}	2470	2480	3500
NO_3^-	866	1230	860
NH_4^+	220	410	310
Net Throughfall for SO_4^{2-}	1870	1740	2730

Net Throughfall = Throughfall - precipitation ; IFS Site and Lower Site are at the same location

ions were very similar for the two periods (data not shown). There was a much greater volume of TF at the upper site versus the lower sampling site, suggesting increased cloudwater input with elevation (Table I). Lindberg and Owens (1993) also reported similar hydrologic trends with increasing elevation at NDW during the summer of 1989.

Positive hydrologic fluxes in net-throughfall (NTF) indicate the presence of measurable amounts of cloudwater input (Lovett *et al.*, 1982) (NTF = the flux in TF minus precipitation). By using all the positive NTF values, we estimate that the lower and upper sites received ~20 and 90 cm yr^{-1} of cloudwater deposition, respectively (Table II). Another way to calculate cloudwater input is to use “conservative” tracers of convenience such as Cl^- , SO_4^{2-} , and Na^+ in TF (Lovett *et al.*, 1982). By combining the results of all methods, we estimate average cloudwater inputs to the lower and upper site of ~40 and ~70 cm respectively (Table II).

Table II. Cloud water volume estimates (cm) at the Noland Divide Watershed during 8/93-7/94

	Lower Site	Upper Site
NTF*	19	89
Cl^- **	43	62
SO_4^{2-} **	47	87
Na^+ **	43	53
Average	40 cm	70 cm

* This estimate is based on positive net throughfall adjusted for evaporation and interception losses (Lindberg and Owens, 1993). ** The estimates for Cl^- , SO_4^{2-} , and Na^+ are based on relationships formulated during the IFS.

3.2 Sulphate Deposition

For the year, there was no significant difference between the upper and lower sites in the amount of sulphate entering the watershed as wet deposition ($p < 0.05$, $n = 48$) (Table I). However, the annual sulphate flux measured in TF was ~40% greater at the upper site and was significantly greater across the entire sampling period ($p < 0.05$, $n = 48$) (Figure 1). Since TF represents a direct estimate of the total atmospheric deposition of sulphate entering a watershed and foliar leaching and/or uptake is minimal (Garten *et al.*, 1988, Lindberg and Garten, 1988), the difference in fluxes between the two sites can be attributed to an increase with elevation of cloud and/or dry deposition. The IFS reported that cloud water represented 45-50% of the total sulphate loading

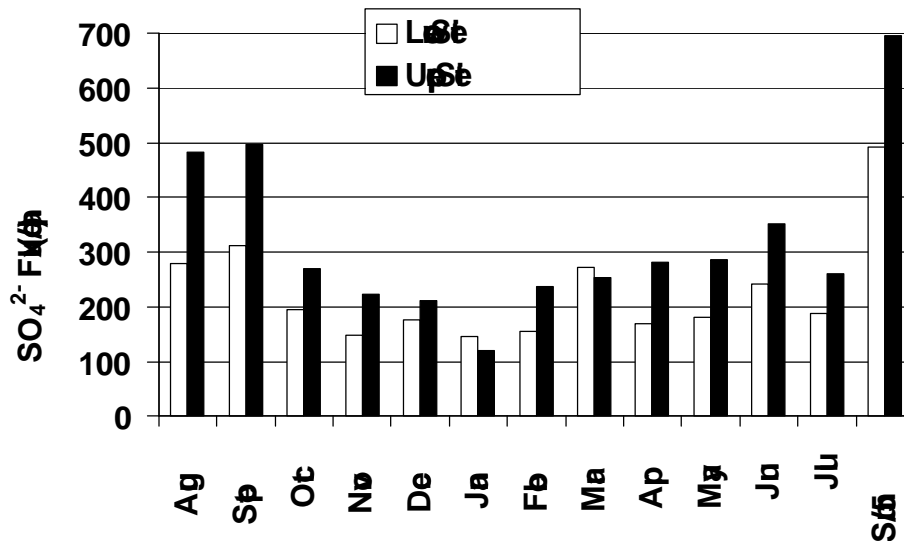


Fig. 1. Monthly total sulphate fluxes based on spatially and temporally composited weekly samples entering the NDW as throughfall. The collections are from 8/93-7/94.

