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Canopy uptake of N deposition in spruce (*Picea abies* L. Karst) stands

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Abstract – In order to quantify the total deposition and canopy uptake of N in spruce, plastic Christmas trees were established next to real spruce trees of approximately similar height in clearings of the Strengbach catchment (Vosges mountains, France). Bulk precipitation and throughfall (TF) composition under the artificial trees, the real spruce trees, and in 3 spruce stands aged 15, 35 and 90 years were monitored during 8.5 months. Fluxes of inorganic N, SO_4^{2-} and Na^+ in throughfall below plastic trees were 50% (N) to 30% (Na^+) higher to those in bulk precipitation, whereas the flux of water was 30% lower. Net throughfall (NTF) fluxes of NH_4^+ were higher below the plastic trees than below the real trees, whereas the reverse was true for NO_3^-. The ratio between total inorganic nitrogen and Na^+ (or SO_4^{2-}) in NTF was higher below plastic trees than below closed spruce stands. Assuming that foliar leaching of Na^+ (or SO_4^{2-}) was negligible, the ratios between Na^+ (or SO_4^{2-}) fluxes in net throughfall under living and plastic canopies were used as deposition indexes, and dry + occult deposition of N was computed for the real spruce trees in the clearings as well as in the 3 closed stands. Results confirmed a significant canopy uptake of dry + occult deposited N and especially NH_4^+ by spruce stands.

dry deposition / nitrogen / throughfall / net throughfall / *Picea abies*

Résumé – Absorption foliaire des dépôts azotés par des peuplements d'épicéa. Afin de quantifier le dépôt et l'absorption foliaire d'azote dans des peuplements d'épicéa (Picea abies L. Karst), nous avons placé des arbres de Noël en plastique à coté de jeunes épicéa de taille identique dans deux clairières du bassin versant du Strengbach. Les pluies à découvert et les pluviolessivats sous les vrais et faux épicéas, et dans trois peuplements fermés de 15, 35 et 90 ans ont été mesurés pendant 8,5 mois. Les flux d'azote minéral, de sulfate et de sodium dans le pluviolessivat récolté sous les arbres artificiels étaient 50 % (N) à 30 % (Na^+) supérieurs à ceux mesurés à découvert pour un flux d'eau 30 % inférieur. Le flux net d' NH_4^+ (pluviolessivat moins pluie) sous les arbres artificiels était supérieur à celui sous les arbres réels, mais inférieur pour le NO_3^-. Le rapport N minéral/ or SO_4^{2-} (ou Na^+) dans le pluviolessivat net sous les arbres artificiels était supérieur à celui mesuré sous les peuplements fermés. En admettant qu'absorption et lessivage foliaire de Na^+ et SO_4^{2-} sont négligeables, nous avons utilisé le rapport entre le pluviolessivat net de Na^+ (ou or SO_4^{2-}) sous arbres (ou peuplements) réels et artificiels comme un index de dépôt occulte, et le rapport N minéral/ Na^+ (ou or SO_4^{2-}) dans le pluviolessivat net sous les arbres artificiels comme un caractéristique intrinsèque du dépôt occulte. Ceci nous a permis de calculer le dépôt occulte d'azote minéral et l'absorption foliaire dans les différents peuplements étudiés. Les résultats confirment une absorption foliaire d'azote, et plus particulièrement d'NH_4^+, très significative pour la nutrition des peuplements forestiers.

