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Deposition of ammonia to a Norway spruce (*Picea abies* (L.) Karst.) stand at Ballyhooly, Co. Cork

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Abstract
Emission of ammonia from manure spreading influences the nitrogen input and acidity of forest ecosystems in agricultural areas. This is mainly due to the interaction between ammonia in ambient air and the forest canopy. The foliage canopy effect can be considered as the difference between precipitation and throughfall fluxes, i.e. net throughfall, and is the result of leaching, dry deposition, uptake and emission. Net throughfall calculated after a heavy shower preceded by a relatively dry period results in total wash-off, which is equivalent to total deposition when no canopy exchange occurs.

Precipitation and forest throughfall fluxes in an agricultural area (Ballyhooly, Co. Cork) have been monitored since 1989. This provides information on dry deposition of ammonia in relation to throughfall composition. Net throughfall fluxes for ammonia at Ballyhooly show a strong seasonal effect. The flux is clearly greater during the summer, which is probably the result of increased dry deposition of ammonia due to manure spreading. Negative net throughfall fluxes during spring indicate an uptake effect. Throughfall fluxes for certain ions can be used as an estimate of total deposition. Due to canopy uptake, however, total deposition of ammonia at Ballyhooly is higher than the throughfall flux.

Keywords: ammonia, dry deposition, canopy uptake, critical loads, forest ecosystems

Introduction
The Forest Ecosystem Research Group (FERG) operates a series of intensive forest monitoring plots in Ireland, with the aim of quantifying the interaction of atmospheric deposition on forest ecosystems (Farrell *et al.*, 1993). In agricultural regions, ammonia from manure spreading may be a significant source of nitrogen (Isermann, 1991). At Ballyhooly, Co. Cork, Ireland, throughfall (precipitation collected beneath the forest canopy) and precipitation fluxes (flux or load is the ionic concentration multiplied by the water volume) have been measured since 1989. This provides information on the behaviour of atmospheric nitrogen in forest ecosystems. The research site is located in a stand of

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Norway spruce (*Picea abies* (L.) Karst.) planted in 1939 on an orthic podzol soil derived from sandstone colluvium (Boyle *et al.*, 1996).

Gaseous and particulate matter in the atmosphere are primarily removed by rainfall (wet deposition) or directly deposited onto terrestrial surfaces (dry deposition). Dry deposition to forests is much greater than to other terrestrial ecosystems, due to the so-called scavenging effect (Farrell *et al.*, 1993). Elevated ammonia concentrations in the air can usually be attributed to local emission sources, as ammonia is removed during long-range transport by dry deposition and solution in water droplets (Asman and Van Jaarsveld, 1992). This implies that nearly all the ammonia detected at Ballyhooly originates from within Ireland. Estimates of ammonia deposition for Ballyhooly are presented in this paper.

Influence of ammonia on throughfall pH

Ammonia (NH_3) has a considerable influence on throughfall chemistry. It neutralises acidity in precipitation and throughfall, forming ammonium (NH_4^+):

$$NH_3 + H^+ \rightarrow NH_4^+ \quad (1)$$

The pH of throughfall may be higher than that of precipitation following dry deposition of ammonia, due to the removal of hydrogen ions according to Equation 1. In the relatively unpolluted environment of Ballyhooly (Farrell *et al.*, 1993), the concentration of acid species is often low and the ionic content is largely neutral seasalts. Therefore, the concentration of ammonia determines the pH to a large extent. In Figure 1, it can be seen that high pH events in throughfall are associated with high ammonia concentrations. This neutralisation is, however, short-lived, as the added ammonium reaching the soil increases soil acidification, either through plant uptake of ammonium or through the microbial transformation of ammonium to nitrate (nitrification) (Reuss and Johnson, 1986).