(Lindberg and Lovett, 1992). We estimated cloudwater inputs of sulphate to the lower and upper sites based on the cloudwater inputs in Table II and cloudwater concentrations measured during the IFS (Johnson and Lindberg, 1992). Our cloudwater estimates are 950 and 1800 eq ha⁻¹ yr⁻¹ and represented ~40 and ~50% of the total deposition in TF to the lower and upper sites, respectively.

Nitrogen Deposition

Unlike sulphate, the fluxes of NO₃⁻ and NH₄⁺ entering as TF were significantly lower ($p < 0.05$, $n = 48$) at the upper site (Figure 2). Lindberg and Owens (1993) noted a similar trend for NO₃⁻ in TF within this watershed. They found that NO₃⁻ fluxes at their lower site (at 1720-m located ~80-m from our lower site) were 30% higher than their upper site (at 1940-m located ~200-m from our upper site). This trend is unusual because one would expect the enhancement of cloudwater at the upper site to also increase the total deposition of N. Since cloudwater contains elevated concentrations of NO₃⁻ and NH₄⁺ (Lovett *et al.*, 1982) and we have determined that the upper site receives 75% more cloud deposition than the lower site, why do we not see higher N fluxes reflected in TF? There are two possible reasons for this “inverse” elevational difference: (1) dry deposition of N is actually much higher at the lower site, and/or (2) foliar N

uptake is much greater at the upper site. Uptake may be estimated from net canopy exchange, defined as “NCE= TF -total deposition” (Johnson and Lindberg, 1992). Although we did not directly measure N dry deposition during the 1993-94 sampling period, we do not expect dry deposition to have been greater at the lower site based on the estimated total sulphate fluxes discussed above and the historical IFS dry deposition data. If anything, we would expect somewhat greater dry deposition to trees at the upper site due to higher wind speeds associated with the ridge-top location and the more open canopy at 1920 m (Lovett and Kinsman, 1990). The lower and upper sites are estimated to have approximately a 70 and 50% canopy closure, respectively (Shubzda, unpublished data). The more canopy openings at the upper site would increase the “edge effect” which is known to enhance both cloud and dry deposition to individual trees (Lindberg and Owens, 1993).

An independent estimate of total inorganic N deposition to these two sites supports the expected trend of increasing N deposition with increasing elevation. Total inorganic N deposition collected under artificial ‘Christmas trees’ located in canopy gaps near ground level within the NDW was ~2 times greater at 1900m than 1700m.

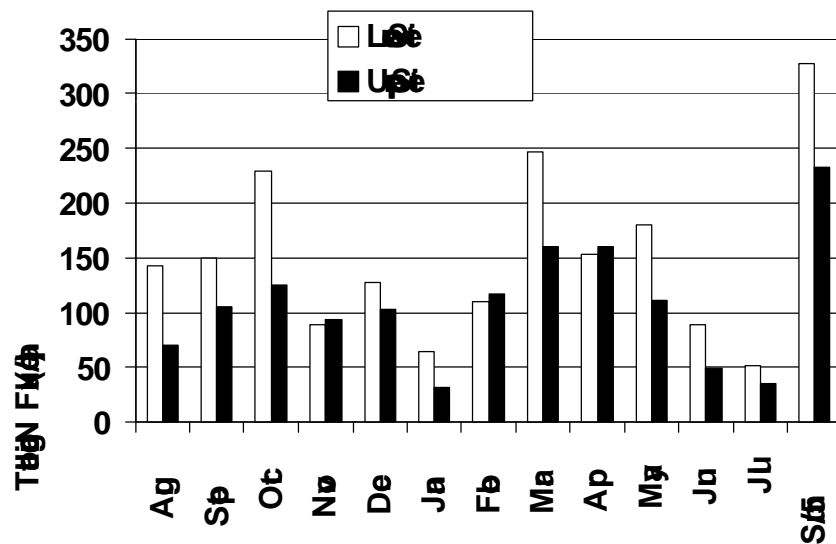


Fig. 2. Monthly total inorganic N fluxes in throughfall entering the NDW.