dépôt sec / azote / pluviolessivat / secrétion / *Picea abiès*

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1. INTRODUCTION

N-compounds from atmospheric pollution may be deposited on forest canopies in different forms: (a) Wet deposition. Away from point sources of pollution, the ratio (eq.eq^{-1}) between NO_3^- and NH_4^+ in wet deposition in the Vosges mountains is close to 1 [6], (b) dry deposition.on leaf surface or on the water film covering needles in wet periods. This can be particulates and gases [7]. HNO_3^- vapour is deposited on any surface with a very high deposition velocity [19, 22]. In comparison, NO_2 deposition velocity is very low, whereas gaseous NH_3 does not occur away from point sources of emission. N occurs in particles as $NO_3^-NH_4^+$, $(NH_4^+)_2$ SO_4^{2-} or in combination to other ions of opposite charge. A major proportion of NH_4^+ and most of NO_3^- occurs in large particles [20]. (c) Occult deposition, this is the deposition resulting from cloudwater and fog. High concentrations of both NO_3^- and NH_4^+ have been measured in fog and clouds at high elevation in the Vosges [6] as in the eastern US [19]. Canopy uptake of N compounds has been shown to occur by the foliage itself, or as retention by micro-organisms or lichens living at the leaf surface. Although analysis of particulates, cloudwater and gases is the most direct way of determining dry deposition, the variability in gas and particle composition in relation to particle size and climatic situations is difficult to handle. Furthermore, the necessity of accurate deposition velocity models [8, 9] and the need of scaling factors to relate measurements to forest canopy surfaces complicate this approach. Mineral N compounds in throughfall mainly originate from deposition [19], but the possibility of NH_4^+ leaching from leaves has also been shown [28]. In remote or low polluted sites, several authors have found lower NH_4^+ and NO_3^- fluxes in throughfall than in rain, suggesting uptake of the N by the canopy. Higher N fluxes in throughfall than in bulk precipitation have often been reported at strongly polluted sites with high deposition, and this obscures a possible canopy uptake. ^{15}N enriched artificial mist has been often used experimentally to quantify direct uptake by leaves and twigs over short periods [3, 4, 17].

A few authors have collected throughfall under artificial and chemically inert canopies in order to mimic the process of deposition in the field, whithout the complicating factor of uptake. Joslin et al. [14] and Joslin and Wolfe [15] have shown that plastic trees could be used to estimate the cloud deposition of water and sulphur on real spruce trees. Bobbink et al. [2], by comparing throughfall composition below plastic and real *Calluna* plants, showed a significant ammonia absorption by the heather foliage.

Bulk precipitation and throughfall has been monitored in various spruce (*Picea abies* (L.) Karst) stands of the Strengbach catchment for 10 years, in the context of reseach on forest decline [6]. The flux of N, as of most mineral elements, has generally increased from rain to throughfall and this increase has been related to stand age. This relation with stand age has been hypothetically linked to an increase in dry deposition with the canopy development of the stand [5].

With the objective of estimating the total deposition and canopy uptake of N, we established a number of plastic Christmas trees in clearings of the catchment and compared throughfall composition below plastic and real trees. The assumptions made were that:

(a) Regarding dry deposition, plastic trees with a similar form could be a reasonable analogue to real trees. This implied that NH_3 and NO_2 deposition, which is controlled by stomatal resistance, would be negligible. Therefore, we assumed that the composition of net throughfall (NTF = throughfall minus bulk precipitation) below plastic trees was close to that of dry + occult deposition;

(b) The ratios between net throughfall fluxes of ions poorly cycled by the tree crowns, as Na (or SO_4^{2-}), below living and artificial canopies could be used as comparative indexes of dry + occult deposition velocities of N.

2. MATERIALS AND METHODS

The study site and sampling method are described in [5]. Briefly, the study site is located on the upper southern oriented slope of the Strengbach catchment [24]. In clearings located on the upper slope of the mountain, within the studied stands, 5 isolated 8-year-old real spruce (R) 1.7–2.2 m in height were selected. 5 artificial (A) trees (1.6 meter-high plastic Christmas trees) were established for 8.5 months in the immediate vicinity of the real trees. Under each tree, 8 polyethylene funnels (diameter 20 cm) were set up, distributed in two concentric rings of 4 funnels each. 4 funnels were also set up to collect bulk deposition (O).