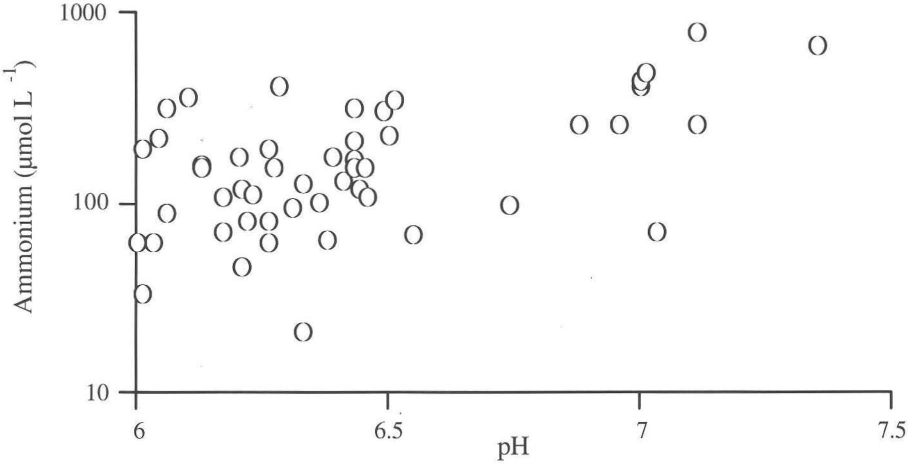

Figure 1. *Ammonium concentration against pH in throughfall at Ballyhooly, 1989-94. High pH events are associated with high concentrations of ammonia.*

Net throughfall of ammonium

Ammonium in throughfall is the result of four processes: (i) wet deposition of ammonia; (ii) dry deposition of ammonia; (iii) wet deposition of ammonium; and (iv) dry deposition of ammonium. The difference between throughfall and precipitation load - net throughfall - only reflects the input of dry deposition:

Net throughfall = Throughfall – Precipitation (2)

It is assumed that ammonia is the main component of dry deposition, due to the observed high atmospheric concentrations (Van den Beuken, 1993). This explains the very high pH and throughfall concentrations of ammonium observed after long dry periods (Figure 1). Mean annual net throughfall of ammonium (230 mol/ha/yr) for Ballyhooly appears to be rather small, lower than the precipitation (310 mol/ha/yr), which is the result of NH_x uptake by the canopy foliage (Boyle *et al.*, 1996). On occasion, however, the net throughfall ammonium flux is high while the mean annual net throughfall flux is low.

Assuming no canopy exchange (direct uptake by canopy vegetation), total deposition is equivalent to net throughfall when all dry deposition is washed off, which only occurs after a heavy shower. Six years of monitoring data, 1989-94, have been divided subjectively into periods of 3 to 9 weeks. Periods were selected so that each ended with a heavy shower, which was assumed to represent total washoff. Net throughfall was estimated according to Equation 2. A plot of net throughfall flux for these periods (Figure 2) shows a strong seasonal effect. The negative net throughfall fluxes in early spring indicate net canopy uptake. This is surprising, as the emission of ammonia is expected to be high due to high ammonia volatilisation rates from slurry spreading during the warm weather of late spring/early summer (April-June). The uptake of ammonium through foliage appears to regulate the net throughfall. In autumn (September-October), when temperatures are still high but tree growth is concentrated in timber and buds rather than in nitrogen-rich foliage, the net throughfall load is greater. This may be the result of dry deposition of ammonia due to manure spreading which is not masked by canopy uptake.

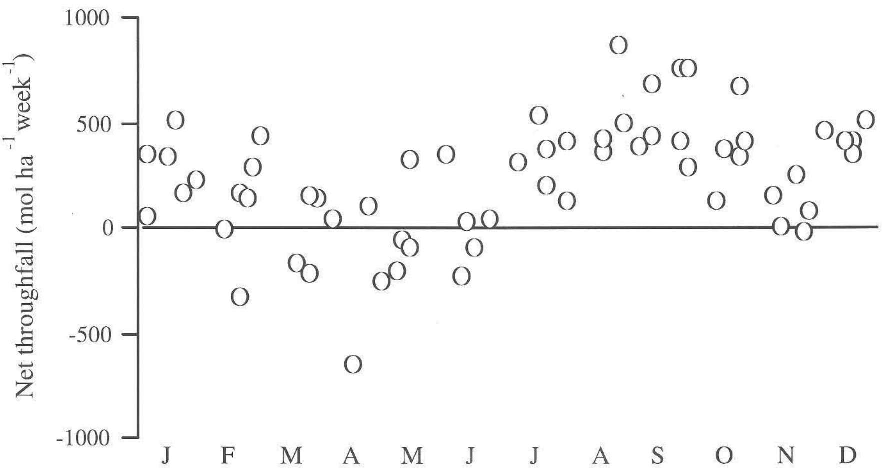

Figure 2. *Net throughfall fluxes for Ballyhooly. Each point on the graph represents mean weekly throughfall for periods of 3-9 weeks from 1989 through to 1994.*