The NH_4^+ flux under the artificial trees showed a large increase (by an order of magnitude) with elevation, while the NO_3^- flux was actually slightly higher at the lower site. We believe that the greater in NH_4^+ flux with elevation is attributable to enhancement of cloud deposition at the upper site but that the trend in NO_3^- can only be explained by somewhat higher dry deposition at the lower site (cf. Lindberg & Owens, 1993). Nevertheless, the artificial tree results support our belief that total N deposition is greater at the upper versus the lower site.

For comparison with the TF flux, we estimated the total inorganic N deposition to the lower and upper sites to be 1950 and 2550 $\text{eq ha}^{-1} \text{yr}^{-1}$, respectively. These estimates are based upon: (1) measured wet deposition rates, (2) the assumption that the IFS 3-year mean dry deposition rates are reasonable approximations for both sites, and (3) calculated N flux in cloud deposition. Compared to the N flux in TF, these estimates

of total N fluxes suggest a canopy uptake rate at the upper site that exceeds that at the lower site by a factor of four (i.e. NCE of -310 and -1380 eq ha¹ yr⁻¹ for the lower and upper sites, respectively). Our estimates are based in part on the assumption that both dry deposition and cloudwater concentrations of N measured during the IFS from 1986-1989 can be applied to these same sites during 1993-1994. Our assumption is supported by the similar SO₄²⁻ fluxes measured in TF at the lower site measured during both periods (Table1).

Conclusions

In general, total deposition increases with increasing elevation (Lovett and Kinsman, 1990). This trend was expected because of the increase in cloudwater input entering the upper reaches of the NDW. The hydrologic and sulphate fluxes in TF support the enhancement of cloudwater input to the upper site. However, the total inorganic N flux in TF was lower at the upper site. Our analysis suggests that the observed difference in TF N flux was due to greater canopy uptake at 1920 m relative to 1740 m elevation in the Noland Divide Watershed. The reasons for the higher apparent uptake are unclear but site specific differences in canopy uptake of N in would confound the use of TF to estimate total N deposition in montane forests.

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References

- Garten, C.T., Bondietti, E.A., Lomax, R.D.: 1988, *Atmospheric Environment*, **22**, 1425-1432.
- Johnson, D.W., and Lindberg, S.E. (Eds): 1992, "Atmospheric deposition and nutrient cycling in forest ecosystems", Springer-Verlag, New York.
- Johnson, D.W., Van Miegroet, H., Lindberg, S.E., Todd, D.E., and Harrison, R.B.: 1991, *Canadian Journal of Forest Research*, **21**, 769-787.
- Joslin, J.D., Mueller, S.F., and MH Wolfe.: 1990, *Atmospheric Environment*, **24A**, 3007-3019.
- Lindberg, S.E., Johnson, D.W., Lovett, G.M., Taylor, G.E., Van Miegroet, H., and Owens, J.G.: 1989, *ORNL/TM*, **11214**, Oak Ridge National Laboratory, Oak Ridge, TN.
- Lindberg, S.E., and Garten, C.T.: 1988, *Nature*, **336**, 148-151.
- Lindberg, S.E., and Lovett, G.M.: 1992, *Atmospheric Environment*, **26A**, 1477-1492.
- Lindberg, S.E., and Owens, J.G.: 1993, *Biogeochemistry*, **10**, 175-194.
- Lovett, G.M., and Kinsman, J.D.: 1990, *Atmospheric Environment*, **24A**, 2767-2786.
- Lovett, G.M., Reiners, W.A., and Olson, R.K.: 1982, *Science*, **218**, 1303-1304.
- Nodvin, S.C., Van Miegroet, H., S.E. Lindberg, N.S. Nicholas, and D.W. Johnson.: 1995, *Water, Air and Soil Pollution*, this volume.