In addition, throughfall was collected for several years in 3 adjacent closed spruce (*Picea abies* Karst) stands aged 15 (S15), 35 (S35) and 90 (S90) years. S90 and S35 are located along the crest, whereas S15 is 50 m lower in elevation. Throughfall was collected with 4 (in S15) or 6 (in S35 and S90) replicates of 2-meter-long polyethylene gutter (individual collection area: 0.2 m^2). No biocide was added to the throughfall collection vessel as NH_4^+ and NO_3^- concentrations have been shown to vary little within a two week time period (10, 23). The main stand characters are indicated in *table I*. Foliar N

Table I. General characters of the spruce stands, foliage biomass and concentration in N and S, and annual return of N in litterfall (data from Le Goaster et al. 1991; Dambrine unpublished).

Stand	age year	Mean height m	density $tree.ha^{-1}$	Needle biomass $tons.ha^{-1}$	Needle N %	Needle S %	Annual litterfall $kg\ N.ha^{-1}$
S15	15	4	3550	24	1.02	0.14	13.7
S35	35	15	2200	27	1.22	0.13	37.7
S90	90	28	500	9	1.4	0.11	29.3

concentrations of the 15, 35, and 90 years-old stands increased from the younger to the elder stand, but the reverse trend was observed for sulphur. No nitrate in soil solution nor nitrification activity were found in young stands, whereas both were observed in the soil of the old stand [16].

Bulk precipitation and throughfall samples were collected twice a month from January 15 to September 30. Samples were filtered in the lab the day after the sampling. Na^+ and S were analysed by ICP (Jobin Yvon 38+), NH_4^+ and NO_3^- by automated colorimetry.

Net throughfall was calculated as the difference between throughfall and bulk precipitation for each type of tree and stand. Differences between chemical fluxes in bulk precipitation (4 replicates) and throughfall below artificial and real trees (5 replicates) have been tested by paired comparisons Student's t test and are statistically significant at the 5% level.

3. RESULTS

3.1 Concentrations

Na and SO_4^{2-}: In all collectors, highest concentrations occured during the period from February to May and in September (*figure 1*). Lowest concentrations were measured in bulk precipitation. In the clearings, concentrations measured in throughfall were higher below real trees than below artificial trees. Below the closed stands, concentrations were higher in the elder stand (S90).

NH_4^+: In all collectors, concentrations were higher during March-April and in September. From June to August, concentrations below real trees and in bulk precipitation were similar and less than the half of that below plastic trees. In the closed stands, concentrations were much higher under S90 than under the younger stands. In the younger stands, concentrations were similar and slightly lower than in bulk precipitation.

NO_3^-: The seasonal variations of NH_4^+ and NO_3^- were similar. NO_3^- concentrations were much higher in throughfall under plastic trees than in bulk precipitation. In the clearings, concentrations under plastic and real trees varied in parallel, but concentrations were higher in spring and autumn and lower in summer below the real trees. In the closed stands, NO_3^- concentration increased with stand age. Below the younger stand (S15), NO_3^- concentration was close to or lower than in bulk precipitation.

3.2. Water fluxes

The amounts of bulk precipitation collected during the study period was 617 mm (*table II*). The relations between bulk precipitation and throughfall amounts were close to those measured over many years in the open and the spruce stands [24, 27] except at S35 for which interception was higher than the mean annual value. Interception under the plastic trees was relatively high especially in comparison to real trees, the foliage of which was much denser. No simple explanation was found for this observation. A few measurements made during the autumn in the clearings indicated that a significant part of the plastic tree throughfall could be lost in stemflow. This may have been due to the fact that plastic needles did not extend along the branches but occured only on the final twigs, so that throughfall droplets could easily be transferred from the branches to the trunk. In the following flux calculations, two values have therefore been presented for the plastic trees: the measured flux and a 10% increased value (*A** in *table II*) calculated to take into account an assumed flux lost in stemflow.

3.3. Mineral fluxes

NO_3^-, SO_4^{2-} and Na fluxes below artificial trees were higher than in bulk precipitation, (*table II*) but lower than below isolated real trees. In contrast, the flux of NH_4^+ in bulk precipitation was close to that in throughfall below real canopies and much lower than that below artificial trees.