Total deposition of NH_x

Throughfall flux is the result of processes which include both dry and wet deposition, but also absorption through foliage and canopy leaching. Canopy interactions must be taken into account when comparing throughfall and total deposition. Due to canopy uptake, throughfall is not a good estimate of total deposition (Figure 2). There are a number of methods which can be used to estimate total deposition of ammonium. Following the method employed by Draaijers and Erisman (1995), the canopy budget model has been applied to Ballyhooly to estimate total deposition of ammonia. The canopy budget model was developed by Ulrich (1983) and extended by Van der Maas and Pape (1991). The model discriminates between canopy exchange and atmospheric deposition using long-term throughfall and precipitation fluxes. Full details are given by Draaijers and Erisman (1995).

It is also possible to estimate dry deposition flux using inferential modelling which is based on air concentrations and deposition velocities:

$$F = V_d.c \quad (3)$$

where F is dry deposition flux, V_d is deposition velocity, and c is air concentration (Wesely, 1989). The relationship between air concentration and dry deposition flux, expressed in the deposition velocity, is however complicated by differences between chemical species and particle size, and is highly dependent on weather conditions. By assuming an appropriate deposition velocity, Van den Beuken (1993) observed that elevated ammonia concentration levels indicated significant dry deposition fluxes at Ballyhooly. Full details on the application of inferential modelling to Ballyhooly are given by Van den Beuken (1993). In Table 1, total deposition estimates for Ballyhooly, derived from the canopy budget model and inferential modelling, are presented along with measured throughfall for ammonia.

Table 1. *Estimates of total deposition of NH_x for Ballyhooly (total deposition = wet deposition + dry deposition).*

	Total deposition estimates mol/ha/yr
Throughfall	540
Canopy budget	978
Inference model	1,280

The estimated total deposition is quite tentative, due to uncertainties associated with the methods used. The canopy budget model is based on several assumptions which are not properly evaluated, and the total deposition derived from inferential modelling is highly dependent on the chosen deposition velocity. The inference model probably results in an over-estimation, as it is not based on a representative set of measured concentrations (Van den Beuken, 1993). Despite the uncertainties, both methods clearly show that throughfall cannot be considered as an estimation of total deposition due to the strong canopy interaction.

Critical loads of N

Based on the canopy budget model (Table 1), a conservative estimate for total deposition of NH_X is 1,000 mol/ha/yr, which is equivalent to 14 kg N/ha/yr. In recent years, pollutant loads of nutrient nitrogen have been described in terms of critical loads as "a quantitative estimate of an exposure to deposition of N as NH_X and/or NO_Y below which empirical detectable changes in ecosystem structure and function do not occur according to present knowledge" (UBA, 1996). Critical loads of nutrient nitrogen for acidic coniferous forests have been set at 10-50 kg N/ha/yr (UNECE, 1994). Taking only the deposition of NH_X into account (i.e. excluding NO_Y), it is clear that there is an exceedance of the critical load of nutrient nitrogen for Ballyhooly at the lower end of the range. The effects of an exceedance of critical load may not be obvious or seem harmful, as trees may grow faster due to the indirect fertilisation by ammonia deposition. Biodiversity may, however, also be affected.

Conclusion

Ammonia originating from agricultural activity has a great effect on the throughfall chemistry in Ballyhooly. Due to canopy interactions, throughfall chemistry is further influenced. Uptake through foliage controls the throughfall load to a large extent. Therefore, throughfall cannot be used as an estimate for total deposition, as it can greatly under-estimate the true value.

Dry deposition of ammonia from agricultural activities is the main input of anthropogenic pollutant species. The total deposition of NH_X is considerably higher than the throughfall load (approximately 45-55% greater). Based on the critical loads of nutrient nitrogen (10-50 kg N/ha/yr), the total deposition of NH_X suggests an exceedance at the lower end of the range for Ballyhooly.

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