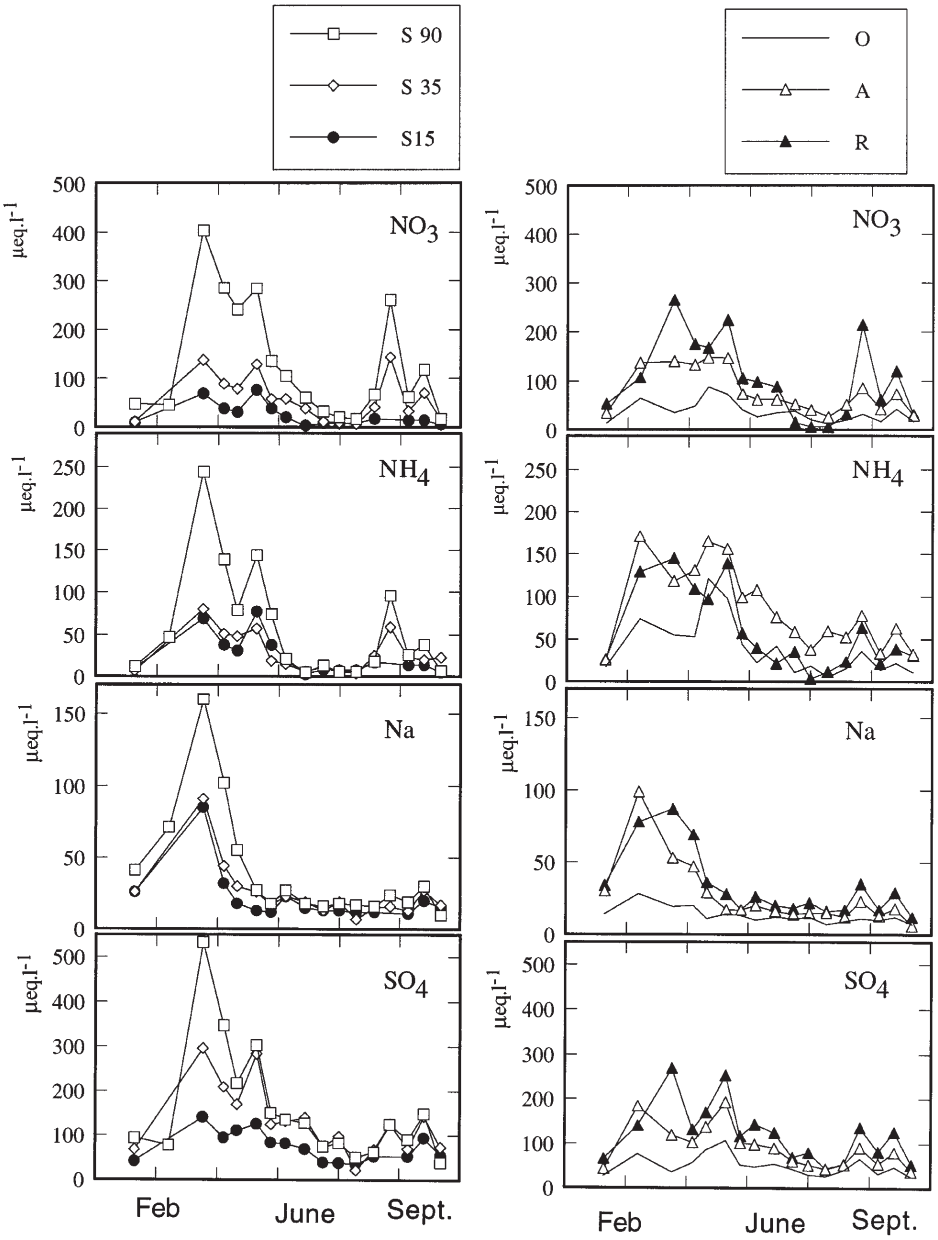

Figure 1. Variation of NO_3^-, NH_4^+, Na^+ and SO_4^{2-} concentrations (µeq.L^{-1}) in bulk precipitation (O) and throughfall under isolated artificial (A) and real (R) trees in clearings, and under three closed stands aged 15 (S15), 35 (S35) and 90 (S90) years over 8.5 months.

In the closed stands, the throughfall fluxes of Na, NO_3^- and NH_4^+ were similar in the S35 and S15 stands, but higher in the S90 stand. The throughfall flux of SO_4^{2-} in the S35 stand was intermediate between that of the S15 and S90 stands. The fluxes of NO_3^-, SO_4^{2-} and Na below real trees in the clearings were similar or lower (SO_4^{2-}) than in the older stand, whereas the flux of NH_4^+ was higher in the clearings.

4. DISCUSSION

Sodium and sulphur$^-$ are poorly recycled by spruce foliage [13]. In fact, input-output budgets worked out for the S90 and S35 stands showed that the annual throughfall inputs of these elements approximately balance the outputs in soil solution below the roots [24]. Airborne Na is present in particles and cloudwater. The difference

Table II. Fluxes of water (mm) and mineral elements ($meq.m^{-2}$) in bulk precipitation, throughfall and net throughfall during the 8.5 months study period. There were little errors in the water fluxes, previously published in [6], which are corrected here. Tables with chemical fluxes had no errors.

	site	water mm	Na	NH_4 $meq.m^{-2}$	NO_3	SO_4
Throughfall	S90	411	13.7	17.3	39.8	54.3
Throughfall	S35	379	9.7	9.3	18.2	45.2
Throughfall	S15	456	10.4	10.3	18.4	31.3
Throughfall	*R*	397	13.8	22.8	40.9	50.6
Throughfall	*A*	431	10.3	32.1	28.8	34.4
Throughfall	A^* = 1.1A	474	11.3	35.3	31.6	37.9
Bulk Precipitation	O	617	7.8	20.9	20.4	29.6
Net Thoughfall	S90		5.8	–3.6	19.5	24.7
Net Thoughfall	S35		1.9	–11.5	–2.1	15.6
Net Thoughfall	S15		2.6	–10.6	–1.9	1.7
Net Thoughfall	*R*		6.0	1.9	20.5	21.0
Net Thoughfall	*A*		2.4	11.2	8.4	4.8
Net Thoughfall	A^* = 1.1A		3.5	14.4	11.3	8.3

Table III. Estimates of dry + occult deposition and canopy uptake of nitrogen during 8.5 months, using SO4 or Na in net throughfall as deposition indexes ($meq.m^{-2}$). Two values are given: the first derives from the measured value of NTF below plastic trees, the second was computed assuming a 10% increase in NTF.

	site	NH_4 (Na based)	NO_3 (Na based)	NH_4 (SO_4 based)	NO_3 (SO_4 based)
		$meq.m^{-2}$			
Dry + occult deposition	*R*	28/25	19/21	36/49	28/36
	S15	11/12	8/9	3/4	2/3
	S35	8/9	6/7	27/36	21/27
	S90	24/27	19/20	42/57	34/43
Canopy uptake	*R*	26/30	0/–1	34/47	8/16
	S15	21/23	10/11	14/15	4/5
	S35	19/20	8/9	38/47	23/29
	S90	27/31	1/–1	46/61	14/23

in NTF fluxes of Na between plastic and real trees should mainly reflect a higher deposition velocity of dry particles and cloud droplets to the real trees directly related to their greater size, as well as their higher leaf area and roughness. Atmospheric S is also deposited as gas (SO_2), or SO_4^{2-} in particles and cloudwater. As SO_2 deposition occurs via the stomata, plastic trees will underestimate dry deposition. The variation of net Na or SO_4^{2-} fluxes between sites can be mainly explained by differences in stand structure and position in the clearings. First, although small, the isolated real trees in the clearings were not protected by other trees and were thus exposed directly to the wind. This effect is similar to the phenomenon described as the forest edge effect [11]. The S90 stand location near the crest, its height, low tree density and high defoliation [1] also allowed a higher deposition velocity in the crown. Foliage of the S15 stand was relatively more protected from the wind because of its high density, moderate height and distance from the crest. The height of the S35 trees would theoretically imply a higher deposition velocity, but NTF flux of Na at S35 was lower than at S15 during the study period, whereas NTF flux of SO_4 was intermediate between that of the S15 and S90 stands. Monitoring of net throughfall for three years at these sites showed that the throughfall flux of Na was higher at the S35 than at the S15 stands. The "anomaly" of the studied period might be related to a temporary leaching of Na from the S15 tree canopies or to the retention of Na on the S35 stand foliage,

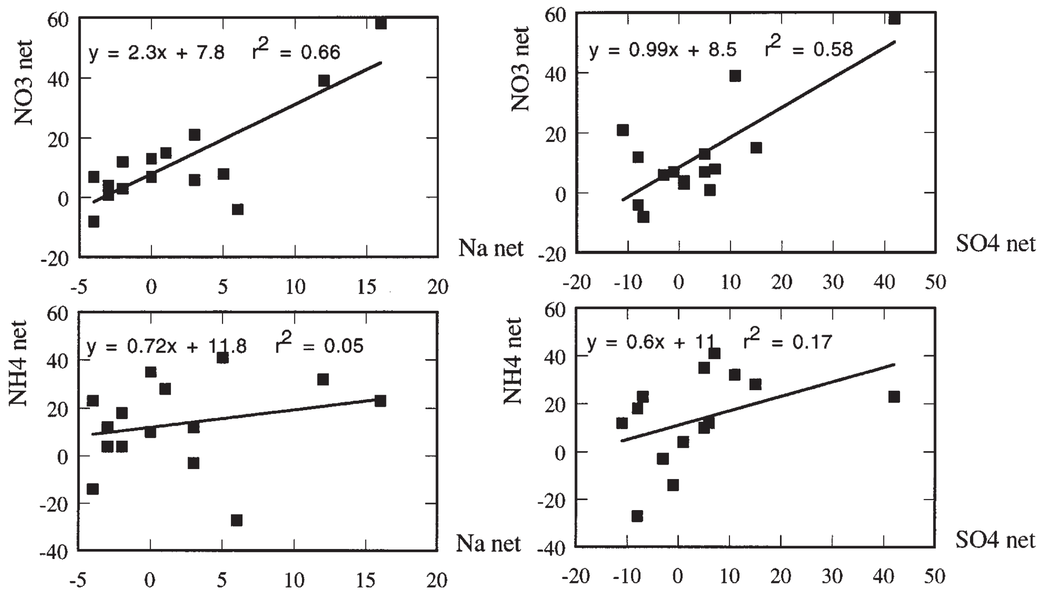

Figure 2. Correlations in net throughfall concentrations (µeq.l^{-1}) between (a) Na^+ and NO_3^-, (b) NO_3^- and SO_4^{2-}, NH_4^+ and Na^+, and (d) NH_4^+ and SO_4^{2-}. Negative concentrations are related to negative net throughfall fluxes. The data point (Na^+: 66 µeq.L^{-1}; SO_4^{2-}: 98 µeq.L^{-1}; NO_3^- 66 µeq.L^{-1}; NH_4^+: 88 µeq.L^{-1} has not been taken into account.

associated with the high interception value measured during the study period at that site.

A relative deposition velocity index was calculated from the ratio between the flux of Na in NTF below living and plastic canopies. Net throughfall of Na was assumed to reflect inputs from dry particles and cloud-water deposition. We assumed that N deposition (as NO_2 or HNO_3) on plastic trees would be very likely small compared to that on living trees. As sulphur is partly deposited as gas, NTF of SO_4^{2-} was also used to calculate an upper estimate for dry + occult deposition of N. To check if NO_3^- or NH_4^+ were better related to either Na or SO_4^{2-} in particulate + occult deposition, we computed the concentrations of particulate + occult deposition by dividing NTF flux of these ions below plastic trees by the flux of water in precipitation. With the exeption of one data point (highest concentration for all ions in March) correlations between NO_3^- and SO_4^{2-} or especially Na were significant (*figure 2*). Correlations with NH_4^+ were absent (Na) or weak (SO_4^{2-}).

To calculate dry occult deposition of N, we assumed that: (a) NTF composition below plastic trees was representative for the major part of dry + occult deposition, and (b) Na^+ and SO_4^{2-} - based relative deposition indexes could be used to compute the dry + occult deposition of NH_4^+ and NO_3^-. The difference between the computed dry + occult deposition of NH_4^+ and NO_3^- and the flux measured in net throughfall was considered to reflect the foliar uptake.

Using Na^+ as a dry + occult deposition index, We computed a dry + occult NO_3^- deposition of about 20 meq.m^{-2} on young trees in the clearing and in the old stand but a much lower deposition (<10 meq.m^{-2}) in the younger (S15 and S35) stands. No NO_3^- was taken up by the crowns at the sites where deposition was high (R and S90), whereas most of the NO_3^- dry deposited was taken up at the sites were deposition was low (S15 and S35). Using SO_4^{2-} as dry + occult deposition index for NO_3^-, the deposition computed for young trees in the clearing and the older stand was higher (respectively about 32 and 38 meq.m^{-2}), and the deposition onto the S35 stand increased to 24 meq.m^{-2}. This increased deposition implied an increased foliar uptake at all sites except at S15. Several arguments suggest that, except for S35 because of the anomaly of Na^+ in NTF during the study period, Na^+ provided a better dry + occult deposition index for NO_3^- than SO_4^{2-}. The reasons are, a) Na^+ is better related to NO_3^- in net throughfall. b) it has been shown, using ^{15}N labelled rain or mist [3, 4], that spruce canopy uptake of NH_4^+ was about 5 times higher than of NO_3^-, which matched our calculations. and c), it seemed unlikely that the older stand, with high foliar N content and low N requirement for growth could take up and metabolise large amounts of NO_3^- in comparison to NH_4^+.

The computed deposition of NH_4^+, using SO_4^{2-} as the dry + occult deposition factor was higher than with Na. From the younger to the older stand, the NH_4^+ deposition increased with age, between 8 to 26 $meq.m^{-2}$ (Na^+-based) or 3 to 48 $meq.m^{-2}$ (SO_4^{2-}based). Most of the NH_4^+ dry + occult deposited at all sites was absorbed by the tree canopies, and a substantial part of wet deposited NH_4^+ was also taken up by the younger stands.

Increasing NTF fluxes of N with stand age have already been reported [12, 21, 26]. From the data presented above, we believe that the increase of N in NTF with stand age reflects two processes, an increase in dry deposition and possibly a decrease of canopy uptake.

Over the 8.5 months period of the study, more than 4 $kg.ha^{-1}$ of inorganic N (Na index) were taken up by the canopies of the different stands. This uptake could make between a 10–30% contribution to the annual requirement of N by foliage (13 and 38 $kg.ha^{-1}.yr^{-1}$) (*table I*). This contribution to the annual requirement for foliage has been computed in other experimental studies at high elevation [4].

5. CONCLUSION

Measurements of throughfall under artificial trees, which mimic real trees, may appear as an easy and economic mean of measuring dry deposition on forest stands (5). However, this method suffers from several inconveniences. First, it is assumed that pollutant deposition on the leaf surface is similar on real and plastic trees. Therefore, the method would not be suitable for areas close to pollution sources, where gas deposition must be taken into account. Second, a scaling factor is needed in order to compare foliages of different surfaces, aerodynamic resistance and roughness. The use of both Na^+ and SO_4^{2-} is practical and provides a range of values in which the true deposition value is likely to be. Chemical monitoring of gas and particles could allow the identification and check of better tracers.

More than 4 $kg.ha^{-1}$ of dry + occult deposited N was taken up by spruce canopies during the 8.5 months study period. The proportion of deposited N which is taken up by the canopies is higher in young, fast growing stands, which have a high N requirement, compared to that of old and poorly growing stands. NH_4^+ was preferentially taken up, but NO_3^- uptake also occurred, at least in the young stands. N uptake was still substantial in the old N-saturated stand. Related to the total annual uptake of N allocated to the foliage, our estimates suggest that canopy uptake may supply 10 to 30% of this flux. This relatively high proportion is likely to alter N allocation within trees, and the equilibrium between nutrients [25].

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